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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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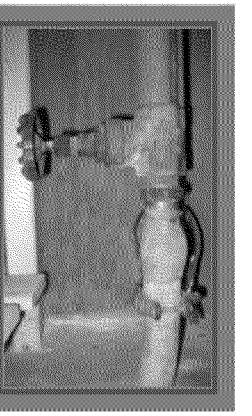
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (µg/L)	34	126	29	113
Fe (µg/L)	<5	<5	<5	34
Mn (µg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 µg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

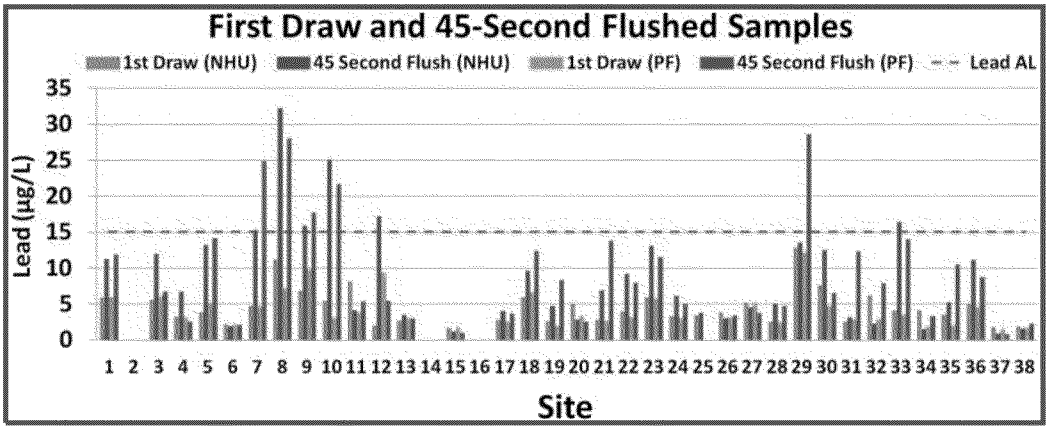

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

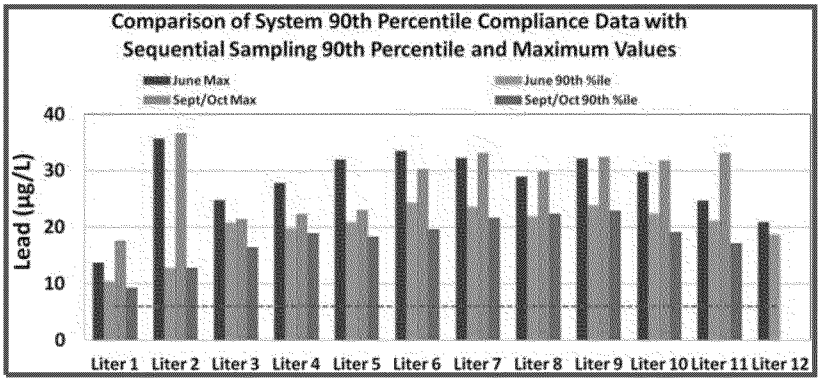

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

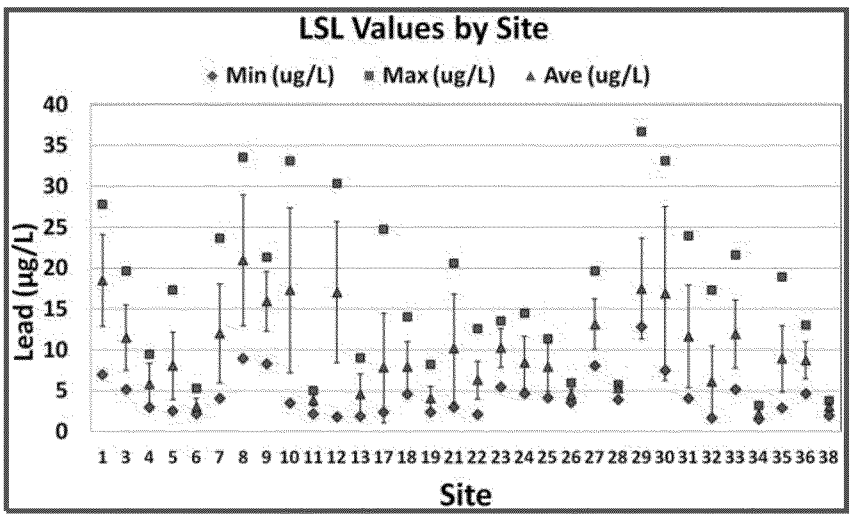

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

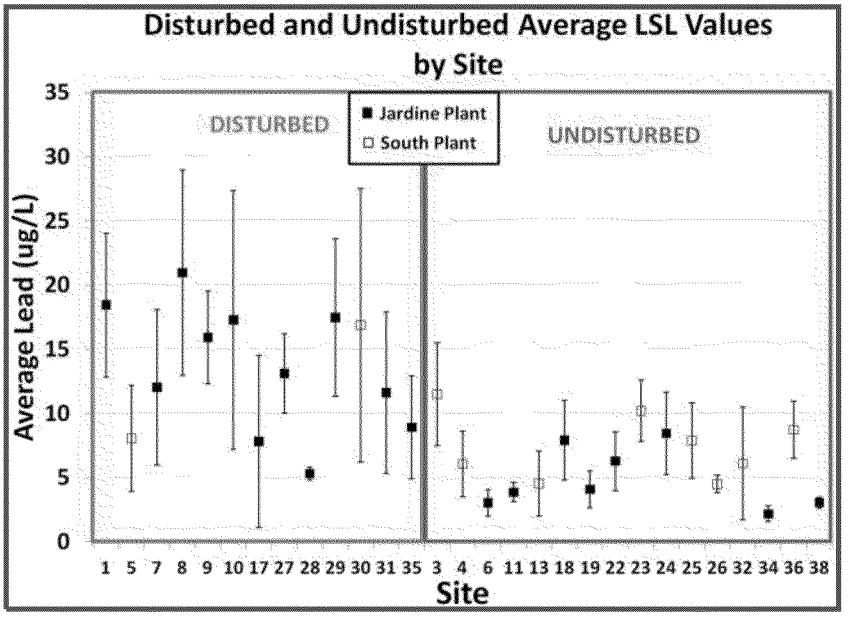

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

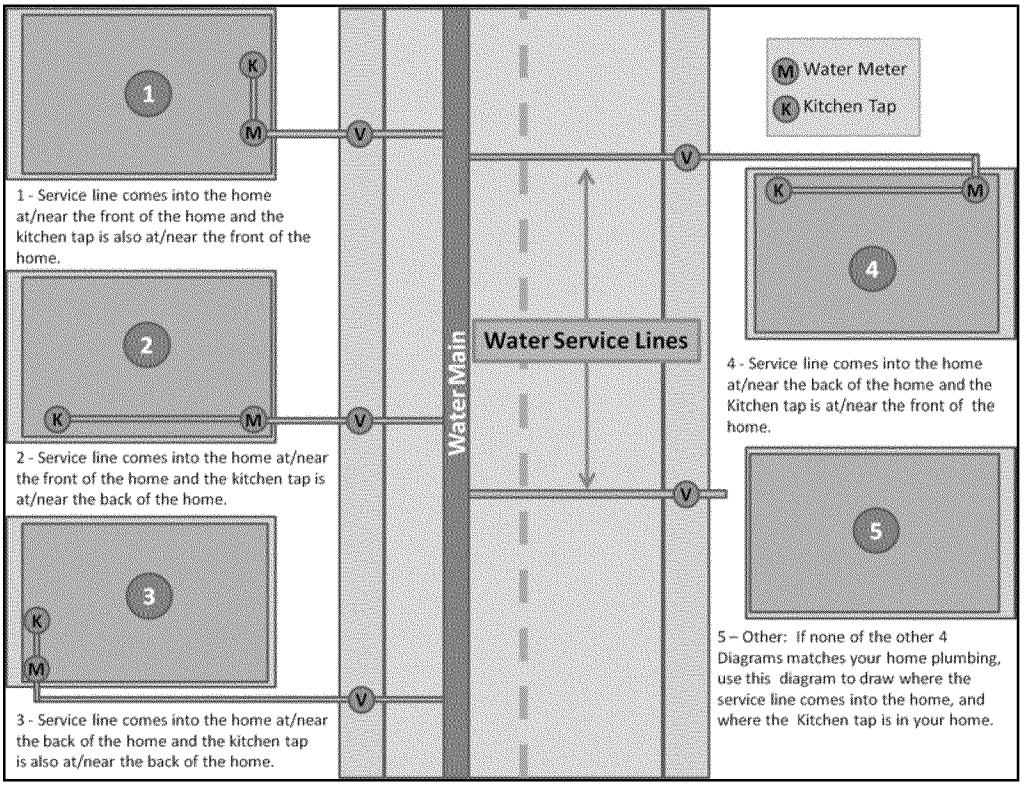


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

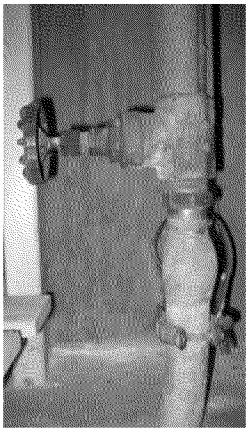


Figure S2: LSL Bulb

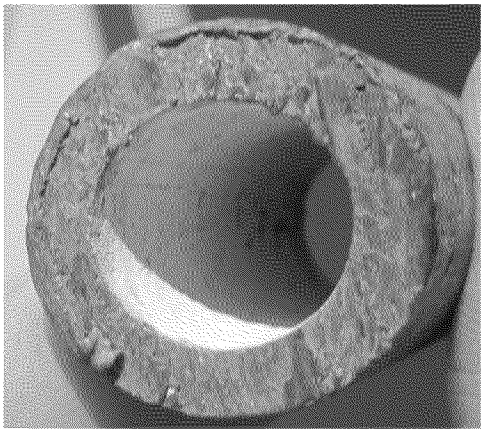


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

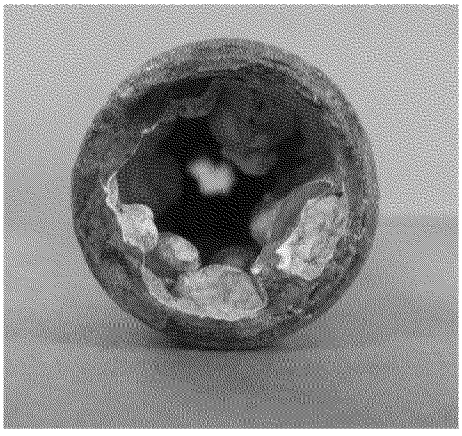


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

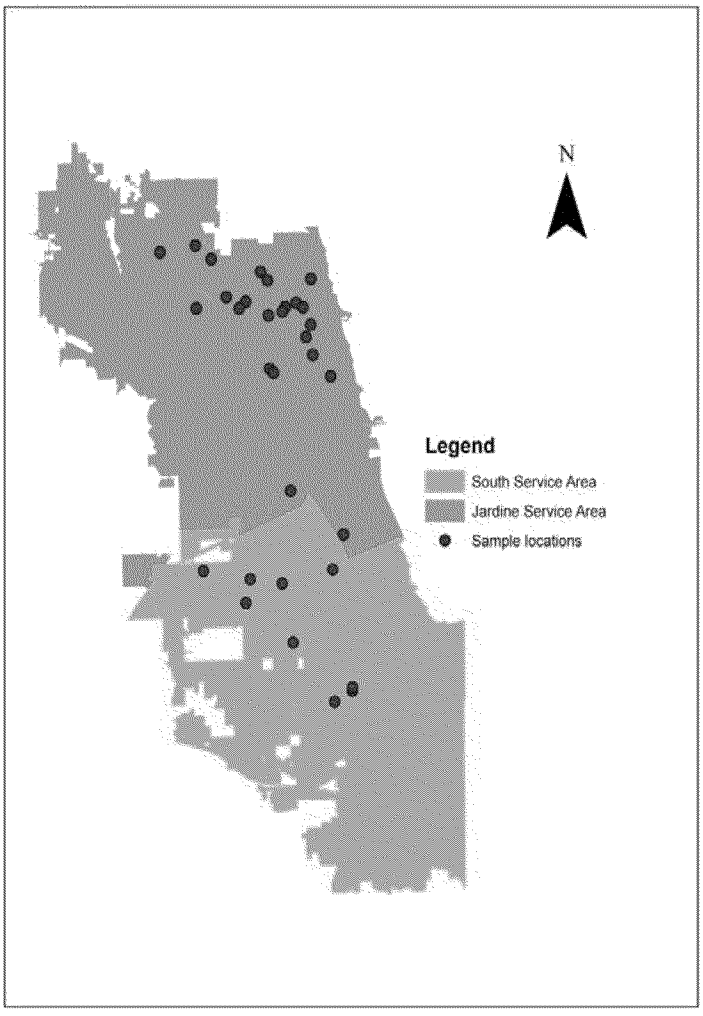


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

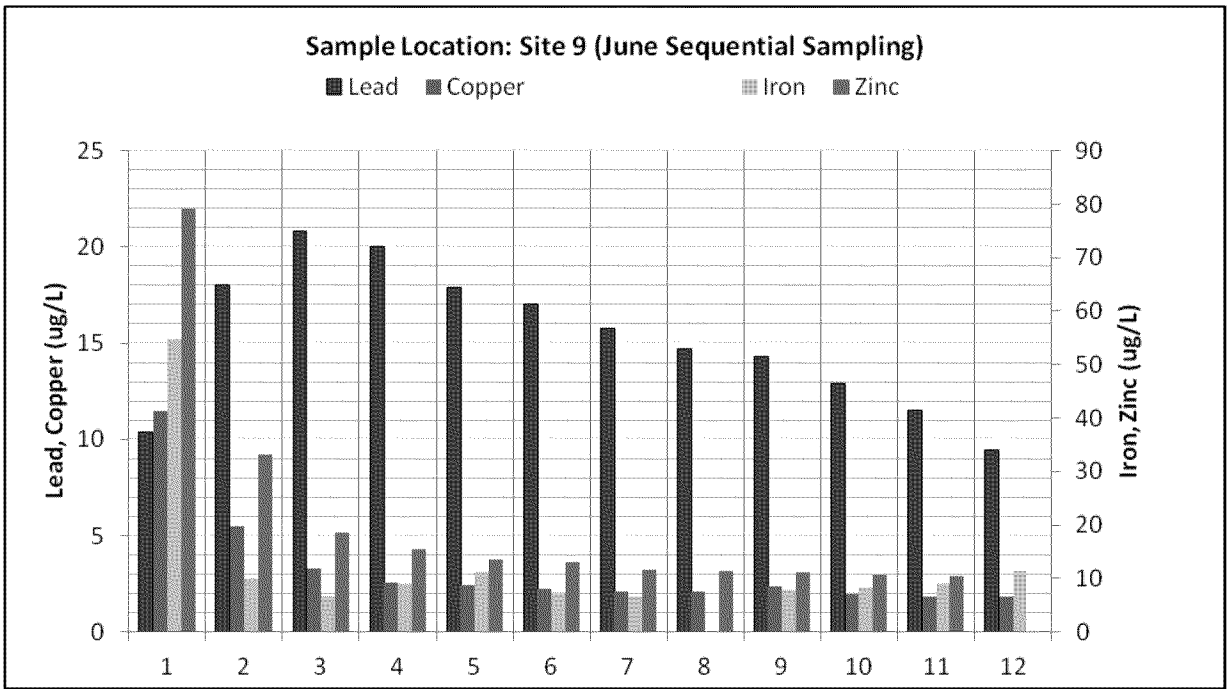


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

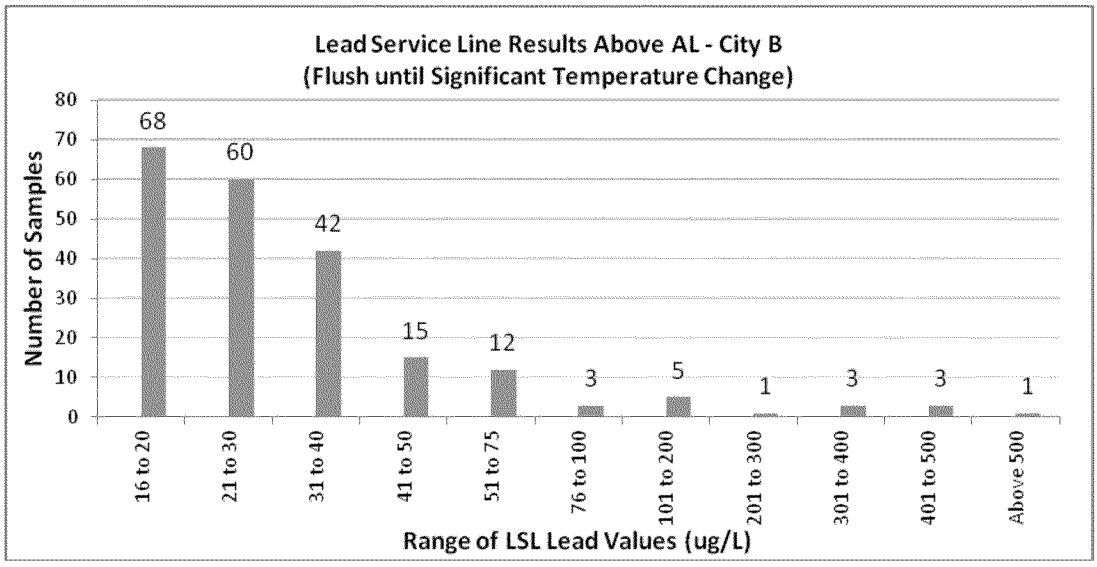
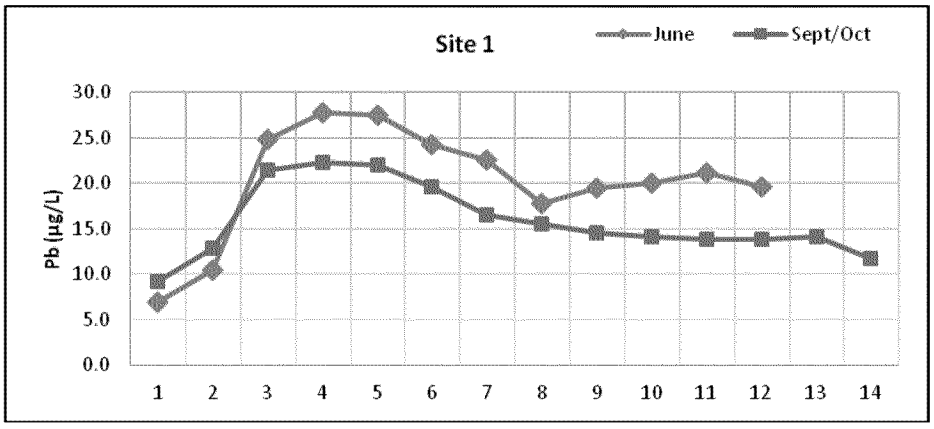


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

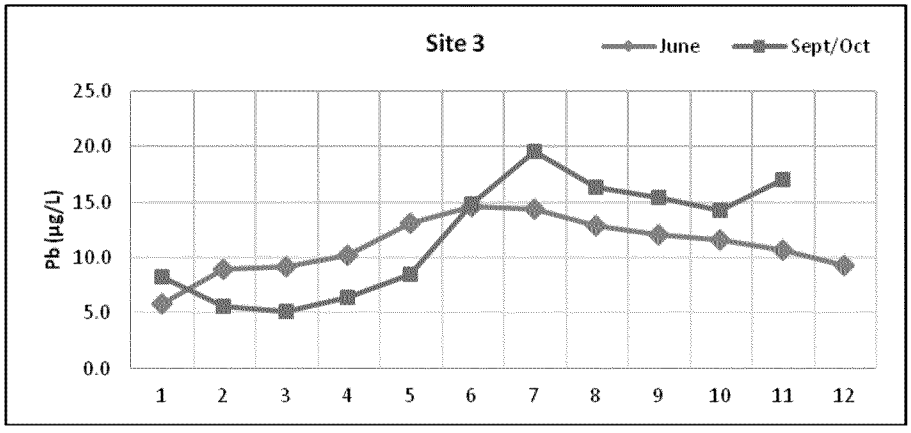
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

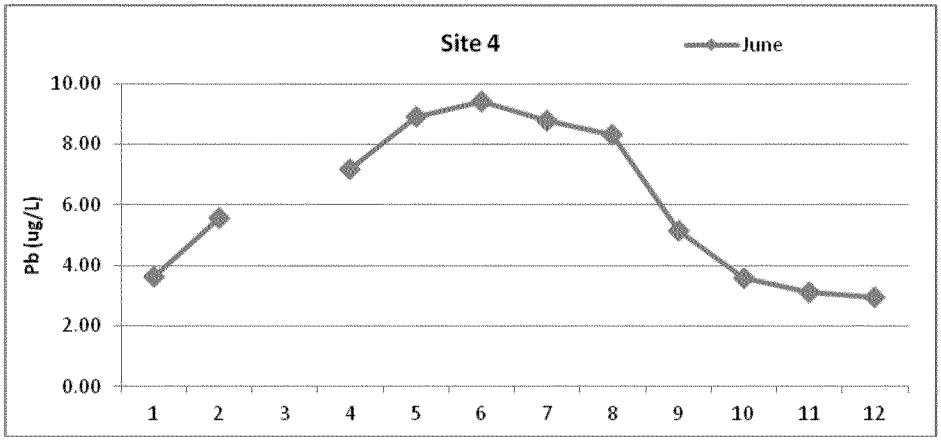
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

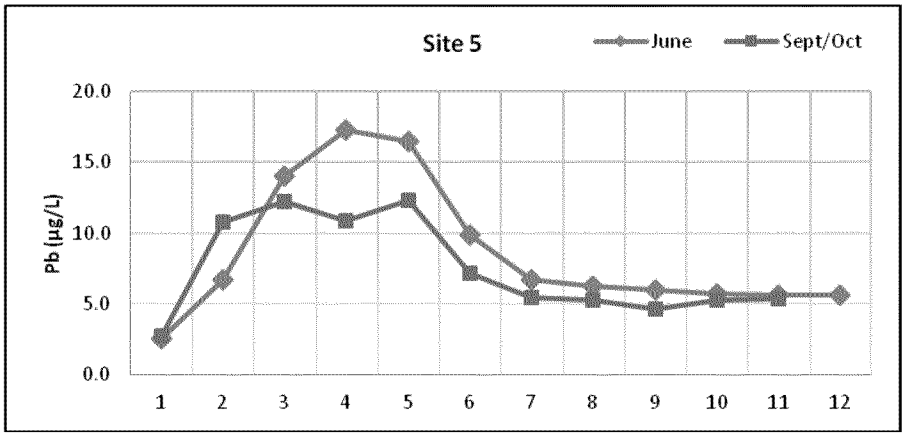
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

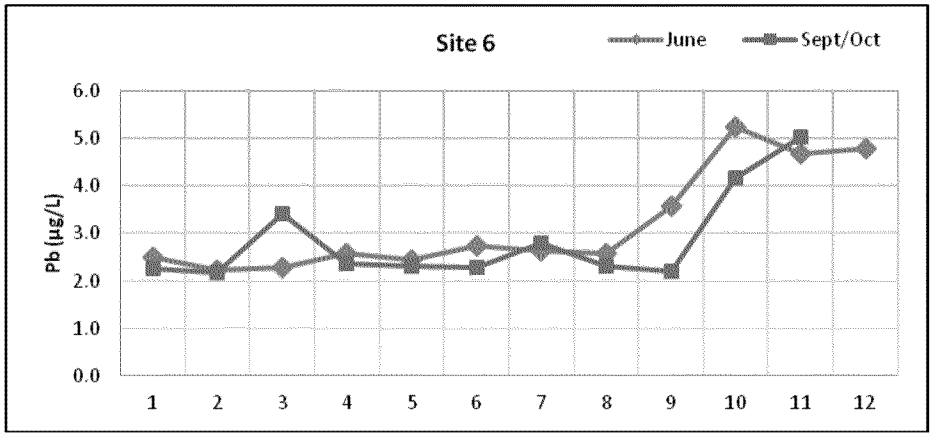
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

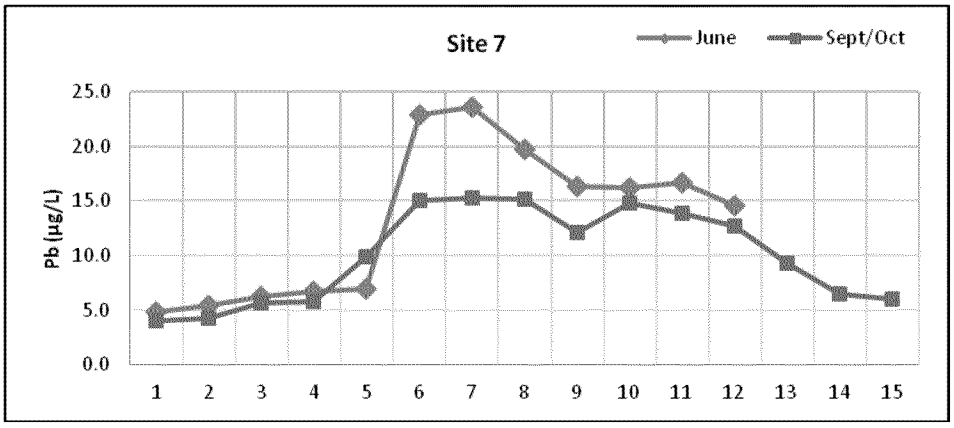
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

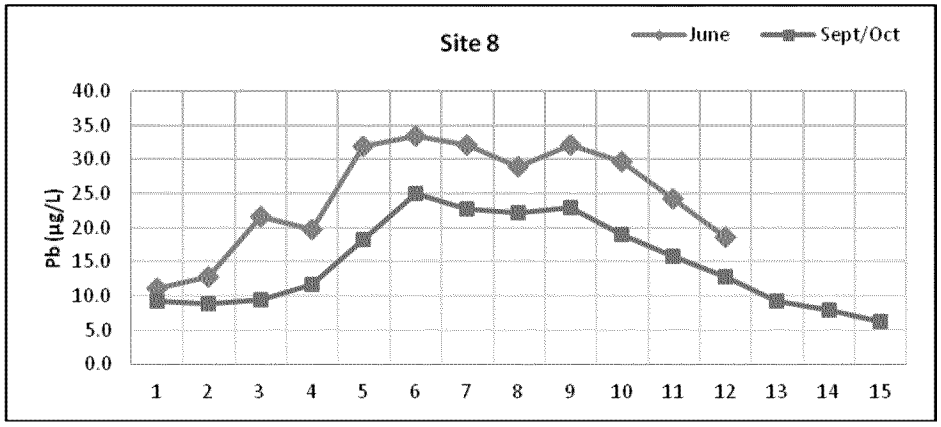
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

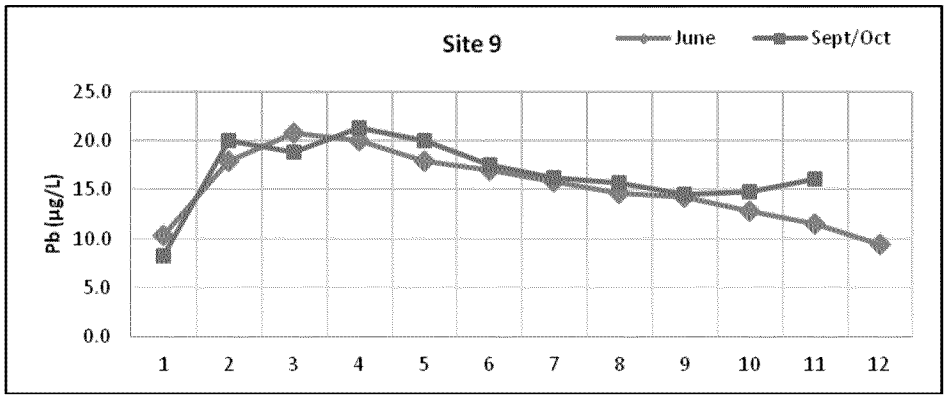
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

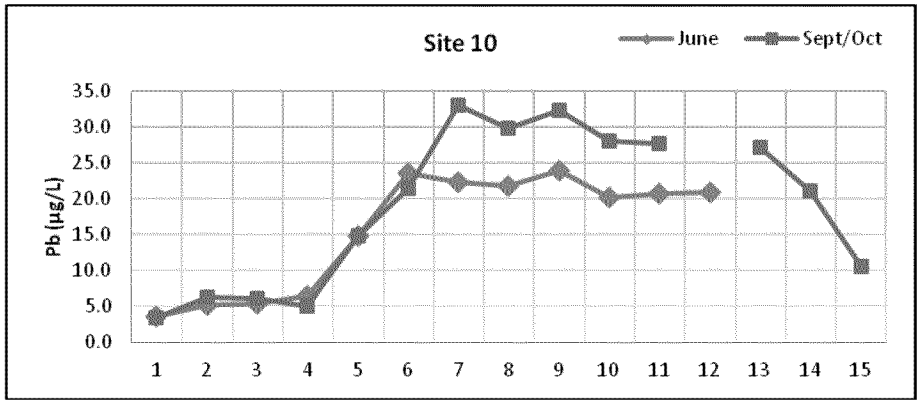
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

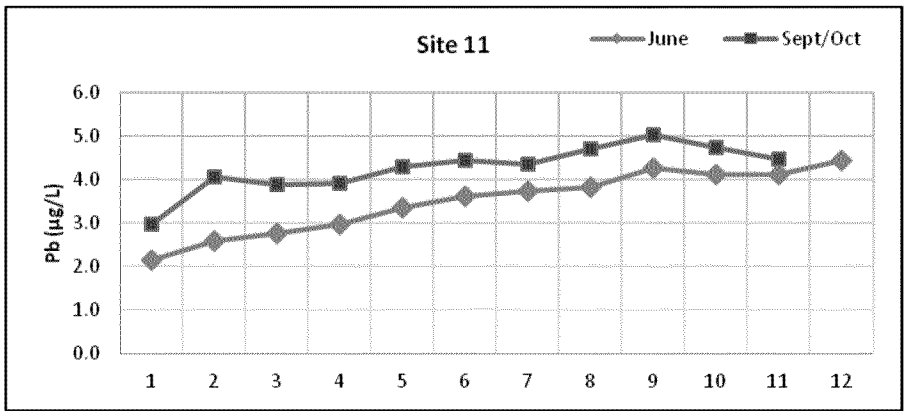
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

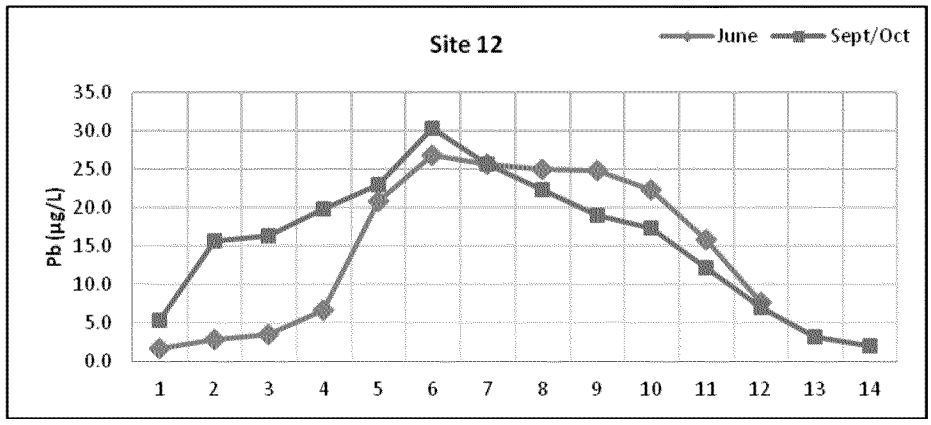
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

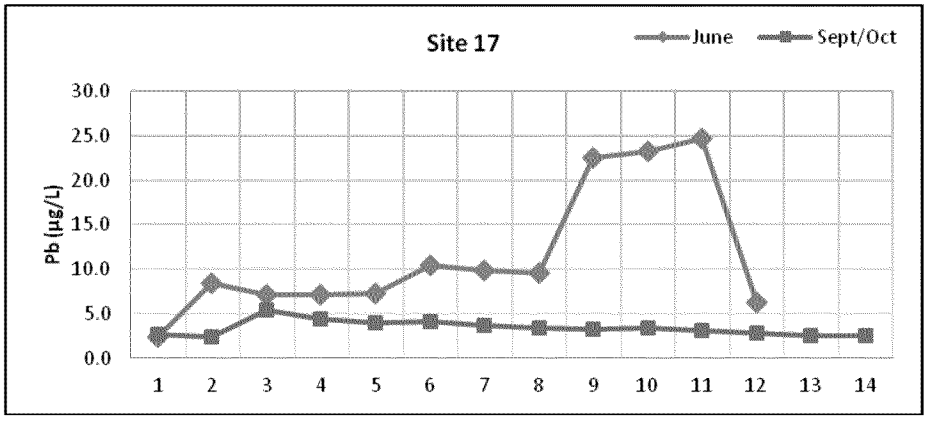
Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7

Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

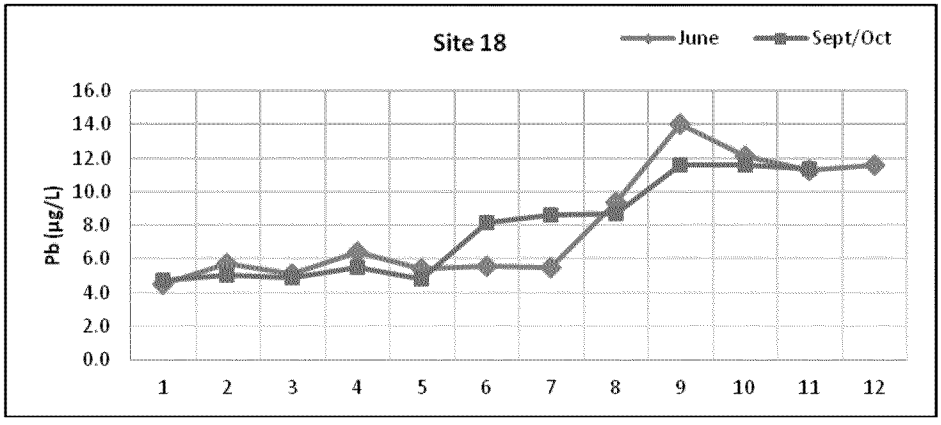
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

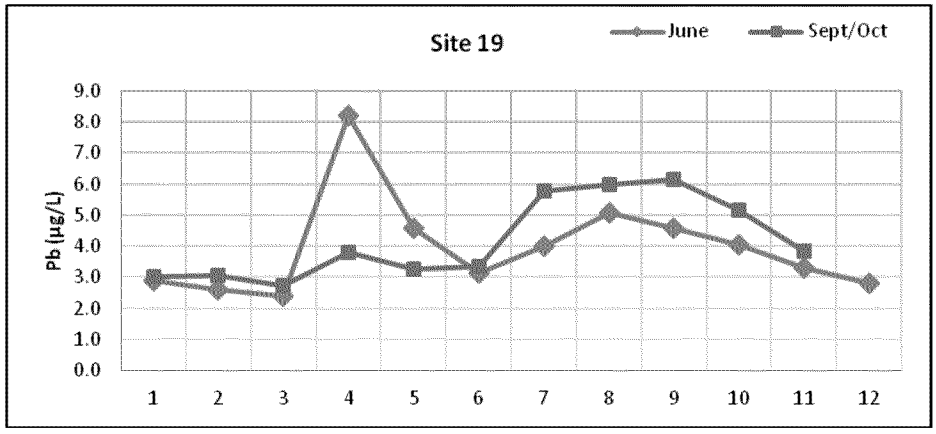
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

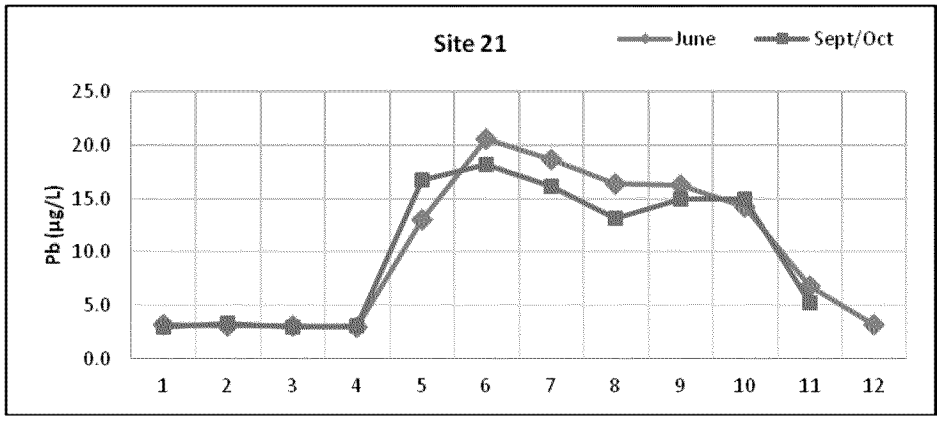
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

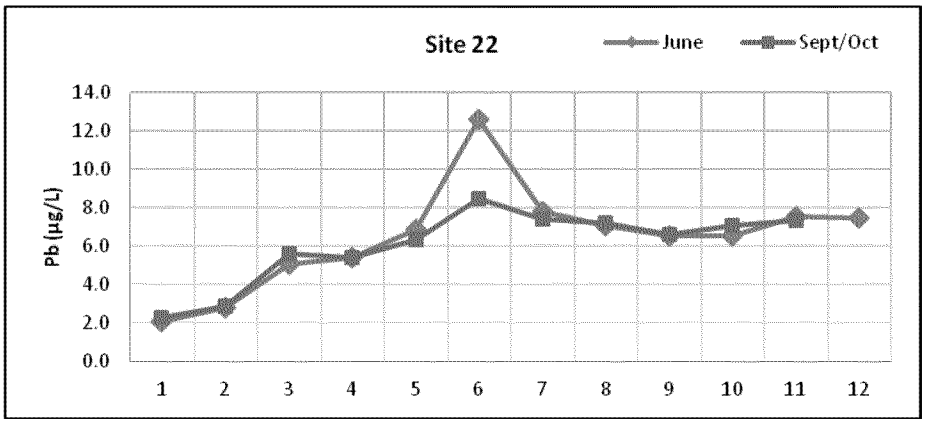
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

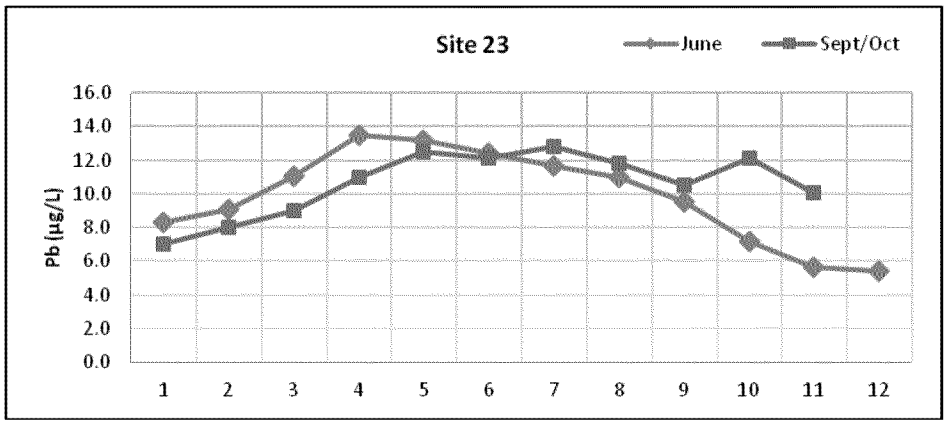
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

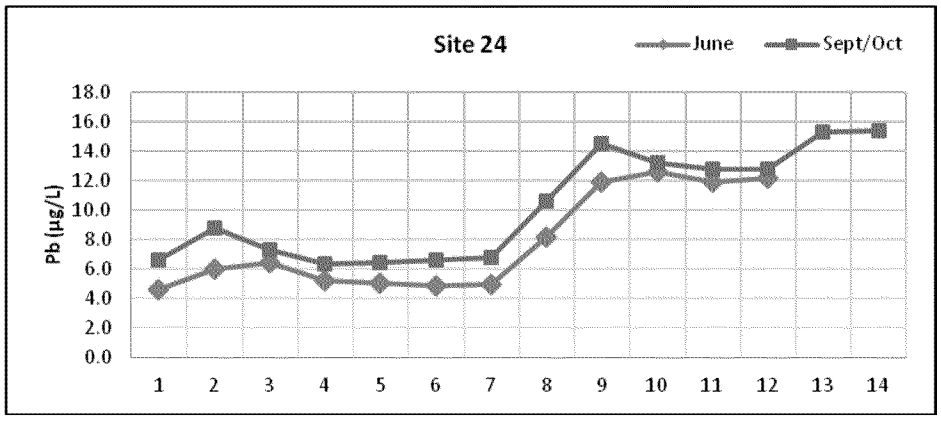
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

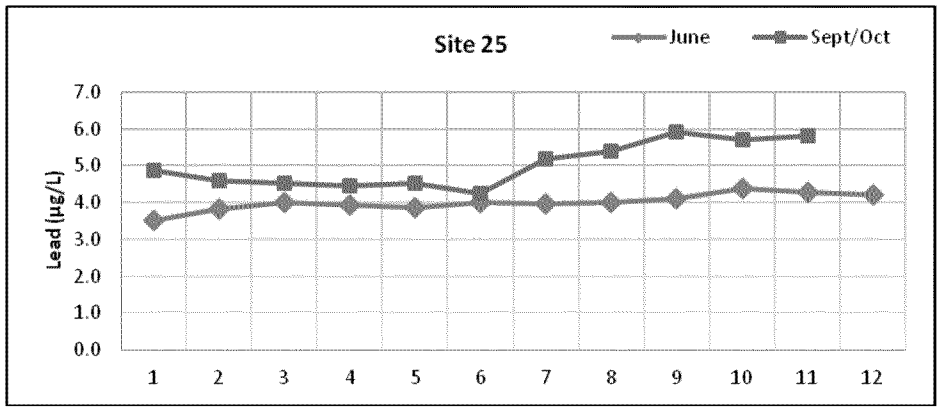
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

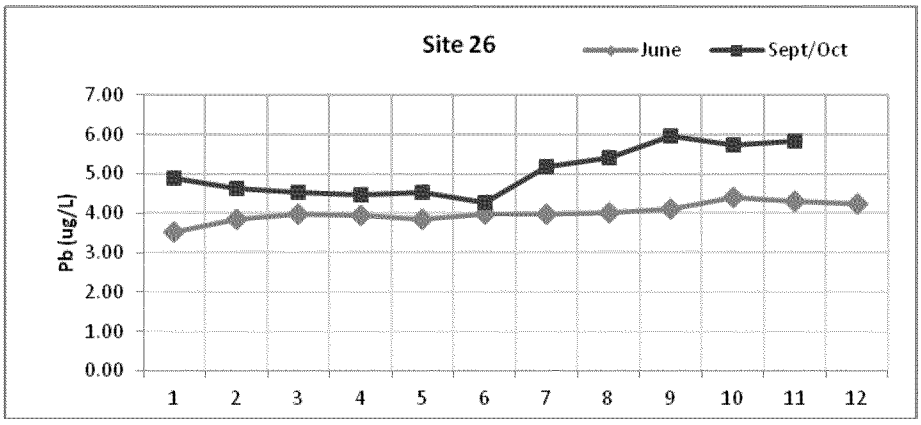
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

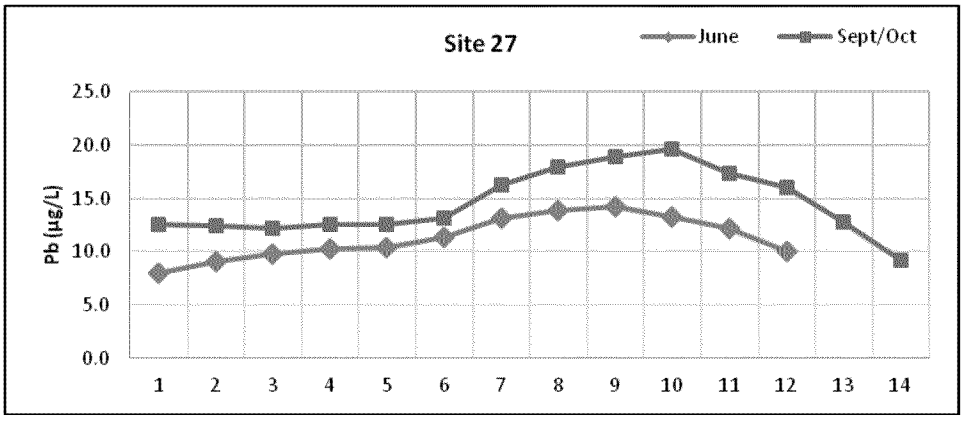
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

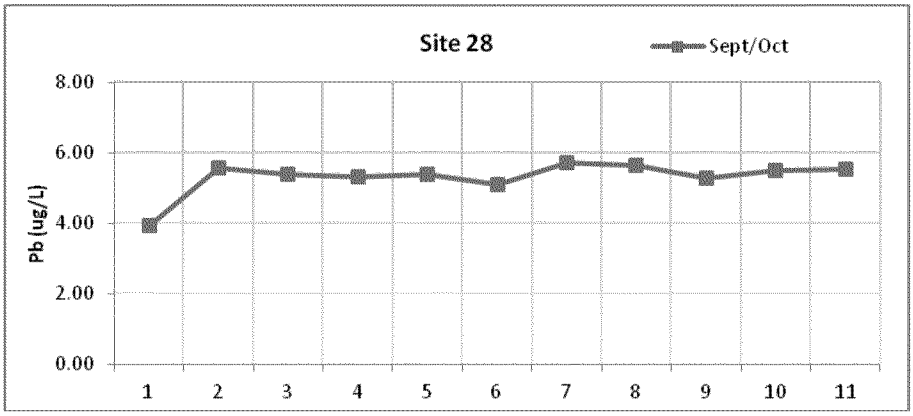
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

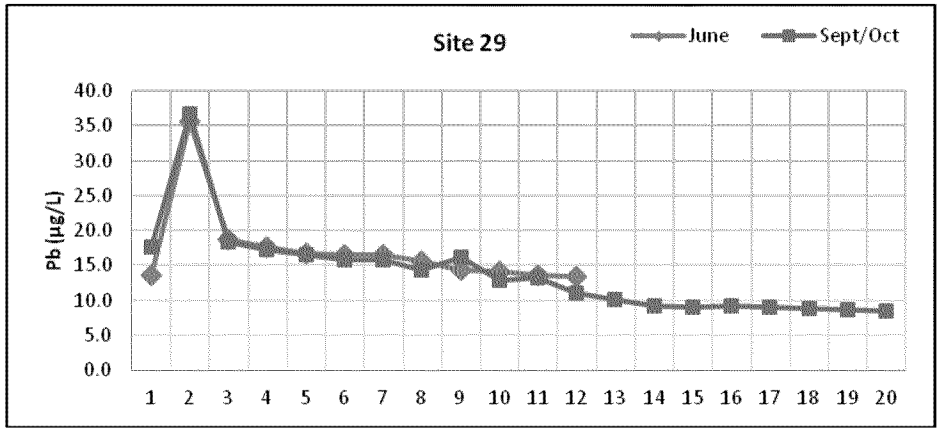
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

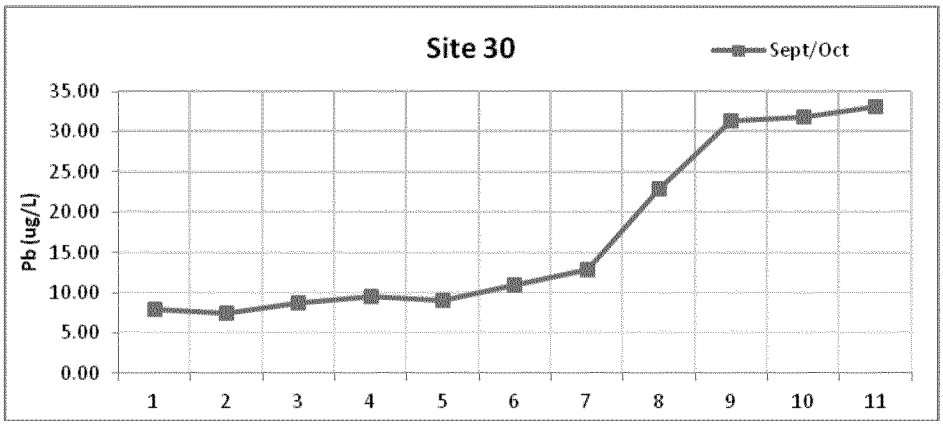
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

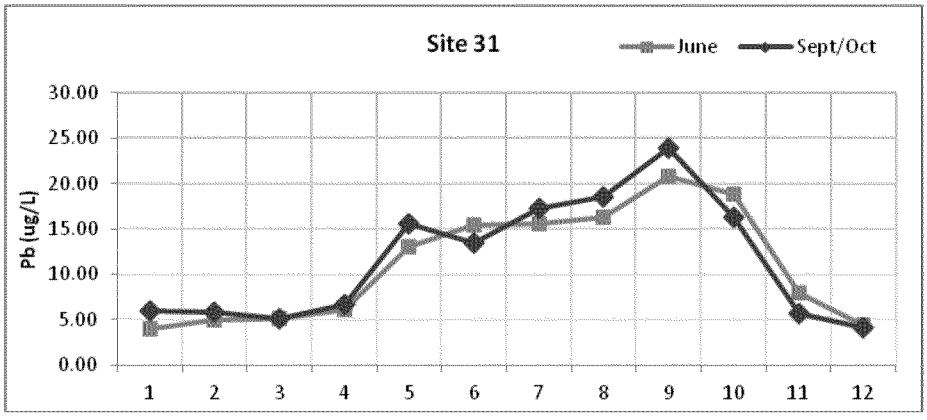
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

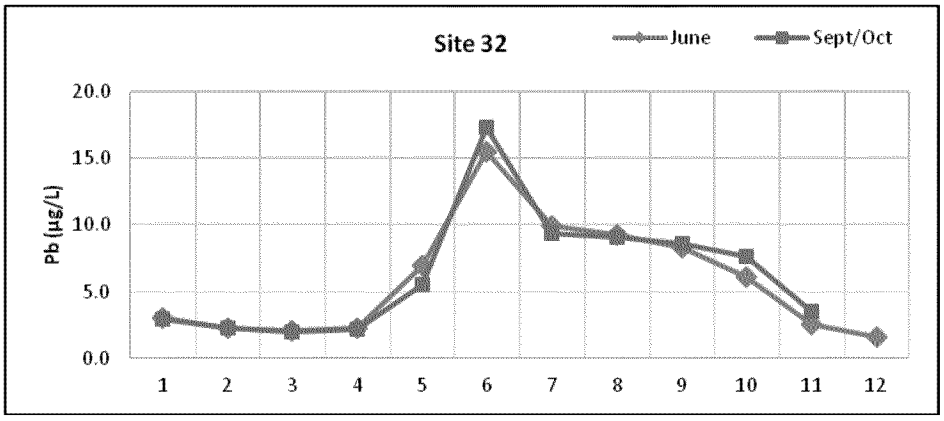
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

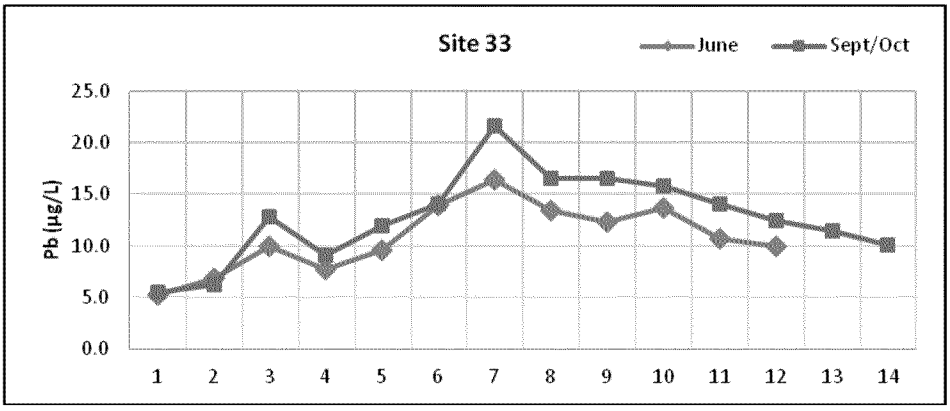
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

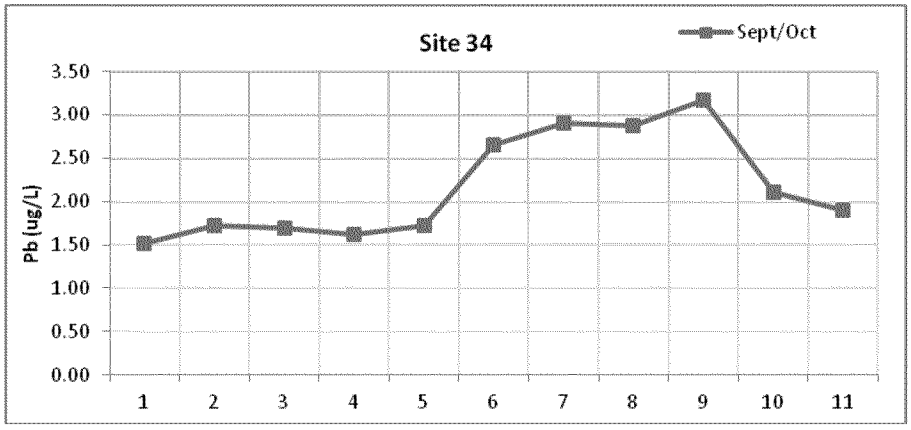
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

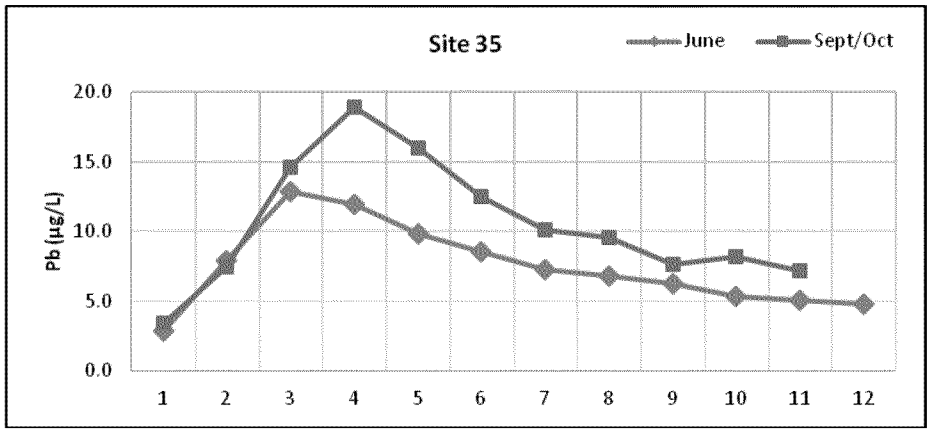
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

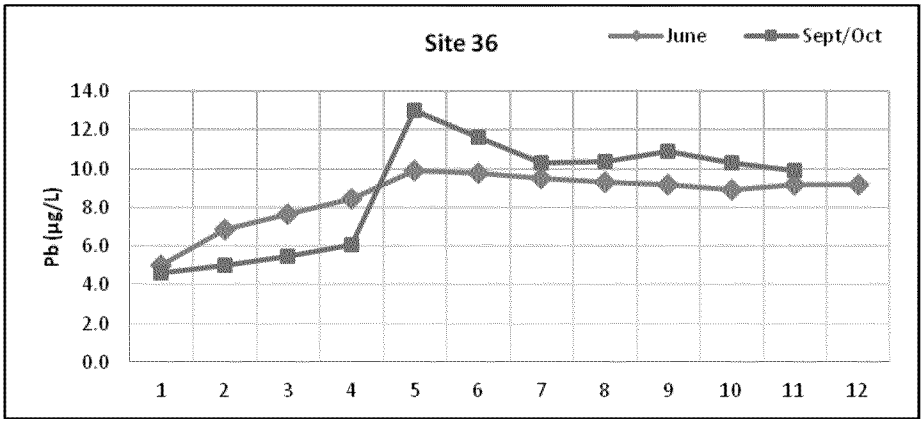
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

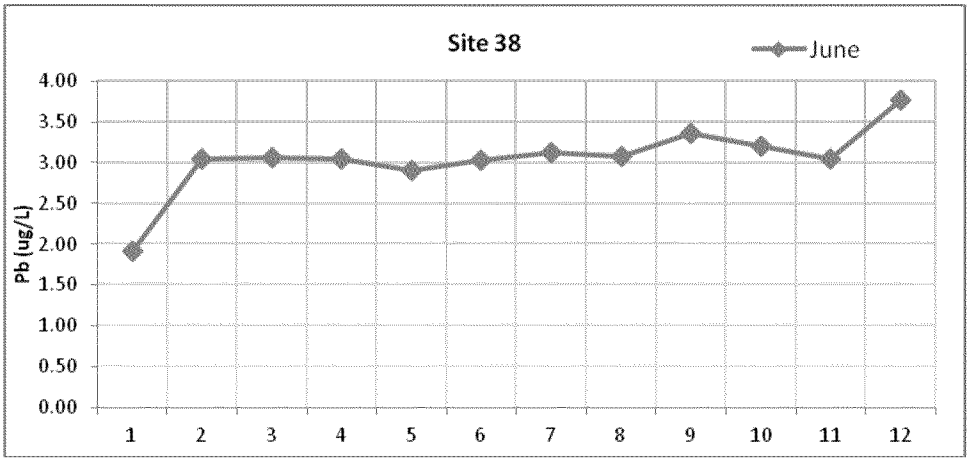
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

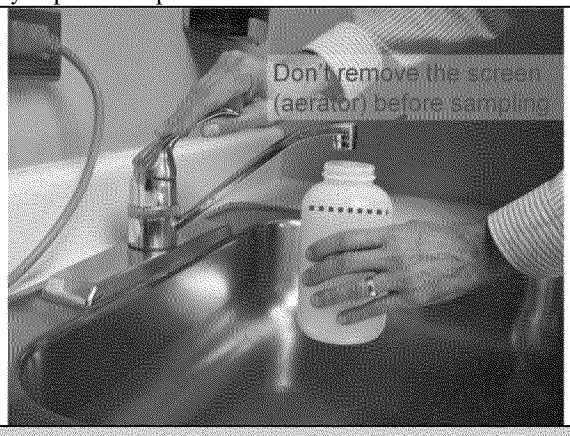
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

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2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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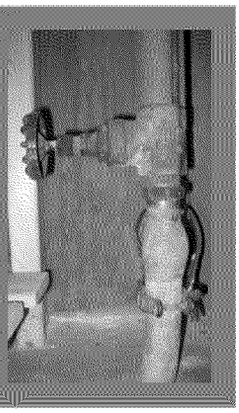
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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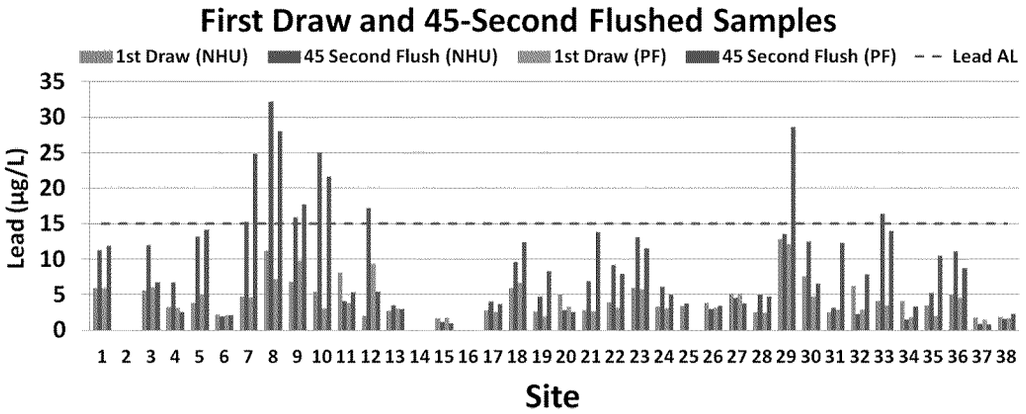

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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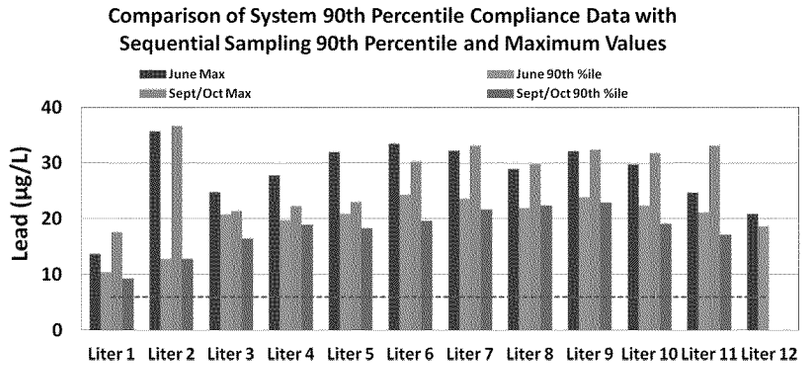

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

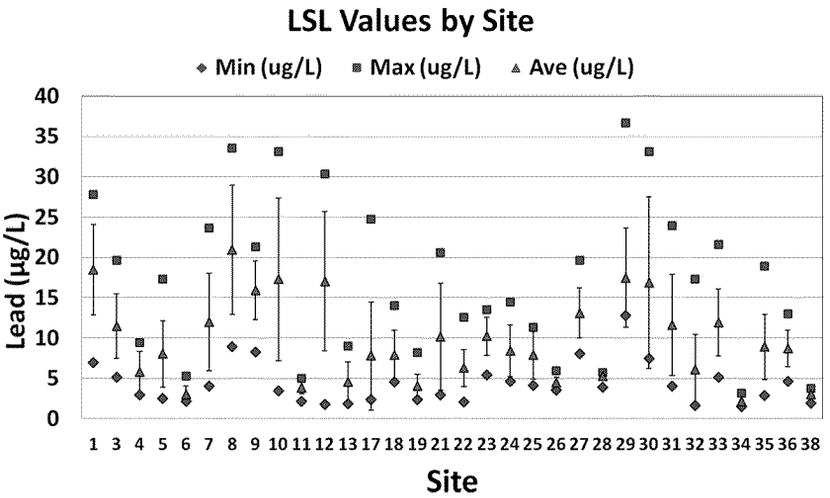

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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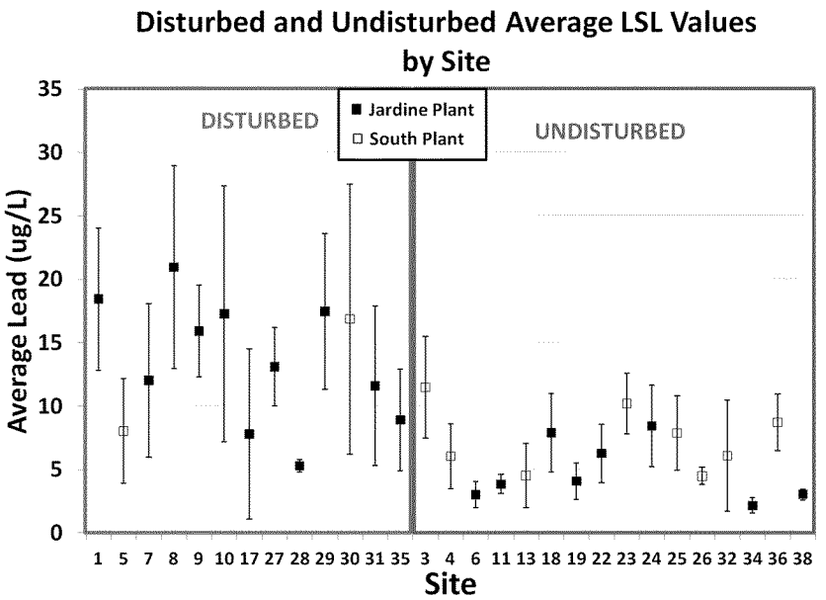

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Figure S2	Photograph of LSL Bulb	Table S2	Chicago Compliance Data
Figure S3	Photograph of LSL Segment	Table S3	Summary of Samples Collected
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Figure S5	Sample Site Map and Home Age	Table S4b	Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.
Figure S6	Graph of Four Metals for Site 9	Table S5	June Sequential Sampling Results
Figure S7	City B Sampling Instructions	Table S6a	Sept/Oct Sequential Sampling Results Used in Analyses
Figure S8	City B LSL Results	Table S6b	Sept/Oct Supplemental Sequential Sampling Results (Not Used in Analyses)
Figure S9-S40	Sequential Sampling Graphs (Lead)	Table S6c	Summary of stagnation times.
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Figure S45	Sept/Oct Sample Collection and Reporting Form		

Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

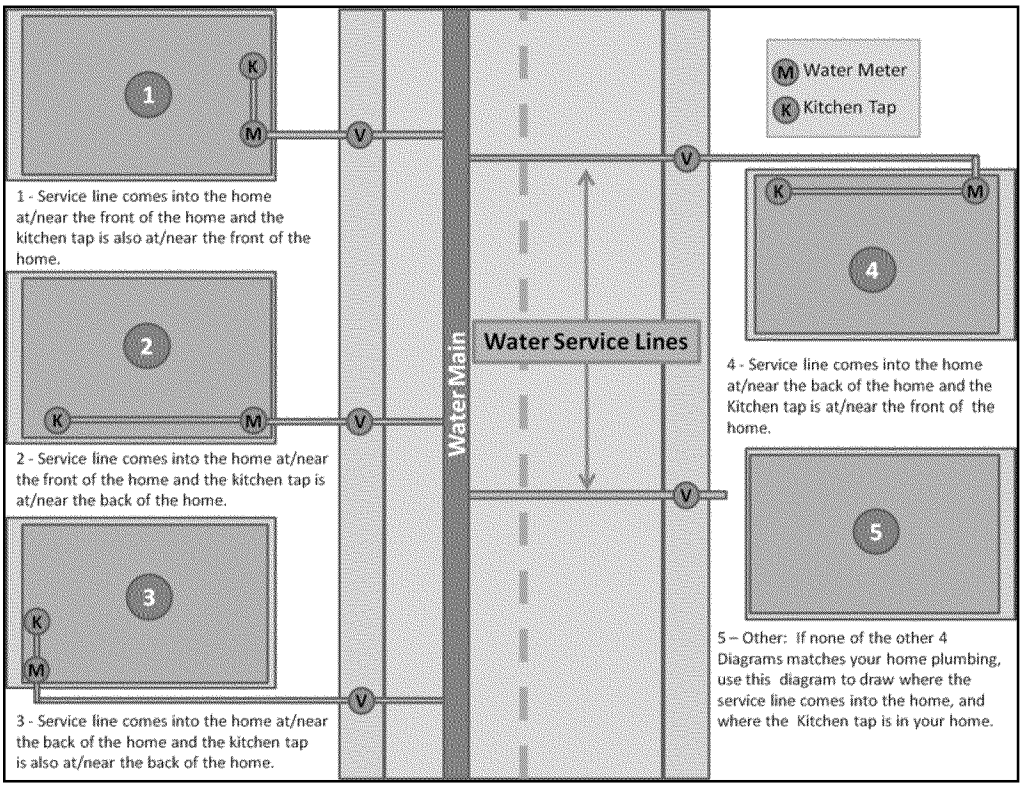


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

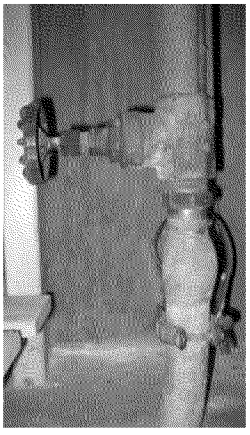


Figure S2: LSL Bulb

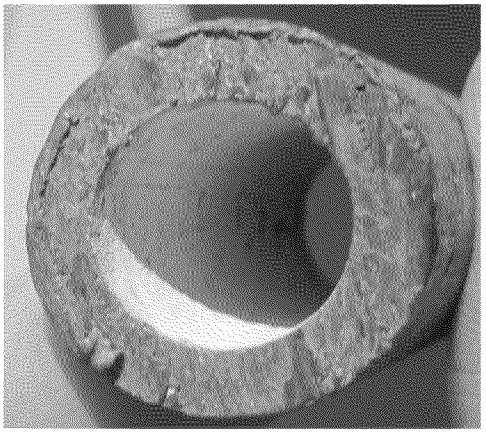


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

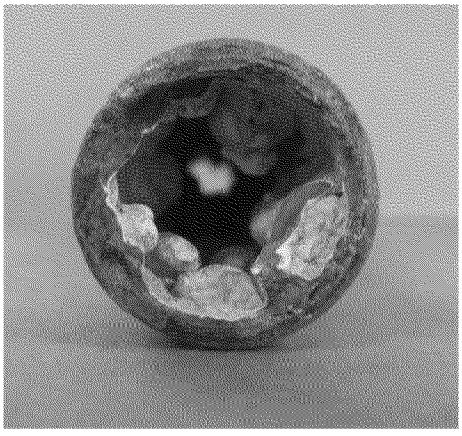


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

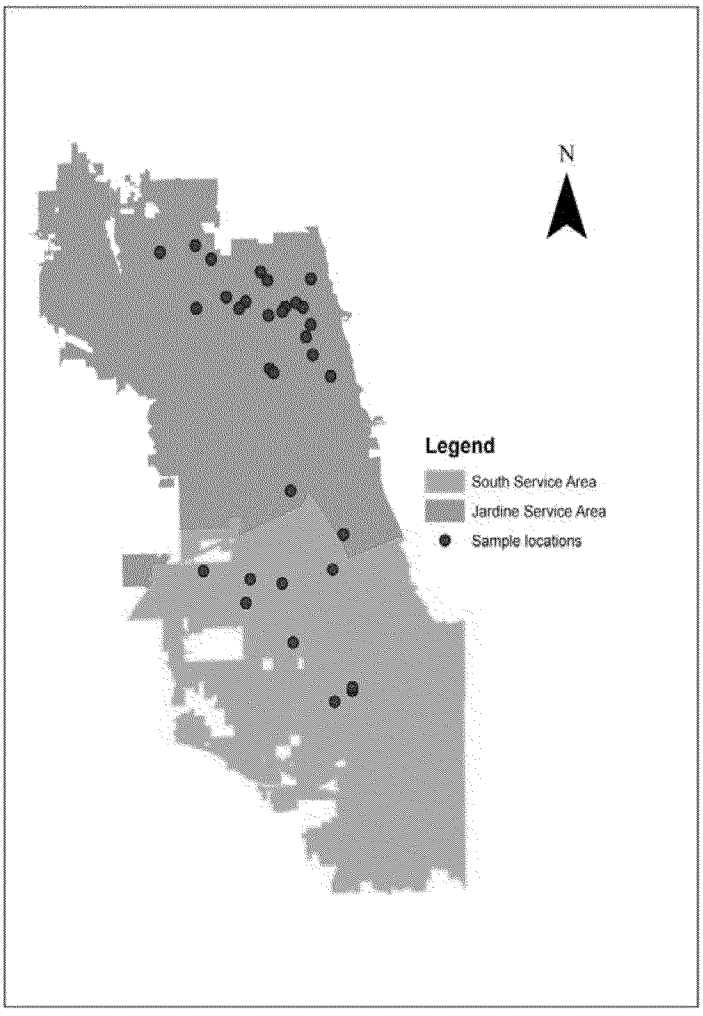


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

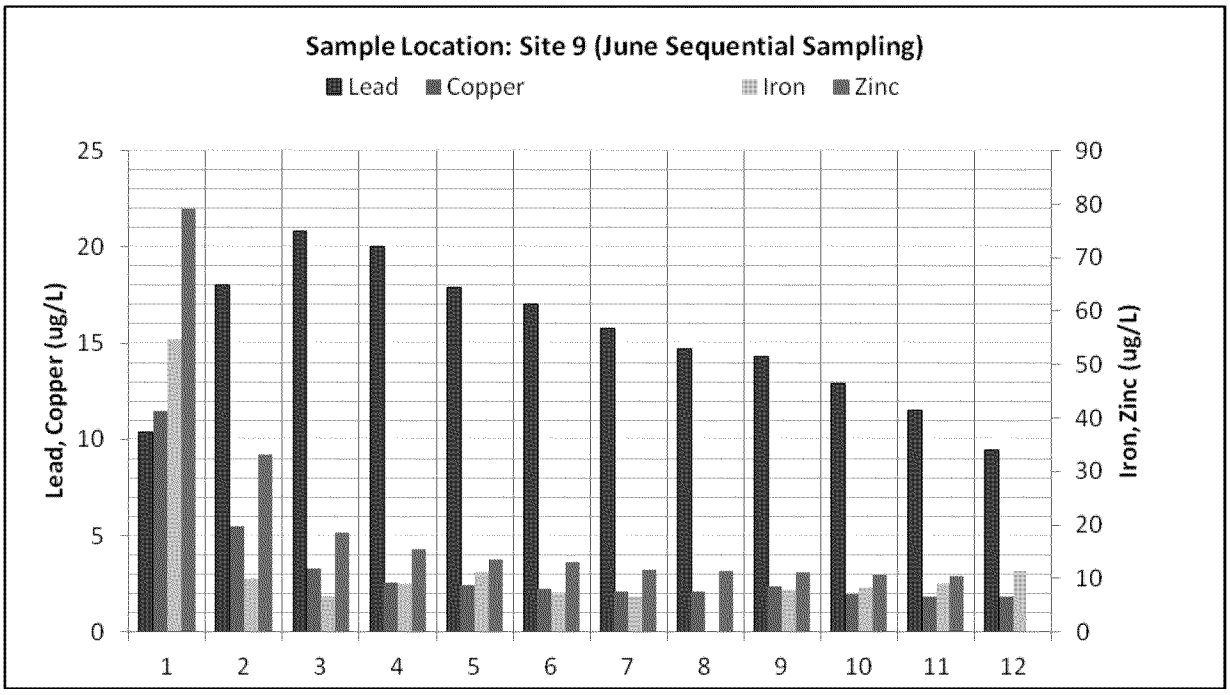


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1 Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2 At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3 When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4 Fill out the enclosed chain of custody form and survey.
5 Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

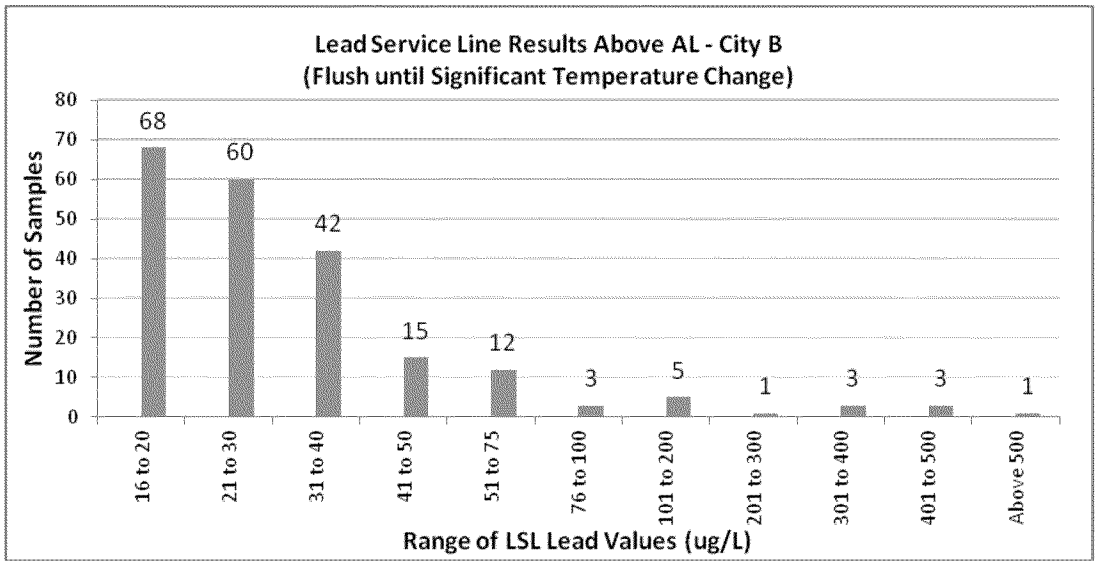
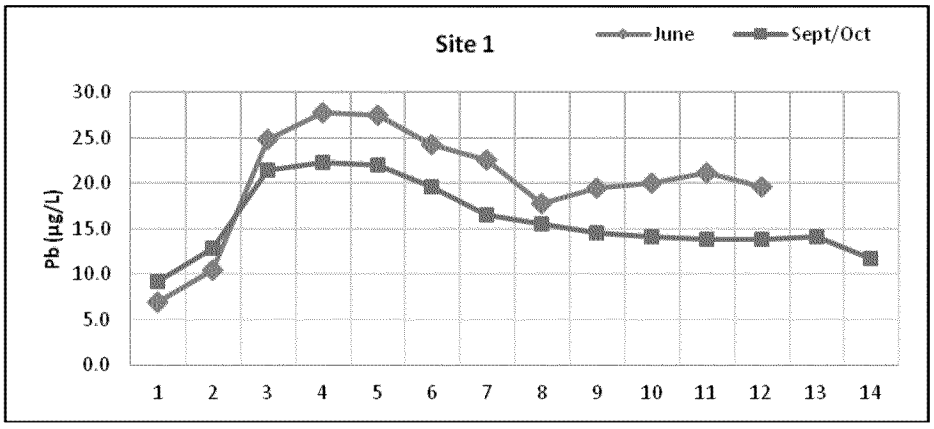


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

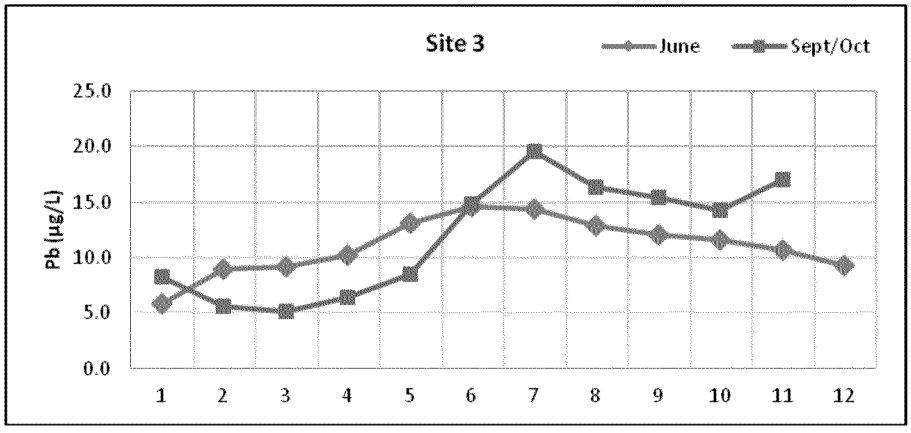
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

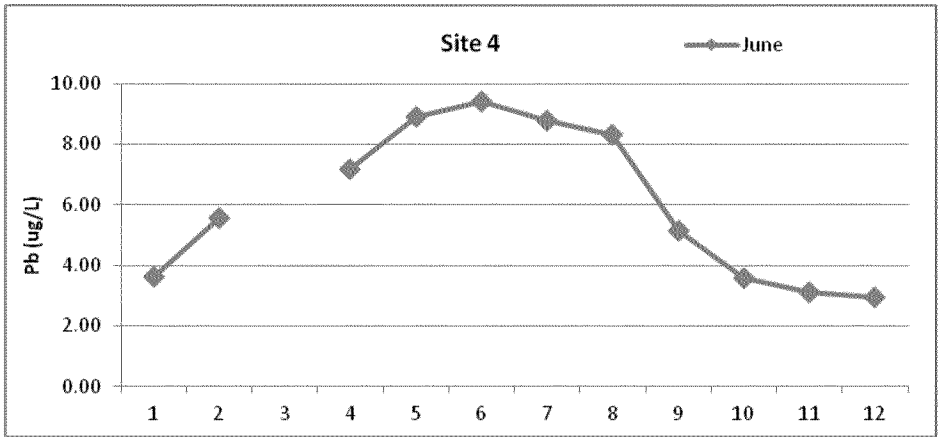
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

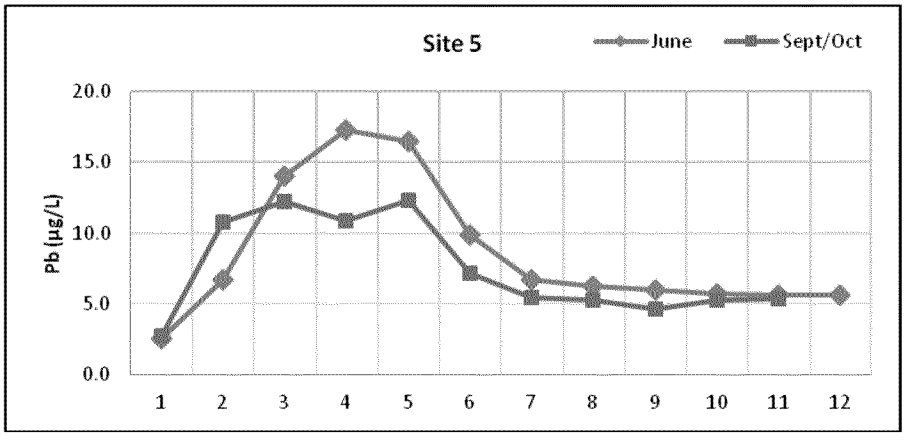
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

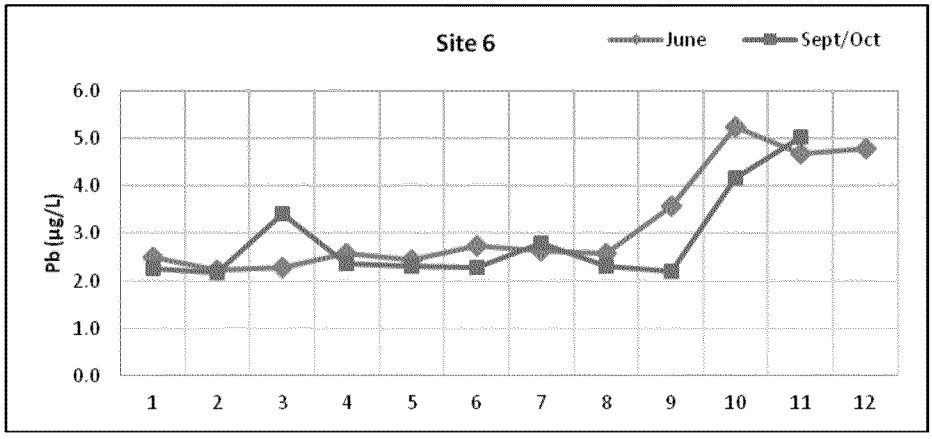
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

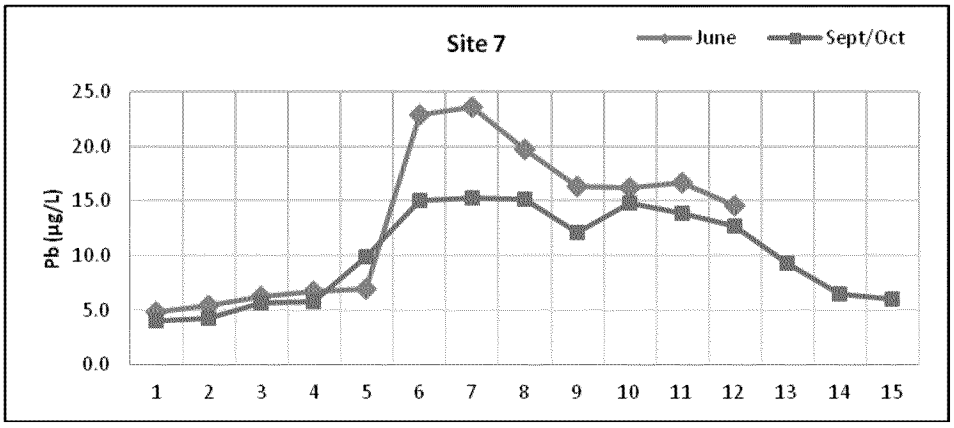
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

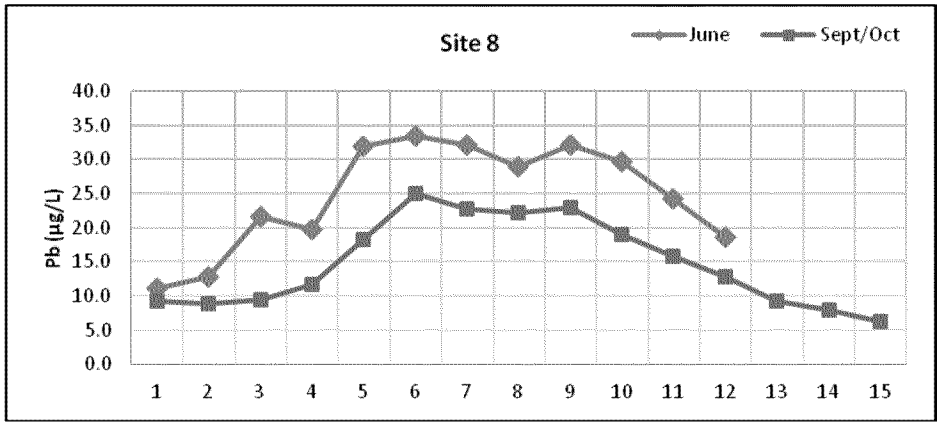
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

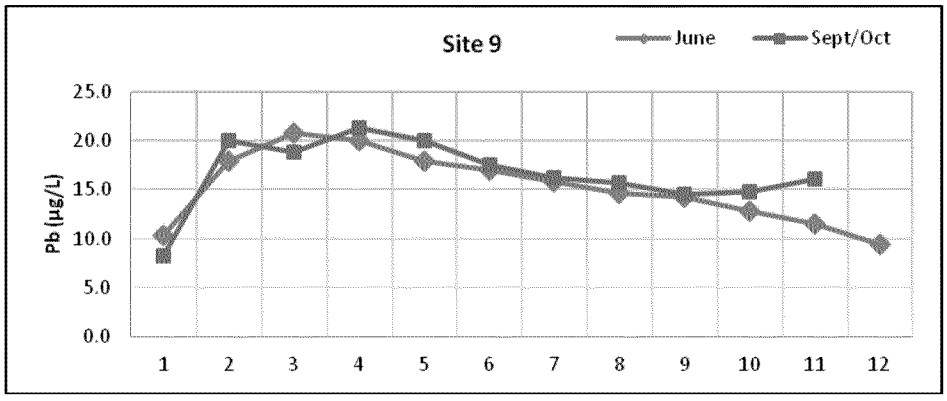
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

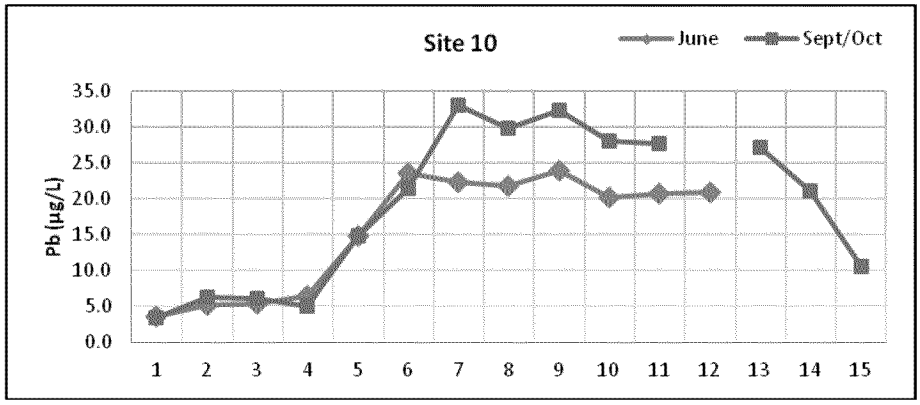
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

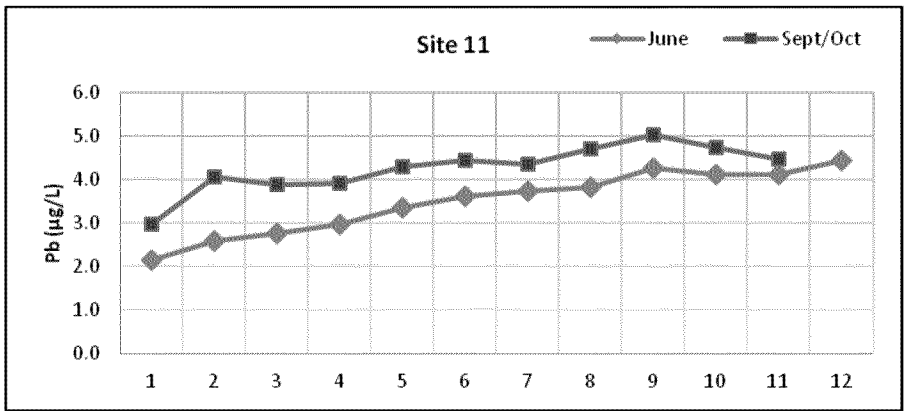
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

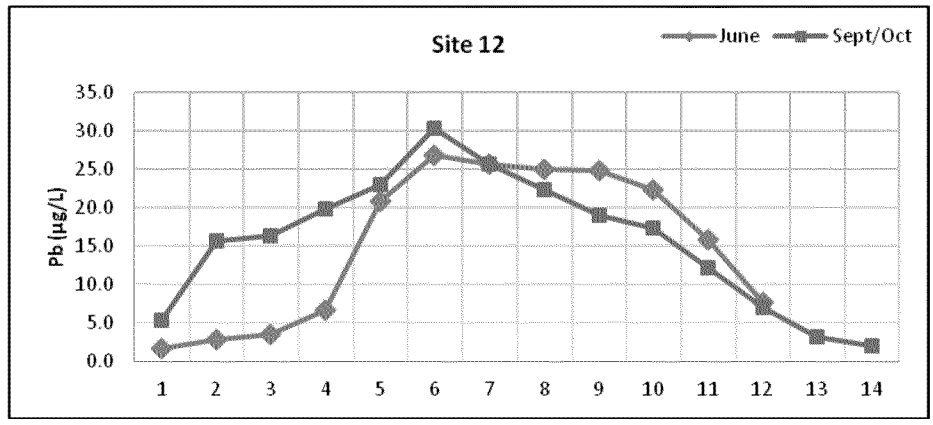
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

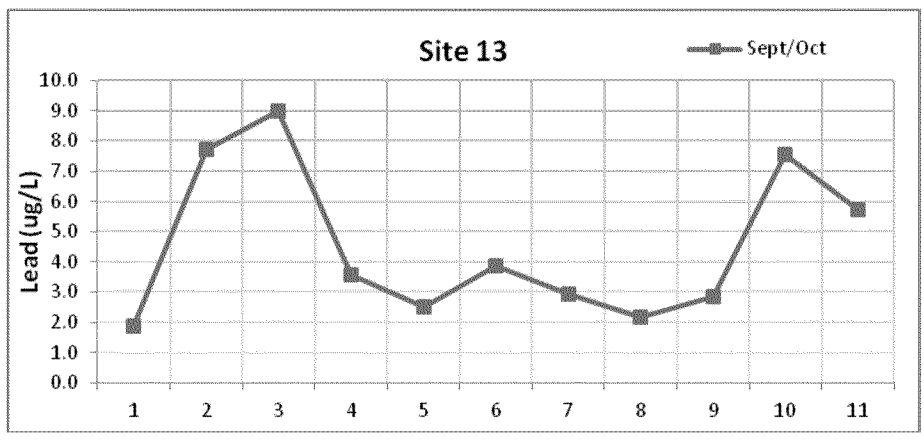
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

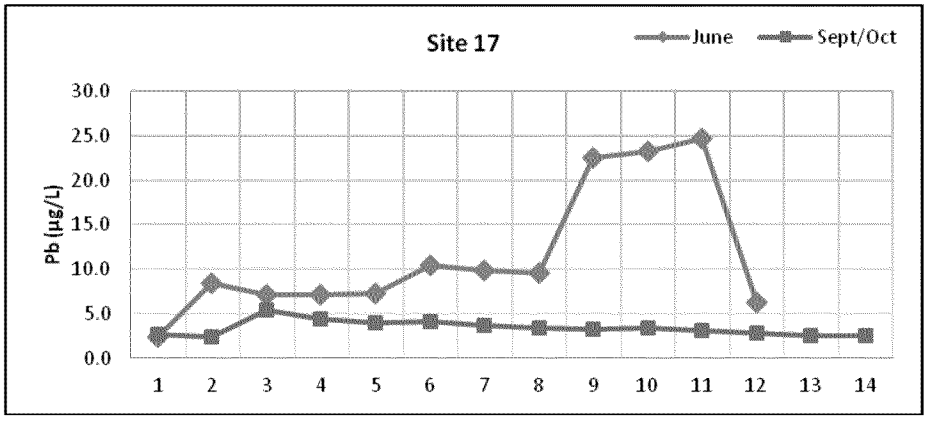
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

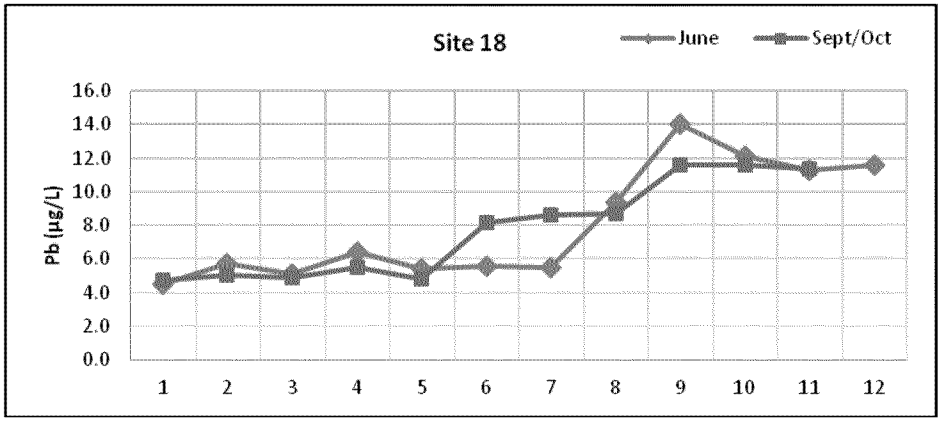
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

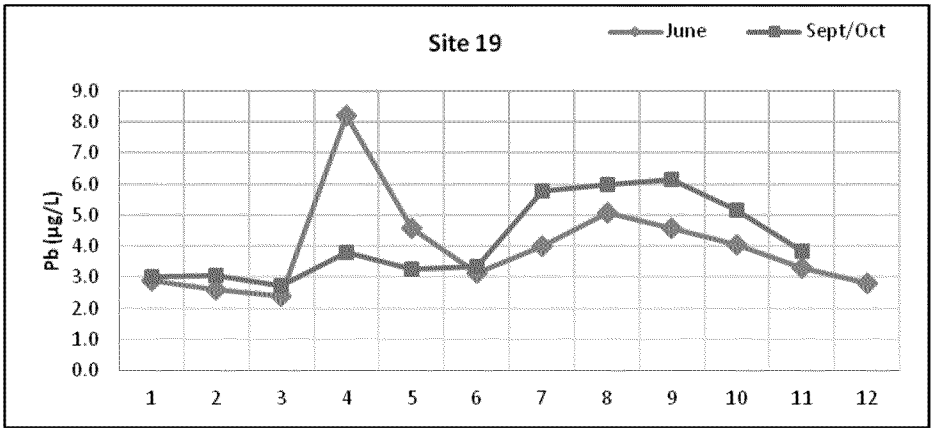
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

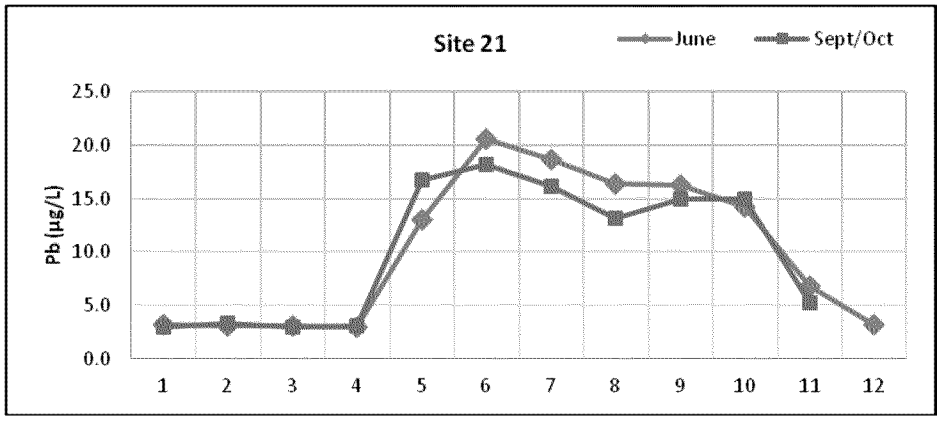
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

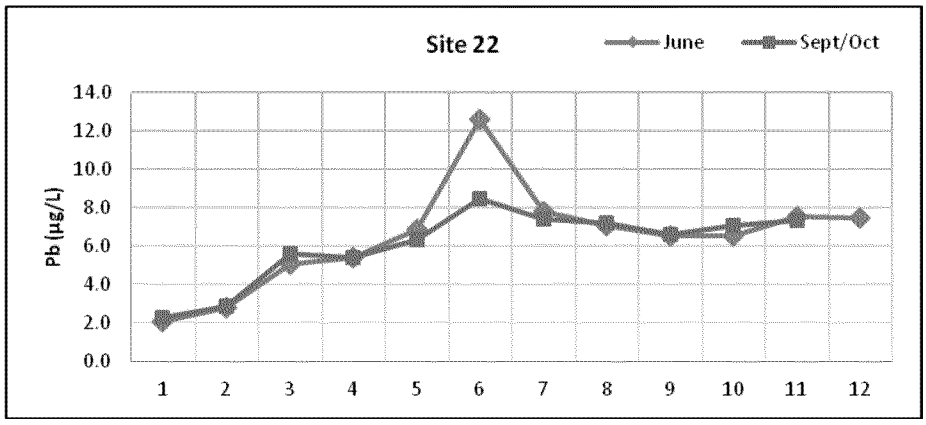
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

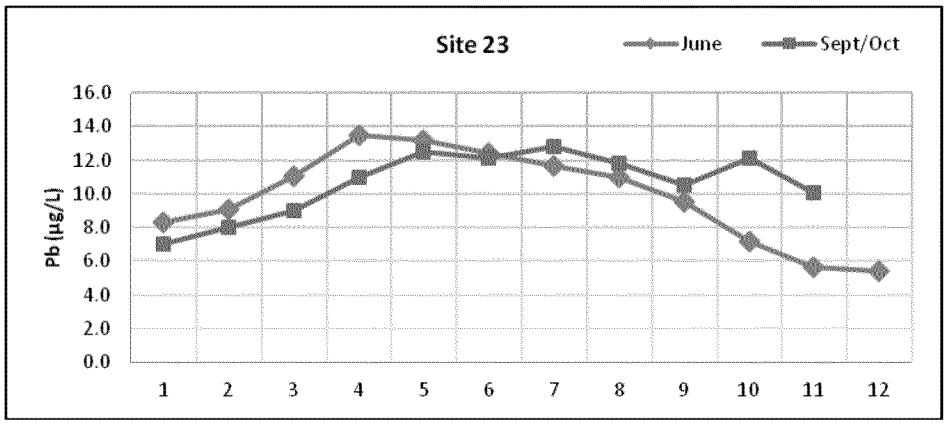
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

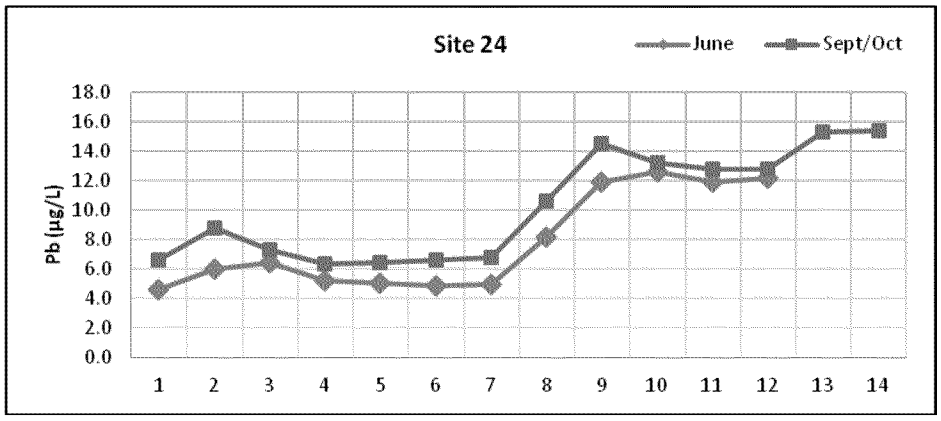
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

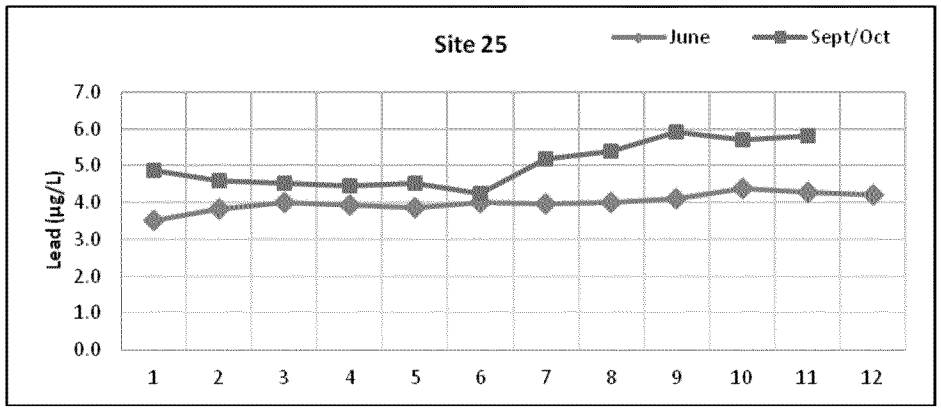
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

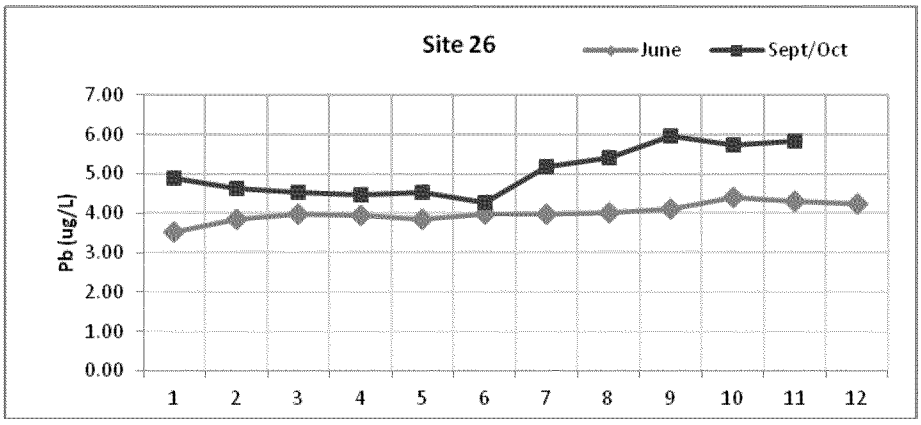
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

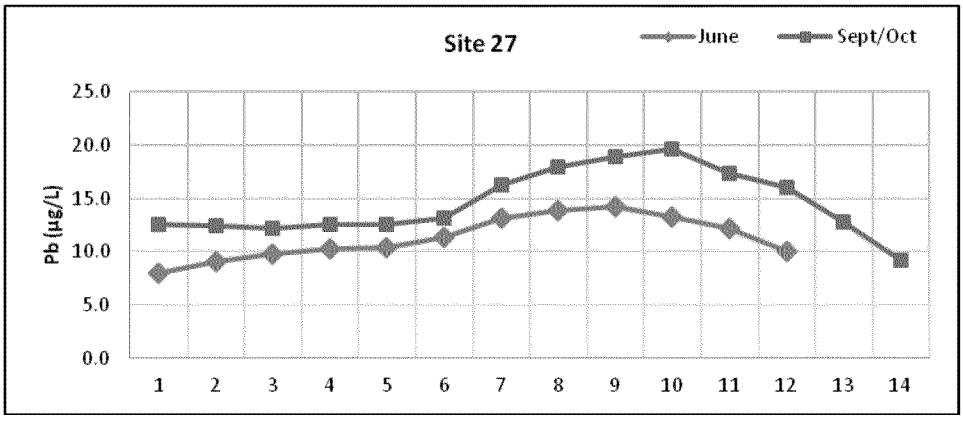
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

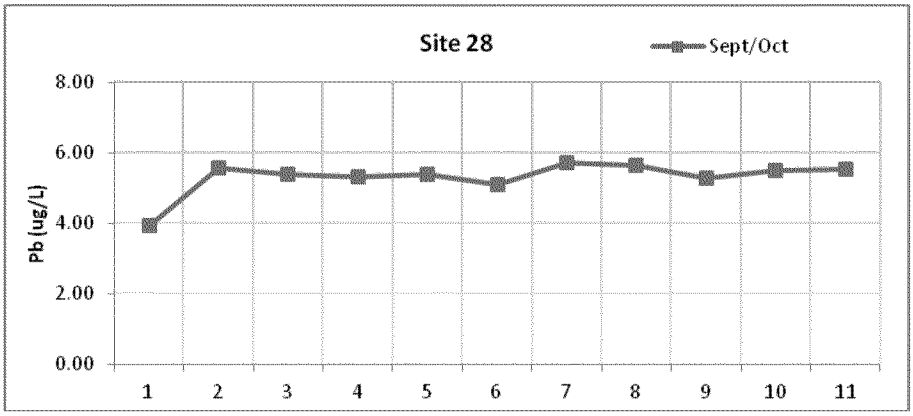
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

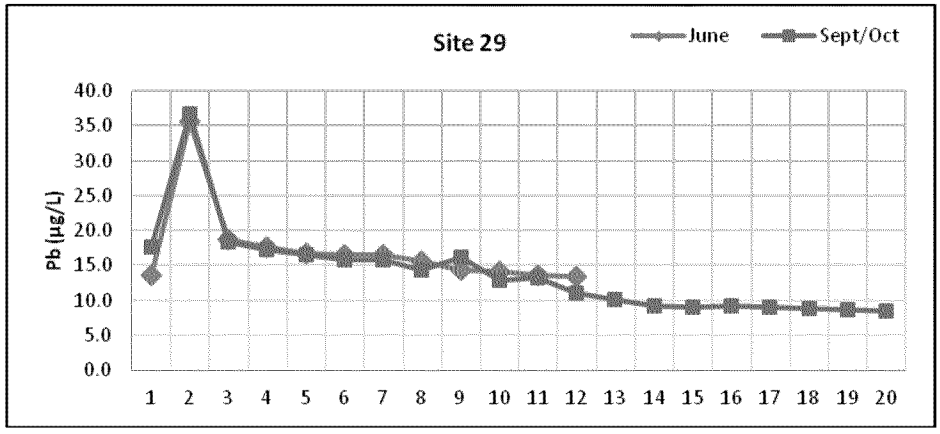
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

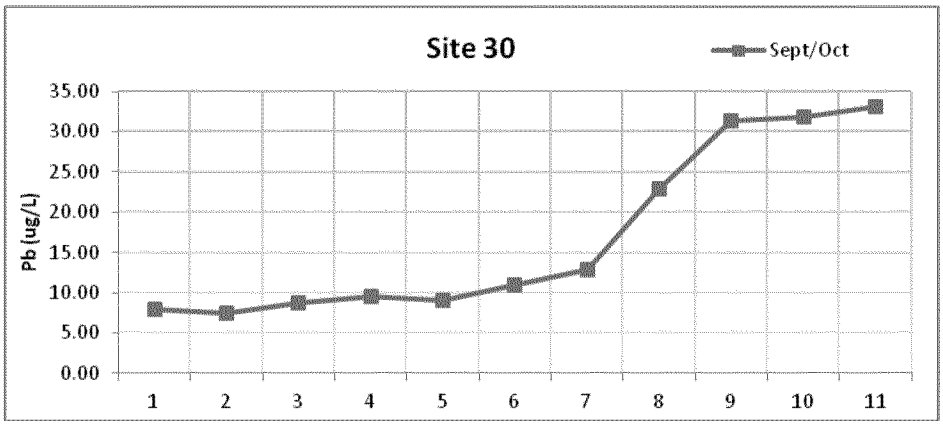
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

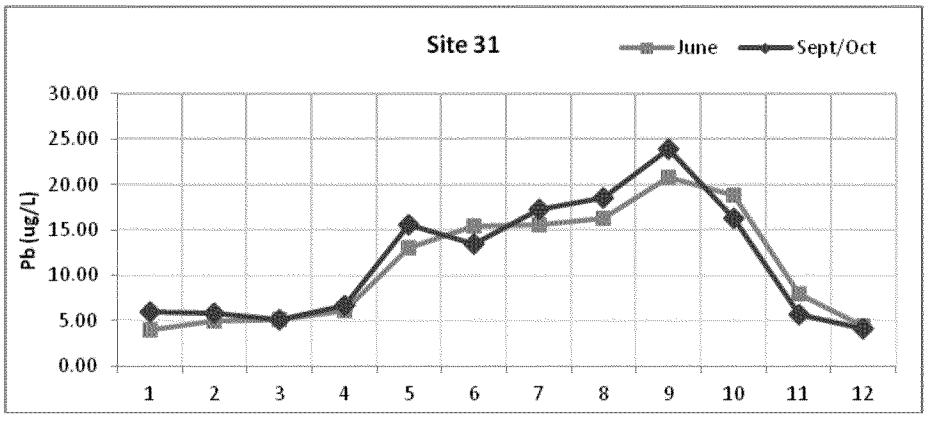
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

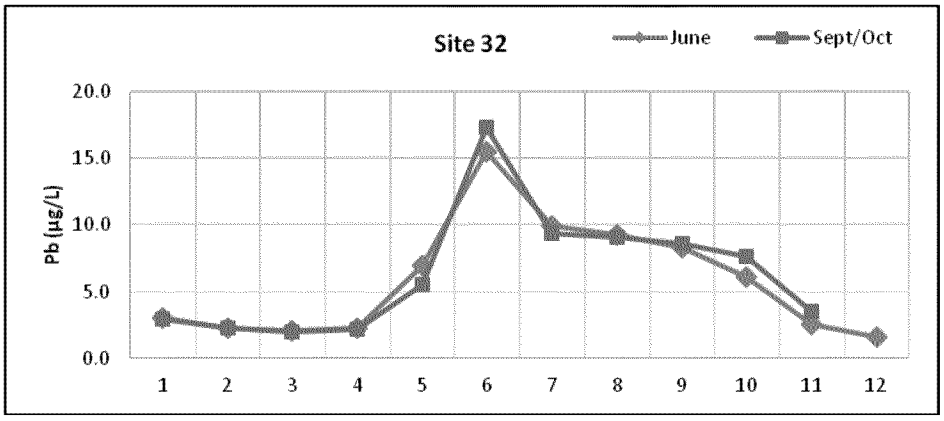
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

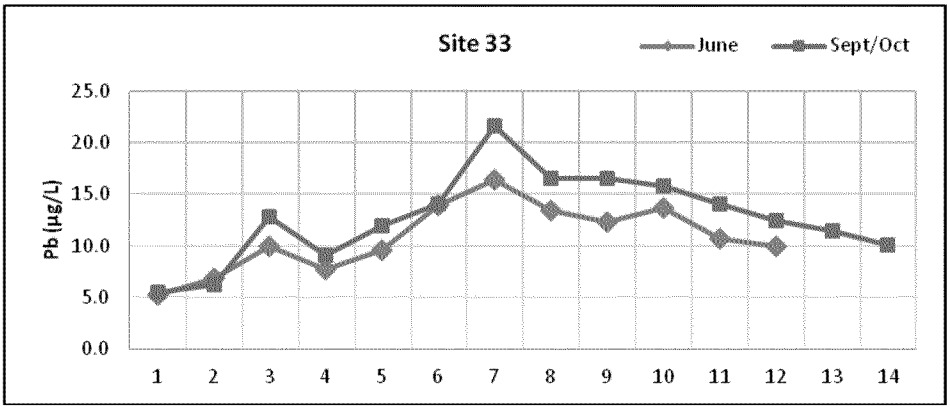
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

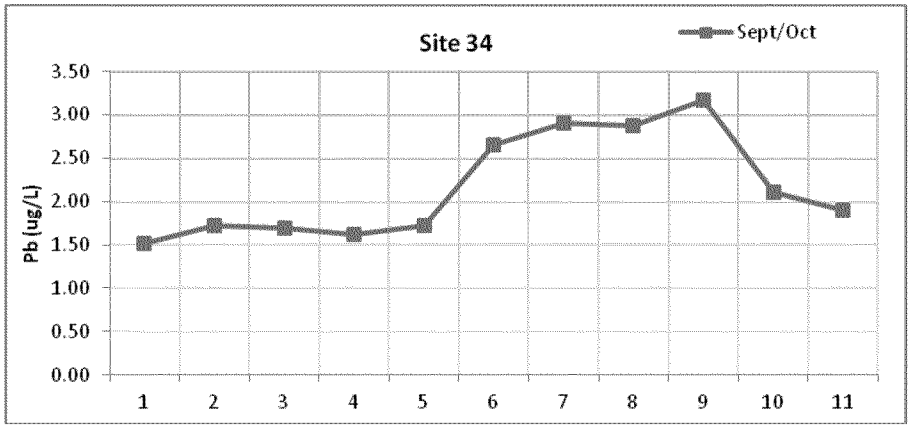
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

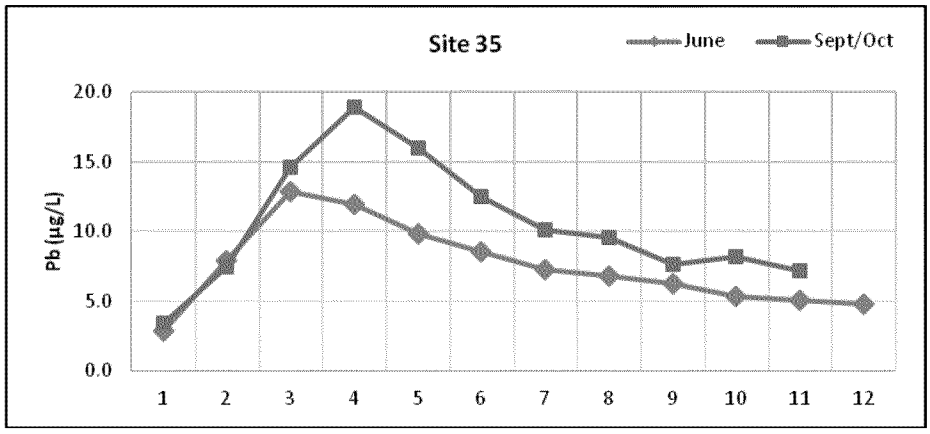
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

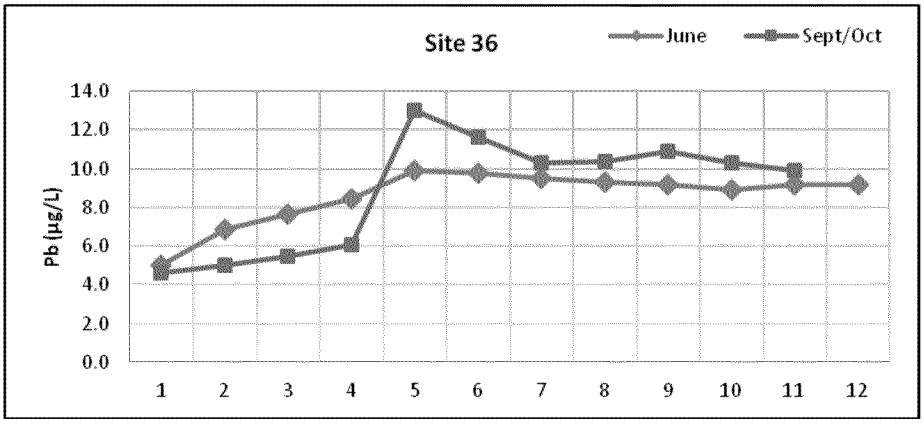
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

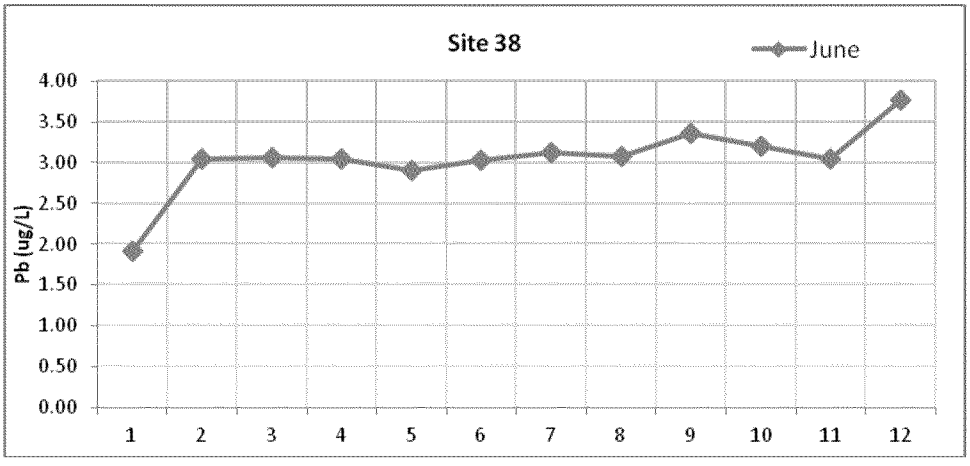
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

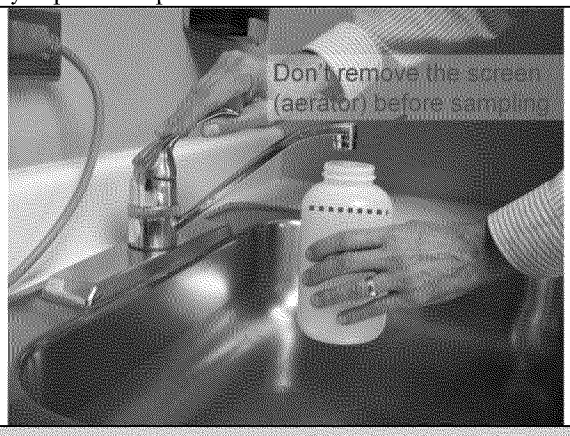
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

Literature Cited/References

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2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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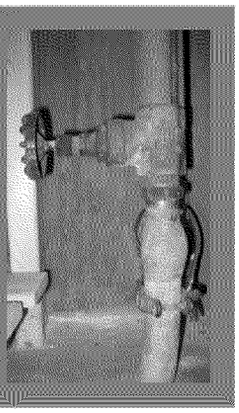
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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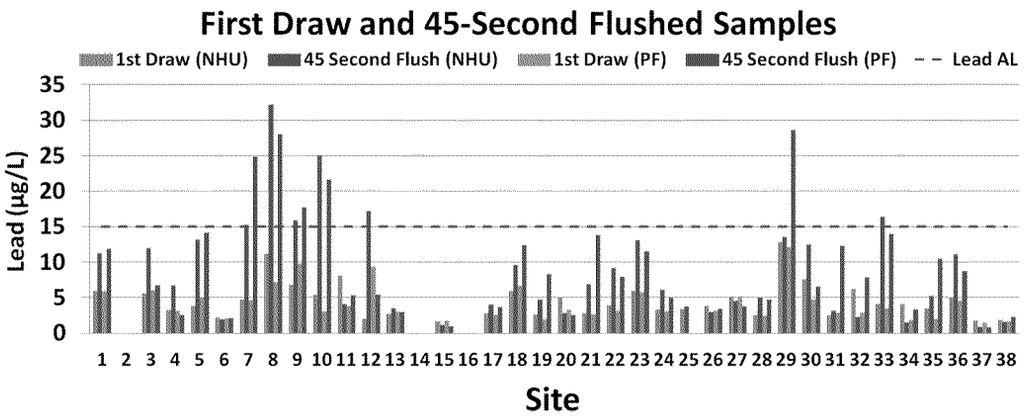

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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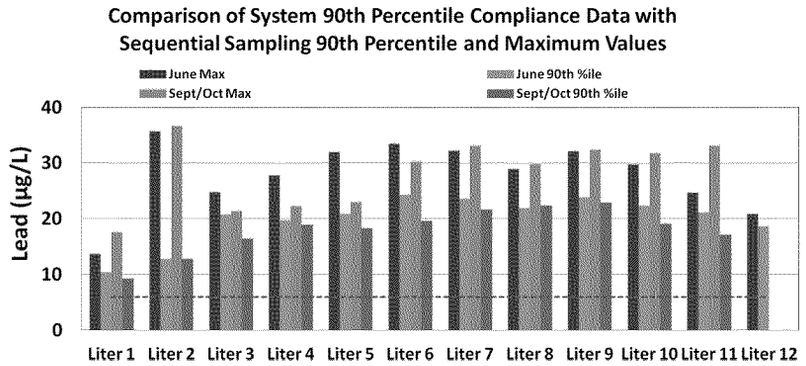

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

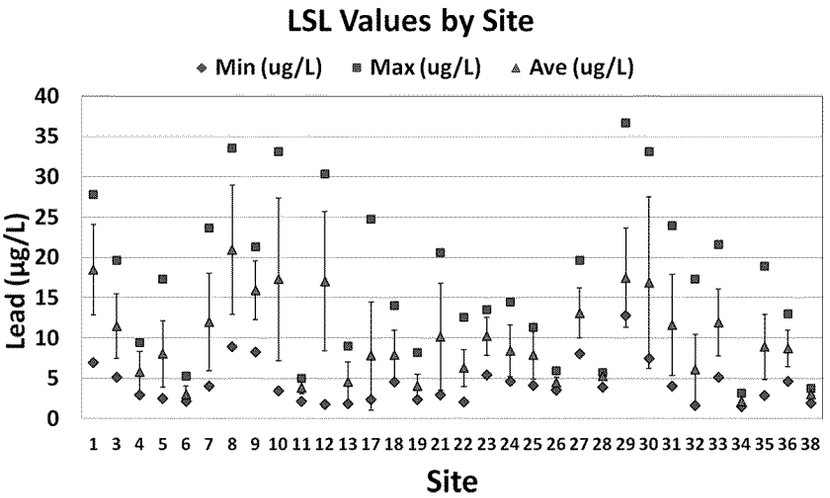

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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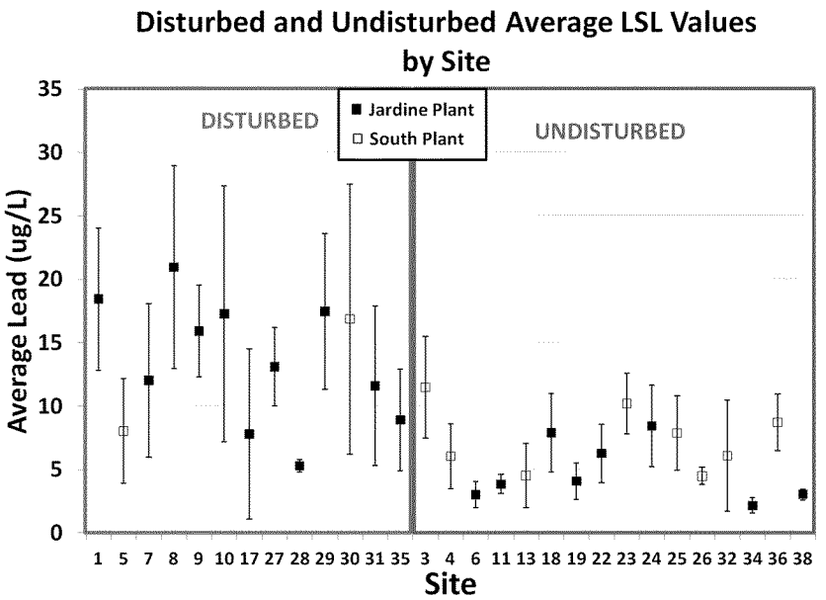

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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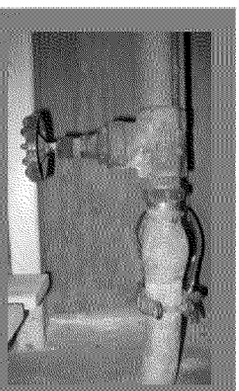
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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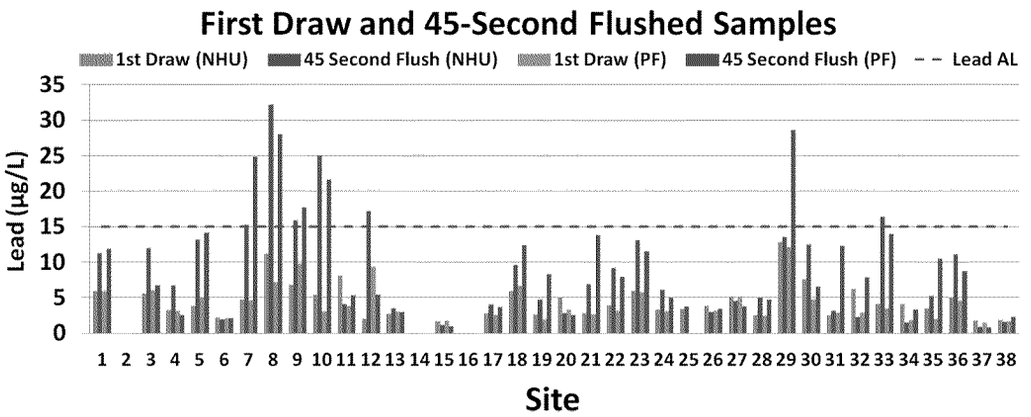

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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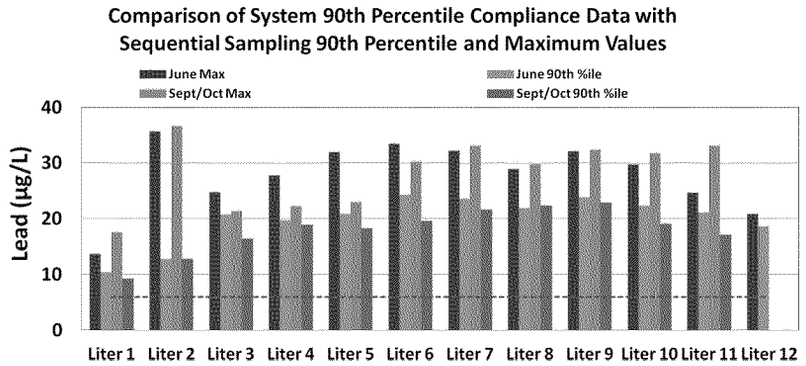

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

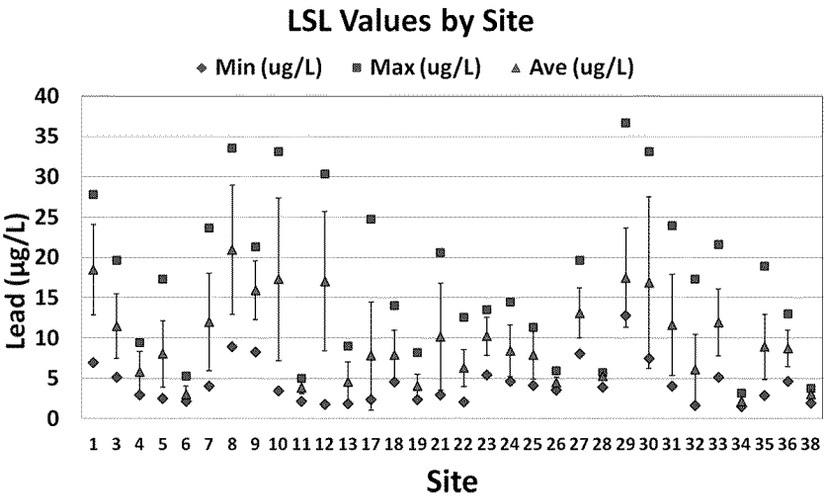

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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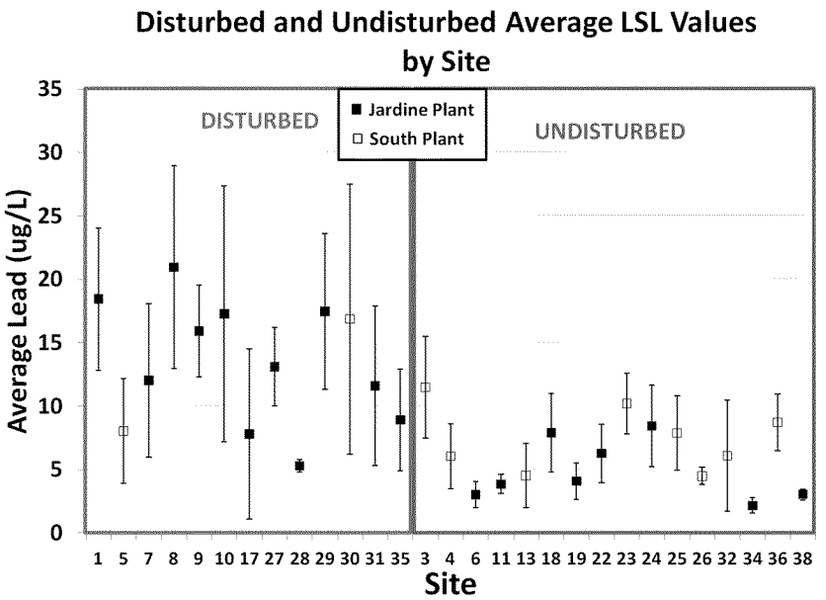

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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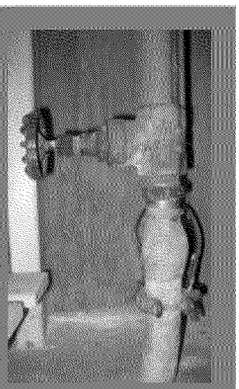
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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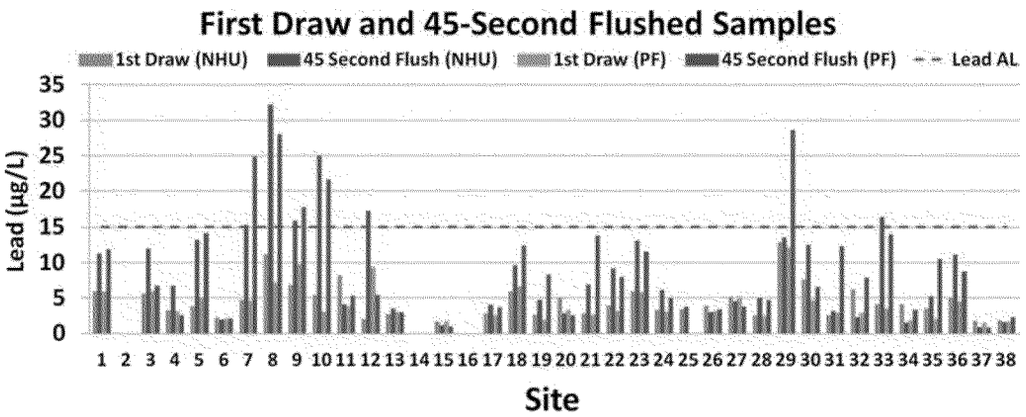

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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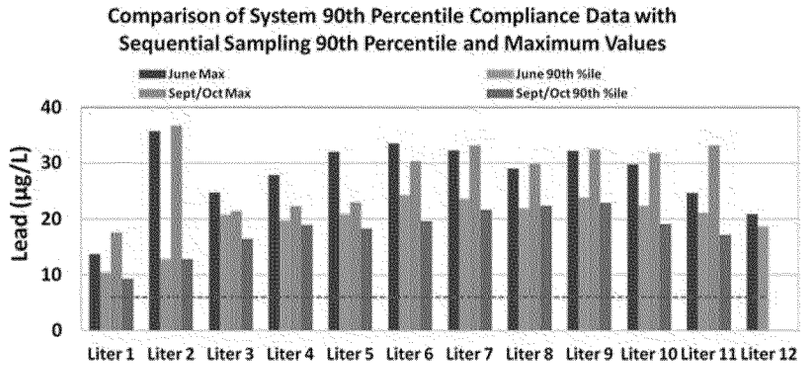

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

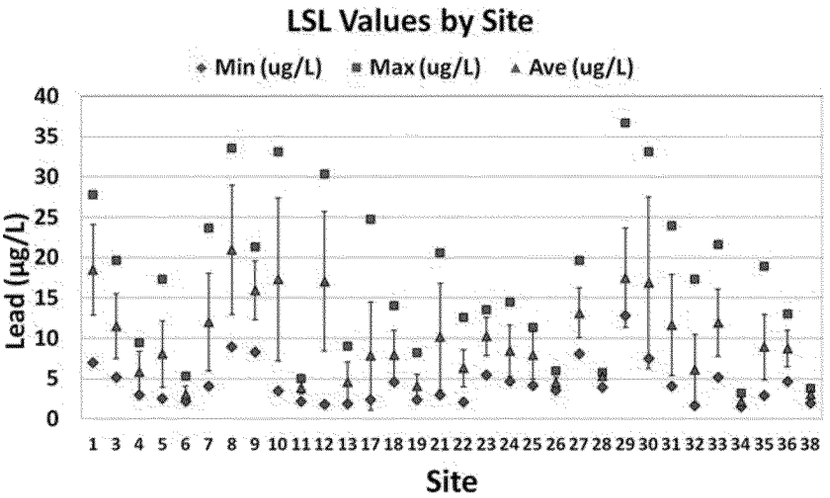

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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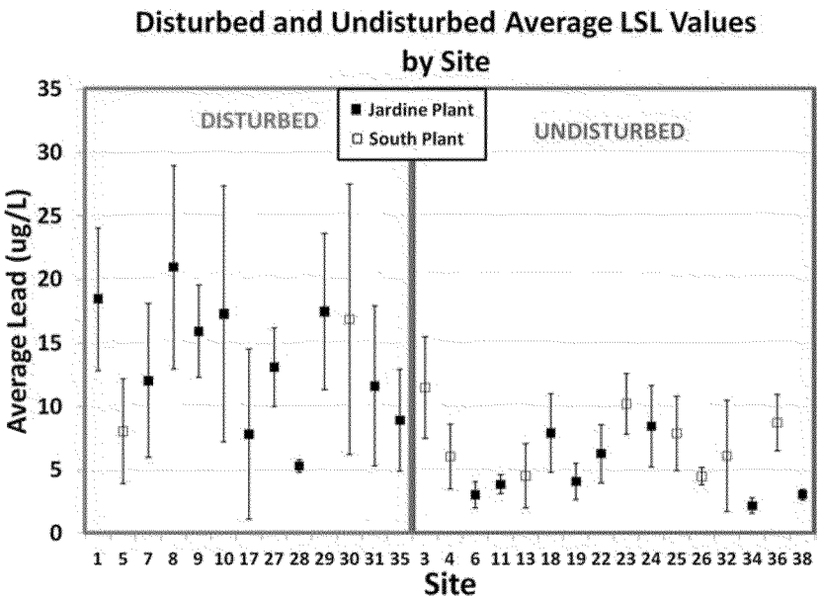

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

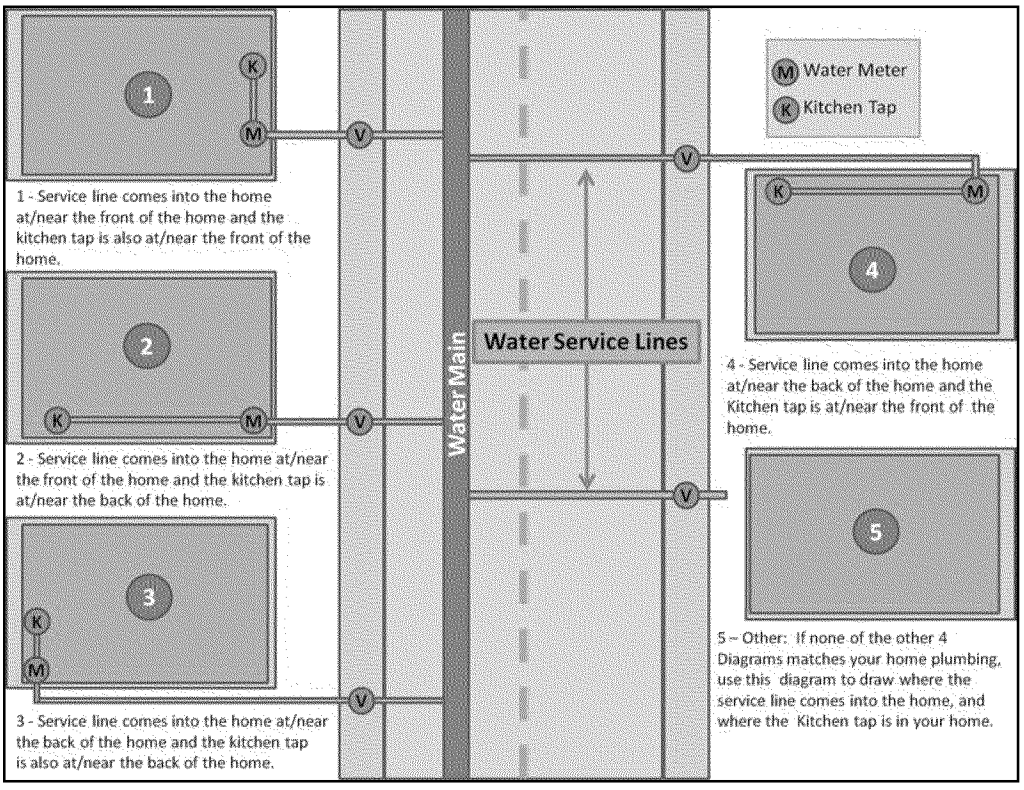


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

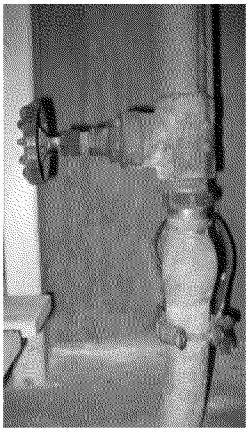


Figure S2: LSL Bulb

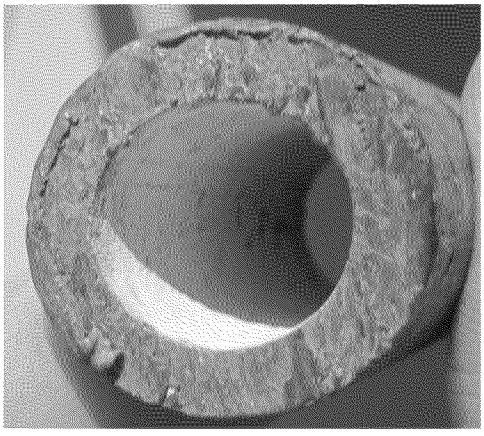


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

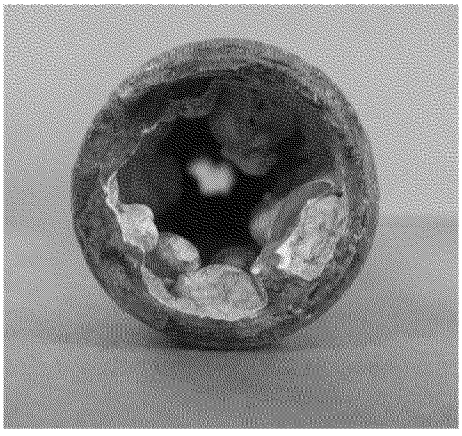


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

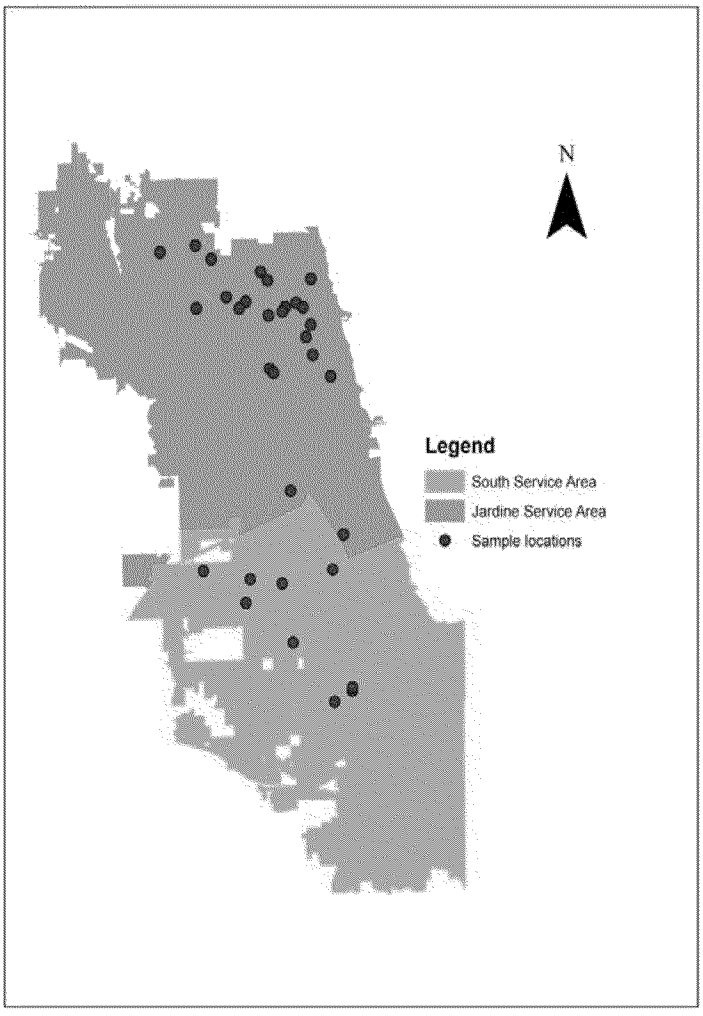


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

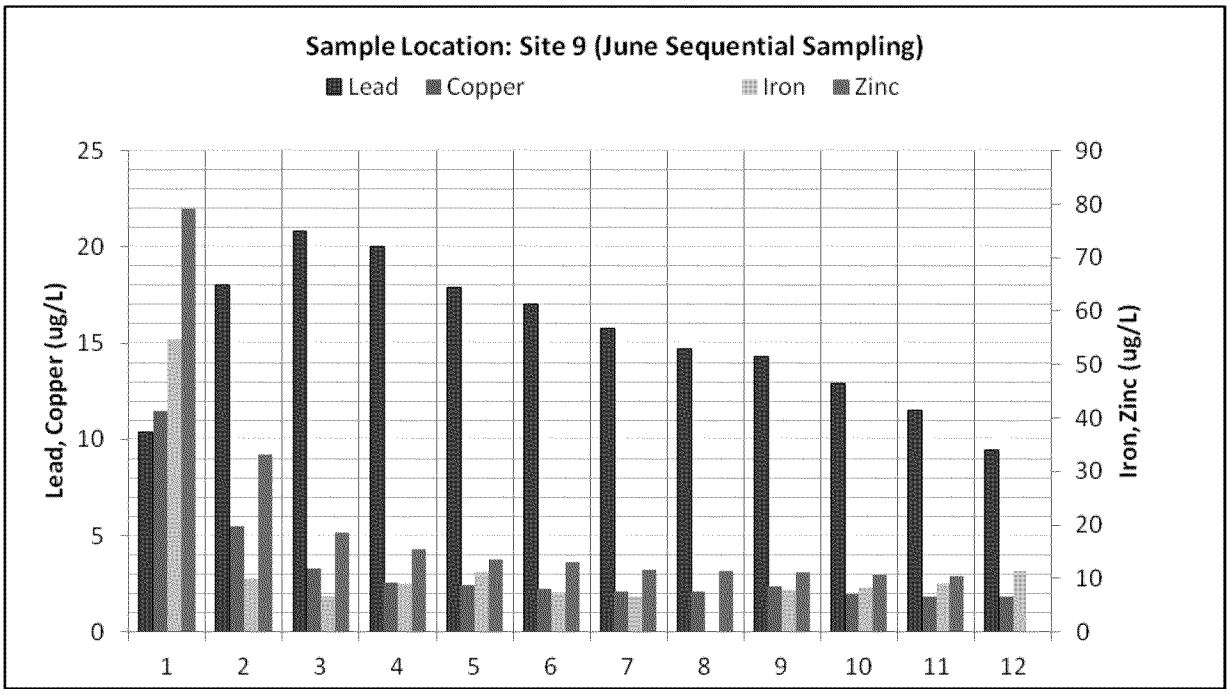


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

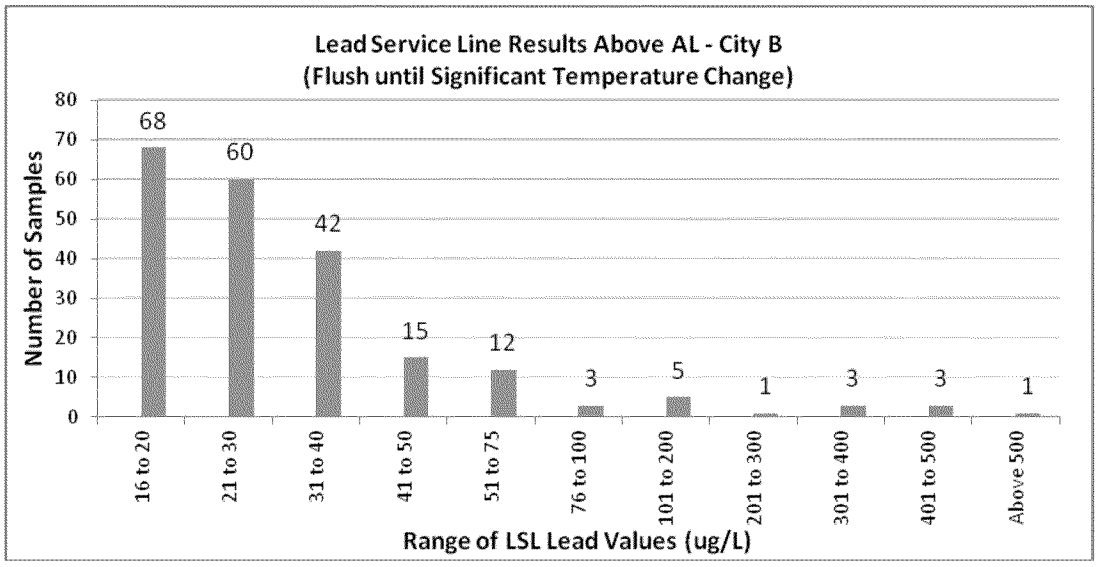
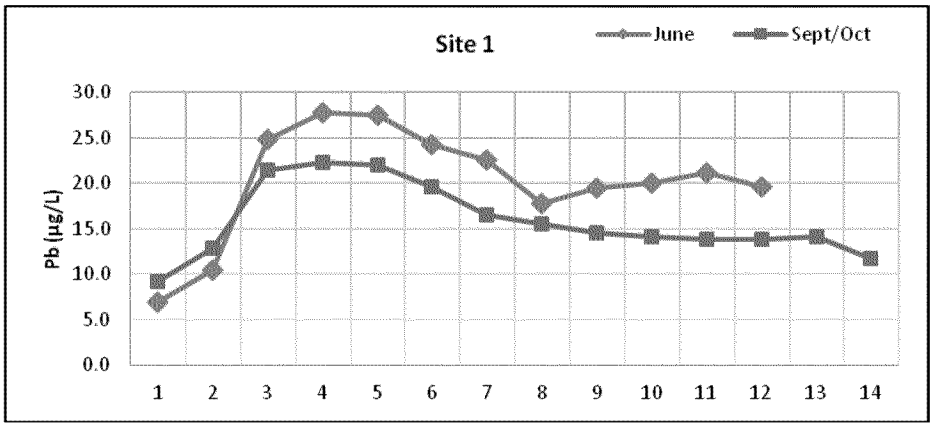


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

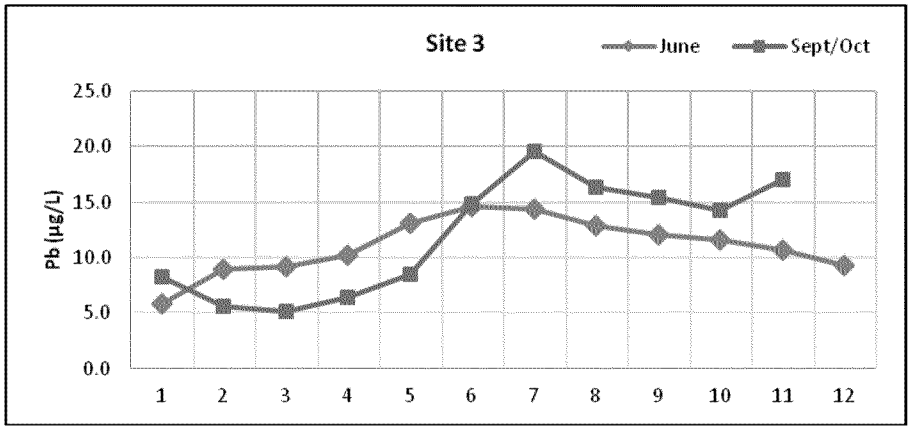
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

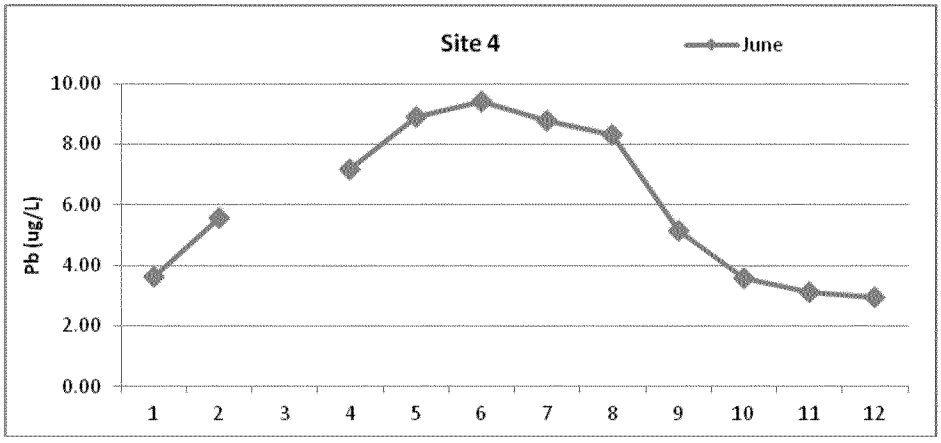
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

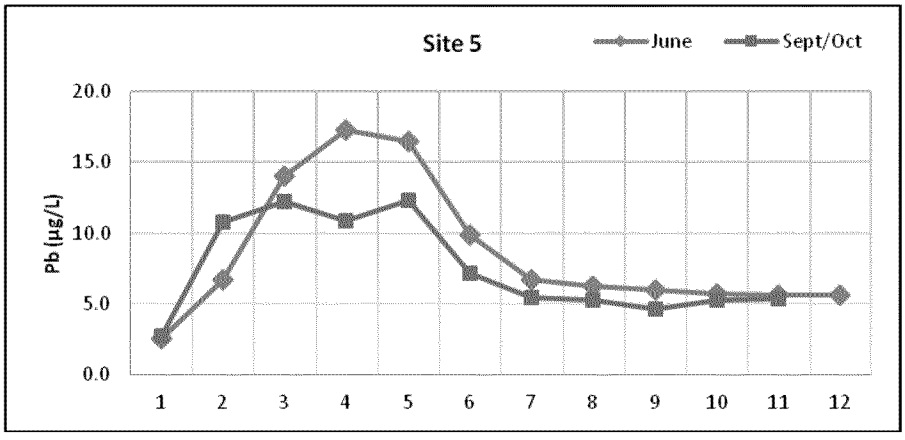
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

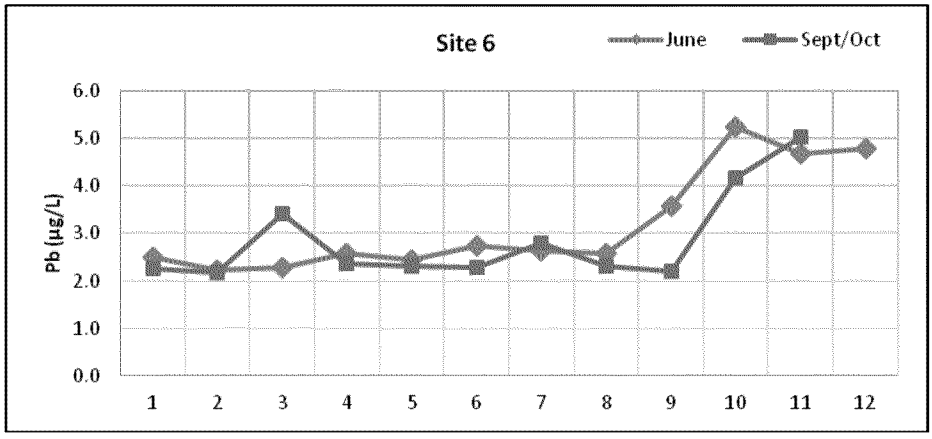
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

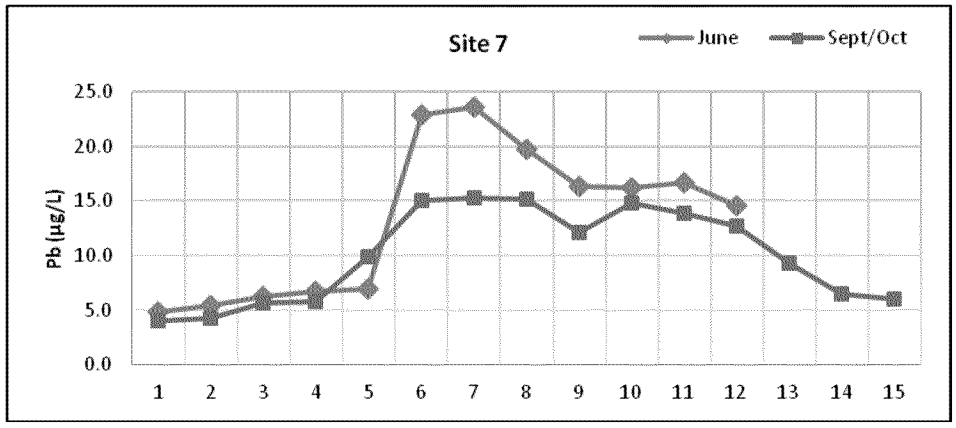
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

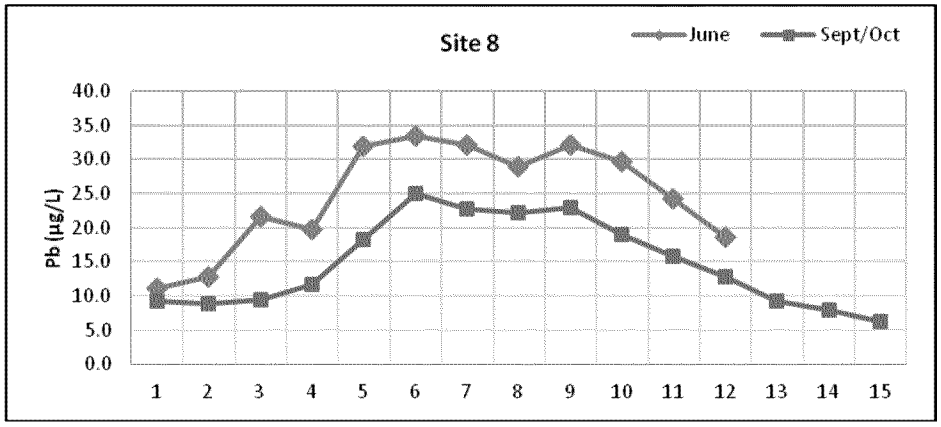
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

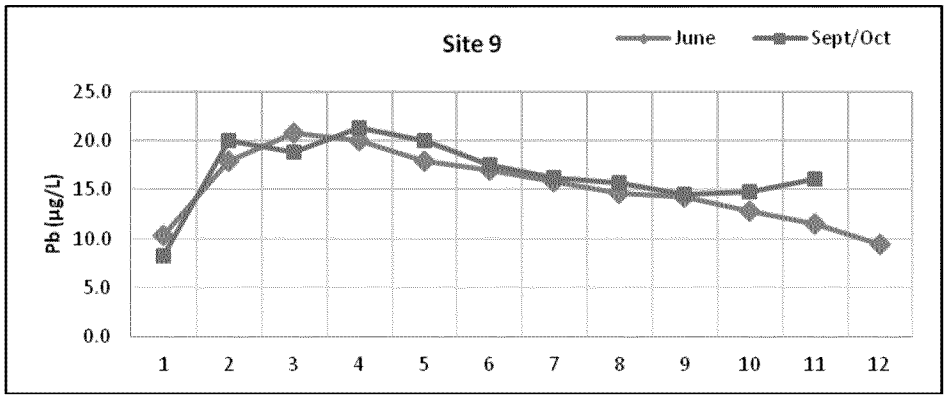
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

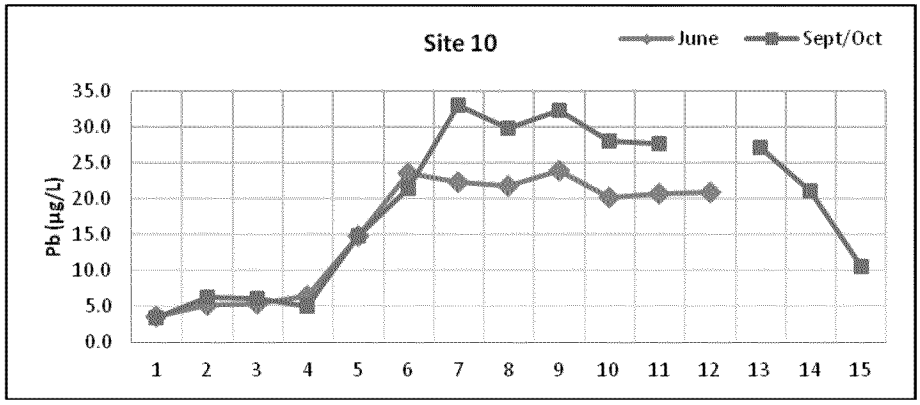
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

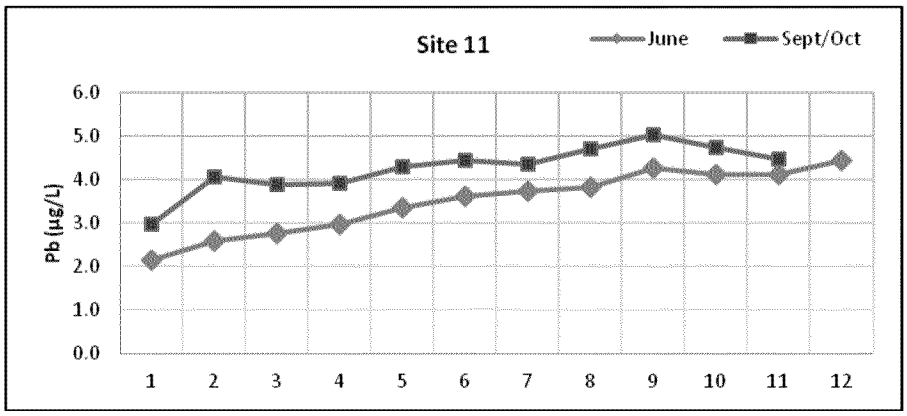
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

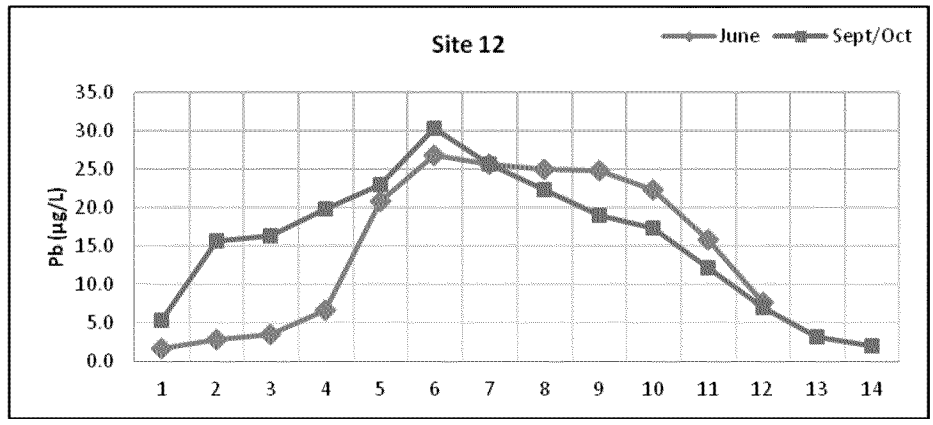
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

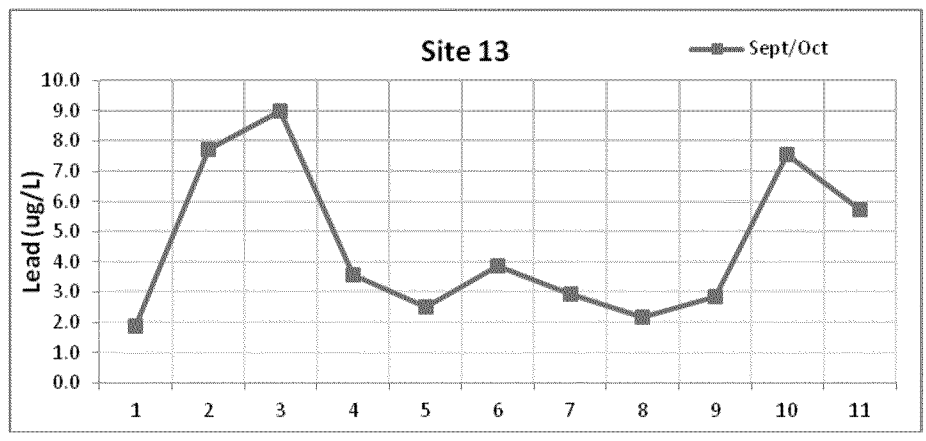
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

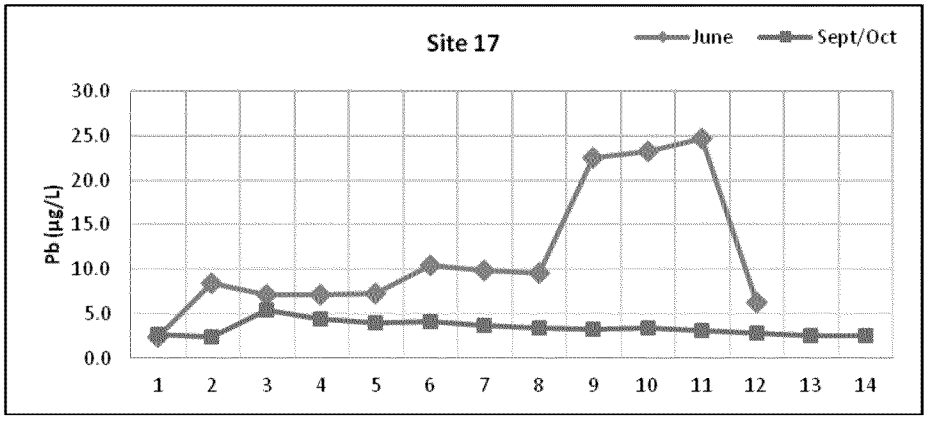
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

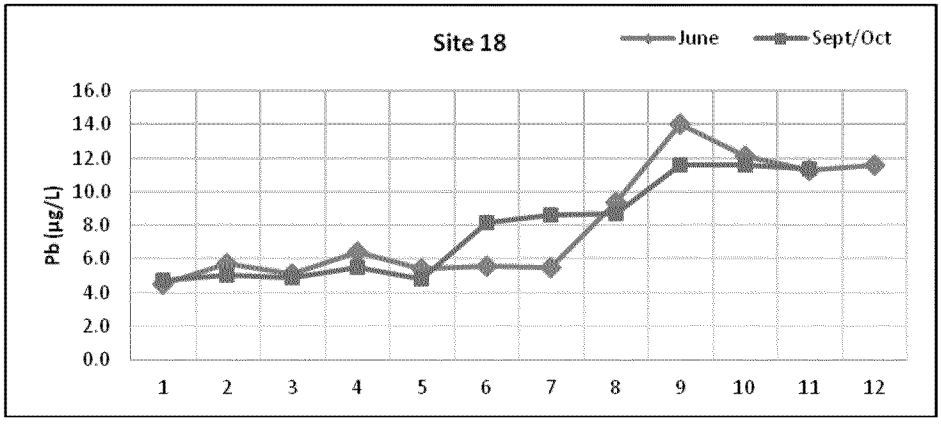
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

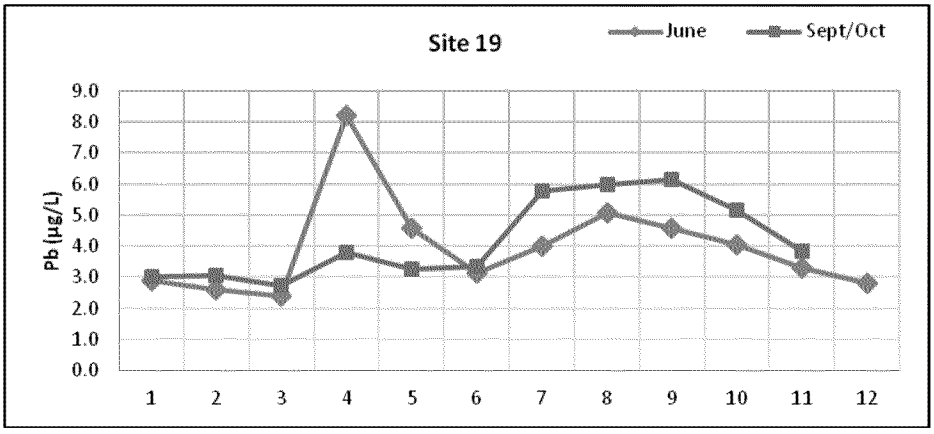
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

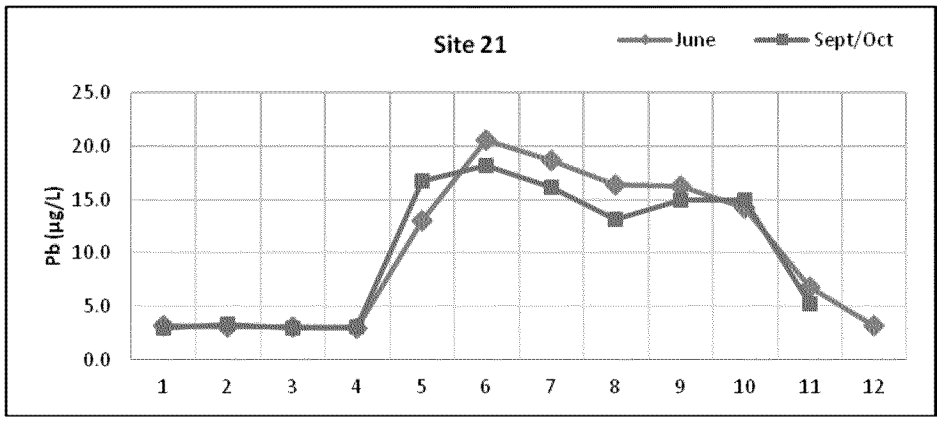
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

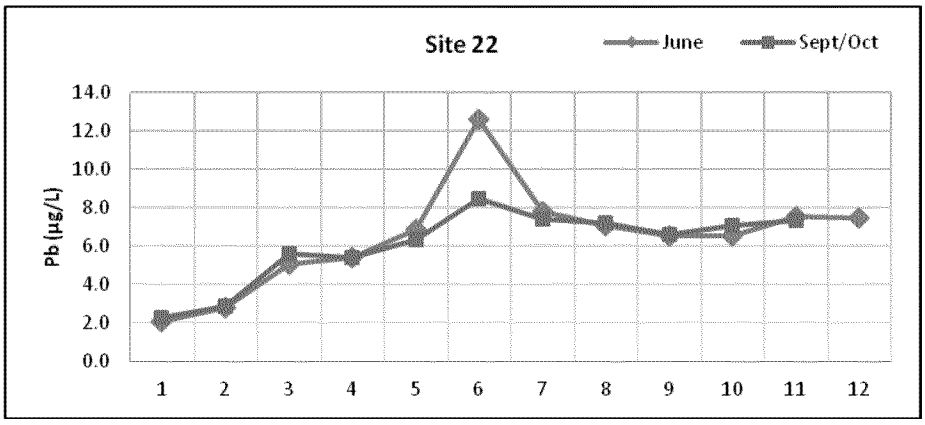
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

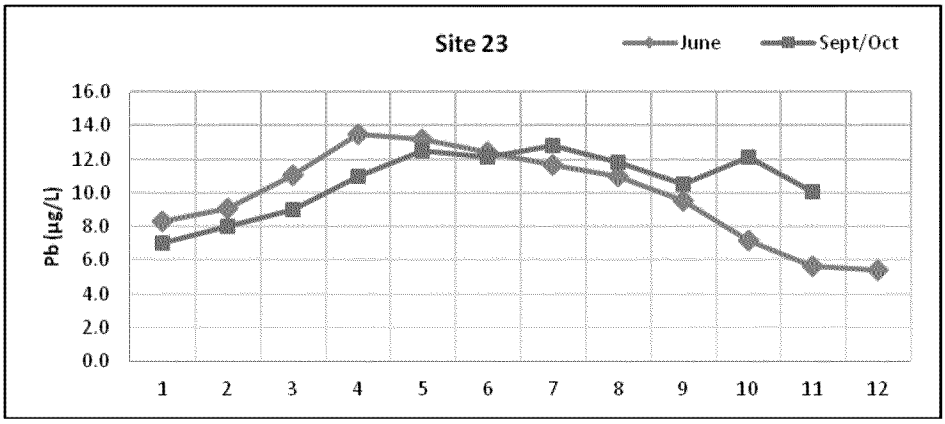
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

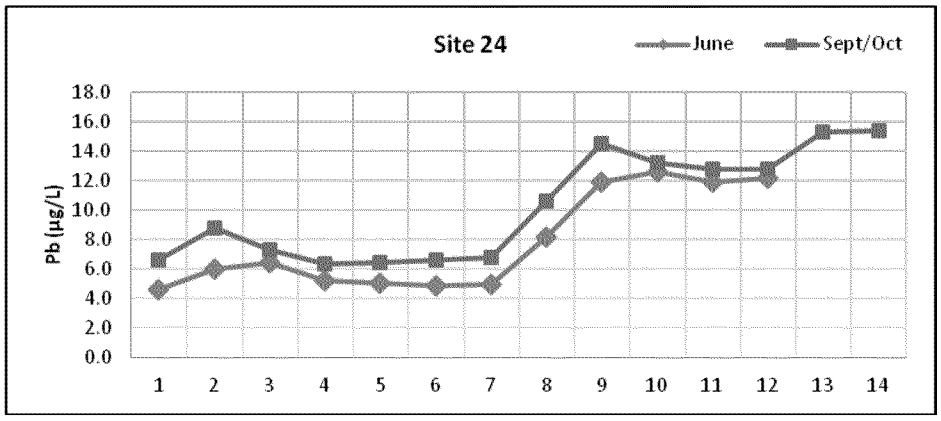
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

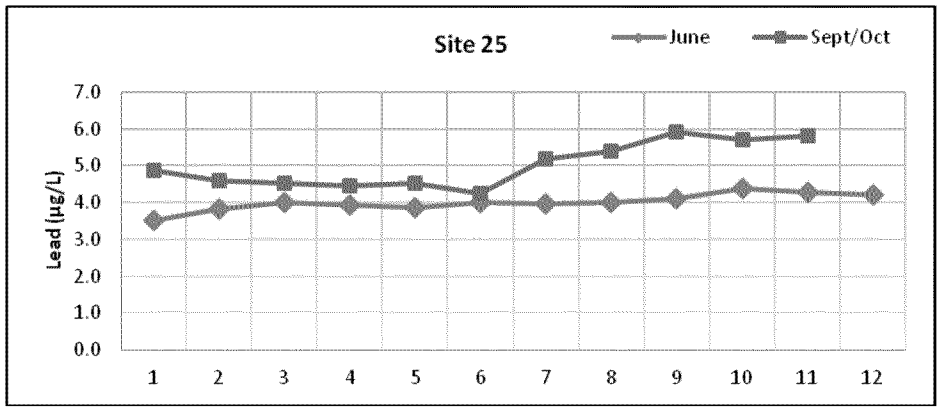
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

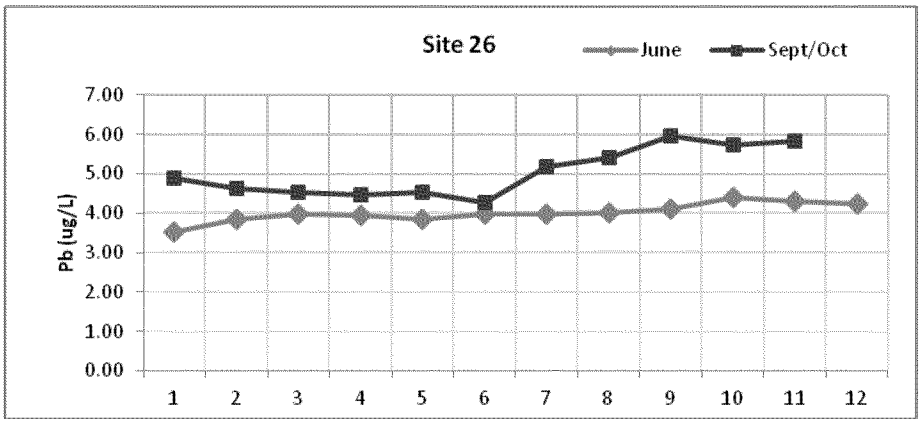
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

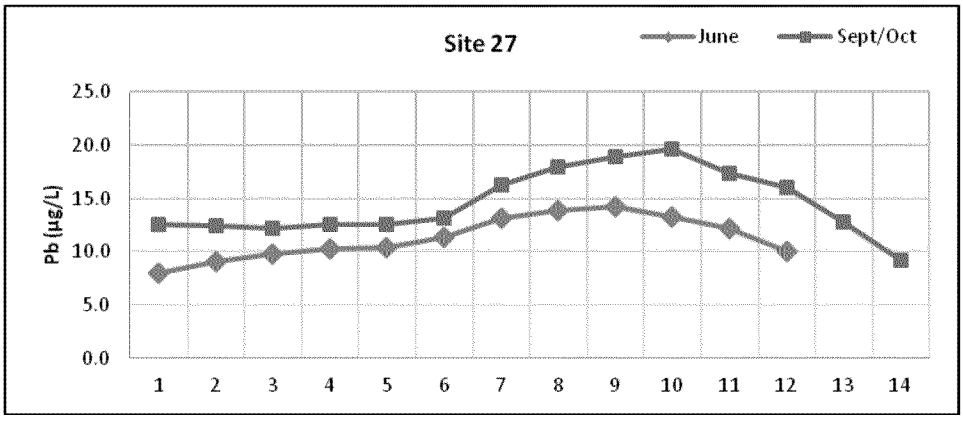
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

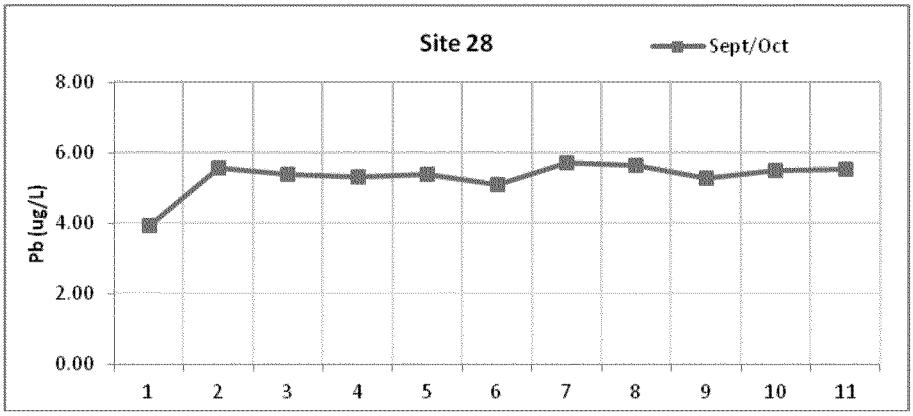
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

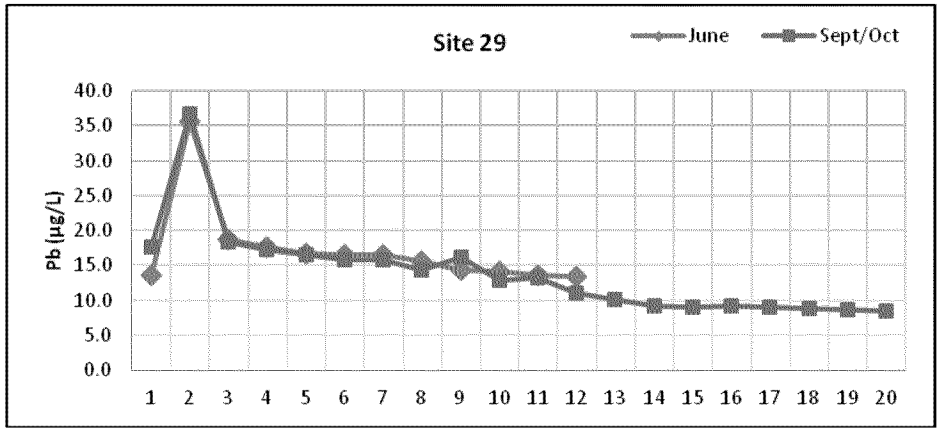
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

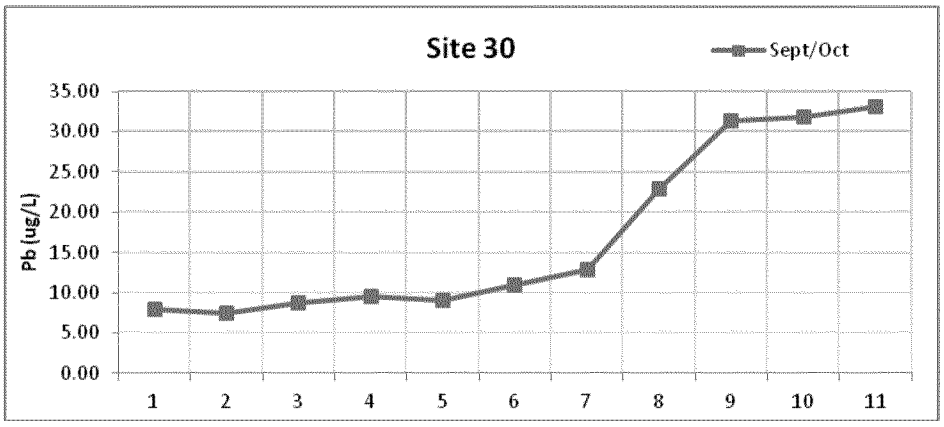
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

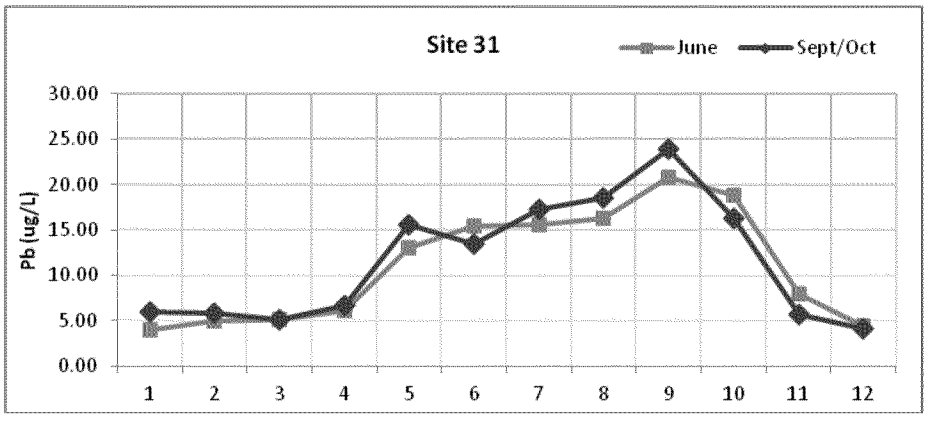
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

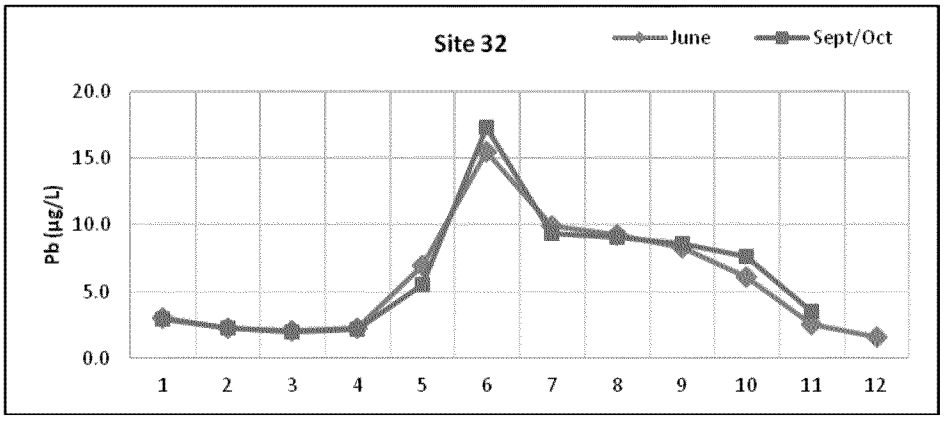
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

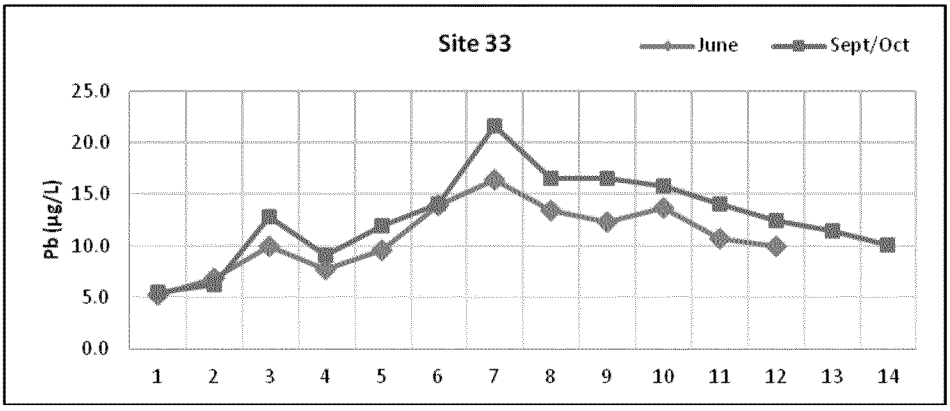
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

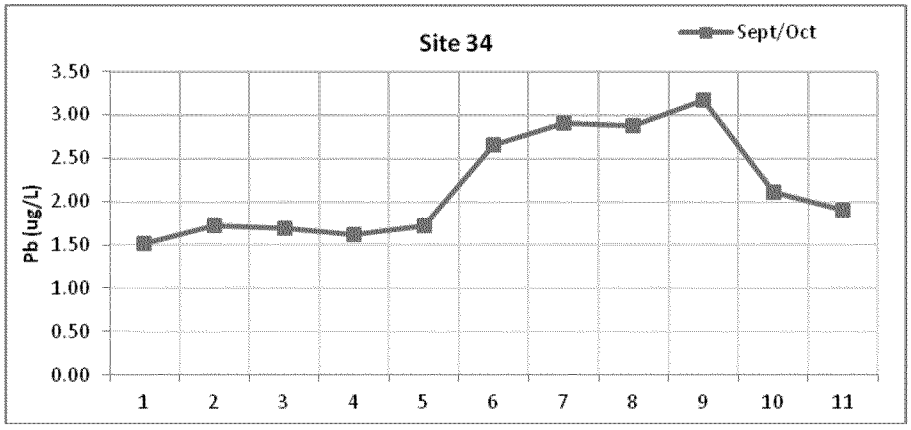
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

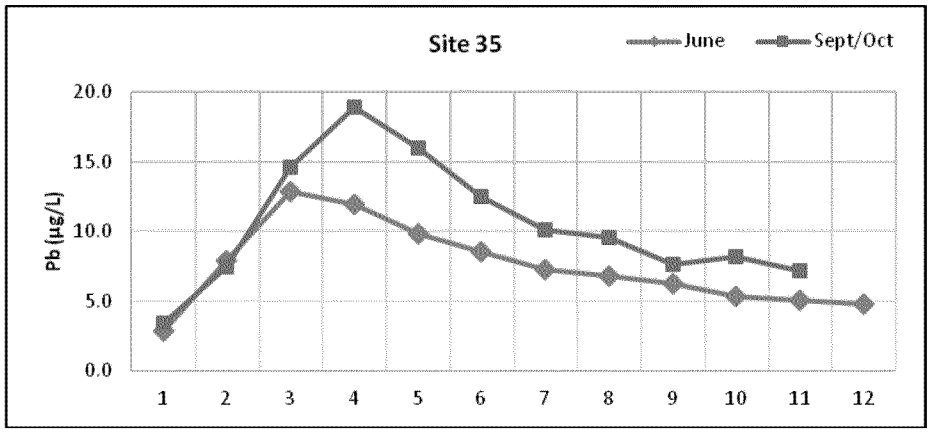
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

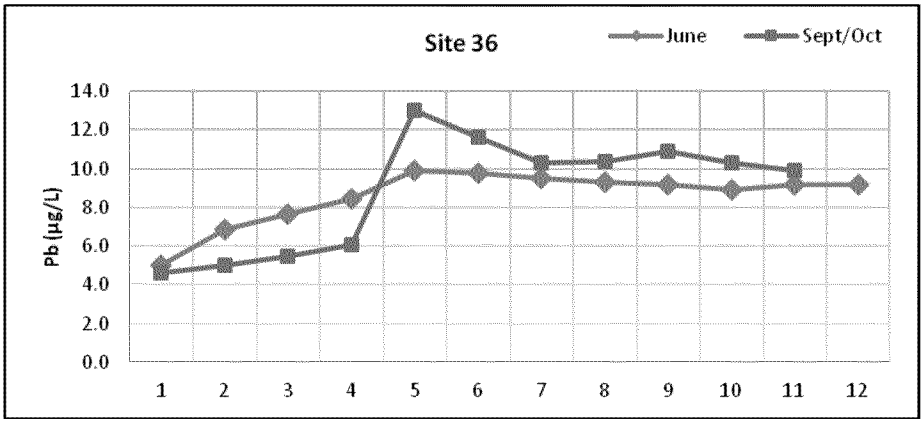
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

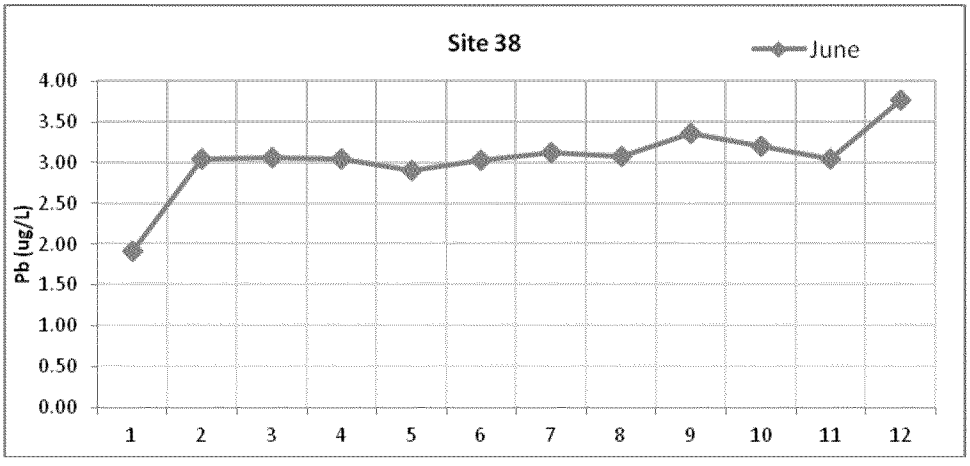
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

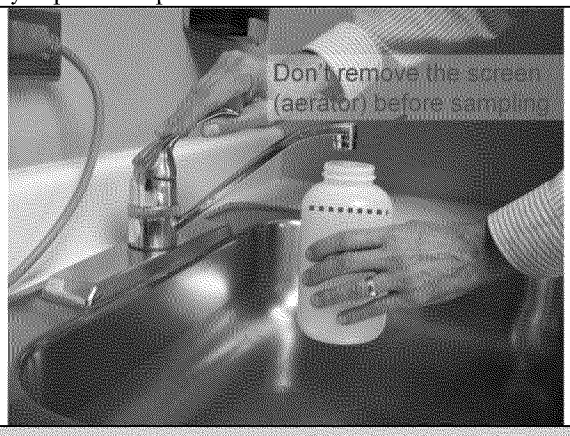
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

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2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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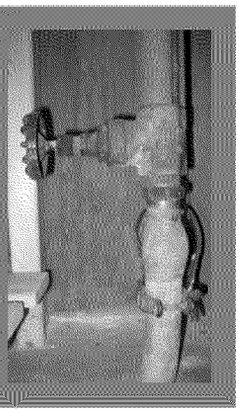
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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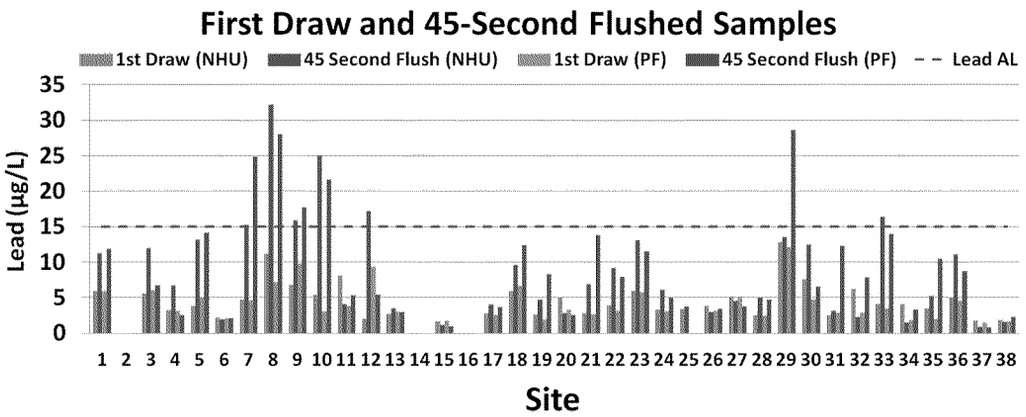

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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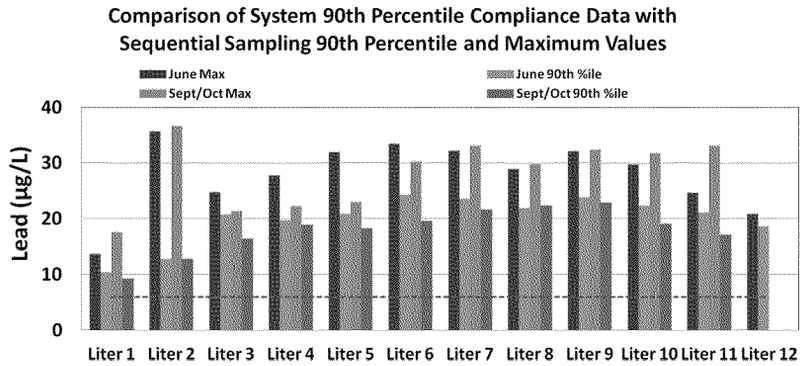

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

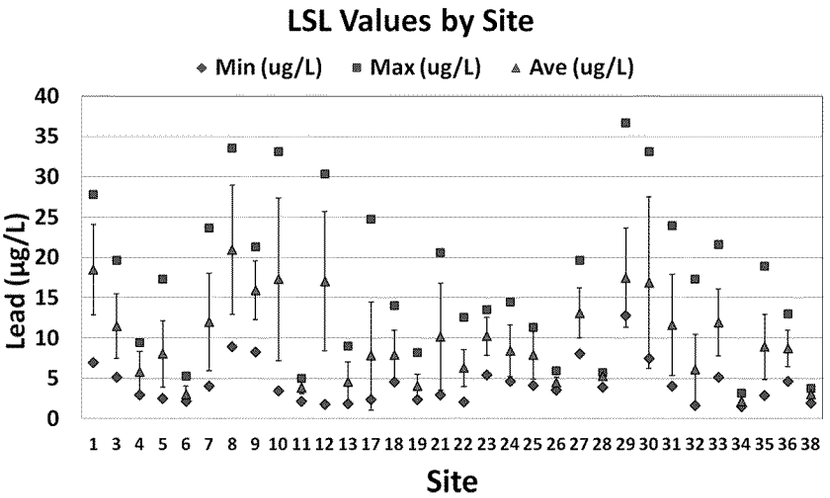

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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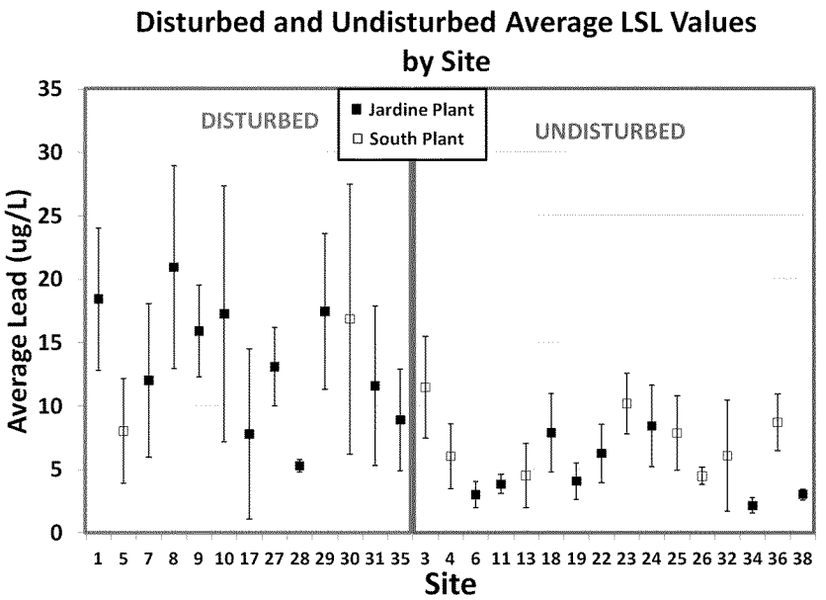

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Figure S2	Photograph of LSL Bulb	Table S2	Chicago Compliance Data
Figure S3	Photograph of LSL Segment	Table S3	Summary of Samples Collected
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Figure S5	Sample Site Map and Home Age	Table S4b	Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.
Figure S6	Graph of Four Metals for Site 9	Table S5	June Sequential Sampling Results
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Figure S8	City B LSL Results	Table S6b	Sept/Oct Supplemental Sequential Sampling Results (Not Used in Analyses)
Figure S9-S40	Sequential Sampling Graphs (Lead)	Table S6c	Summary of stagnation times.
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Figure S45	Sept/Oct Sample Collection and Reporting Form		

Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

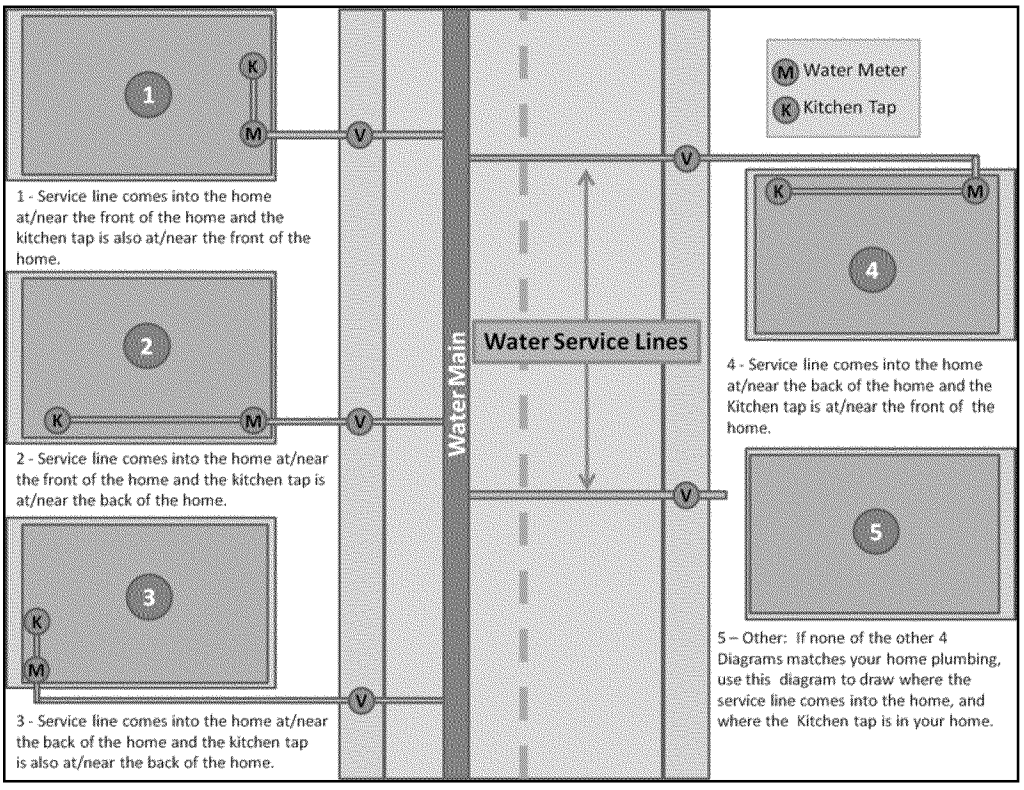


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

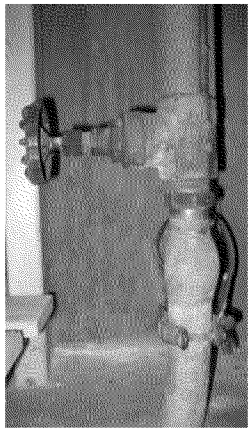


Figure S2: LSL Bulb

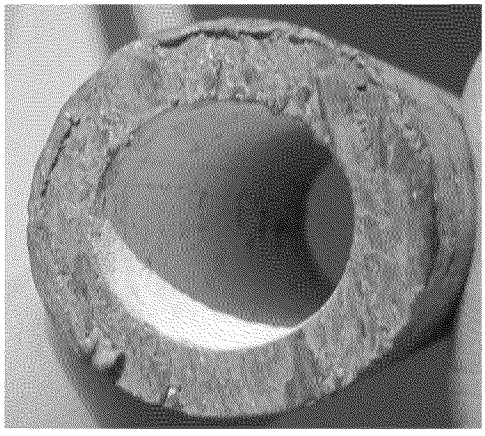


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

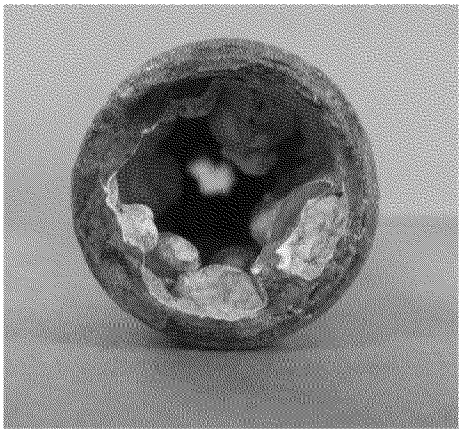


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

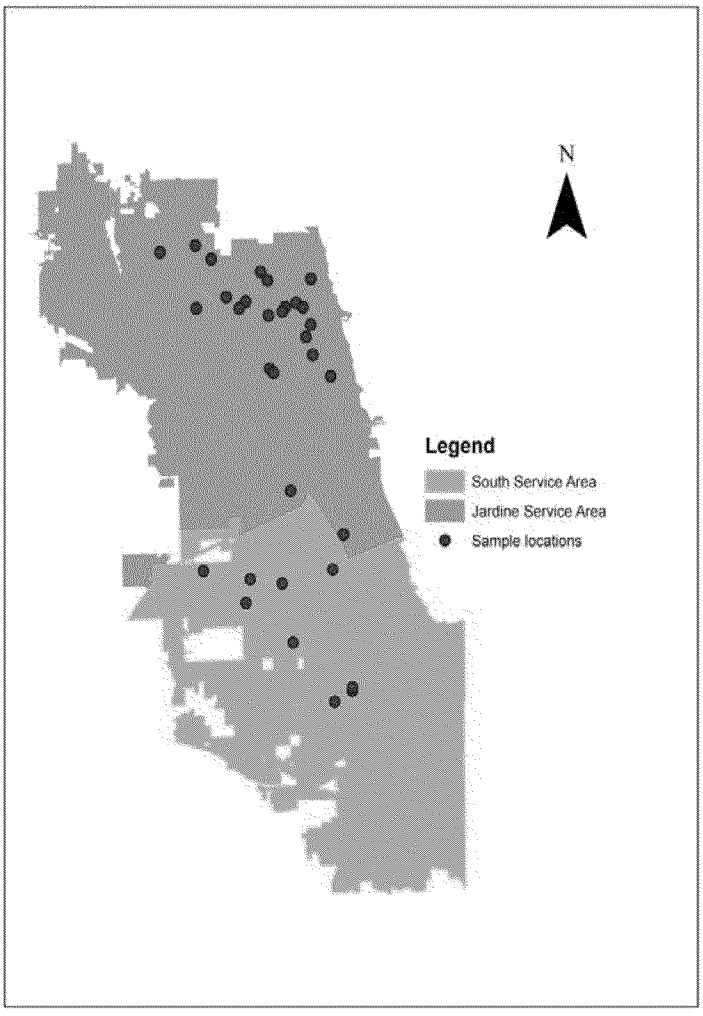


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

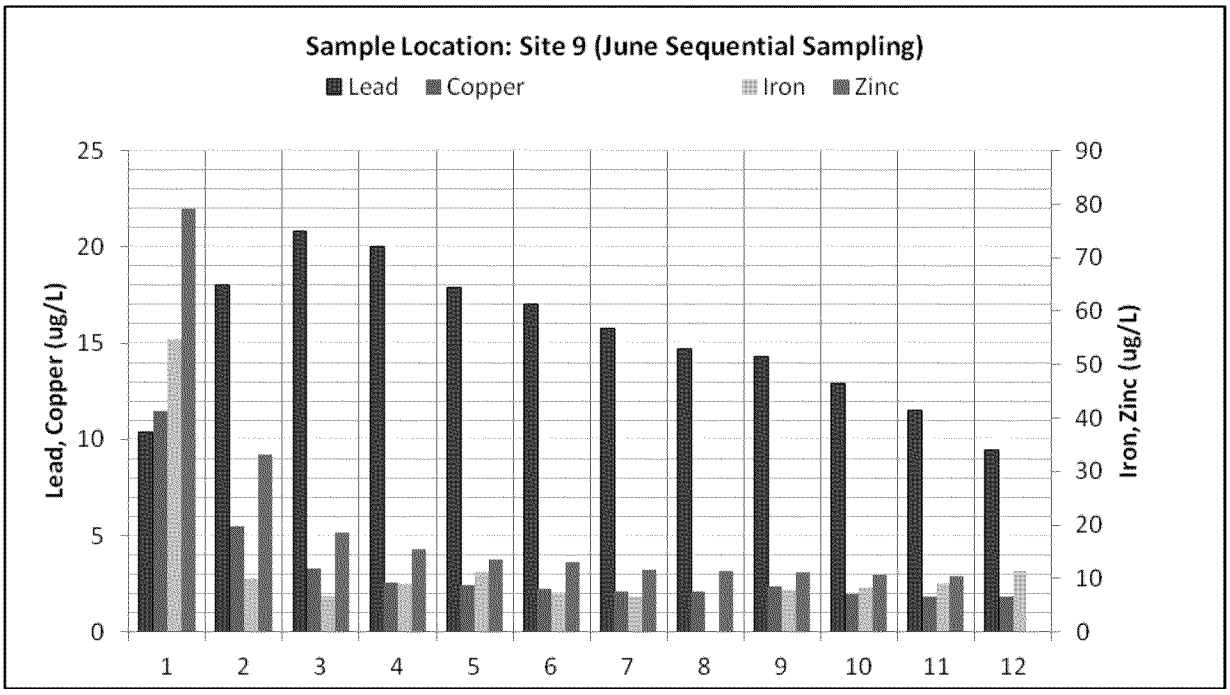


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

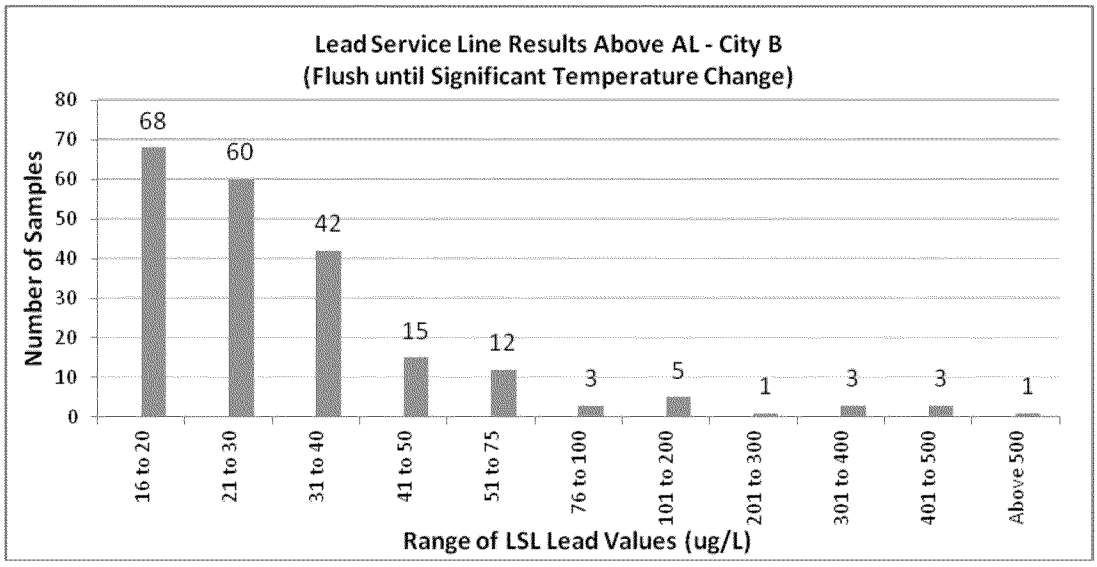
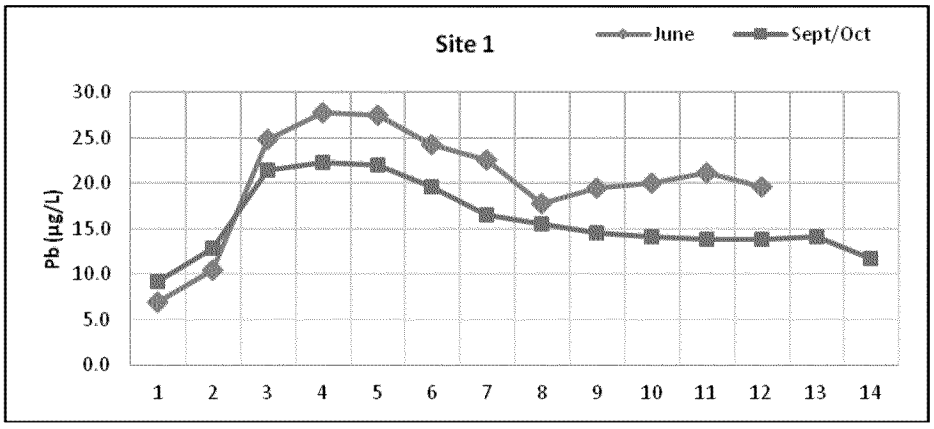


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

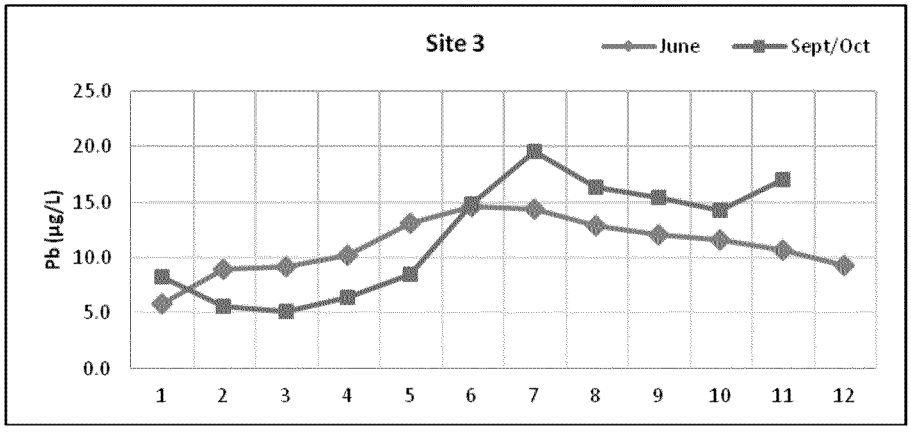
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

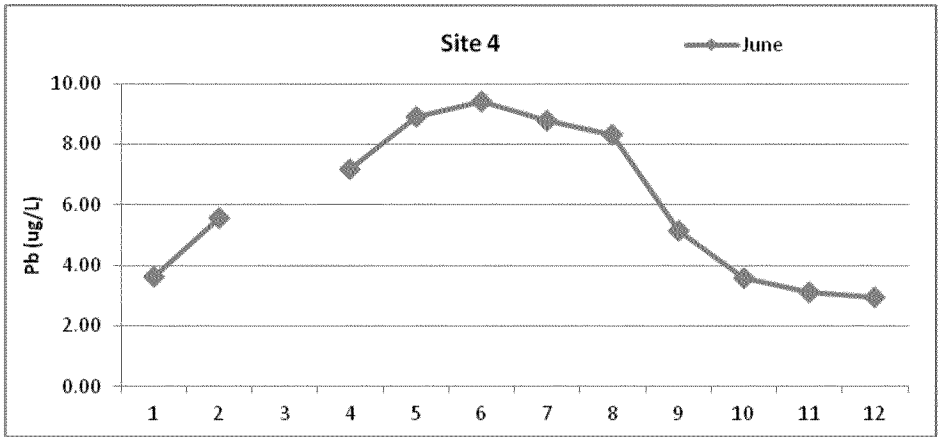
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

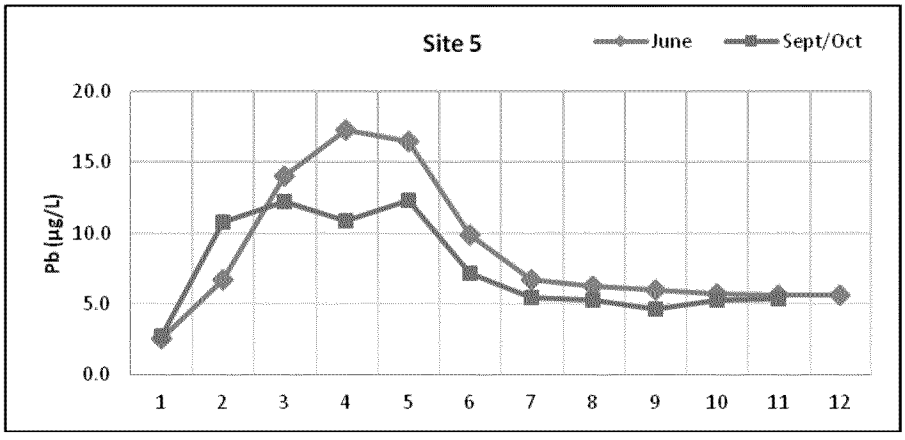
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

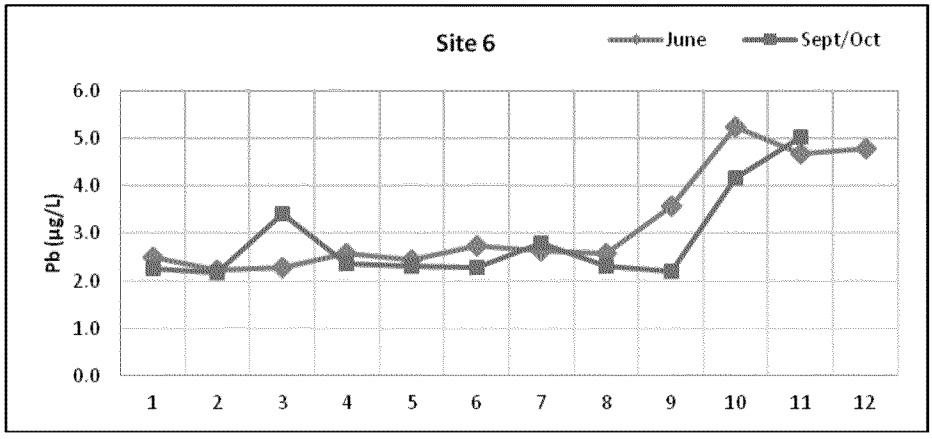
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

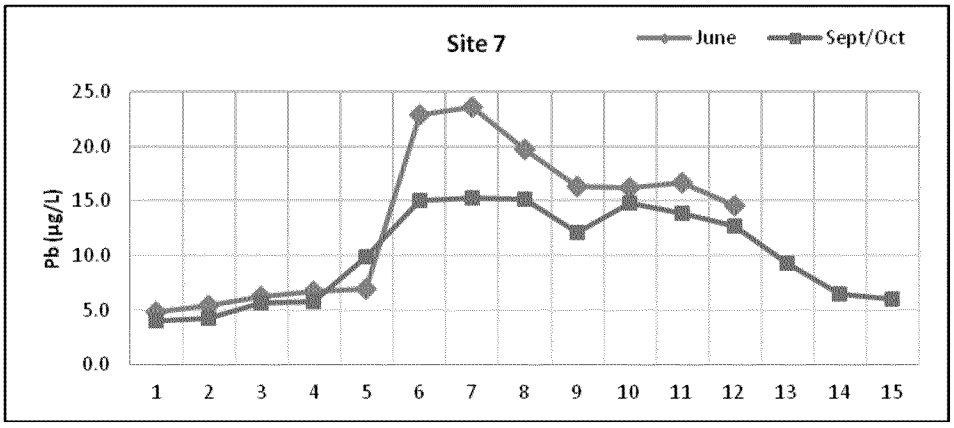
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

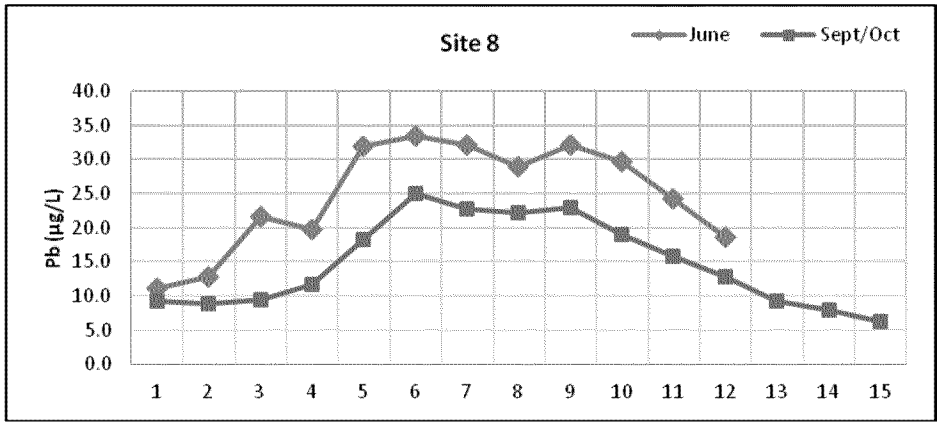
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

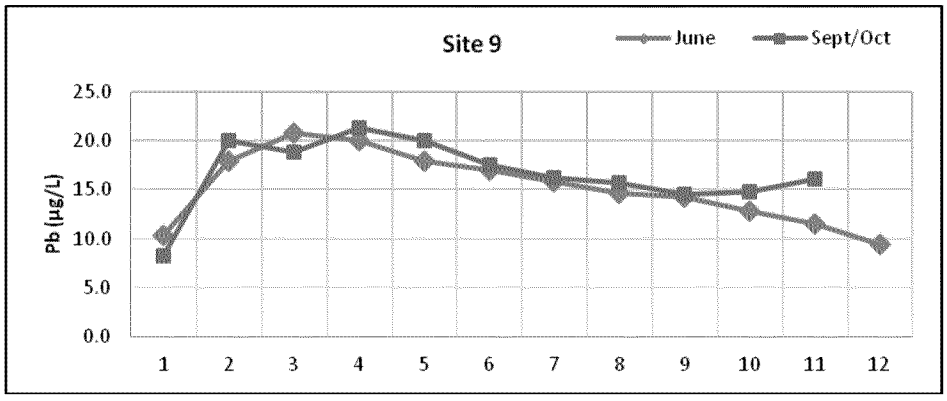
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

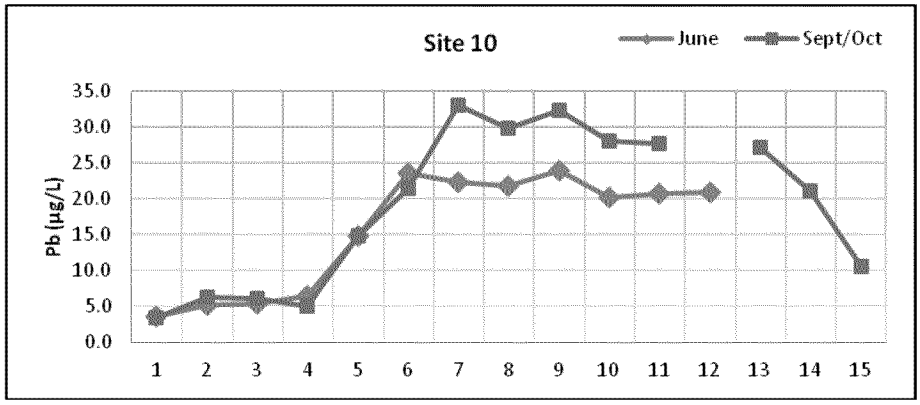
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

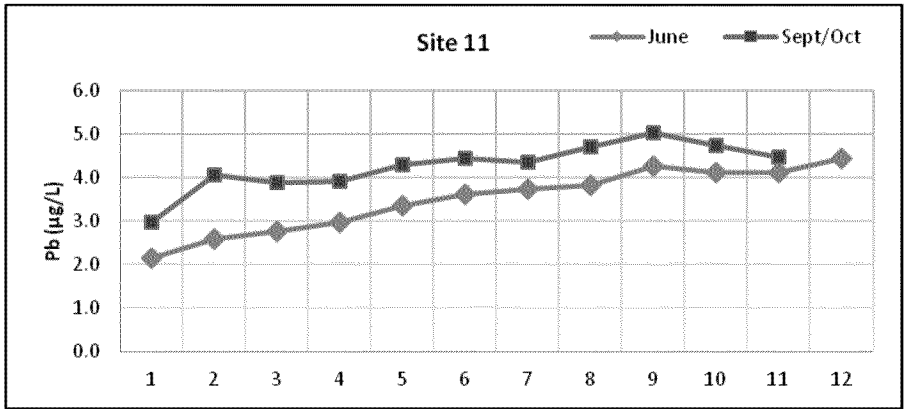
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

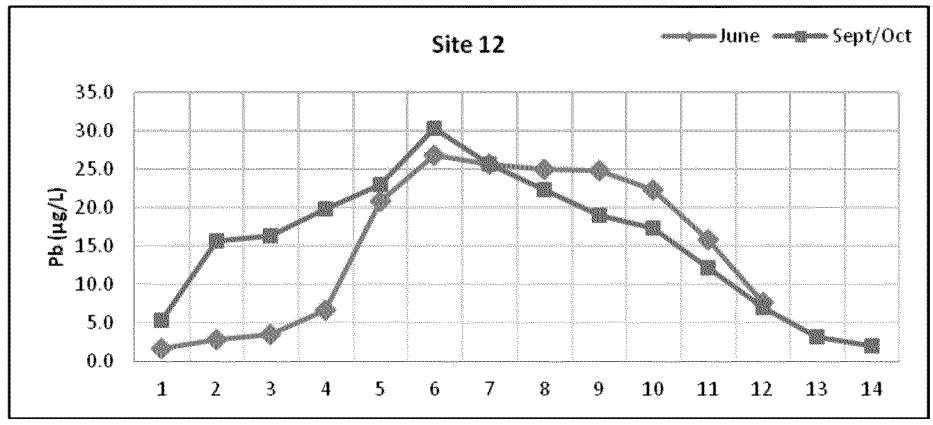
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

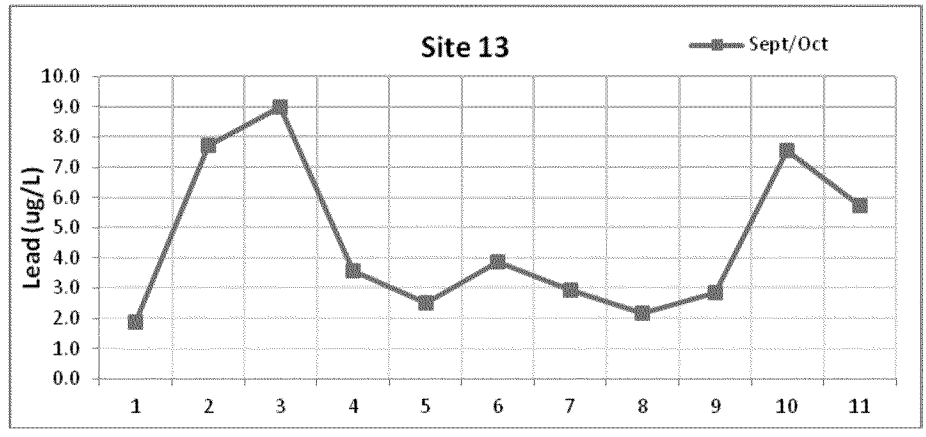
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

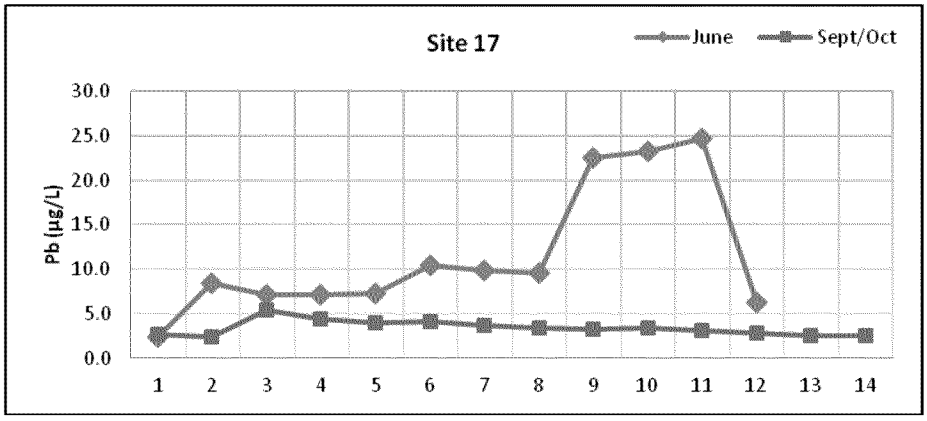
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

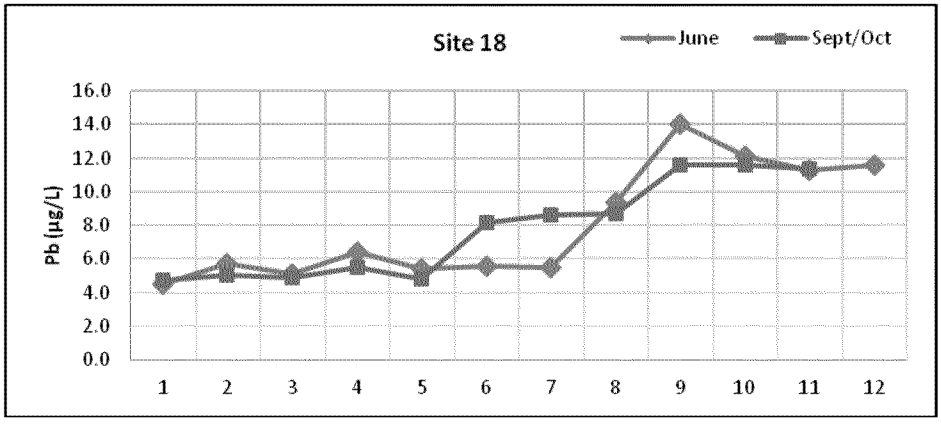
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

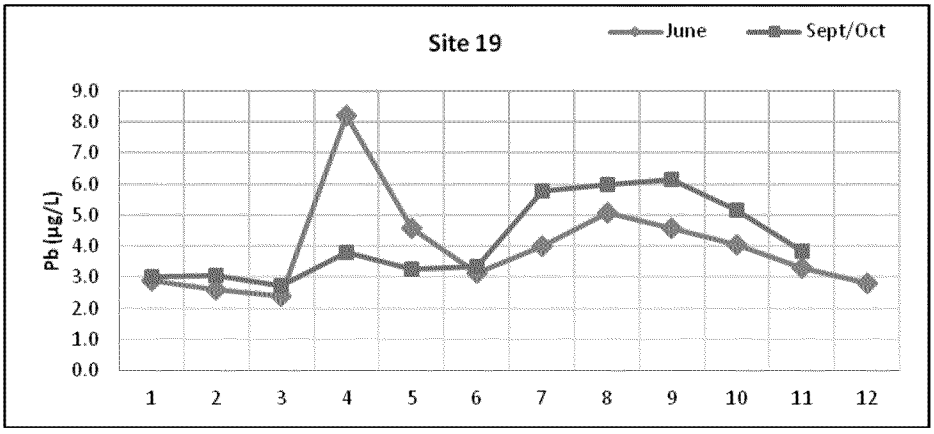
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

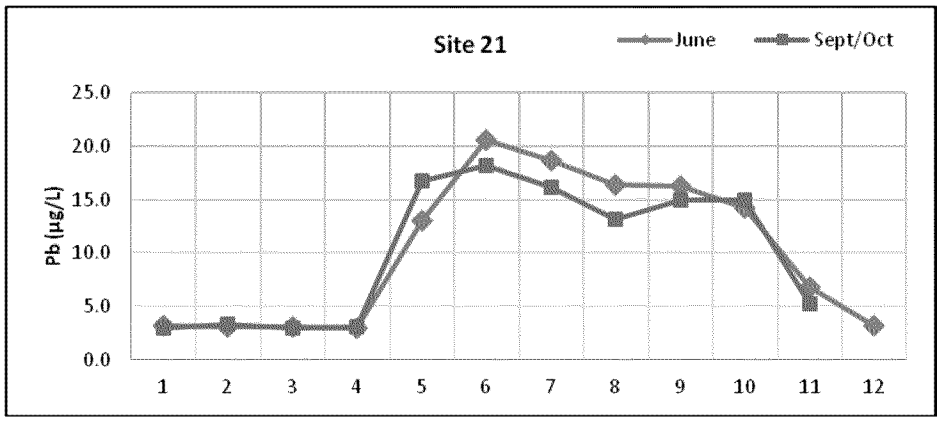
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

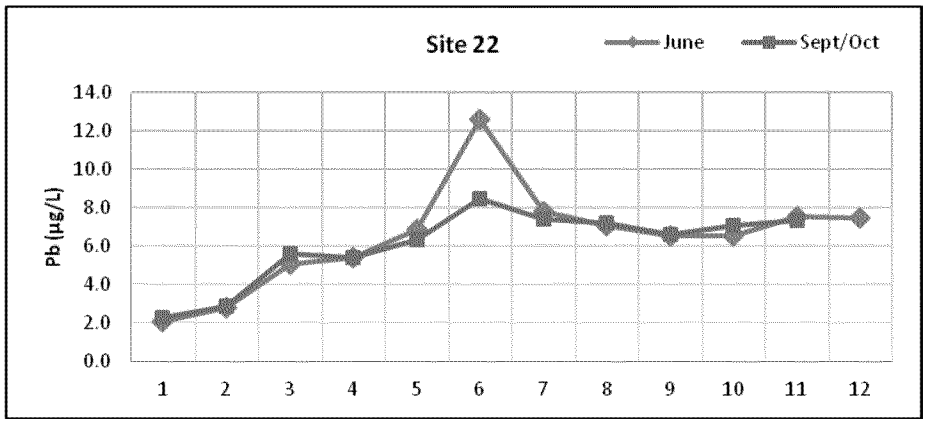
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

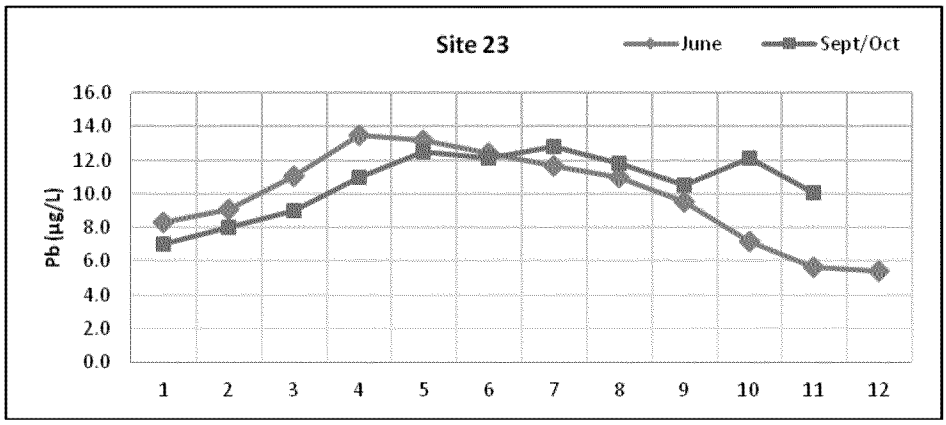
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

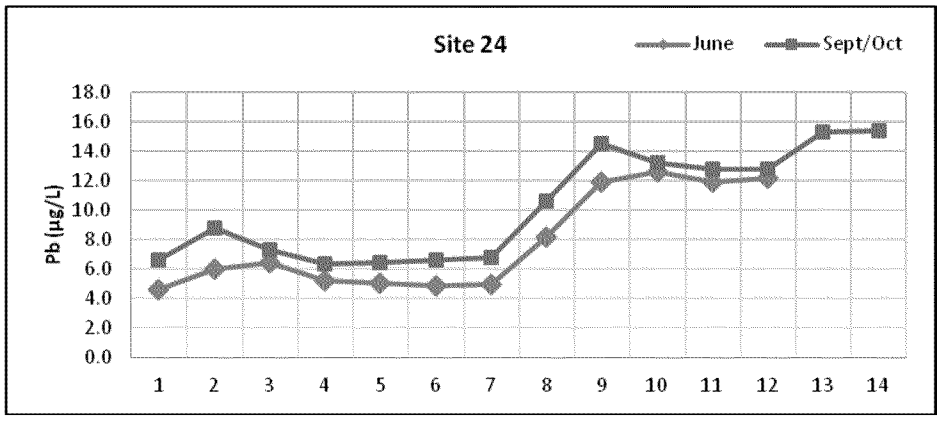
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

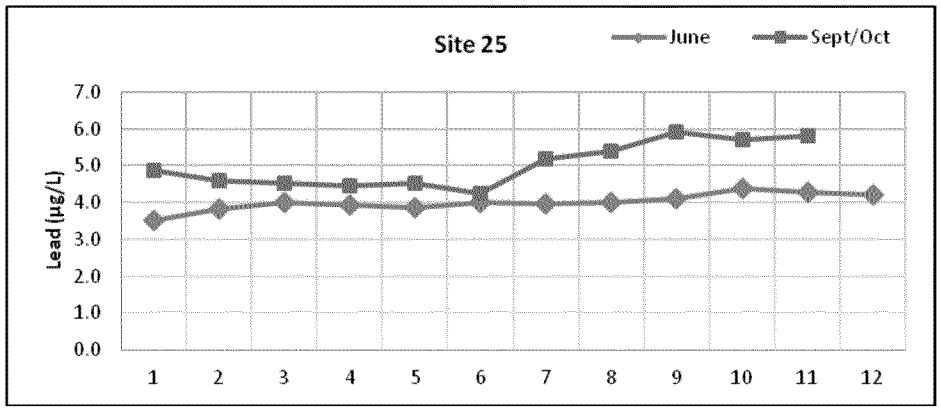
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

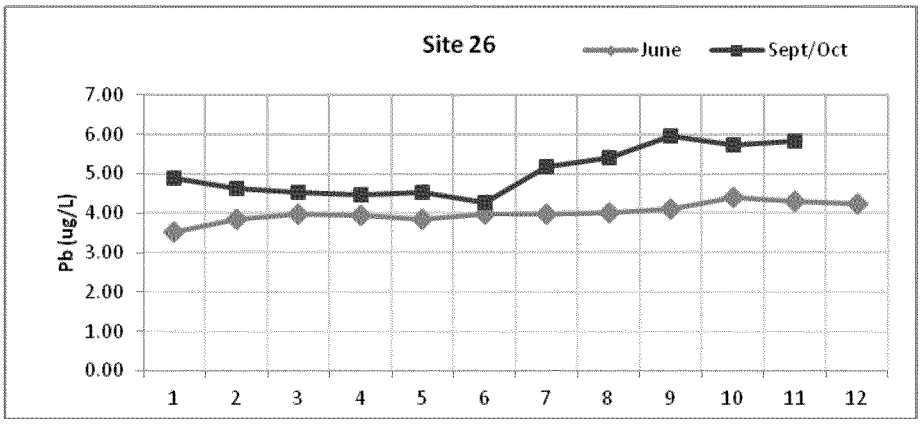
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

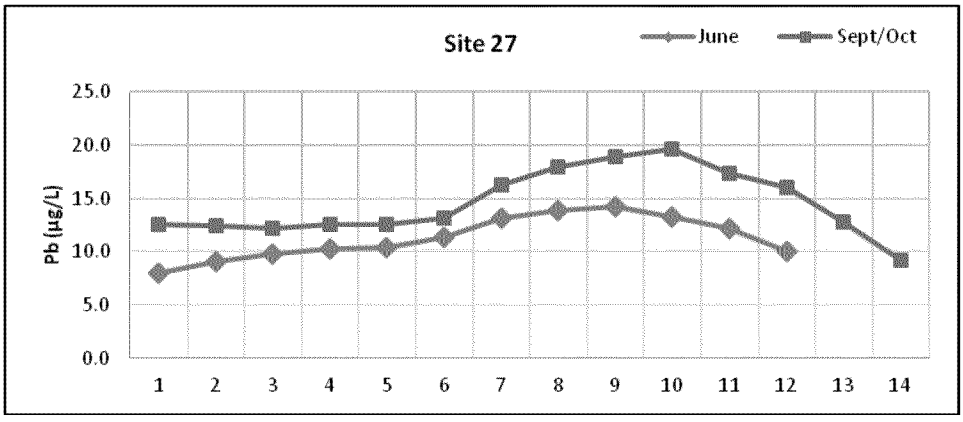
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

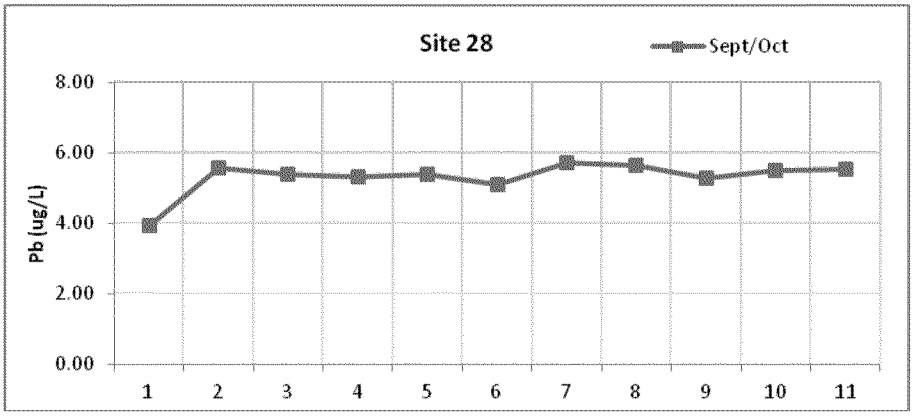
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

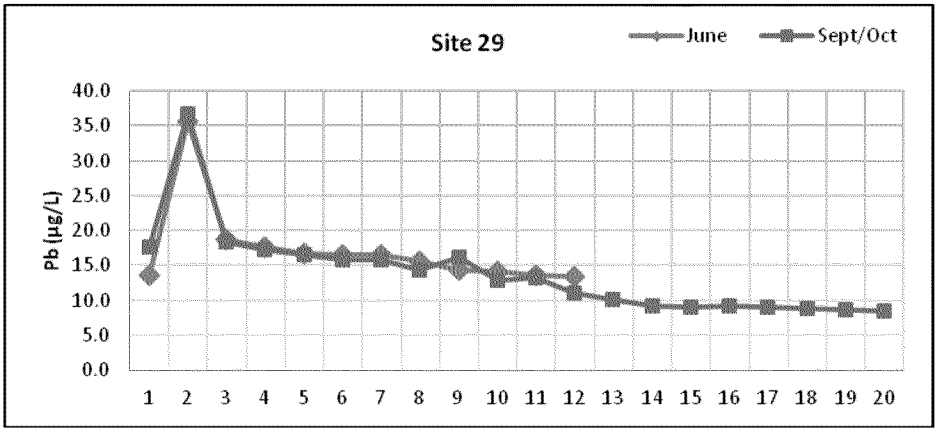
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

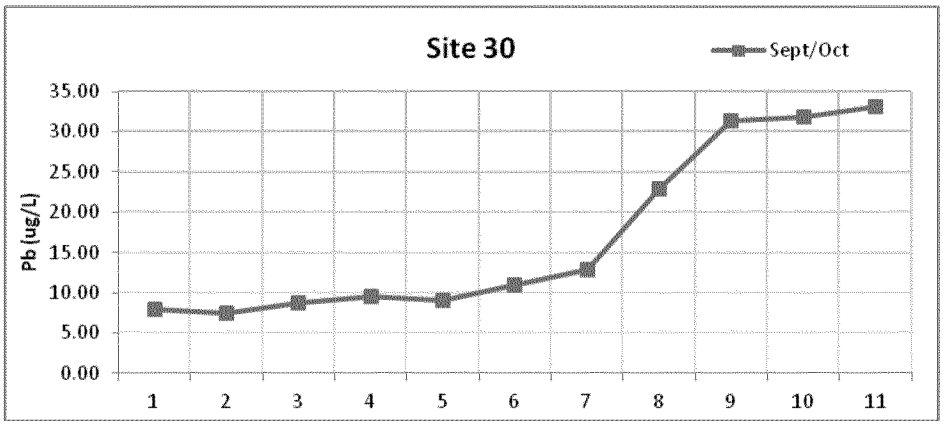
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

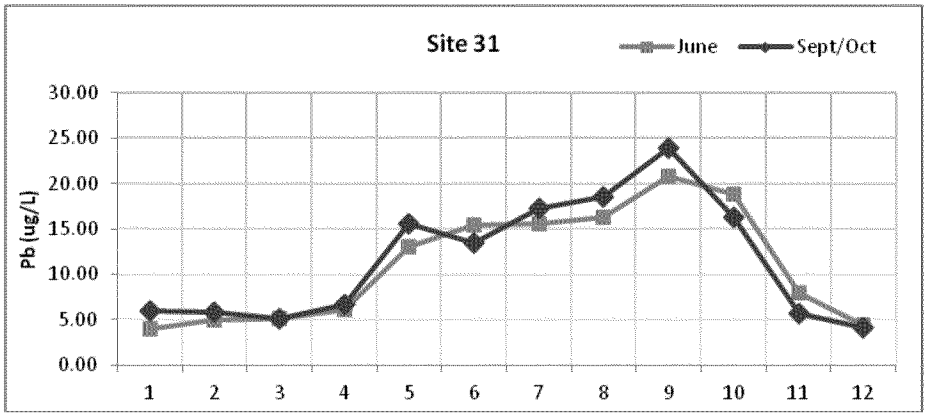
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

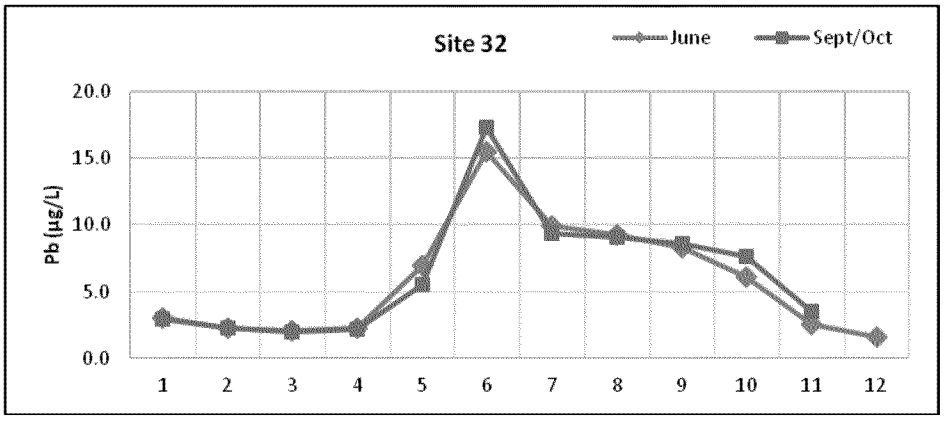
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

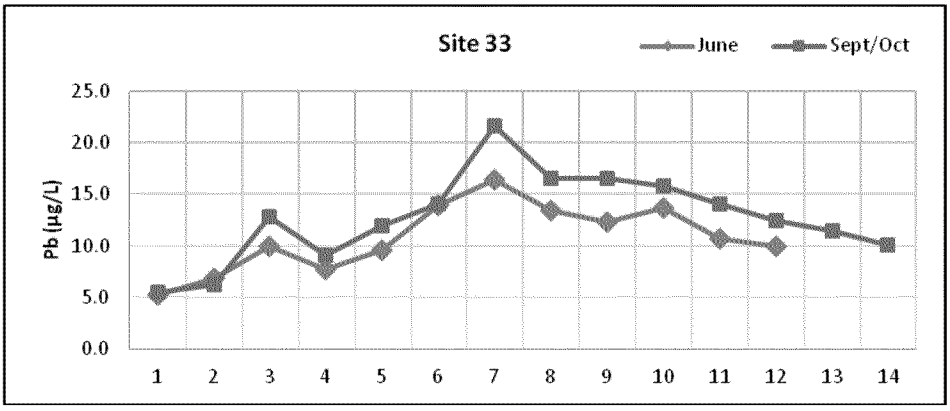
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

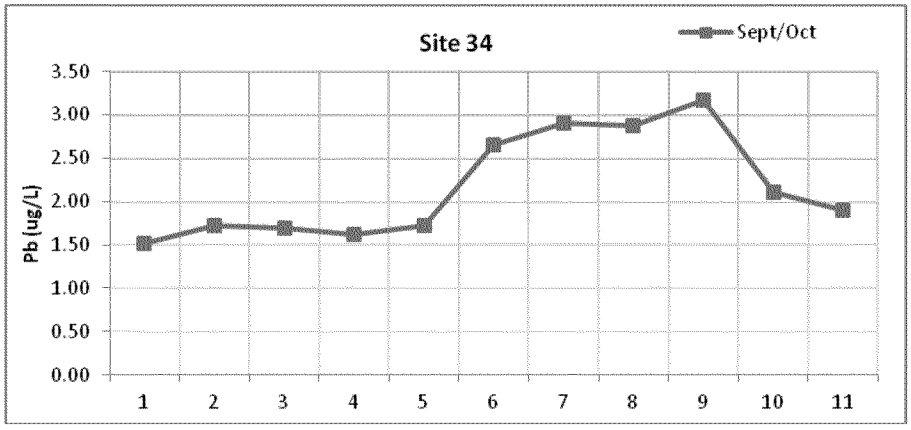
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

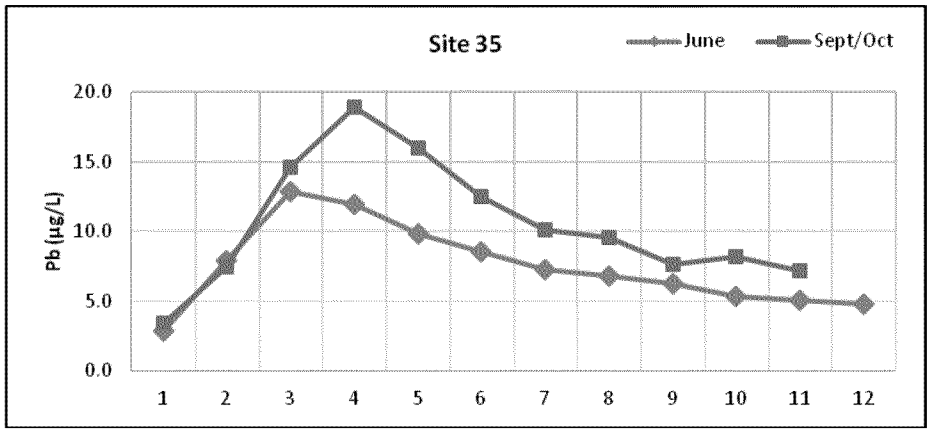
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

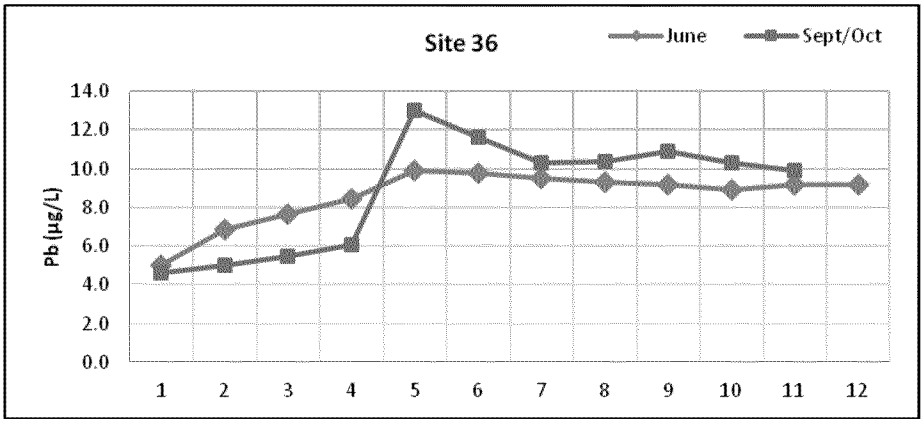
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

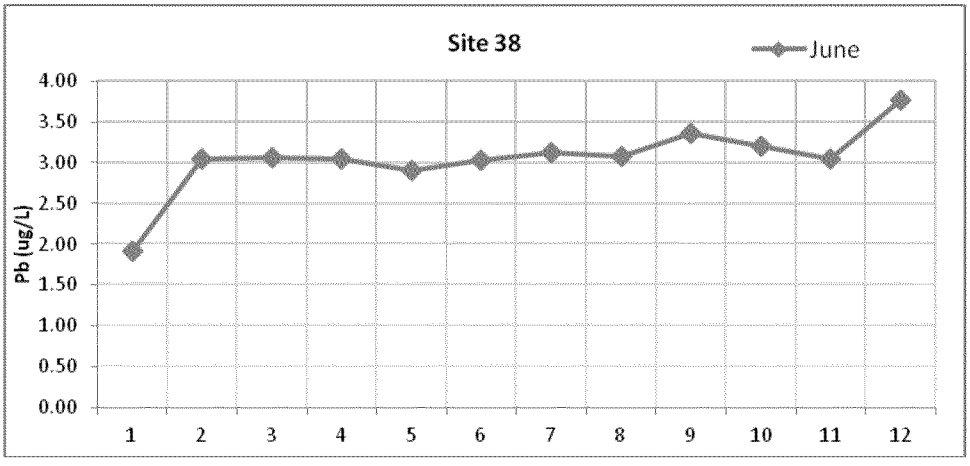
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

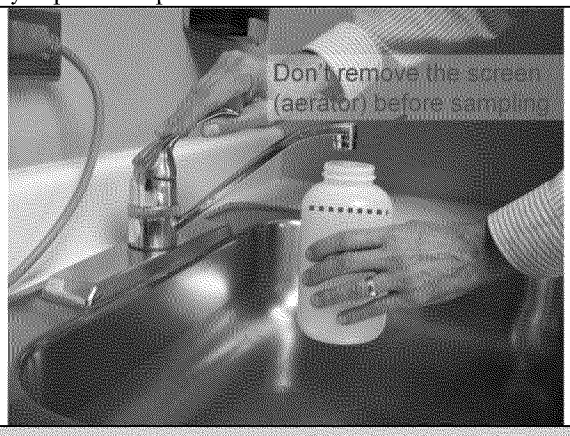
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

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EPA warns modernizing water system may boost levels of lead

Chicago study finds high levels of toxic metal in areas of street work or plumbing repairs

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A section of pipe is guided into place Friday as new water mains are installed along Thorndale Avenue at Glenwood Avenue on the city's North Side. An EPA study found that such projects can interfere with anti-lead protections in older pipes and boost levels of lead in drinking water. (Chris Walker, Chicago Tribune / September 20, 2013)

By Michael Hawthorne, Chicago Tribune reporter
September 25, 2013

Dangerous levels of lead are turning up in Chicago homes where pipes made of the toxic metal were disturbed by

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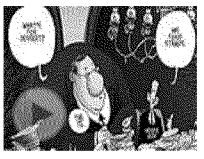
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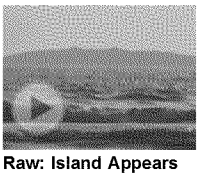
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street work or plumbing repairs, according to a new federal study that suggests the city's aggressive efforts to modernize its water system could inadvertently pose health risks.

The problem starts with lead service lines that Chicago installed across the city until the mid-1980s to connect water mains with homes. Researchers at the U.S. Environmental Protection Agency found that spikes of lead can leach into tap water when those pipes are altered by water main replacements, meter installations or street work.

Wall Chart
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High levels can be found in tap water for years afterward, the EPA study found, raising concerns that other cities with lead pipes could face similar problems.

Most homeowners likely are unaware they could be drinking tainted water. Under federal rules, utilities rarely are required to warn residents that work is being done or tell them they can take steps to reduce their exposure to lead. A potent neurotoxin, lead can damage the brains of young children, lower IQ and trigger learning disabilities, aggression and criminal behavior later in life.

Lead is so hazardous that the EPA and the Centers for Disease Control and Prevention say there is no safe level of exposure. The metal has been phased out of gasoline, removed from paint and banned in children's toys. But the widespread use of lead pipes during the last century has left a festering problem nationwide.

"We owe it to people to tell them that their water might not be safe to drink," said Marc Edwards, a professor of civil and environmental engineering at Virginia Tech University who wasn't involved with the EPA study but has reached similar conclusions in his own research.

The new study comes as Mayor Rahm Emanuel pushes to speed up the replacement of aging water mains and increase the number of water meters installed citywide — two activities that EPA researchers found to be linked to high amounts of lead in tap water.

Lead levels exceeded federal standards at all but one of 13 Chicago homes tested where the service line had been physically disturbed between 2005 and 2011, according to results published last month in the peer-reviewed journal Environmental Science & Technology.

At 16 other homes where there was no known disturbance of the service line, only three had high lead levels.

The study also found that normal federal testing procedures would not have detected most of these spikes, a problem the Tribune described in a 2011 story based on preliminary data. Under the EPA's current testing protocol, only the first liter of water that flows out of a household faucet is checked, but the researchers took multiple consecutive samples.

Their testing showed that lead levels exceeded the federal limit of 15 parts per billion at 15 of 29 homes used in the study, but only one home showed high levels in the first liter of water drawn. The high levels didn't taper off until the 12th liter drawn from the taps, according to the researchers. Clearing pipes of that much water requires a continuous flow for up to five minutes, they said.

In response to the research, the city in July started advising residents to flush their pipes after a water main has been replaced on their street. "We're trying to be proactive," said Andrea Putz, a research specialist at the Chicago Department of Water Management.

The city's warning, which appears at the end of a packet of information handed out by work crews, says opening all household taps for three to five minutes will remove "any lead particulates that may have come loose from your property's service line as a result of the water main replacement."

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But the EPA says it is important for households with lead service lines to flush their pipes for several minutes any time household water hasn't been used for several hours — not just one time after a repair.

Two simple ways to do that are to take a shower or do a load of laundry. Then, before drawing water to drink or to prepare baby formula, the tap should be flushed for another 35 to 45 seconds to clear any remaining water sitting in the home's pipes, according to the EPA's recommendations.

Some household devices can filter out lead. NSF International provides a list of certified devices on its website under the heading "Consumer Resources."

Tinka Hyde, director of the water division in the EPA's Chicago office, said the study could influence a long-running debate about the Lead and Copper Rule, a 1991 regulation that by many accounts fails to adequately protect Americans from the harmful effects of lead in water.

Chicago has a sprawling network of pipes laid before the harmful effects of lead were fully accepted. The city's building code required lead service lines as recently as 1986. Yet Chicago has not recorded a violation of the Lead and Copper Rule for nearly 20 years. Nor have most other cities.

Under the rule's testing protocols, cities aren't required to take action unless water in more than 10 percent of the homes tested have lead levels exceeding 15 parts per billion, a limit that wasn't based on science about lead hazards but rather was considered cost-effective for water utilities to meet.

Water rarely contains lead when it leaves treatment plants, but under certain conditions the metal can leach into water while it sits in or flows through pipes and taps. Pipes and faucets inside homes also can contain lead, as can solder and brass plumbing devices.

Treatment plants add orthophosphate and other chemicals to form a white coating inside pipes that can prevent lead from leaching into the water. But street work and plumbing repairs can interfere with those protective measures.

Soon after becoming mayor, Emanuel won approval for a series of water rate increases to replace leaky pipes that in many cases are decades old. About 75 miles of new water mains are expected to be laid this year, up from 30 in 2010, according to city records. The city also plans to install 12,000 new meters this year to encourage residents to save water. But such fixes generally leave in place the lead service pipes that often connect water mains with homes.

Scores of other cities are embarking on similar efforts to overhaul aging water systems, a task the American Water Works Association estimates could cost at least $1 trillion nationwide during the next quarter-century.

The EPA study is part of growing amount of research suggesting that the work, while much-needed to stem leaks and conserve water, could have unintended consequences.

One of the challenges for regulators is finding solutions that are broadly effective. Water from two homes on the same block could have widely different lead levels depending on the type of service lines, the amount of lead in brass plumbing fittings and the extent of repair work.

"There is no magic bullet," said Putz, the city water specialist. "Everybody's plumbing is different, and that can have a big effect on lead levels."

City officials who assisted the EPA in its research plan to conduct their own study, testing lead levels in more than two dozen homes before and after water main replacements and

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meter installations, Putz said.

The presence of so many lead service lines across Chicago and other cities remains a concern. So does the staggering cost to eliminate the health risks.

"It's so huge that utilities and the states don't want to talk about it unless there are federal funds to fix it," said Bruce Lanphear, a noted lead expert at Simon Fraser University in Vancouver, British Columbia, who served on an EPA scientific advisory panel that studied the issue. "But if we don't start to do something, we're sacrificing another generation of children to the hazards of lead."

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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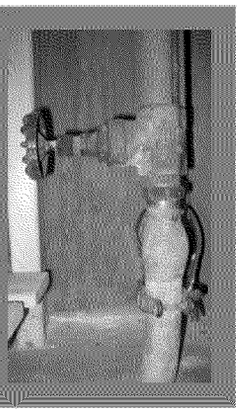
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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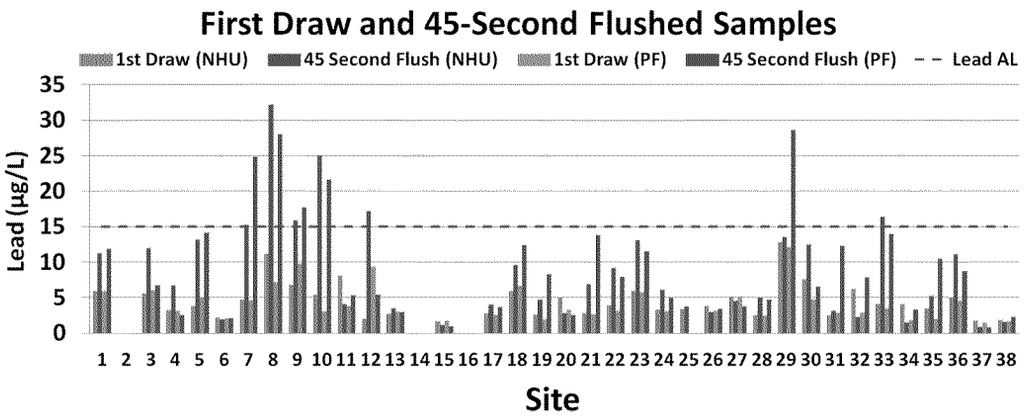

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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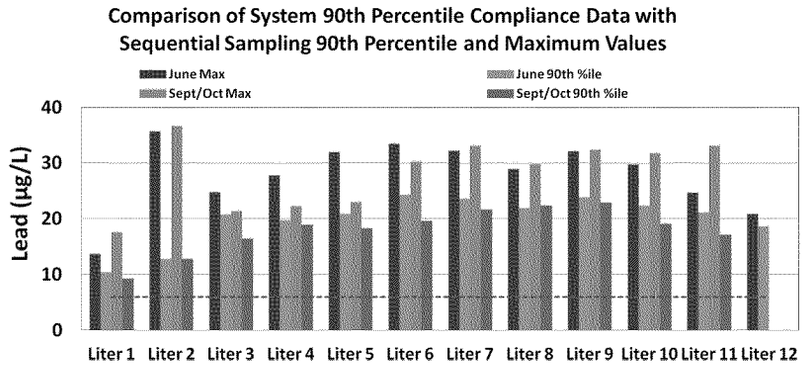

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

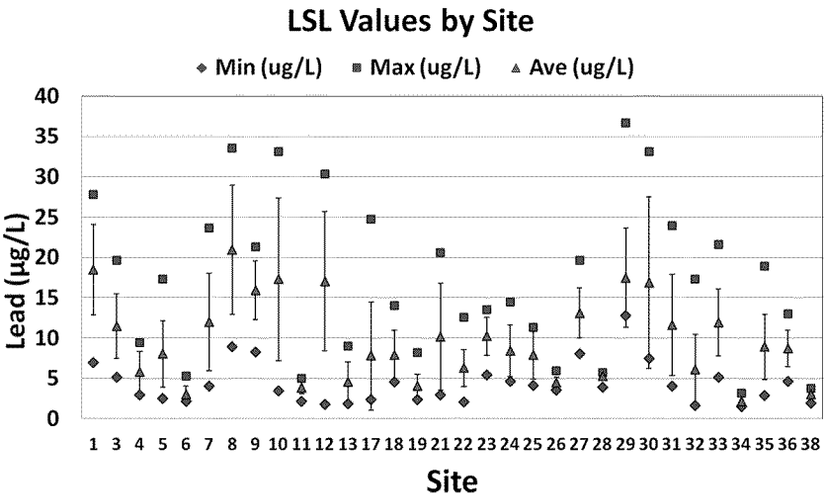

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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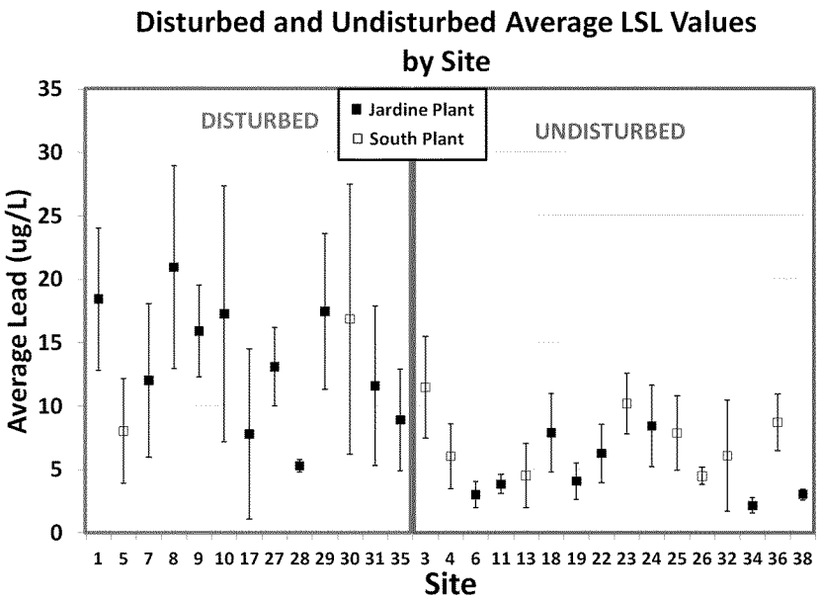

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

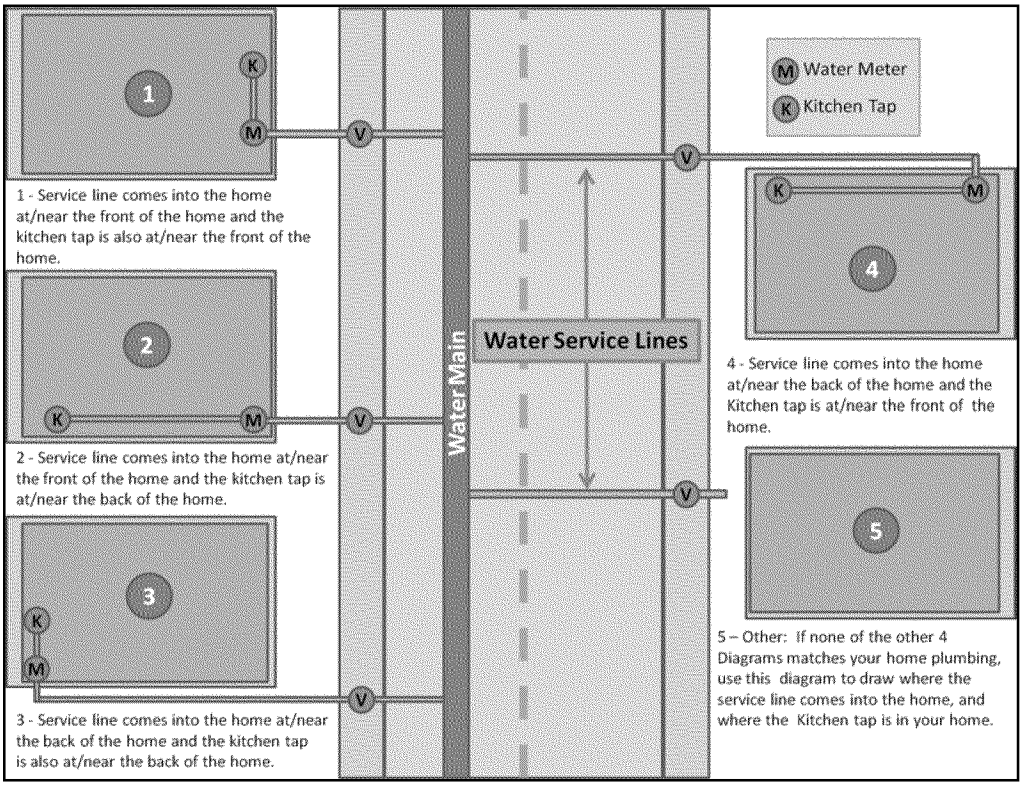


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

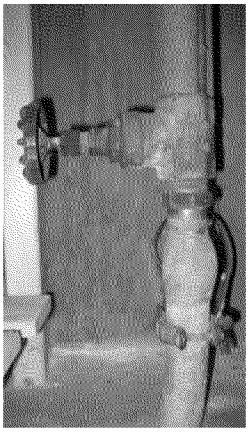


Figure S2: LSL Bulb

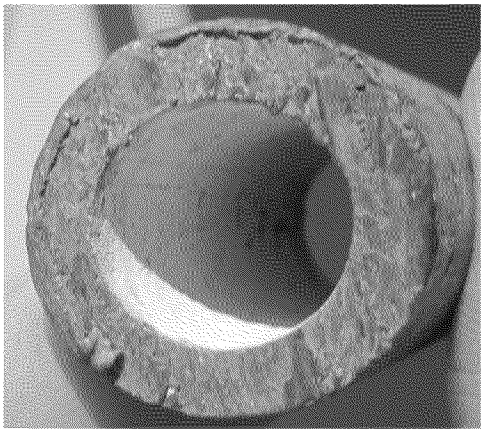


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

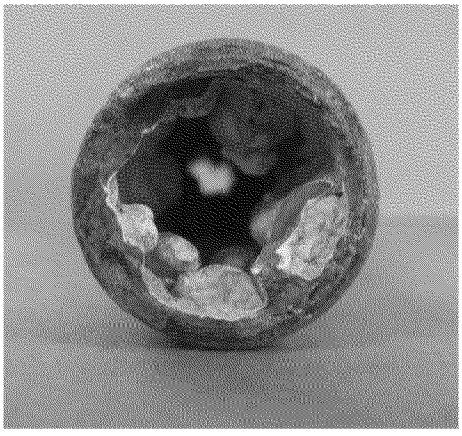


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

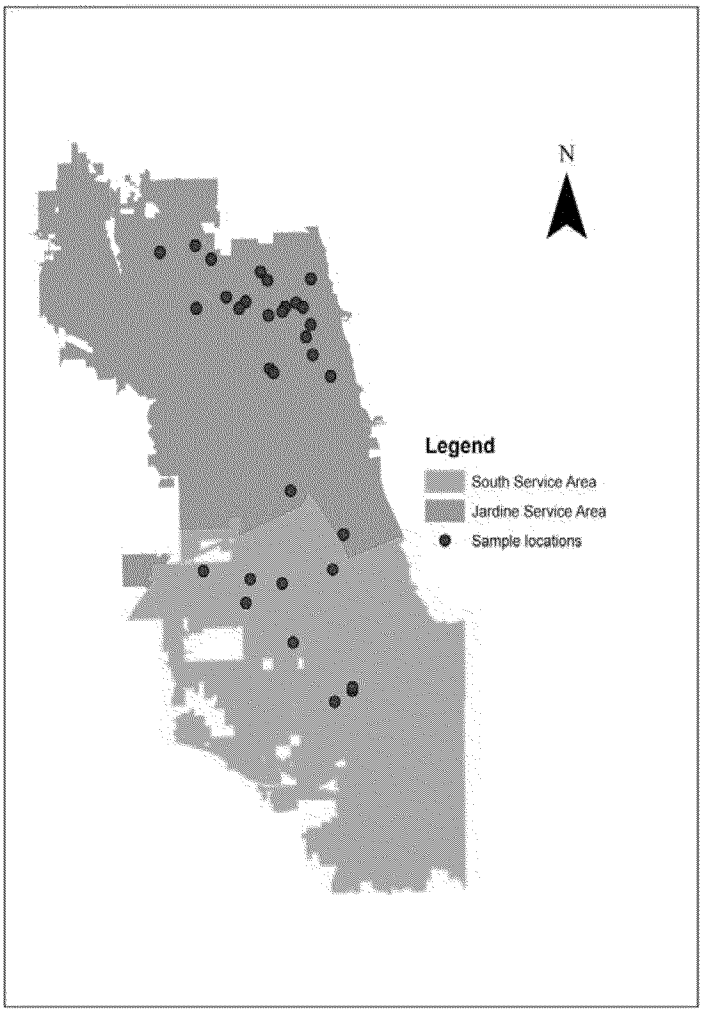


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

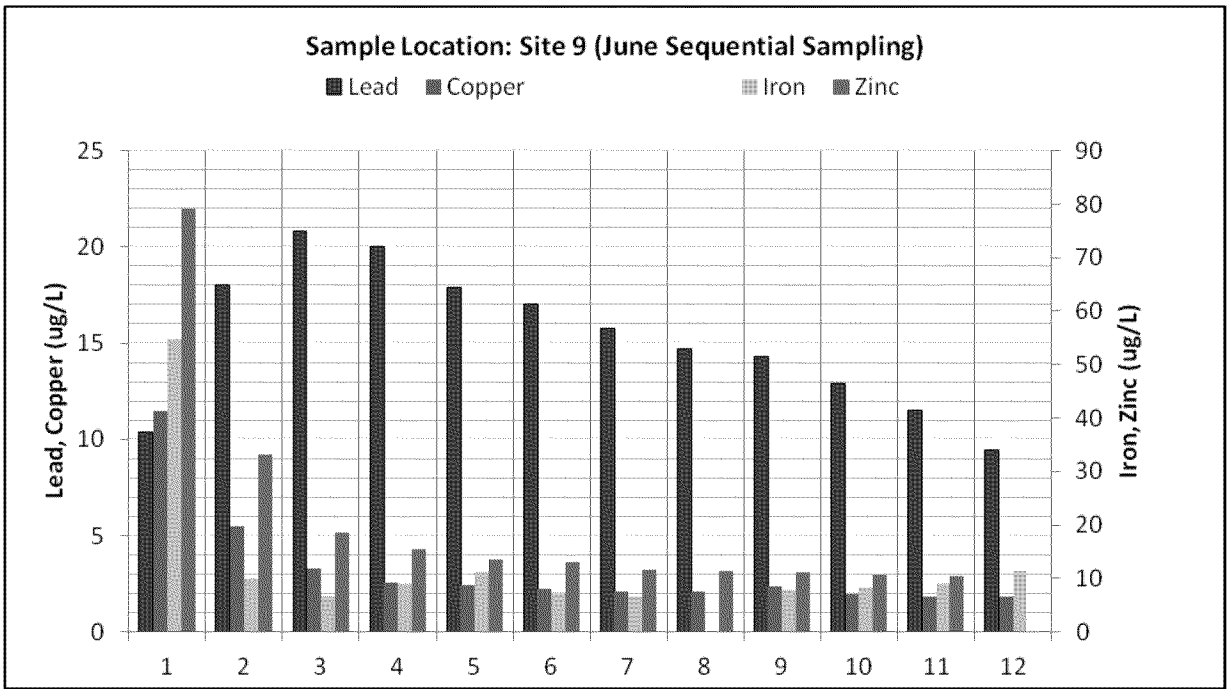


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

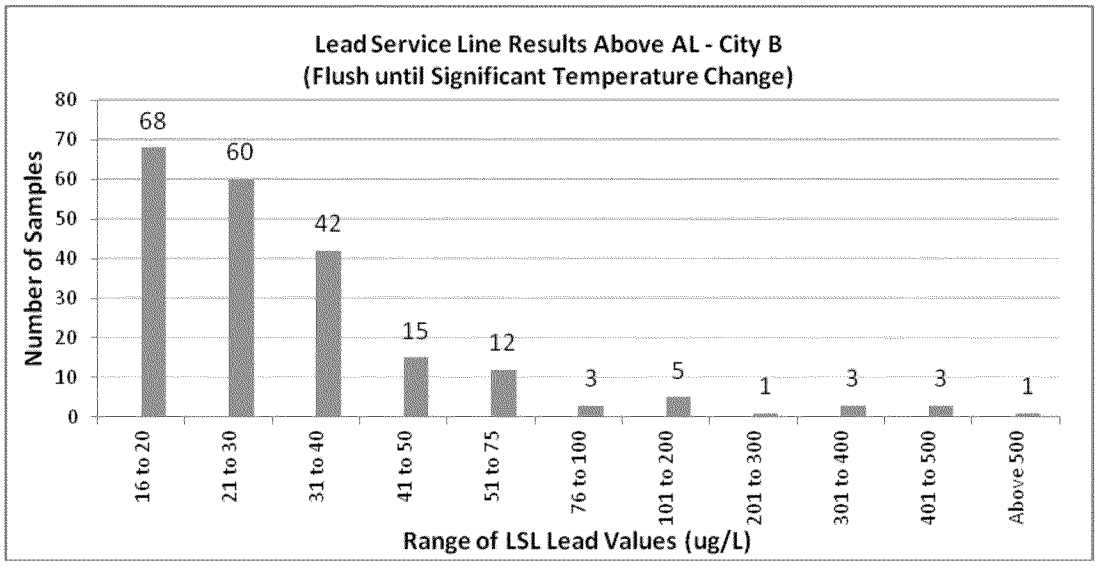
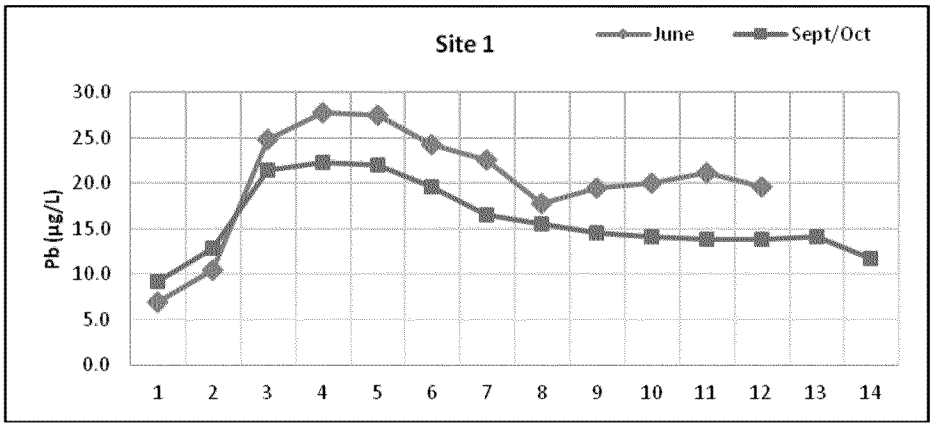


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

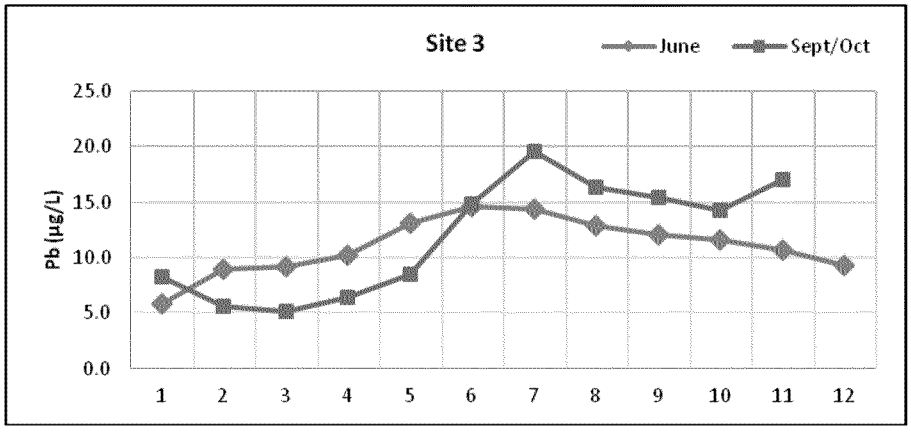
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

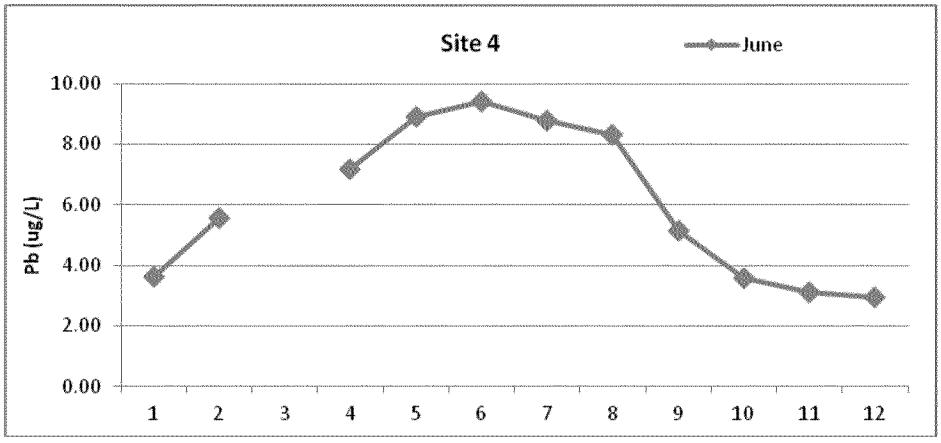
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

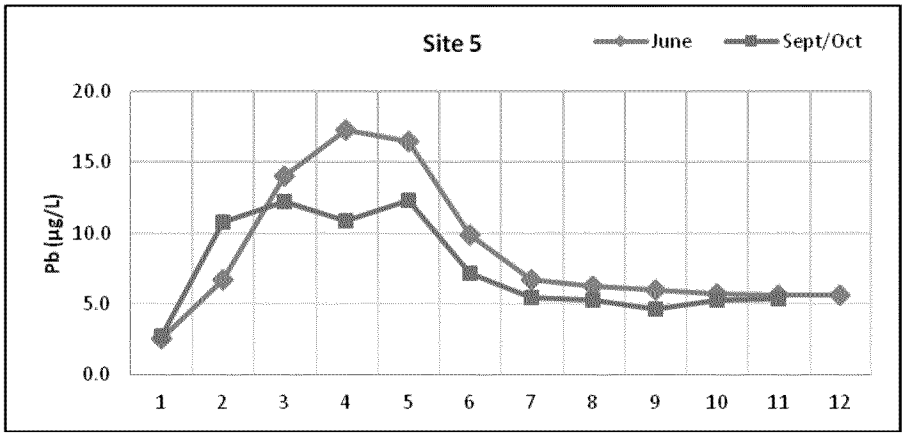
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

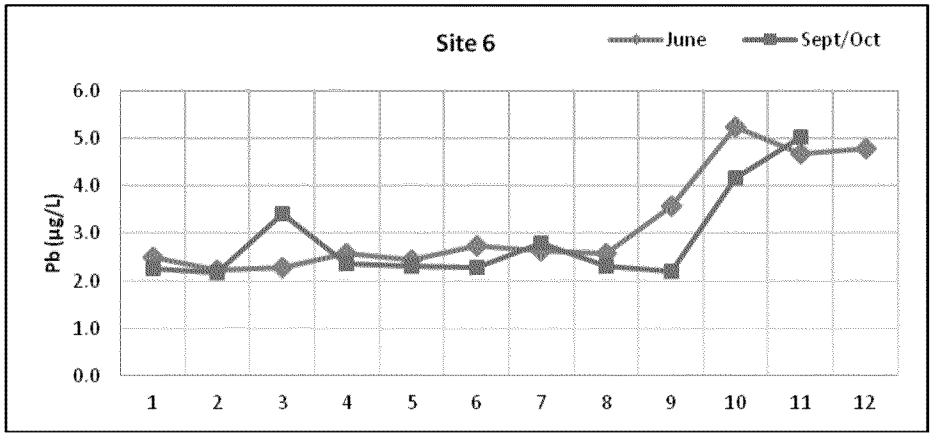
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

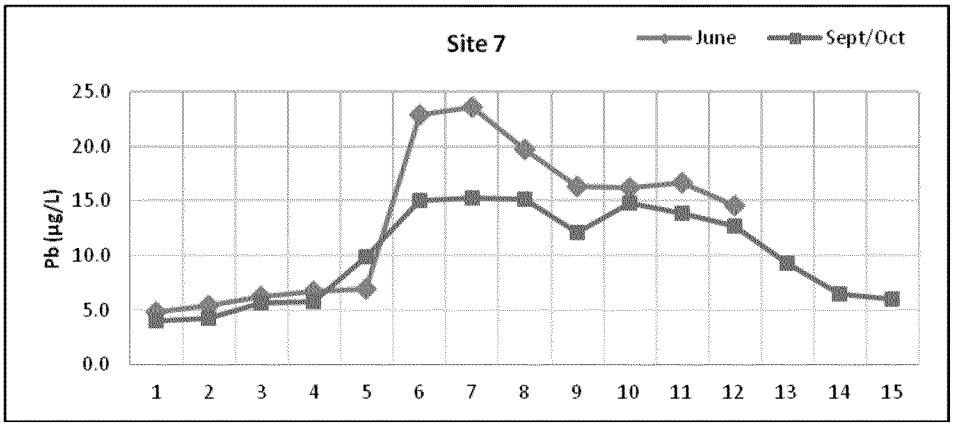
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

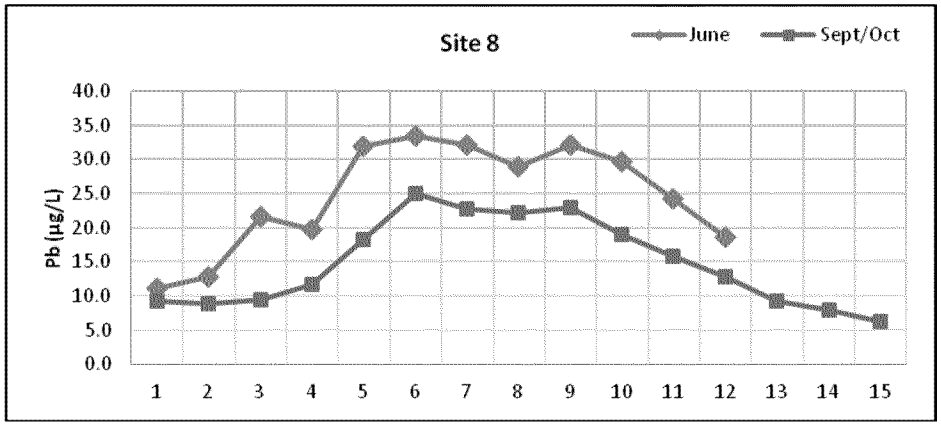
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

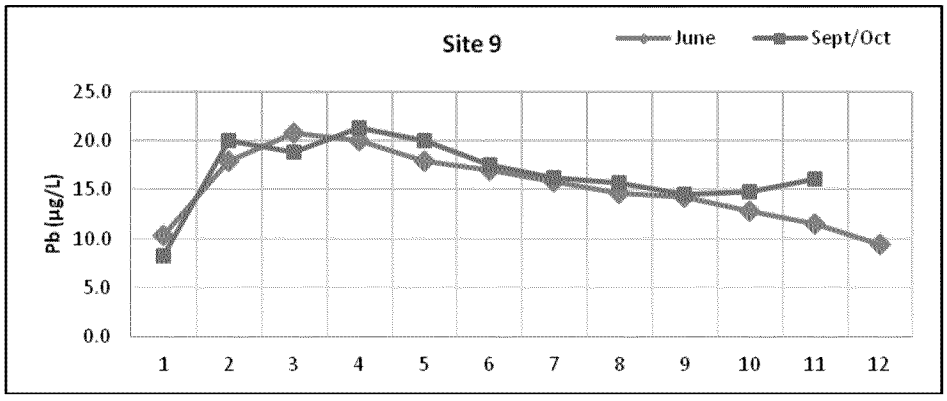
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

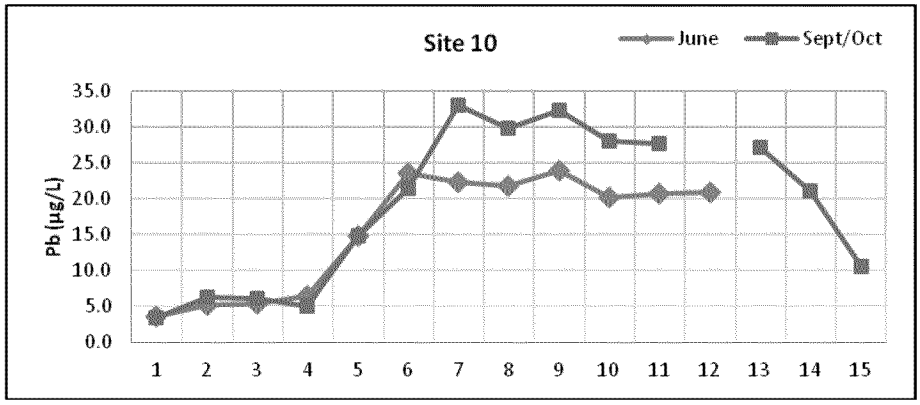
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

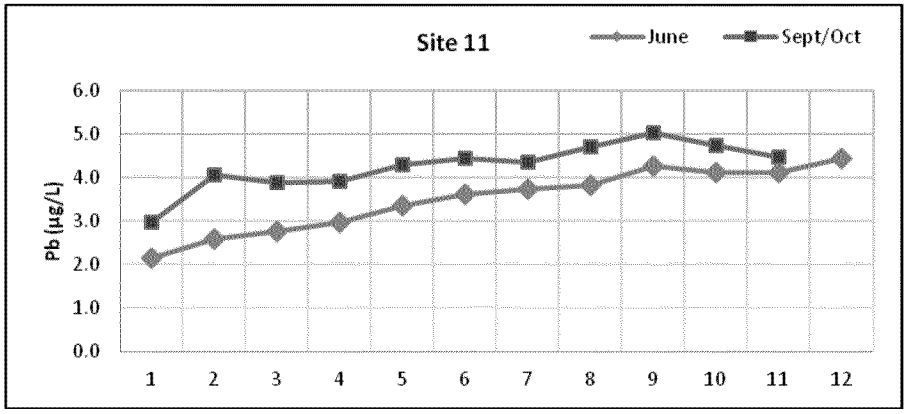
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

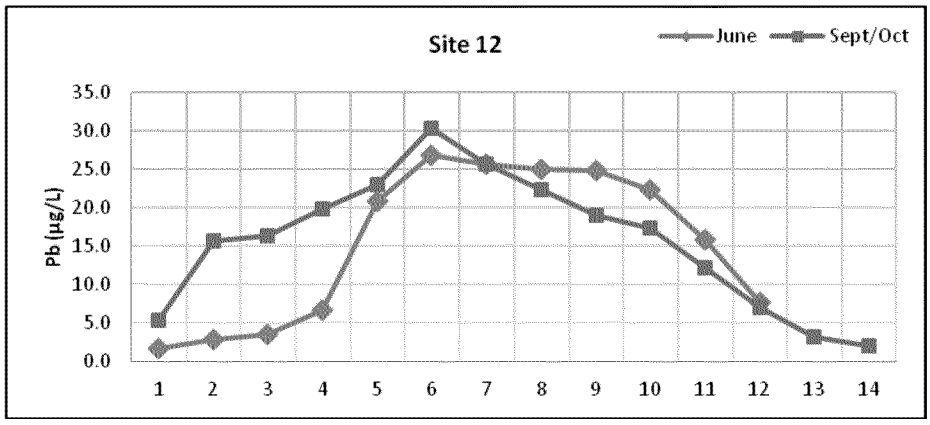
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

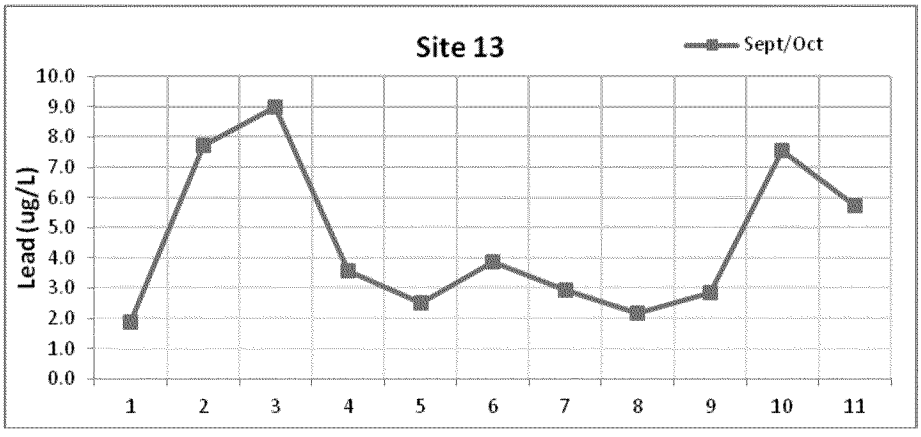
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

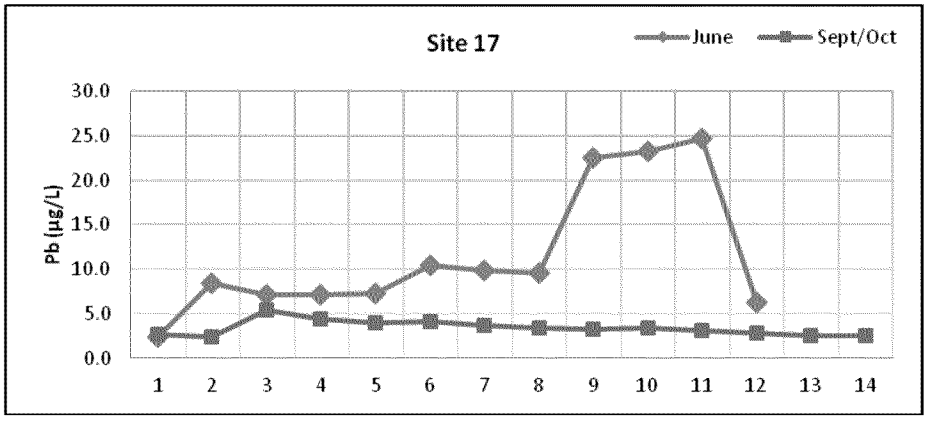
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

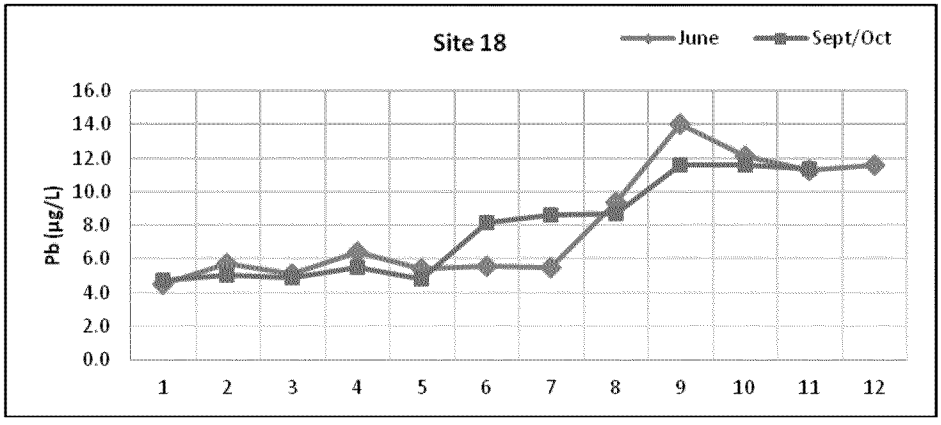
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

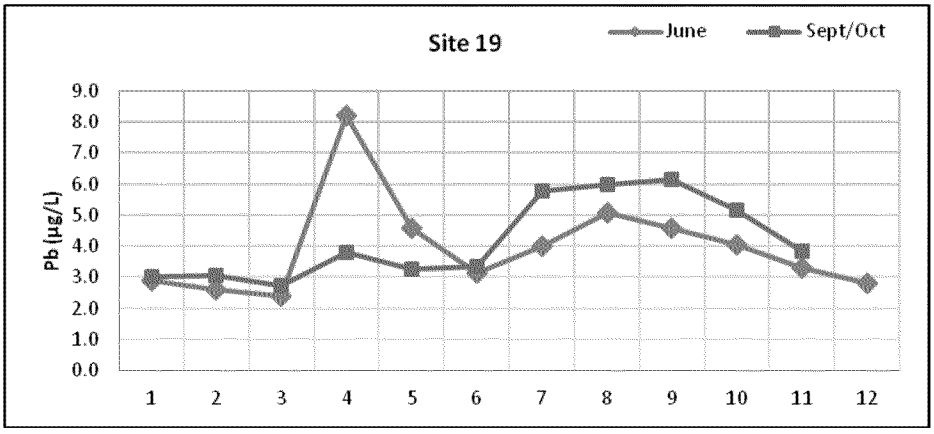
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

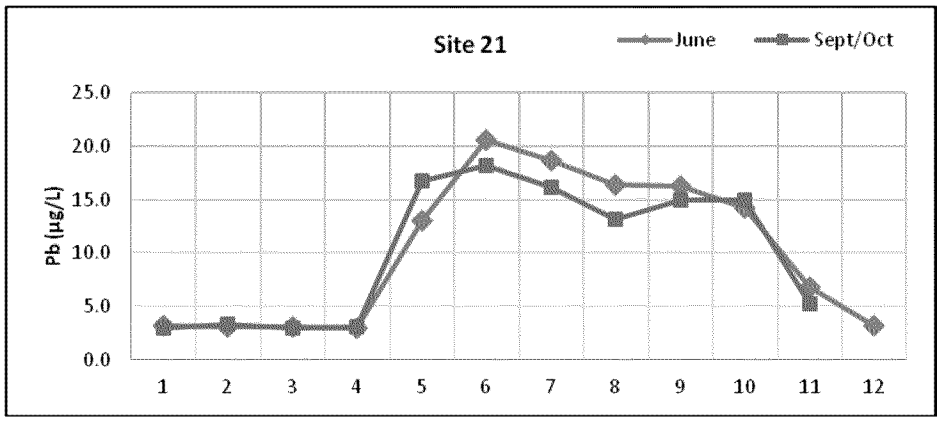
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

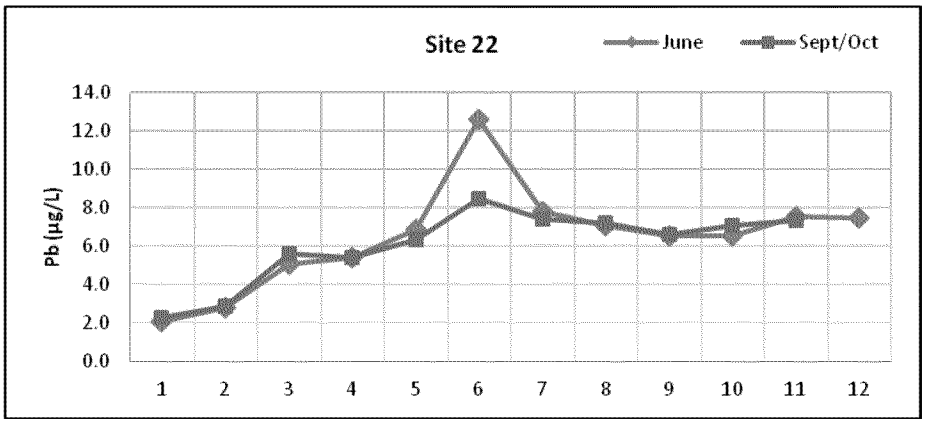
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

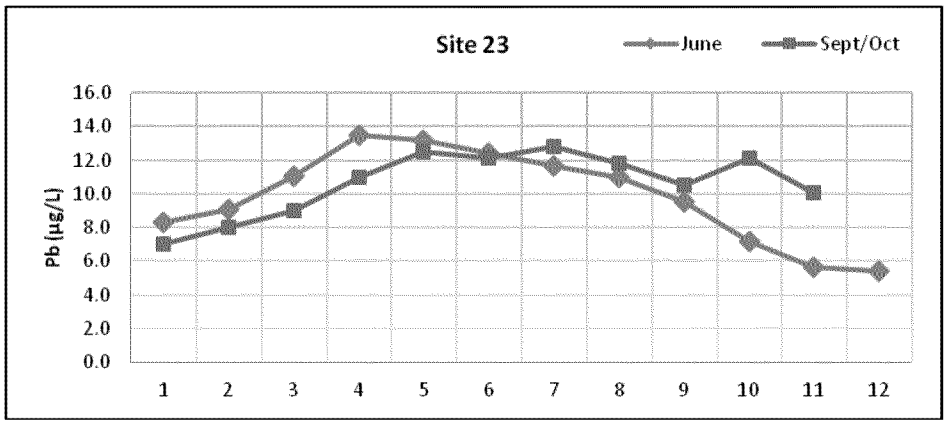
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

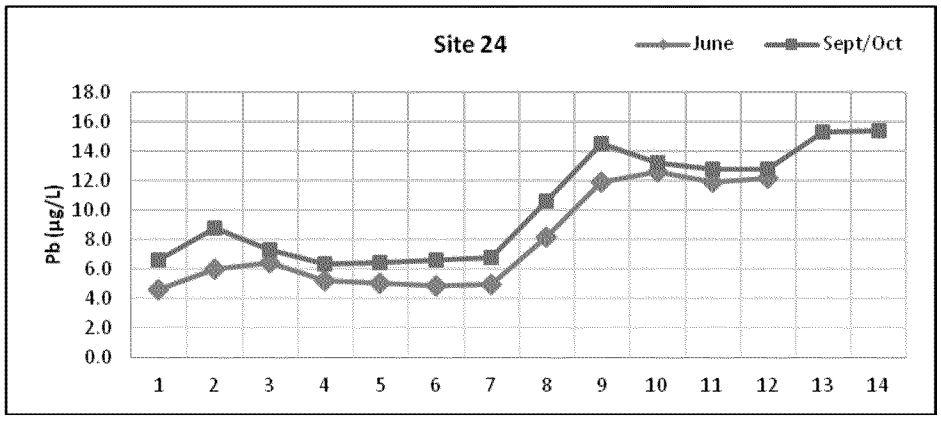
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

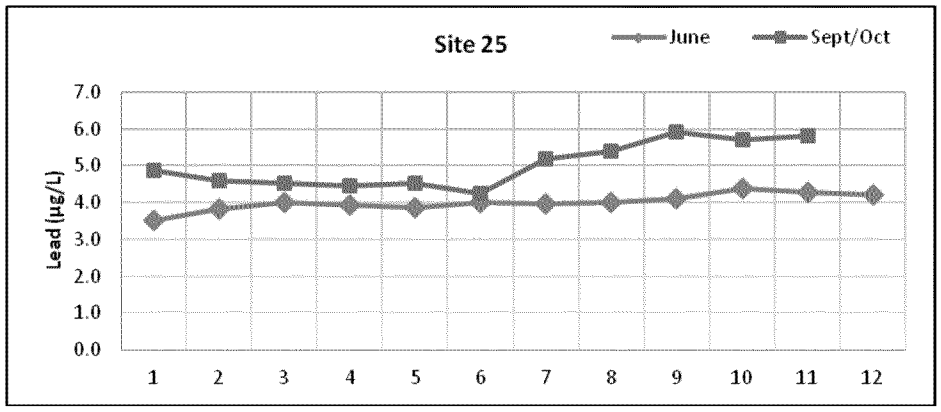
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

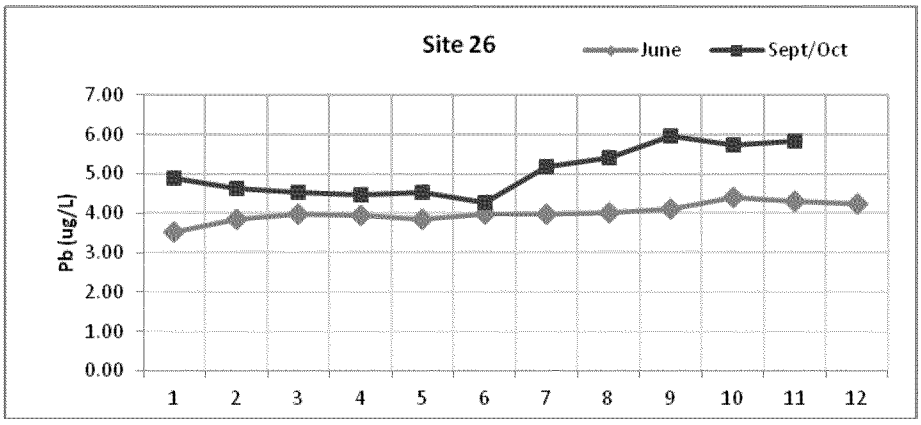
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

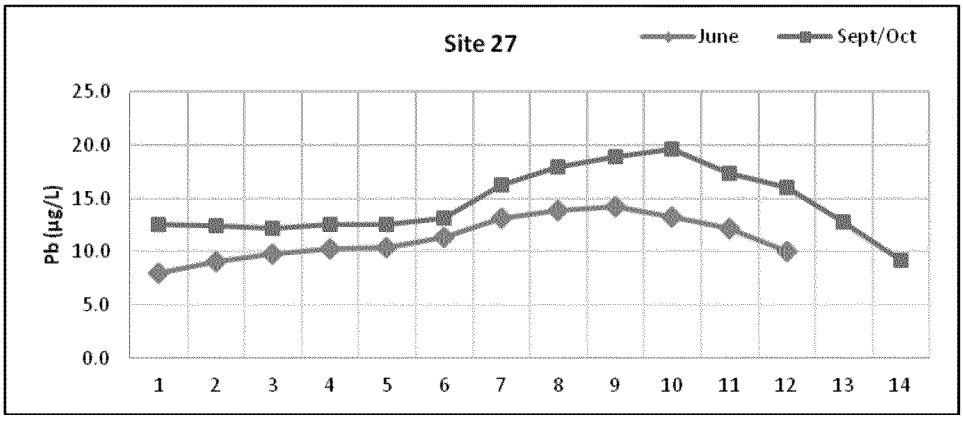
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

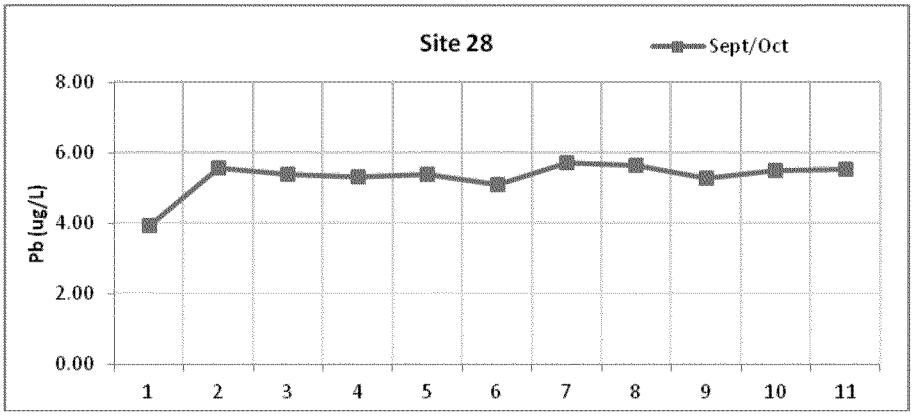
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

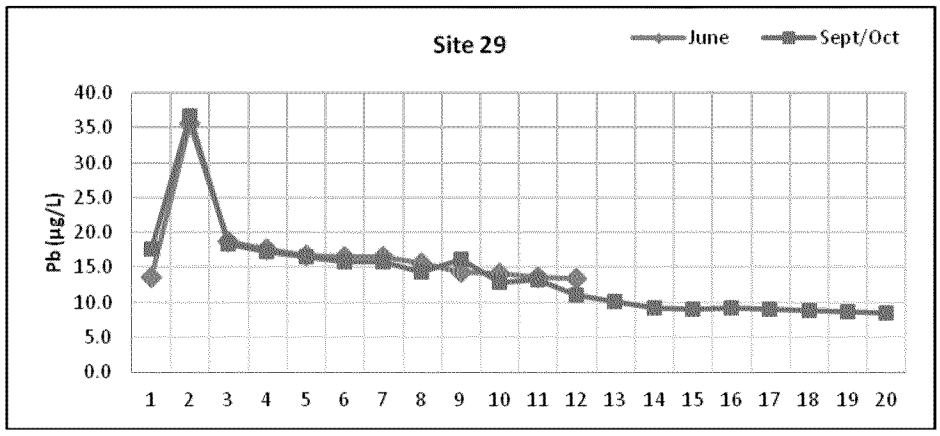
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

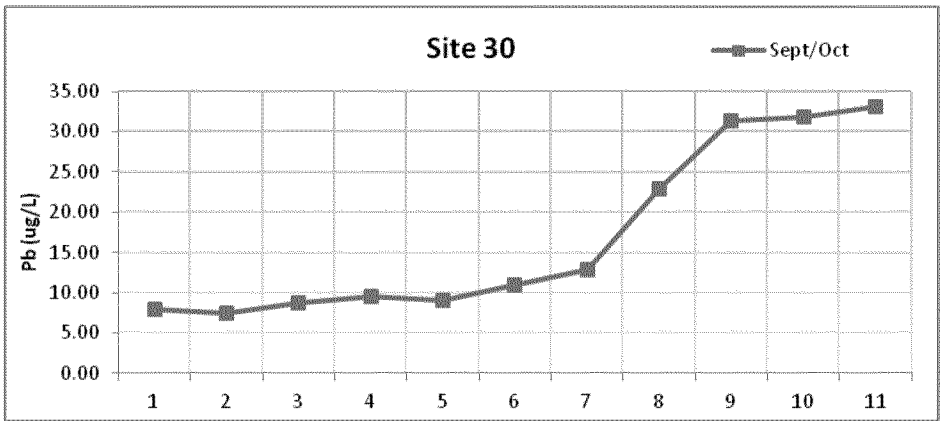
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

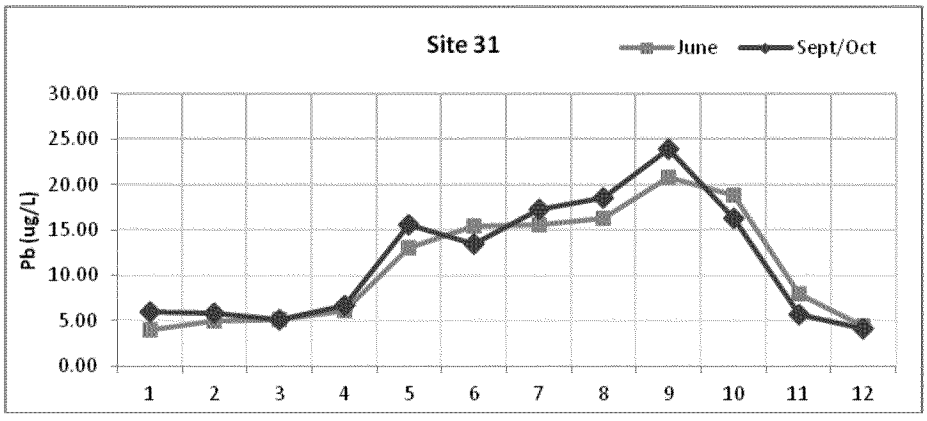
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

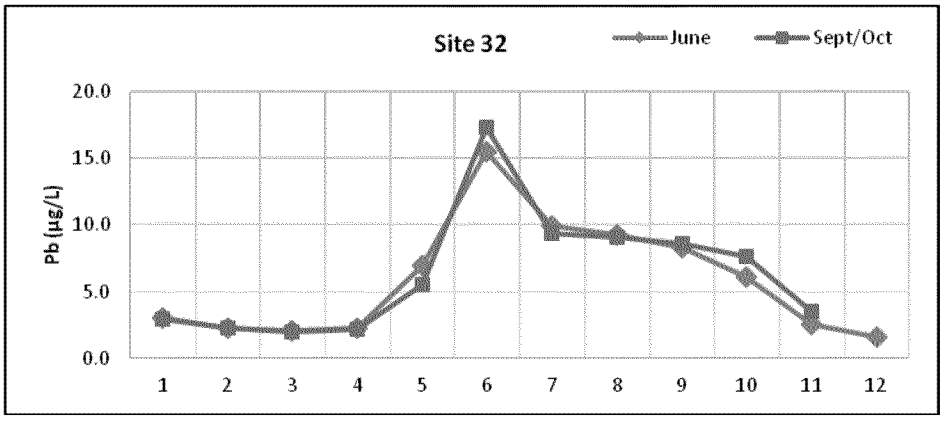
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

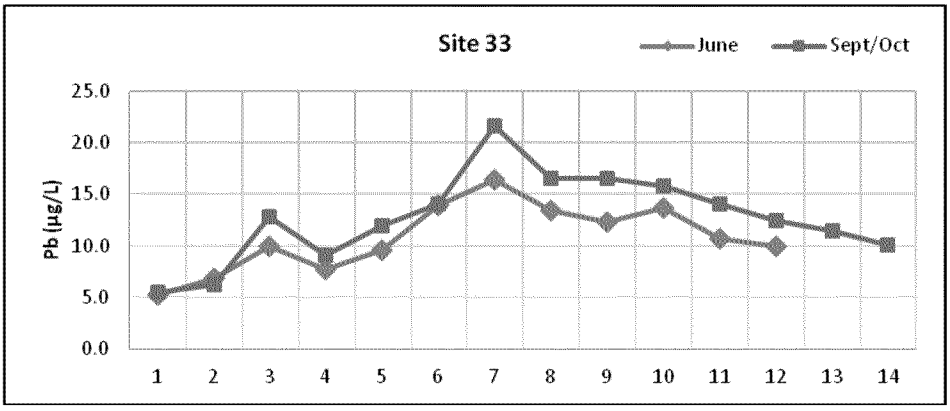
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

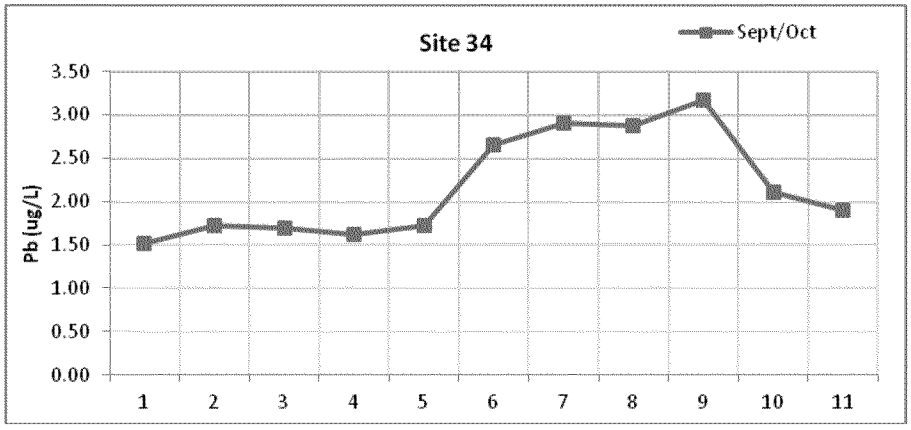
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

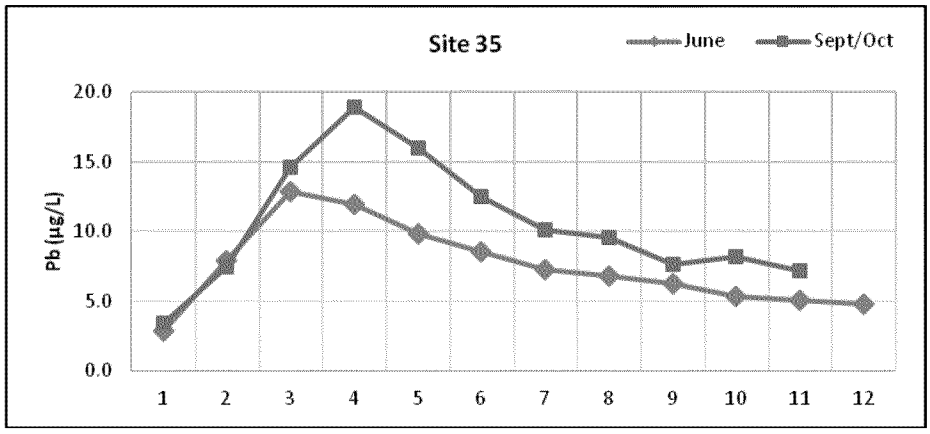
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

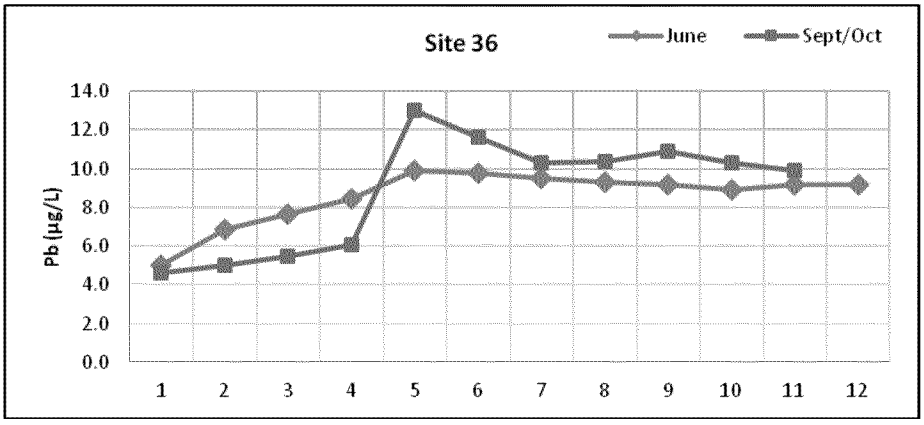
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

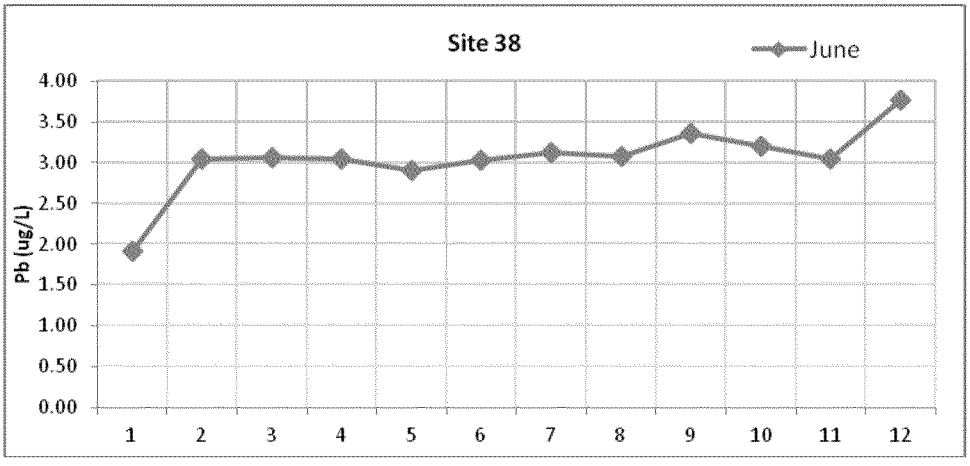
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

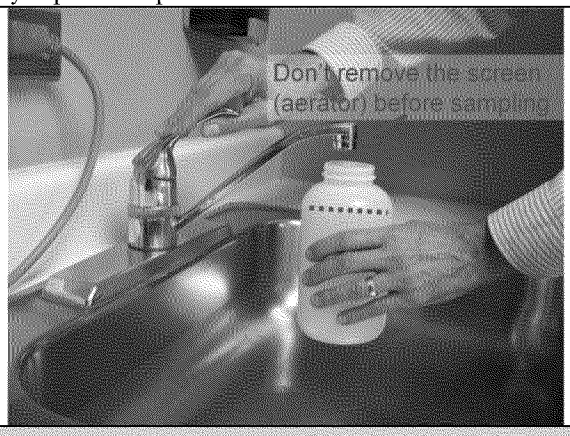
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

Literature Cited/References

1. Triantafyllidou, S.; Edwards, M., Galvanic corrosion after simulated small-scale partial lead service line replacements. *Journal American Water Works Association* **2011,** *103*, (9), 85-+.
2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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Chicago study finds high levels of toxic metal in areas of street work or plumbing repairs

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A section of pipe is guided into place Friday as new water mains are installed along Thorndale Avenue at Glenwood Avenue on the city's North Side. An EPA study found that such projects can interfere with anti-lead protections in older pipes and boost levels of lead in drinking water. (Chris Walker, Chicago Tribune / September 20, 2013)

By Michael Hawthorne, Chicago Tribune reporter
September 25, 2013

Dangerous levels of lead are turning up in Chicago homes where pipes made of the toxic metal were disturbed by

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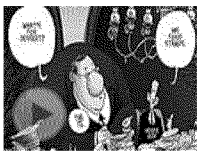
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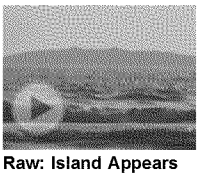
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street work or plumbing repairs, according to a new federal study that suggests the city's aggressive efforts to modernize its water system could inadvertently pose health risks.

Wall Chart
www.DuraLabel.com
Free wall chart - pipe color coding standards set by ANSI and ASME

The problem starts with lead service lines that Chicago installed across the city until the mid-1980s to connect water mains with homes. Researchers at the U.S. Environmental Protection Agency found that spikes of lead can leach into tap water when those pipes are altered by water main replacements, meter installations or street work.

High levels can be found in tap water for years afterward, the EPA study found, raising concerns that other cities with lead pipes could face similar problems.

Most homeowners likely are unaware they could be drinking tainted water. Under federal rules, utilities rarely are required to warn residents that work is being done or tell them they can take steps to reduce their exposure to lead. A potent neurotoxin, lead can damage the brains of young children, lower IQ and trigger learning disabilities, aggression and criminal behavior later in life.

Lead is so hazardous that the EPA and the Centers for Disease Control and Prevention say there is no safe level of exposure. The metal has been phased out of gasoline, removed from paint and banned in children's toys. But the widespread use of lead pipes during the last century has left a festering problem nationwide.

"We owe it to people to tell them that their water might not be safe to drink," said Marc Edwards, a professor of civil and environmental engineering at Virginia Tech University who wasn't involved with the EPA study but has reached similar conclusions in his own research.

The new study comes as Mayor Rahm Emanuel pushes to speed up the replacement of aging water mains and increase the number of water meters installed citywide — two activities that EPA researchers found to be linked to high amounts of lead in tap water.

Lead levels exceeded federal standards at all but one of 13 Chicago homes tested where the service line had been physically disturbed between 2005 and 2011, according to results published last month in the peer-reviewed journal Environmental Science & Technology.

At 16 other homes where there was no known disturbance of the service line, only three had high lead levels.

The study also found that normal federal testing procedures would not have detected most of these spikes, a problem the Tribune described in a 2011 story based on preliminary data. Under the EPA's current testing protocol, only the first liter of water that flows out of a household faucet is checked, but the researchers took multiple consecutive samples.

Their testing showed that lead levels exceeded the federal limit of 15 parts per billion at 15 of 29 homes used in the study, but only one home showed high levels in the first liter of water drawn. The high levels didn't taper off until the 12th liter drawn from the taps, according to the researchers. Clearing pipes of that much water requires a continuous flow for up to five minutes, they said.

In response to the research, the city in July started advising residents to flush their pipes after a water main has been replaced on their street. "We're trying to be proactive," said Andrea Putz, a research specialist at the Chicago Department of Water Management.

The city's warning, which appears at the end of a packet of information handed out by work crews, says opening all household taps for three to five minutes will remove "any lead particulates that may have come loose from your property's service line as a result of the water main replacement."

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But the EPA says it is important for households with lead service lines to flush their pipes for several minutes any time household water hasn't been used for several hours — not just one time after a repair.

Two simple ways to do that are to take a shower or do a load of laundry. Then, before drawing water to drink or to prepare baby formula, the tap should be flushed for another 35 to 45 seconds to clear any remaining water sitting in the home's pipes, according to the EPA's recommendations.

Some household devices can filter out lead. NSF International provides a list of certified devices on its website under the heading "Consumer Resources."

Tinka Hyde, director of the water division in the EPA's Chicago office, said the study could influence a long-running debate about the Lead and Copper Rule, a 1991 regulation that by many accounts fails to adequately protect Americans from the harmful effects of lead in water.

Chicago has a sprawling network of pipes laid before the harmful effects of lead were fully accepted. The city's building code required lead service lines as recently as 1986. Yet Chicago has not recorded a violation of the Lead and Copper Rule for nearly 20 years. Nor have most other cities.

Under the rule's testing protocols, cities aren't required to take action unless water in more than 10 percent of the homes tested have lead levels exceeding 15 parts per billion, a limit that wasn't based on science about lead hazards but rather was considered cost-effective for water utilities to meet.

Water rarely contains lead when it leaves treatment plants, but under certain conditions the metal can leach into water while it sits in or flows through pipes and taps. Pipes and faucets inside homes also can contain lead, as can solder and brass plumbing devices.

Treatment plants add orthophosphate and other chemicals to form a white coating inside pipes that can prevent lead from leaching into the water. But street work and plumbing repairs can interfere with those protective measures.

Soon after becoming mayor, Emanuel won approval for a series of water rate increases to replace leaky pipes that in many cases are decades old. About 75 miles of new water mains are expected to be laid this year, up from 30 in 2010, according to city records. The city also plans to install 12,000 new meters this year to encourage residents to save water. But such fixes generally leave in place the lead service pipes that often connect water mains with homes.

Scores of other cities are embarking on similar efforts to overhaul aging water systems, a task the American Water Works Association estimates could cost at least $1 trillion nationwide during the next quarter-century.

The EPA study is part of growing amount of research suggesting that the work, while much-needed to stem leaks and conserve water, could have unintended consequences.

One of the challenges for regulators is finding solutions that are broadly effective. Water from two homes on the same block could have widely different lead levels depending on the type of service lines, the amount of lead in brass plumbing fittings and the extent of repair work.

"There is no magic bullet," said Putz, the city water specialist. "Everybody's plumbing is different, and that can have a big effect on lead levels."

City officials who assisted the EPA in its research plan to conduct their own study, testing lead levels in more than two dozen homes before and after water main replacements and

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meter installations, Putz said.

The presence of so many lead service lines across Chicago and other cities remains a concern. So does the staggering cost to eliminate the health risks.

"It's so huge that utilities and the states don't want to talk about it unless there are federal funds to fix it," said Bruce Lanphear, a noted lead expert at Simon Fraser University in Vancouver, British Columbia, who served on an EPA scientific advisory panel that studied the issue. "But if we don't start to do something, we're sacrificing another generation of children to the hazards of lead."

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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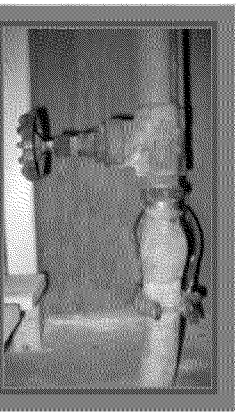
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (µg/L)	34	126	29	113
Fe (µg/L)	<5	<5	<5	34
Mn (µg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 µg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

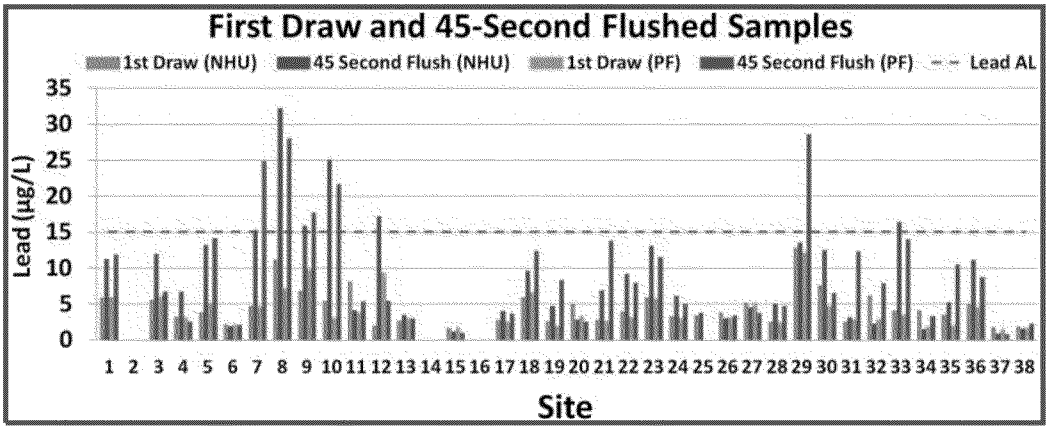

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

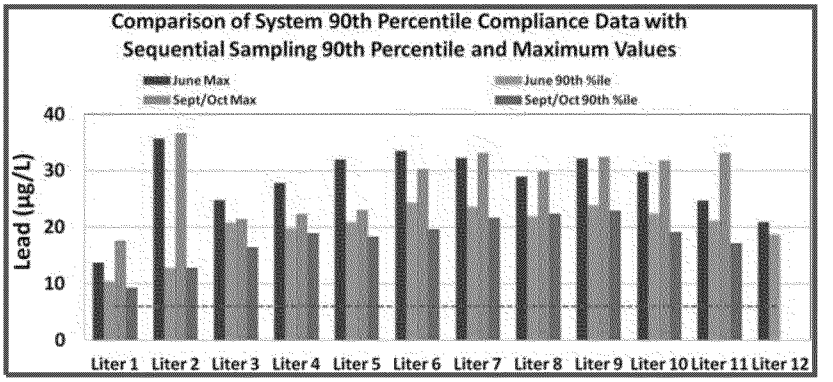

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

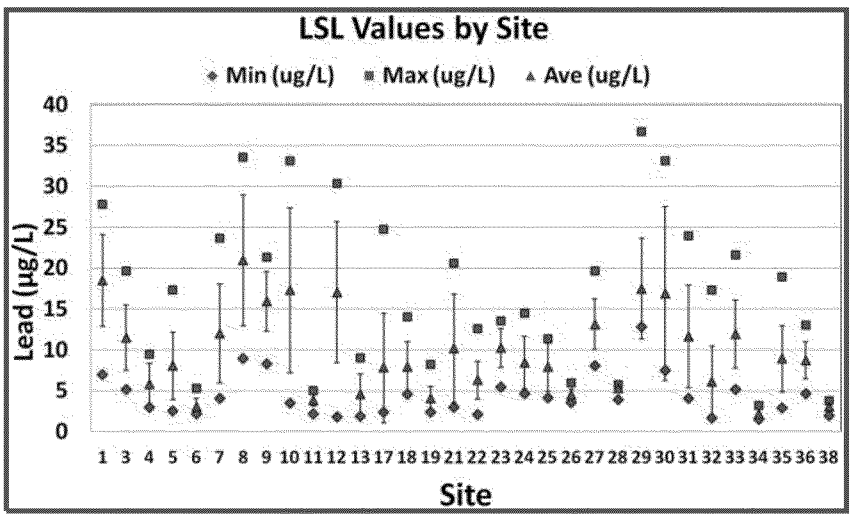

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

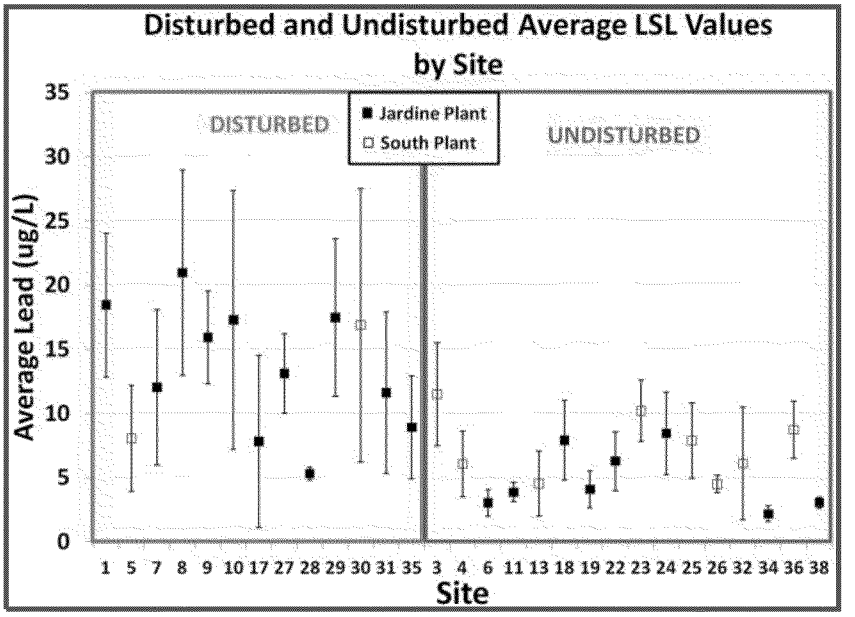

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Figure S2	Photograph of LSL Bulb	Table S2	Chicago Compliance Data
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Figure S5	Sample Site Map and Home Age	Table S4b	Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.
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Figure S9-S40	Sequential Sampling Graphs (Lead)	Table S6c	Summary of stagnation times.
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Figure S45	Sept/Oct Sample Collection and Reporting Form		

Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

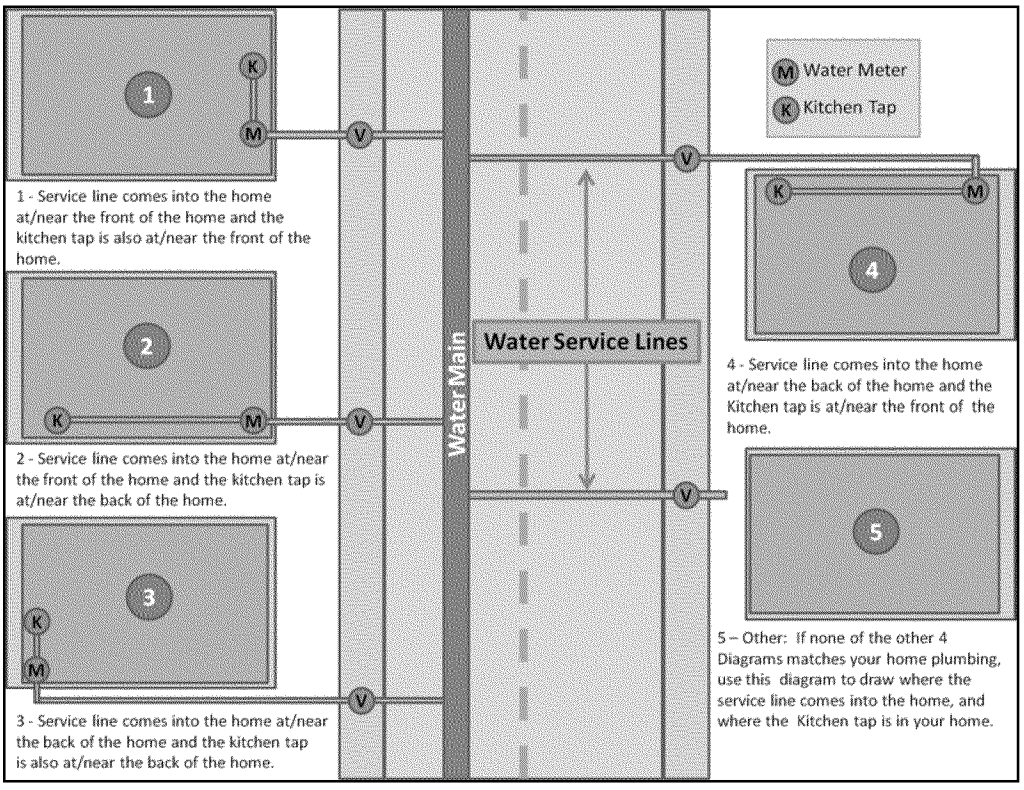


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

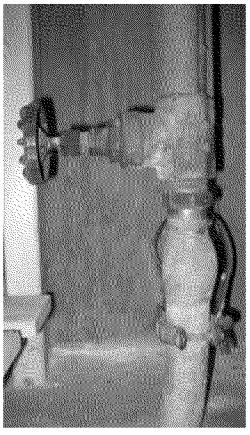


Figure S2: LSL Bulb

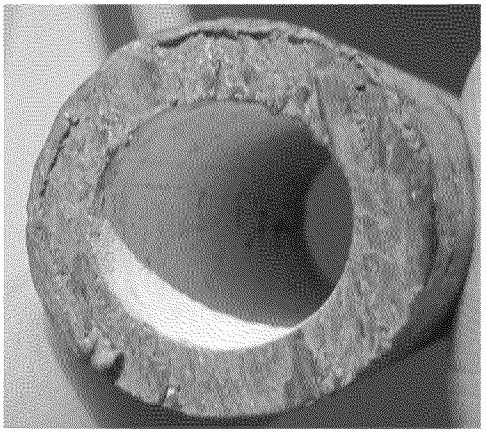


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

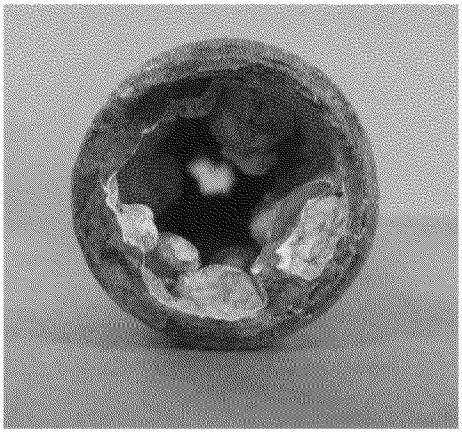


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

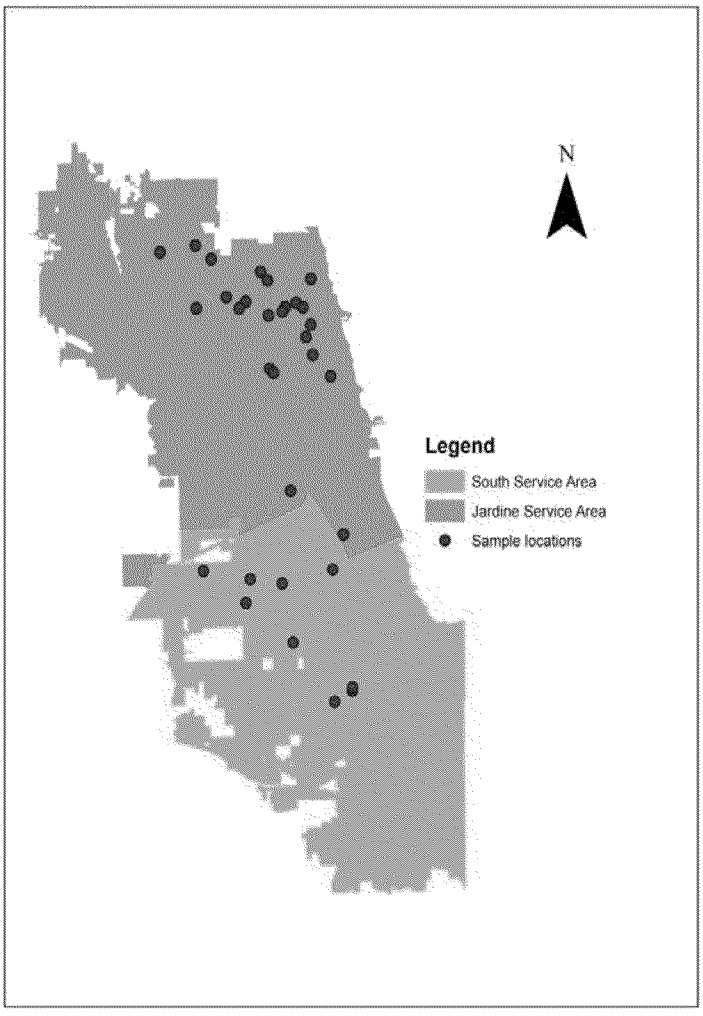


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 μg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

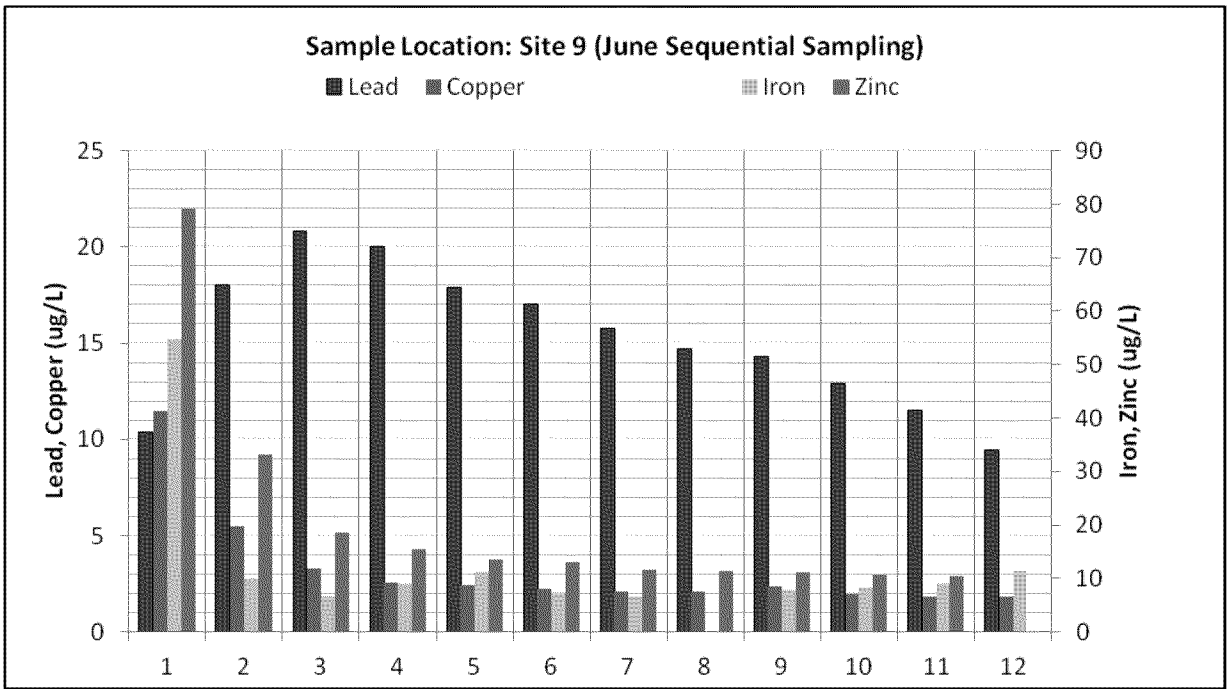


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

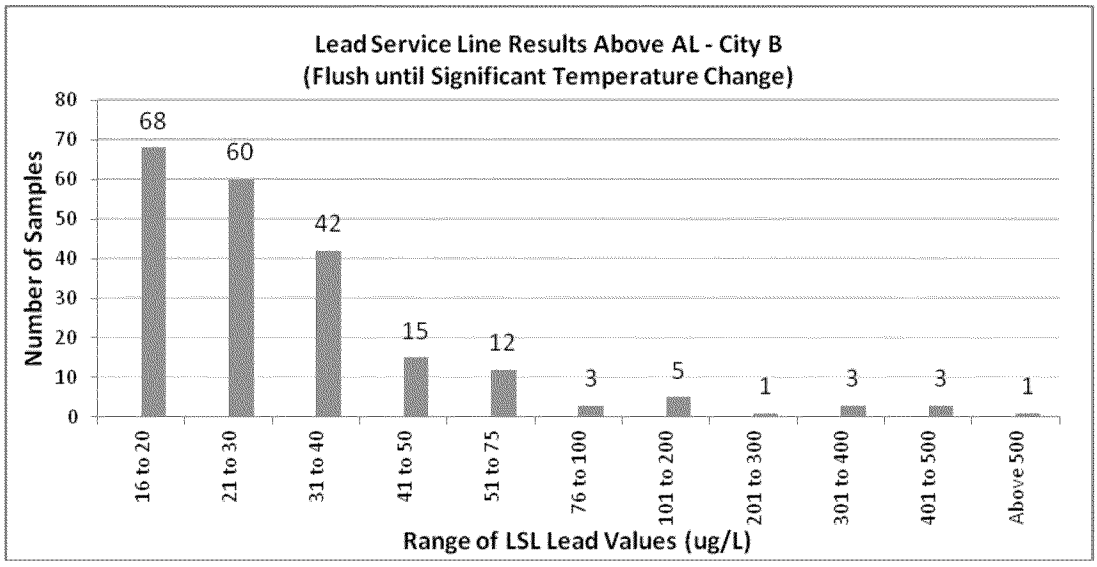
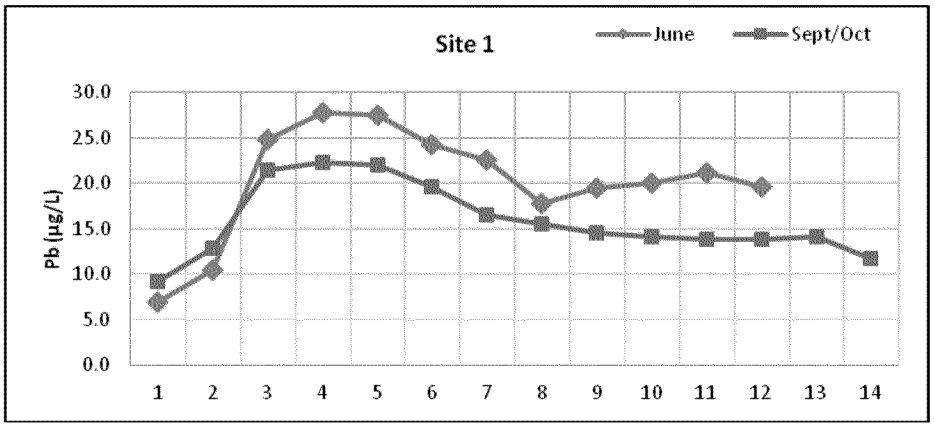


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

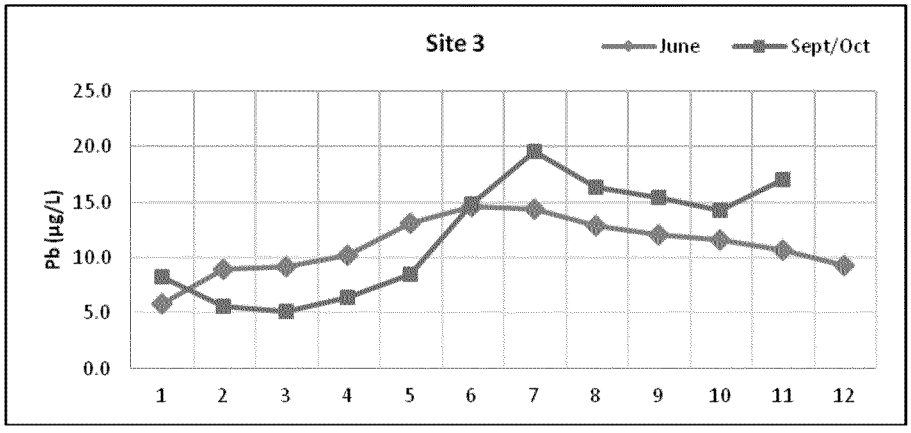
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

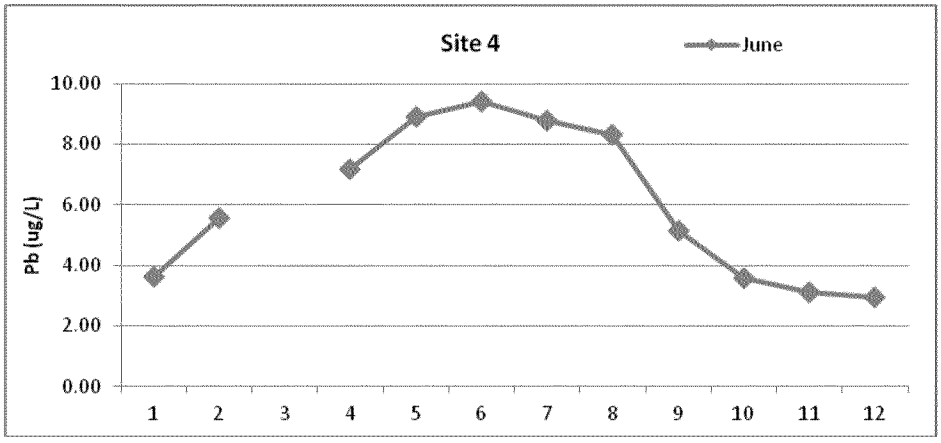
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

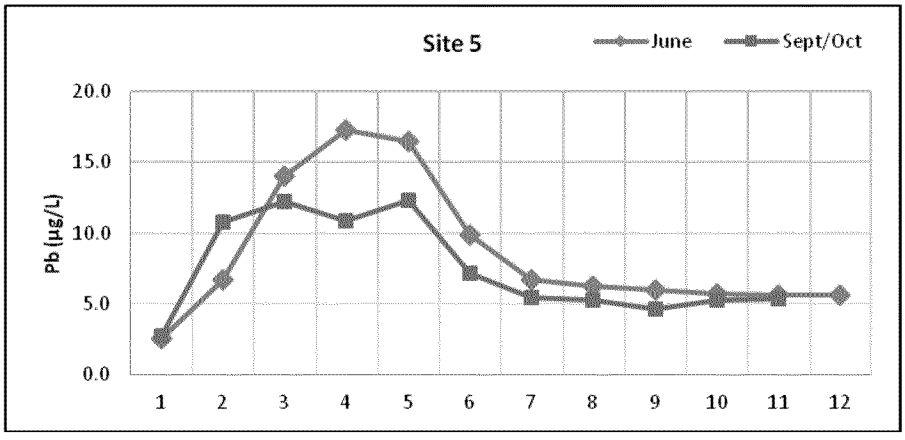
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

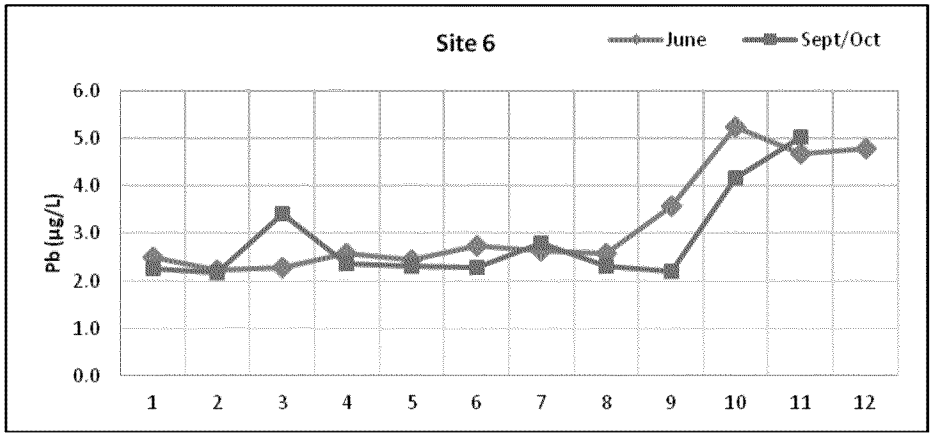
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

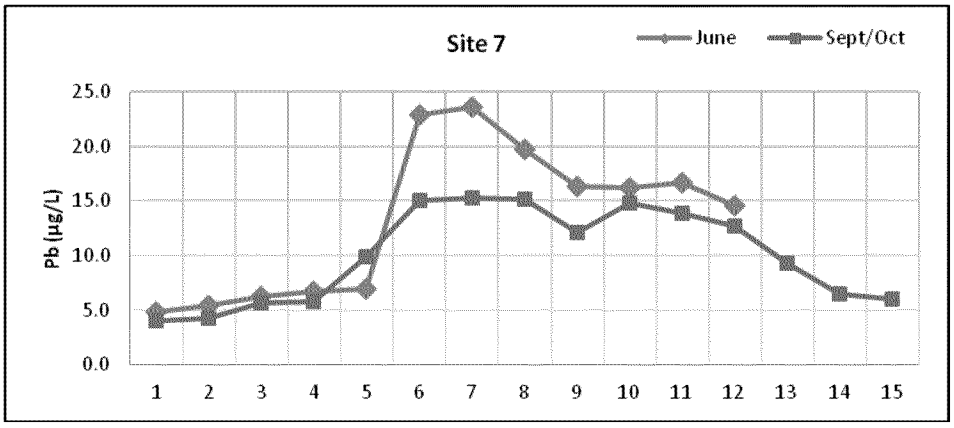
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

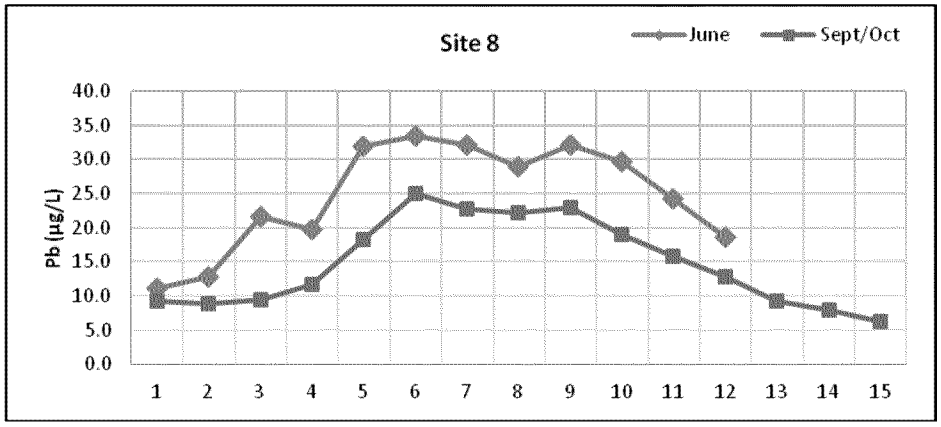
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

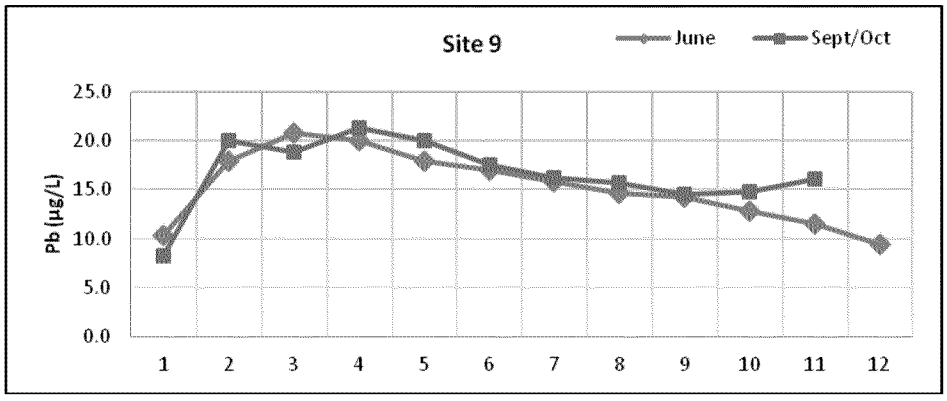
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

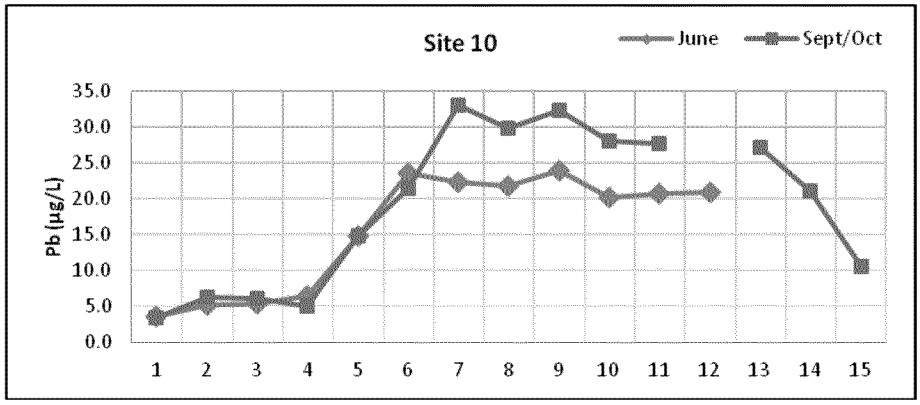
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

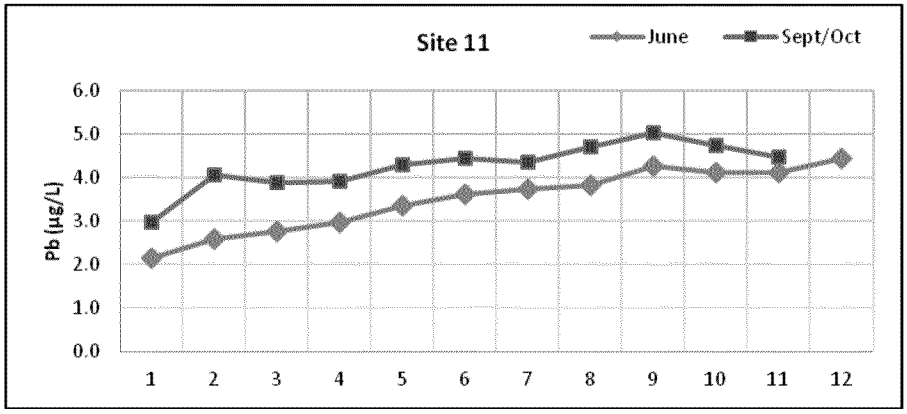
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

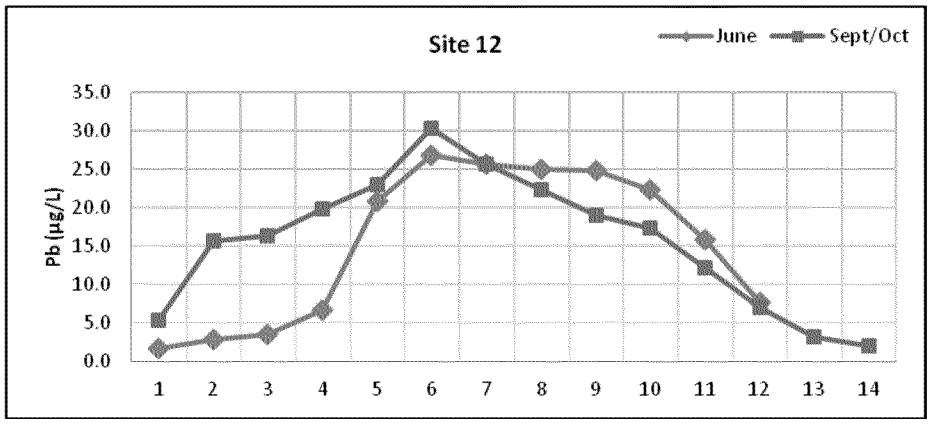
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

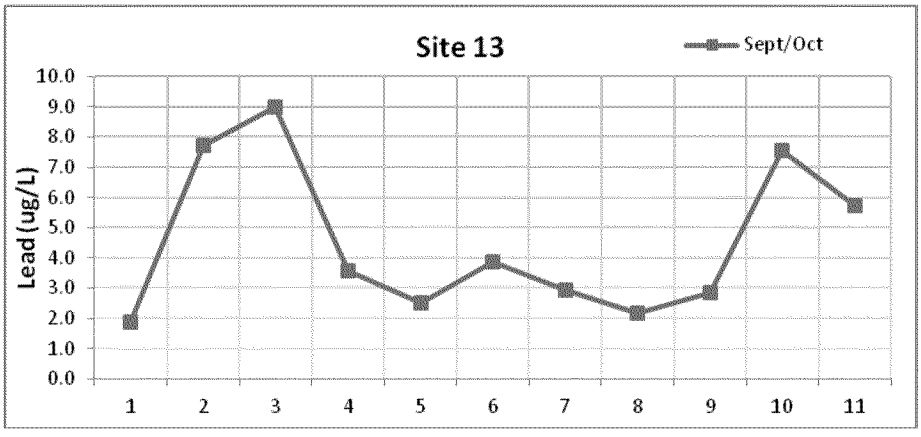
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

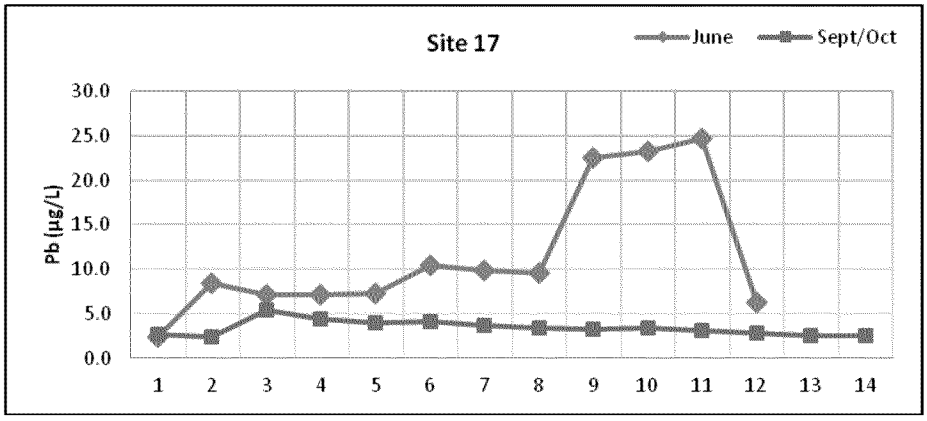
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

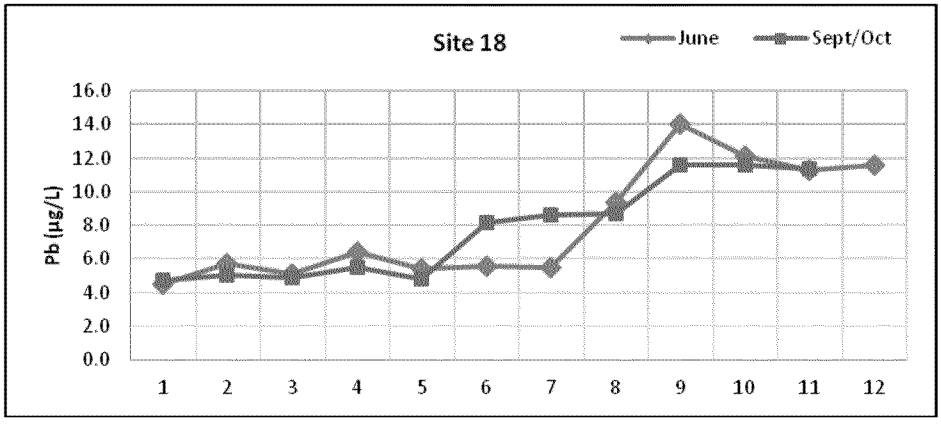
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

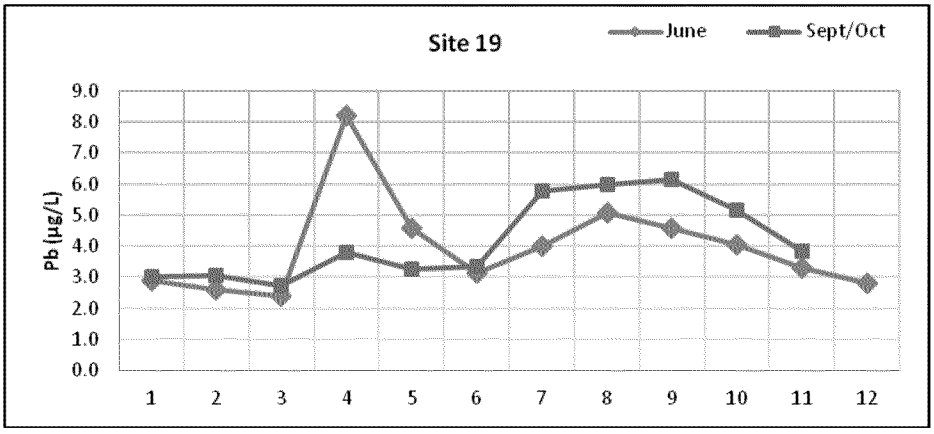
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

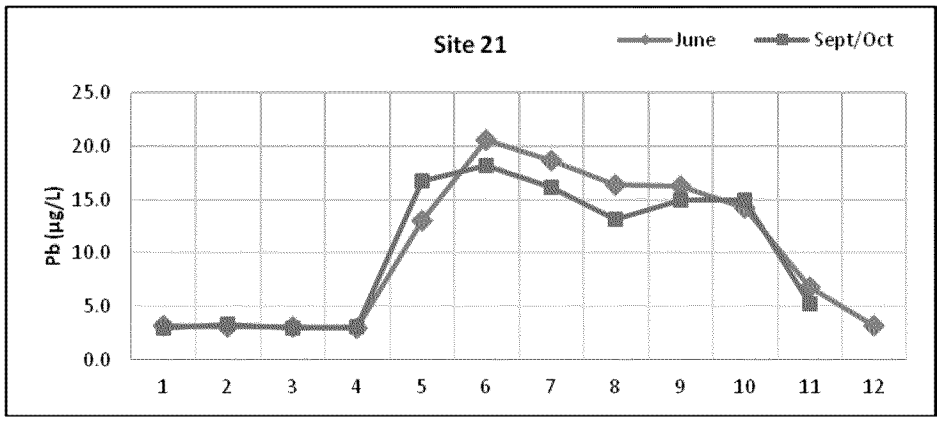
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

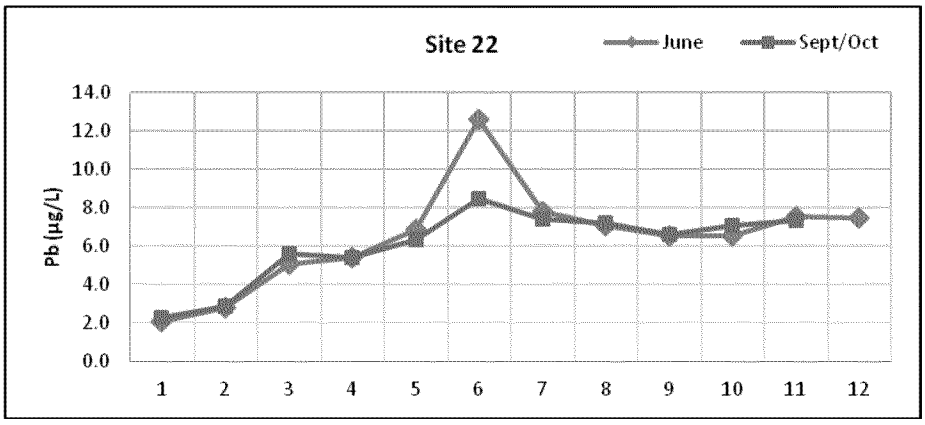
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

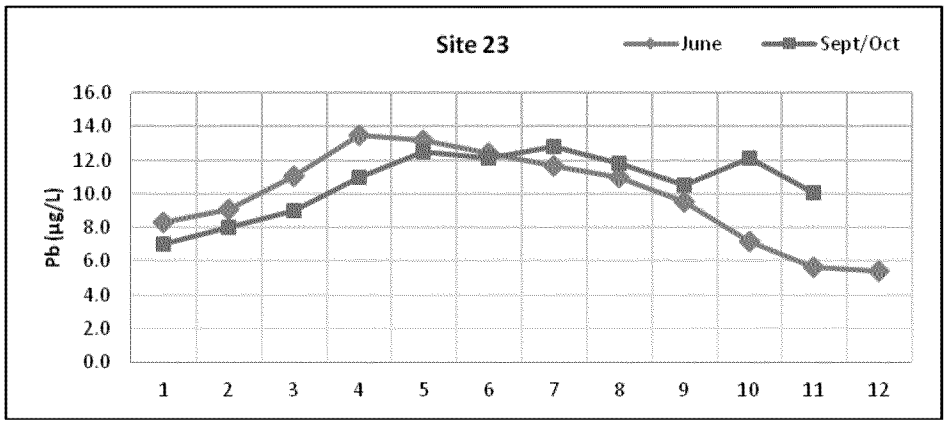
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

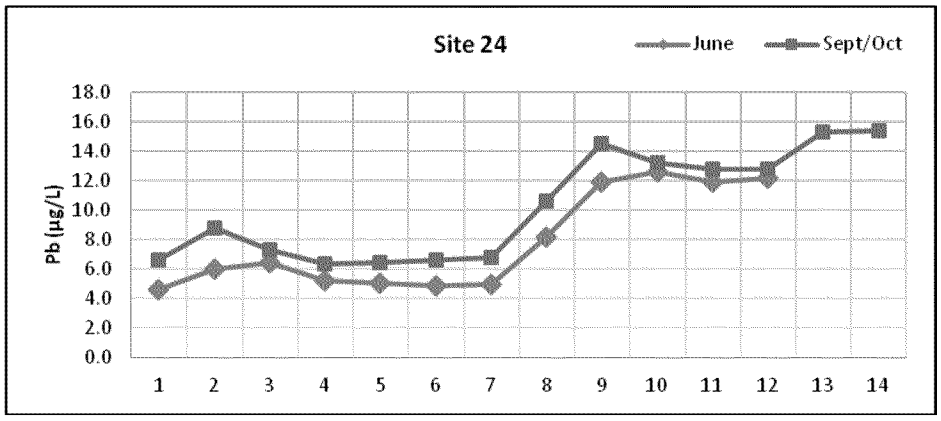
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

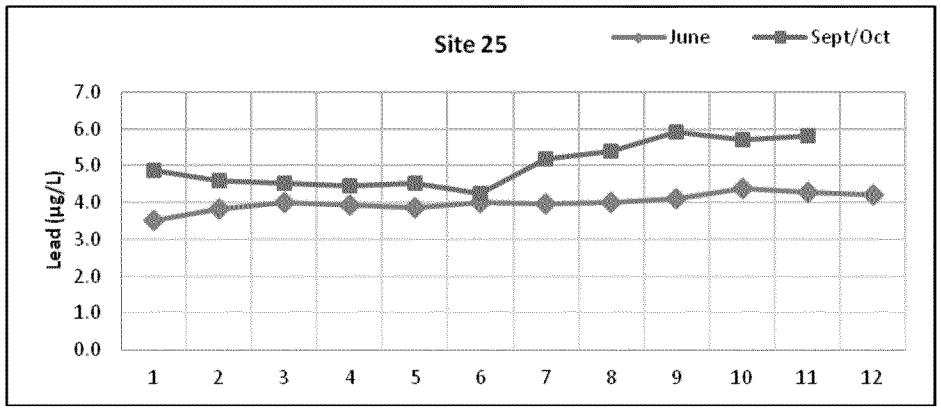
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

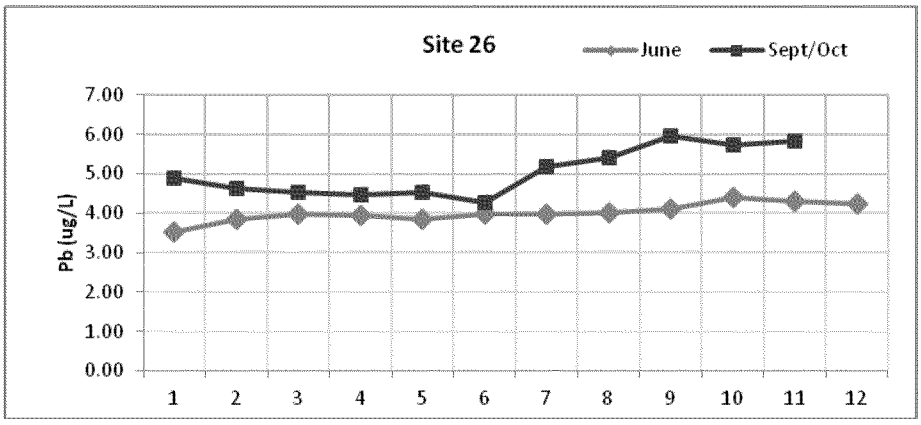
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

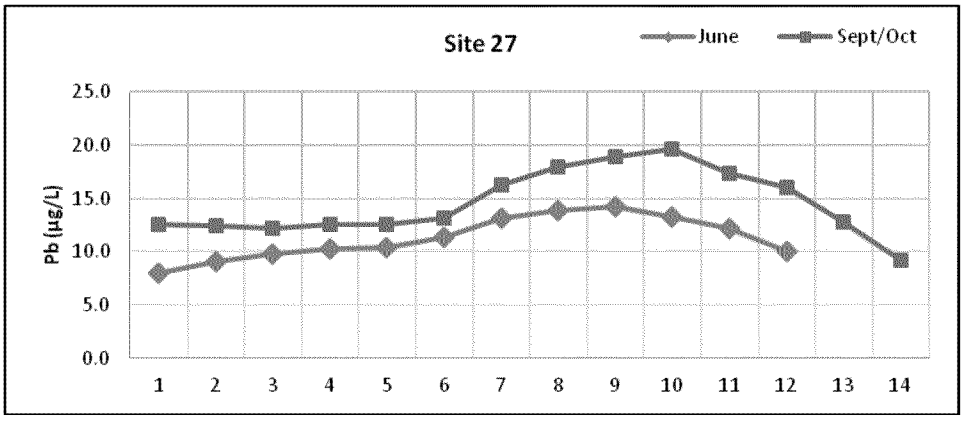
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

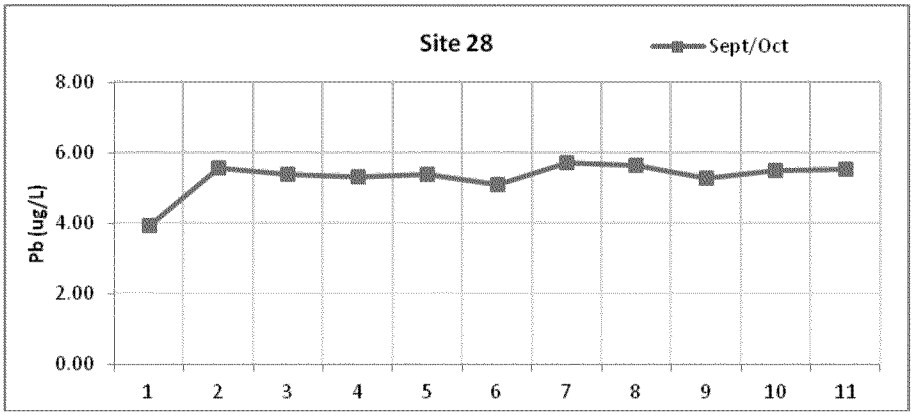
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

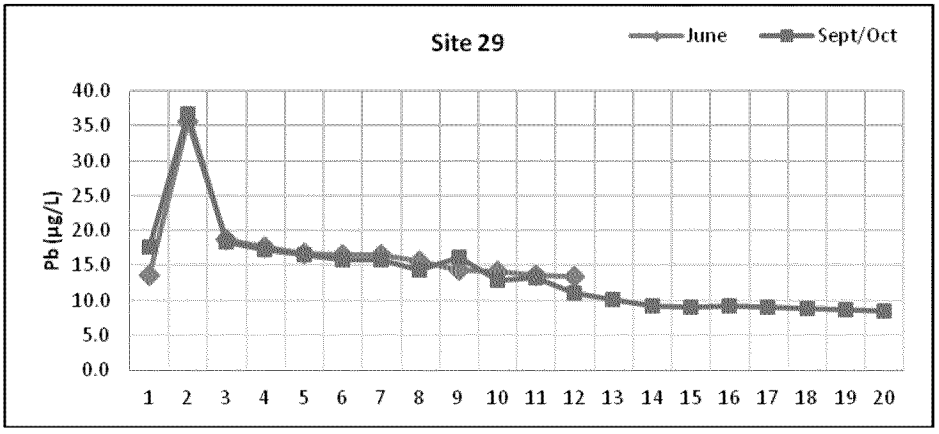
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

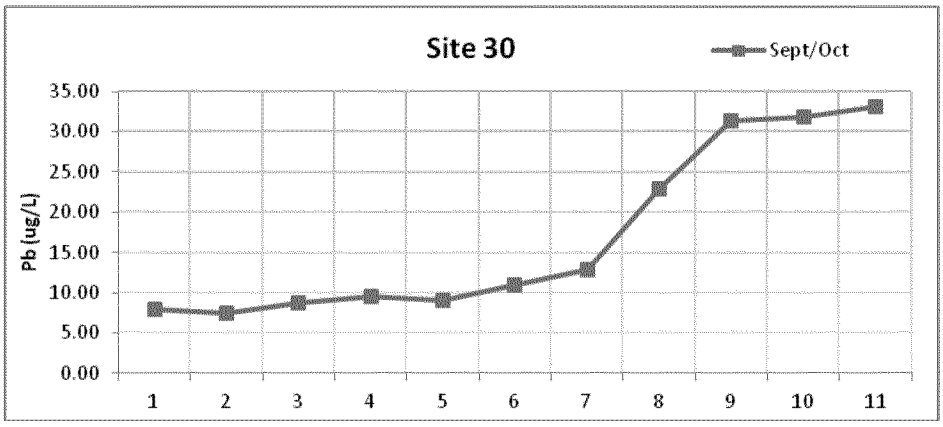
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

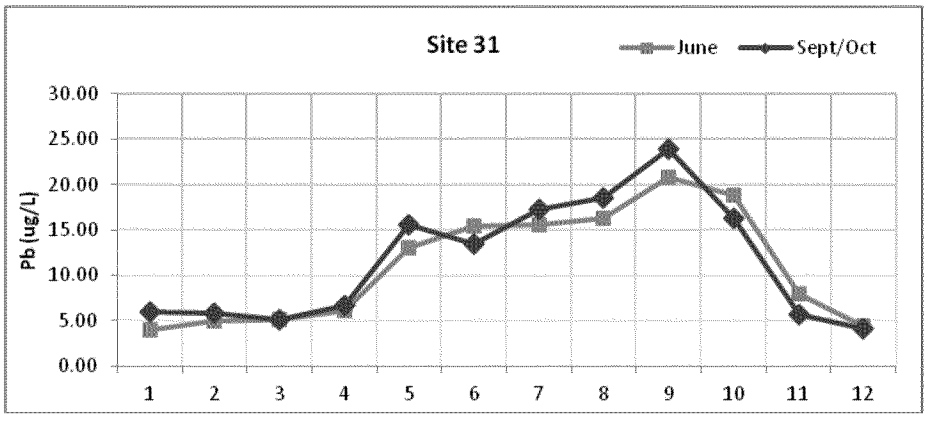
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

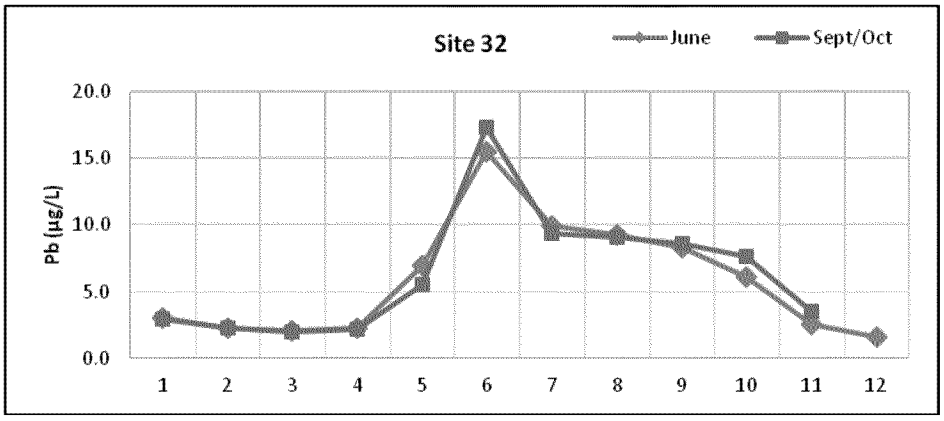
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

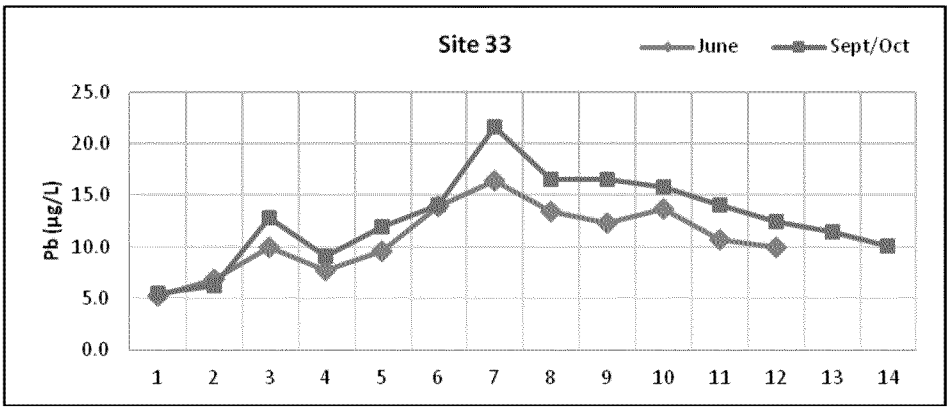
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

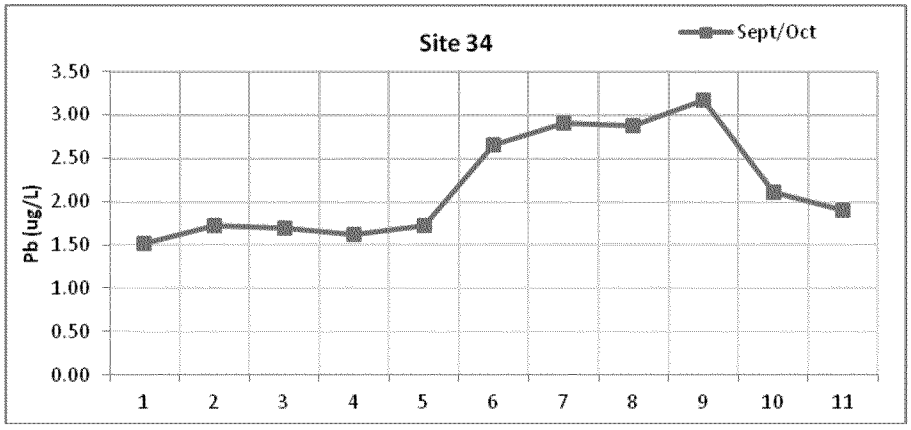
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

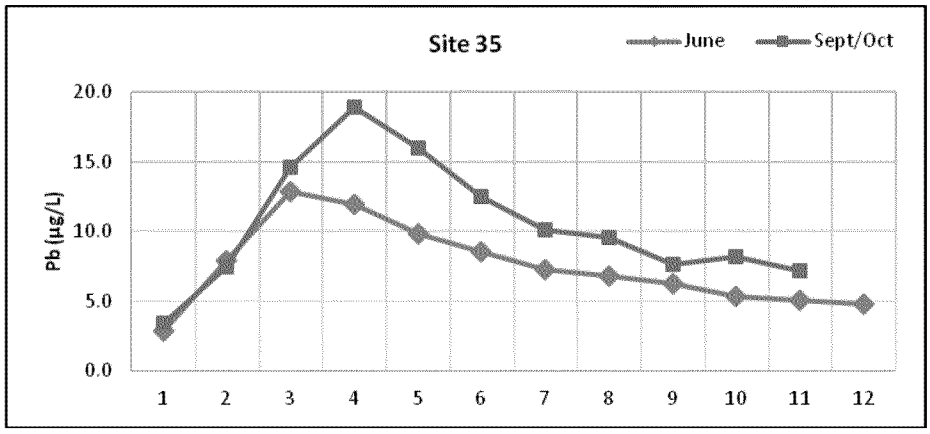
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

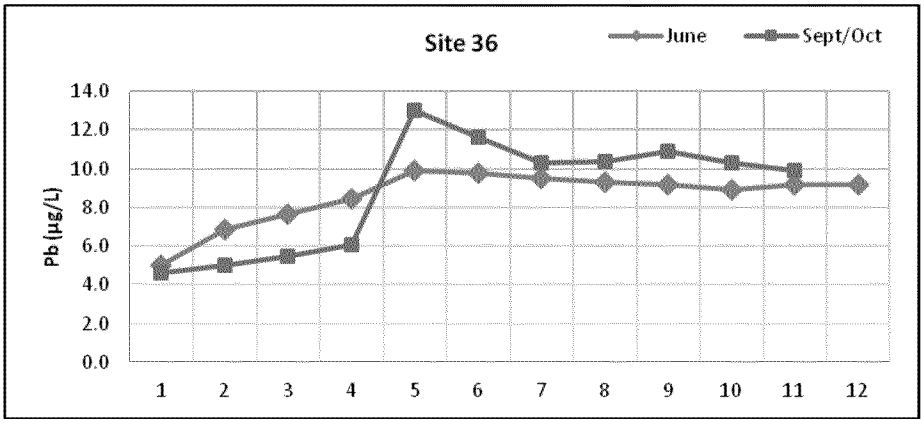
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

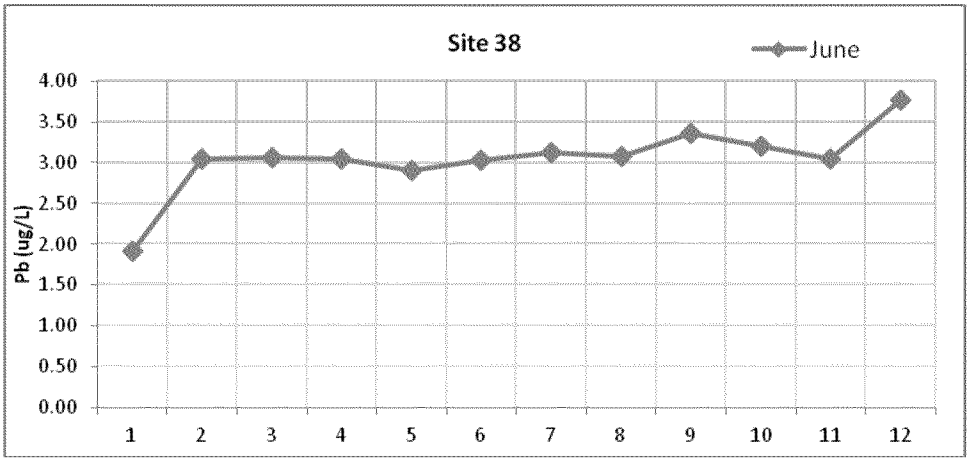
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

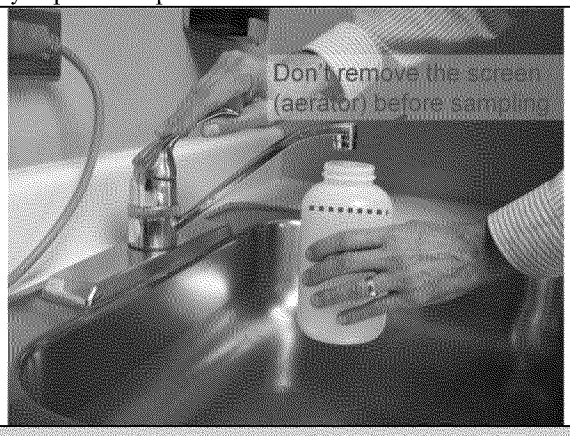
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

Literature Cited/References

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2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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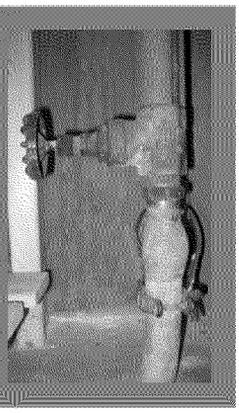
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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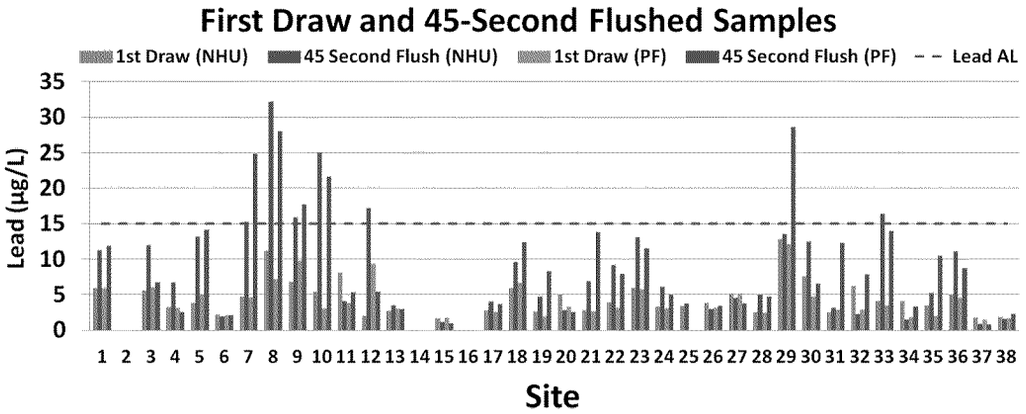

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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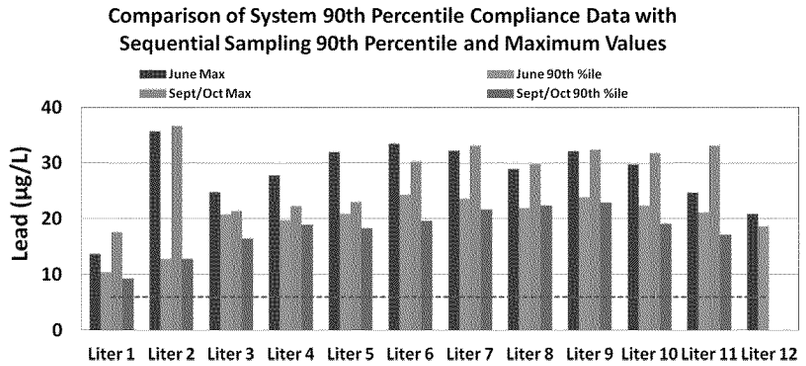

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

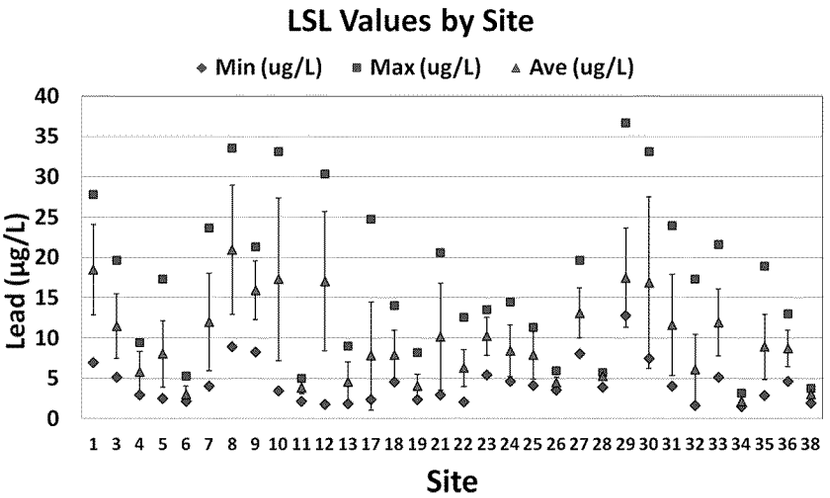

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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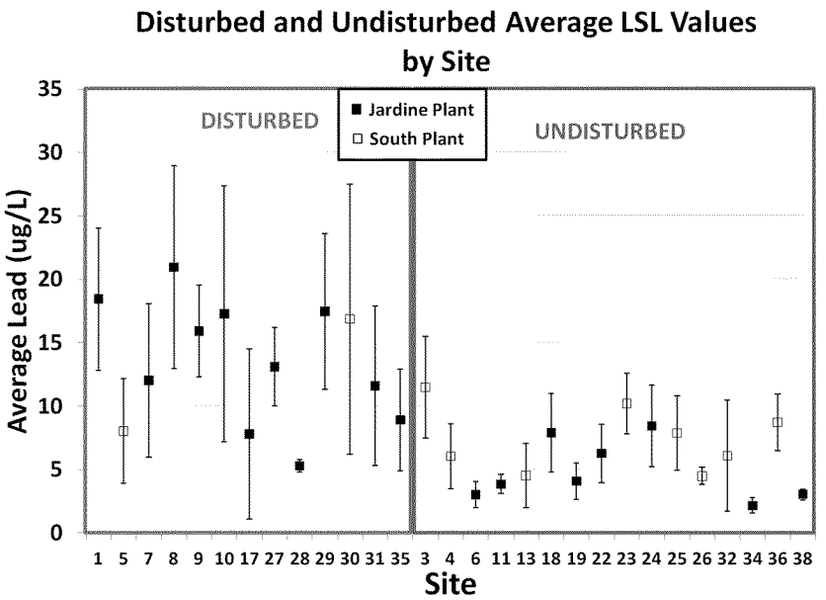

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Figure S3	Photograph of LSL Segment	Table S3	Summary of Samples Collected
Figure S4	Photograph of Corroded Galvanized Pipe	Table S4a	First-draw and Second-draw (45 second flushed) Sample Results
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Figure S6	Graph of Four Metals for Site 9	Table S5	June Sequential Sampling Results
Figure S7	City B Sampling Instructions	Table S6a	Sept/Oct Sequential Sampling Results Used in Analyses
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Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

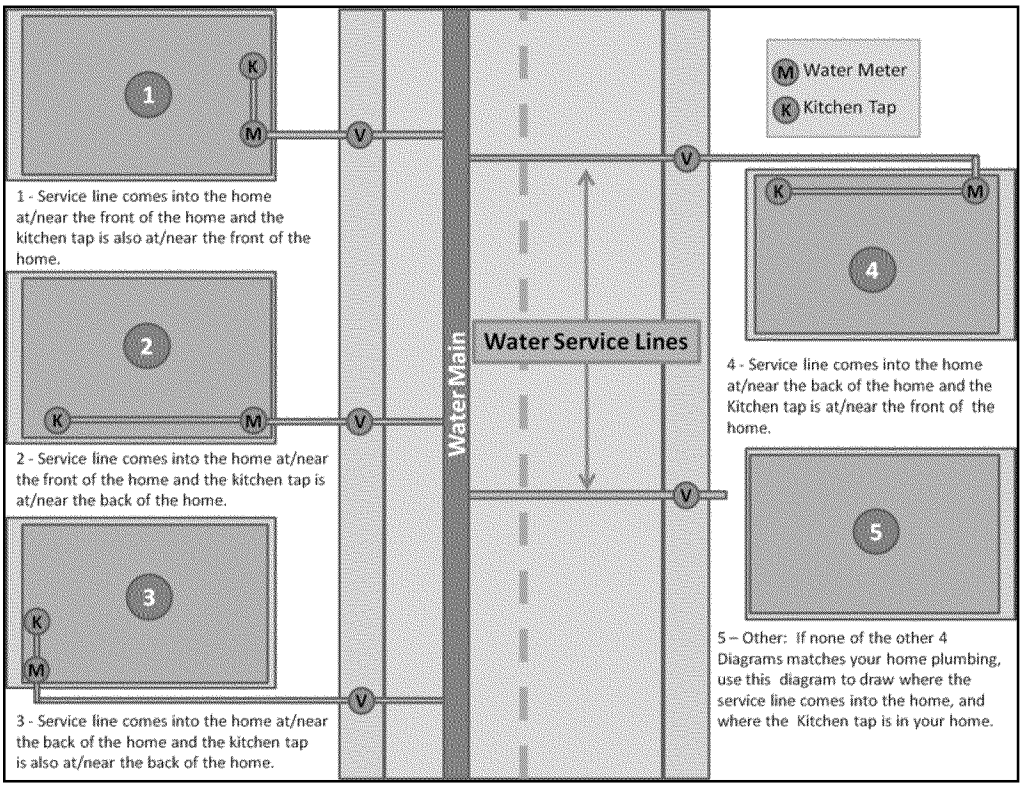


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

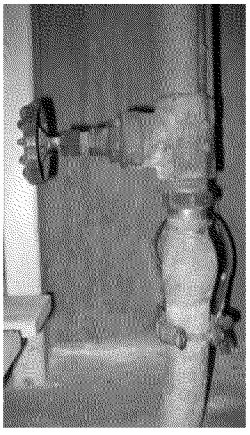


Figure S2: LSL Bulb

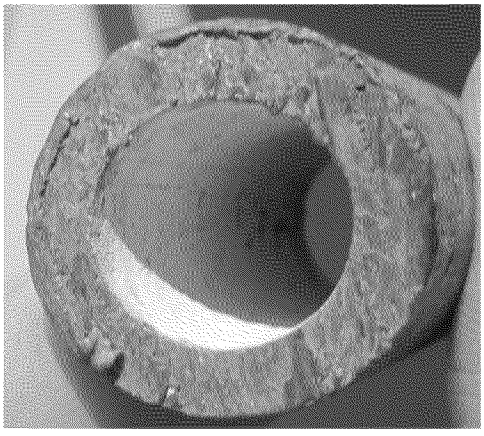


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

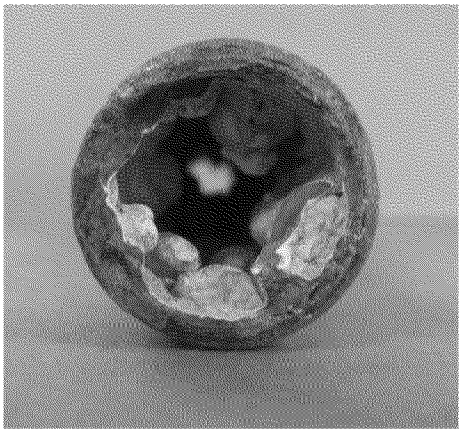


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

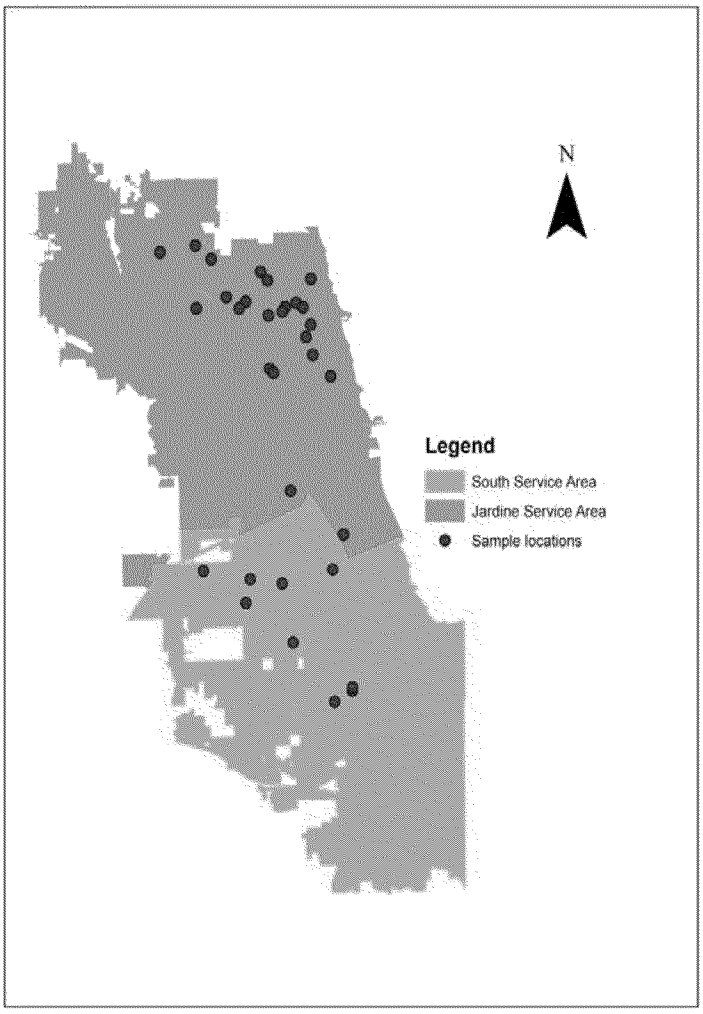


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

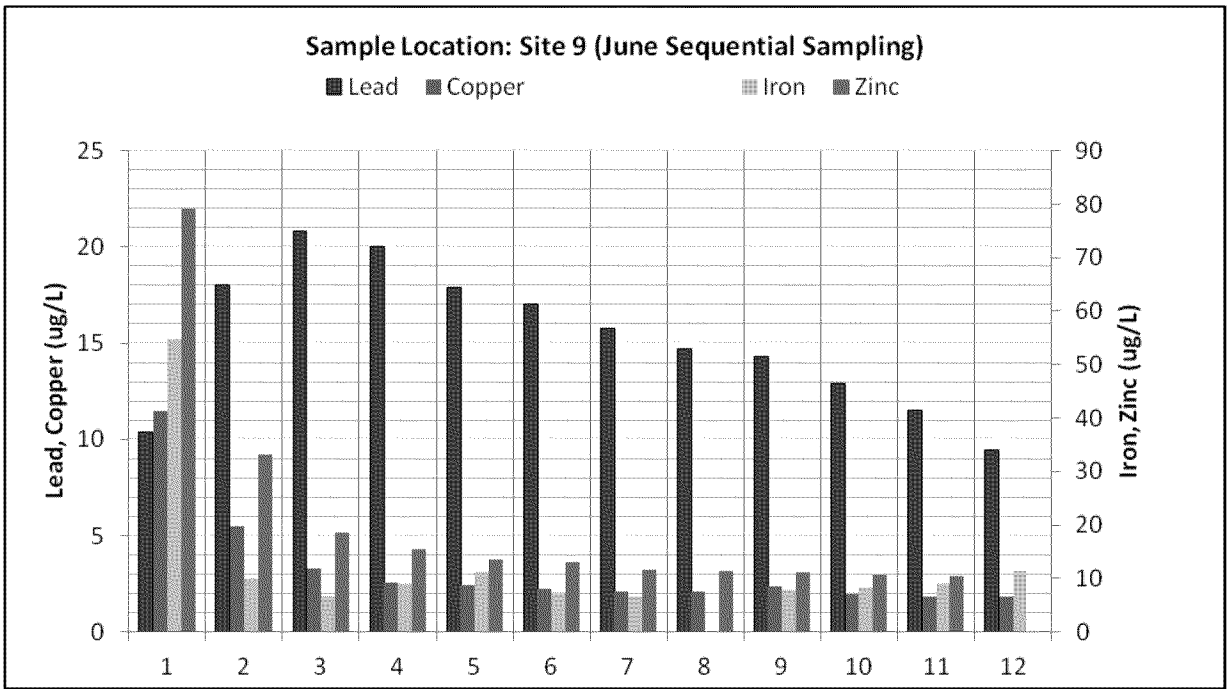


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

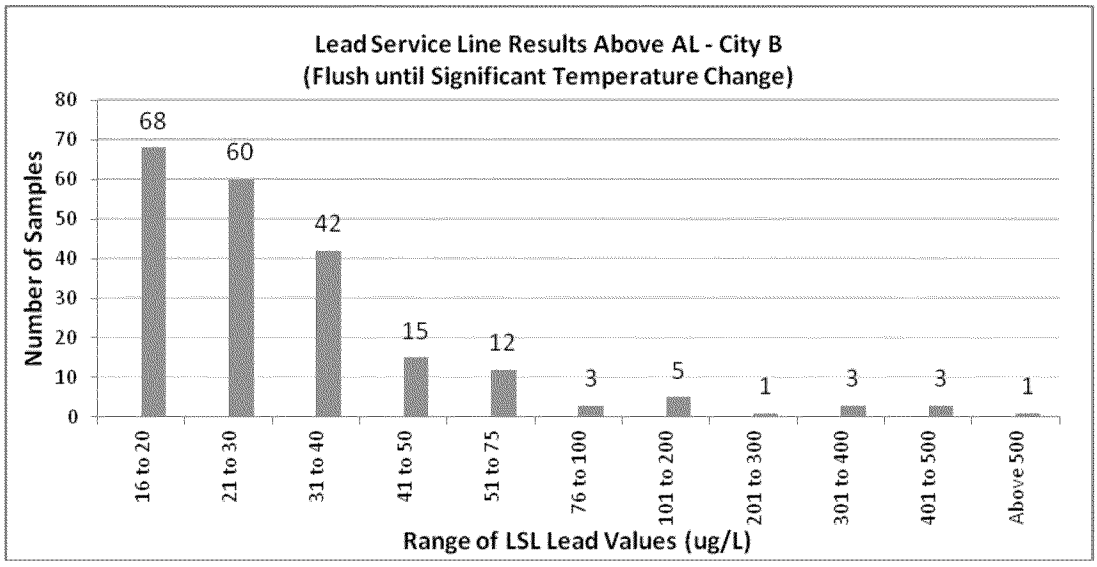
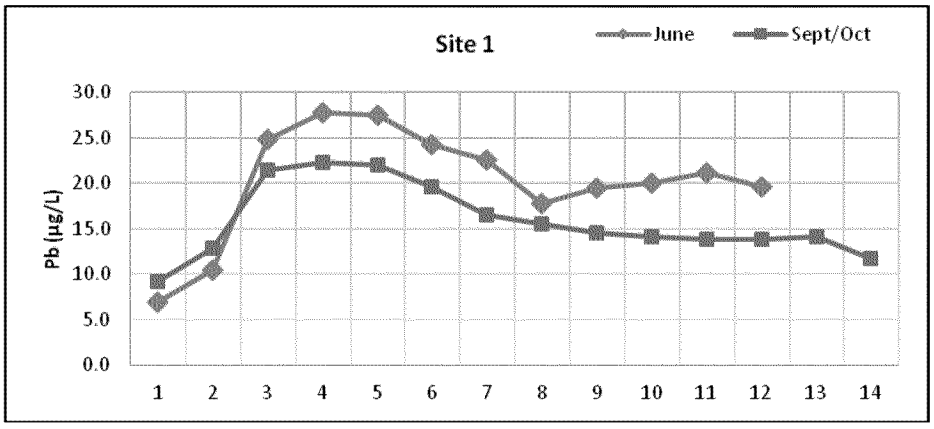


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

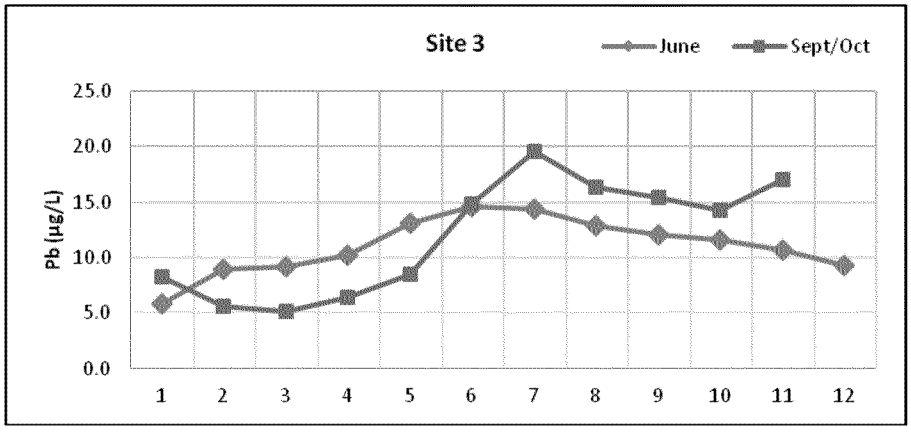
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

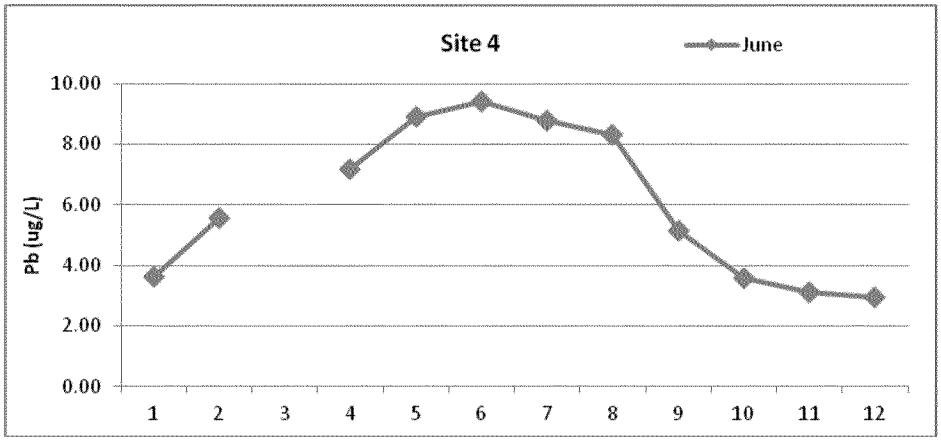
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

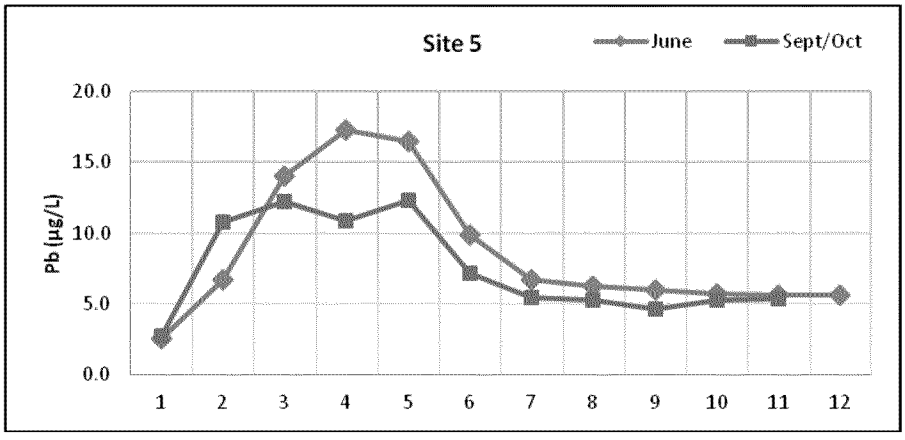
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

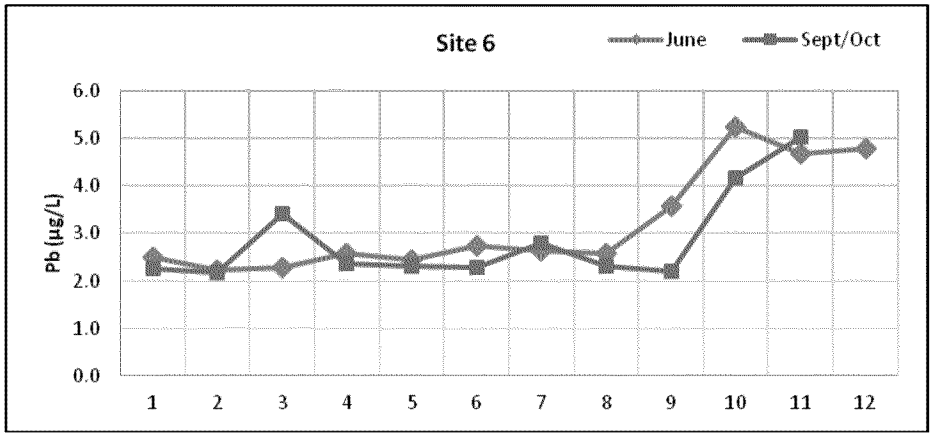
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

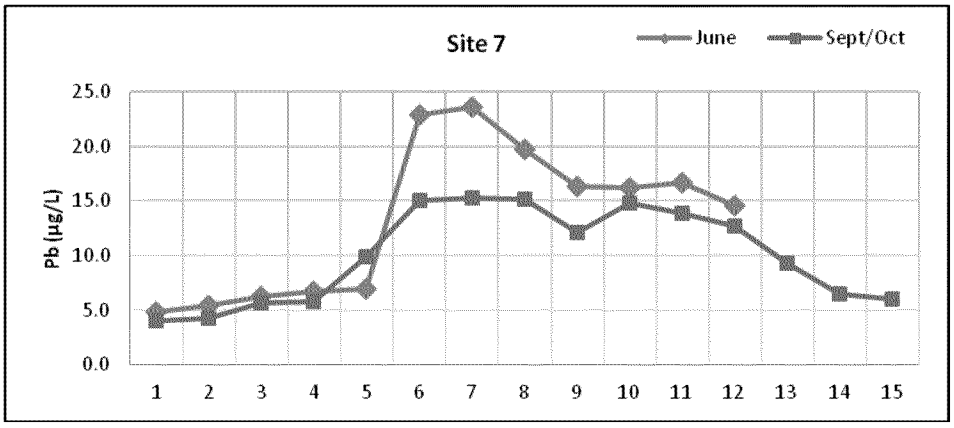
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

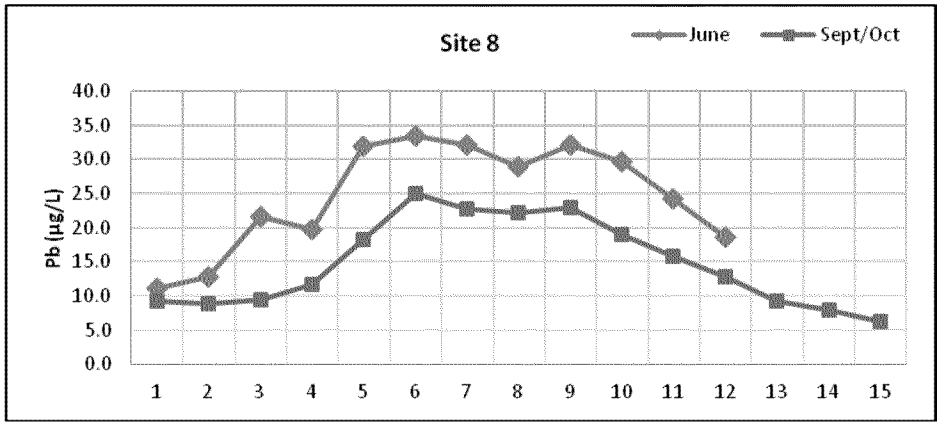
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

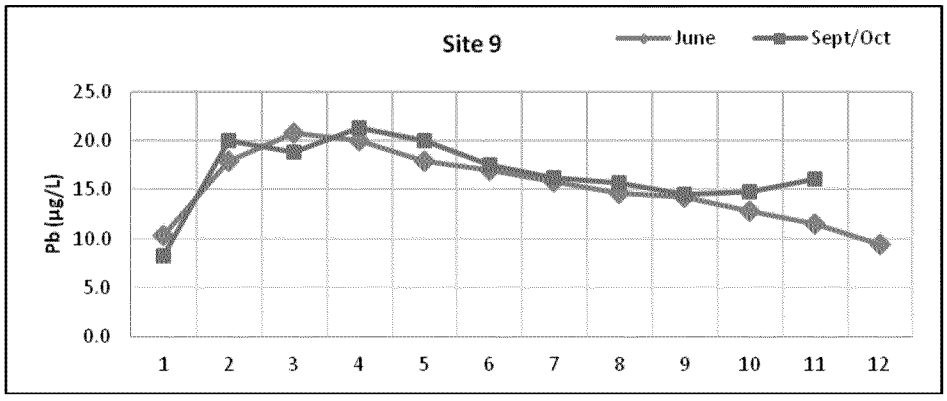
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

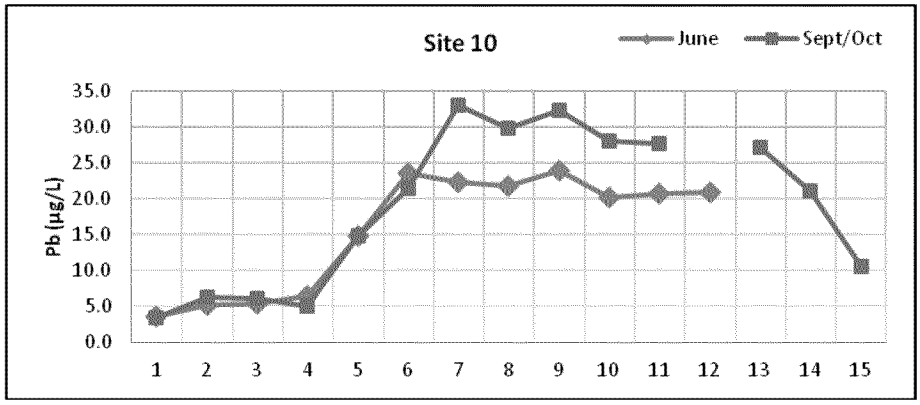
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

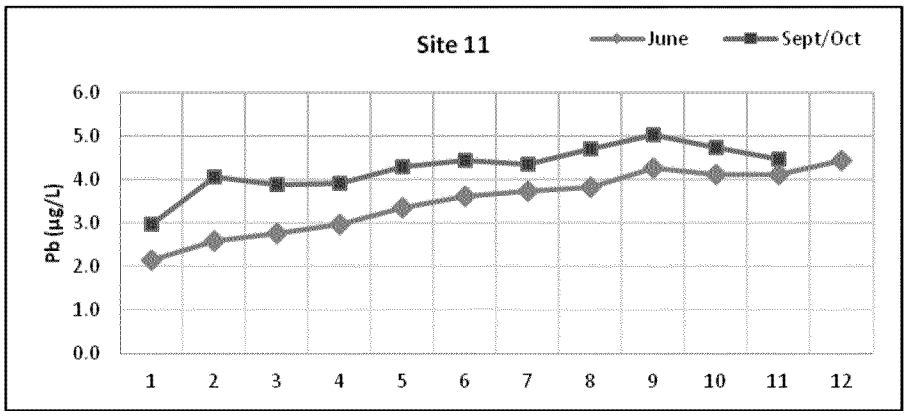
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

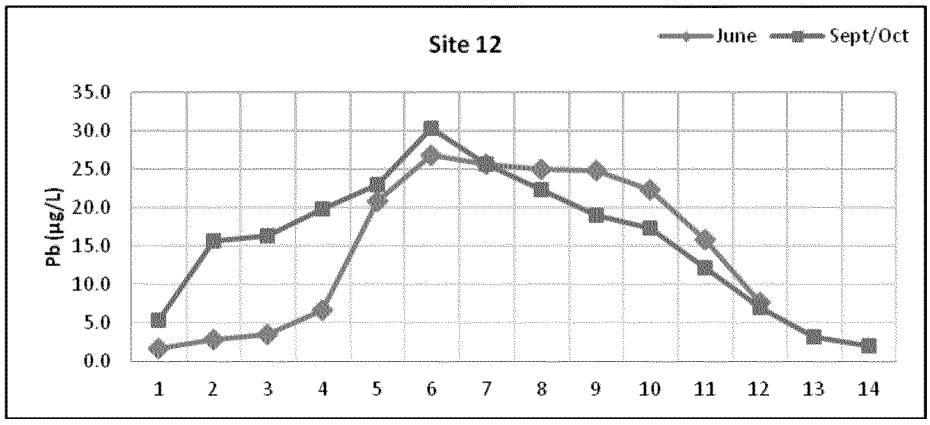
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

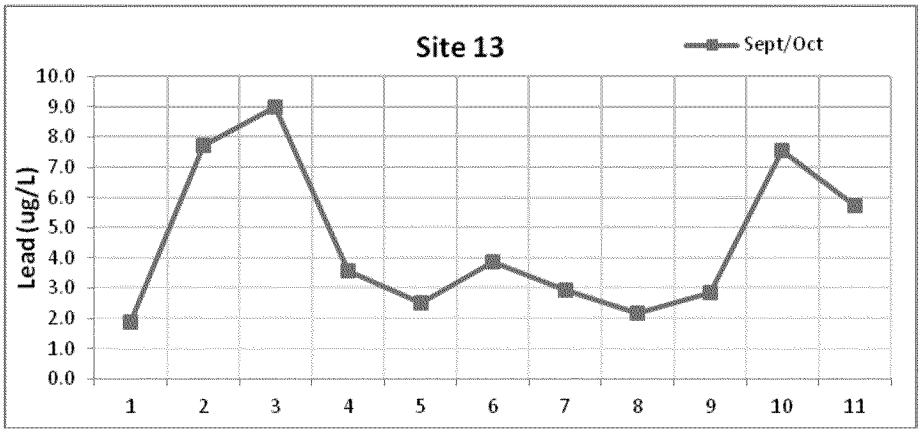
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

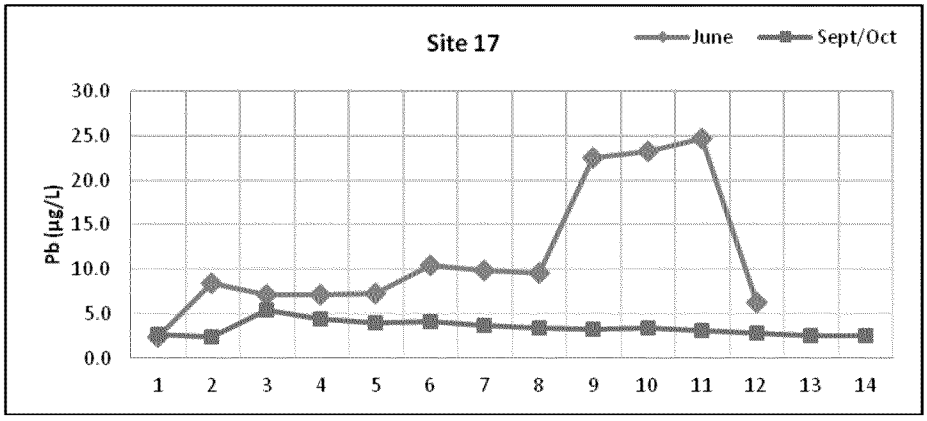
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

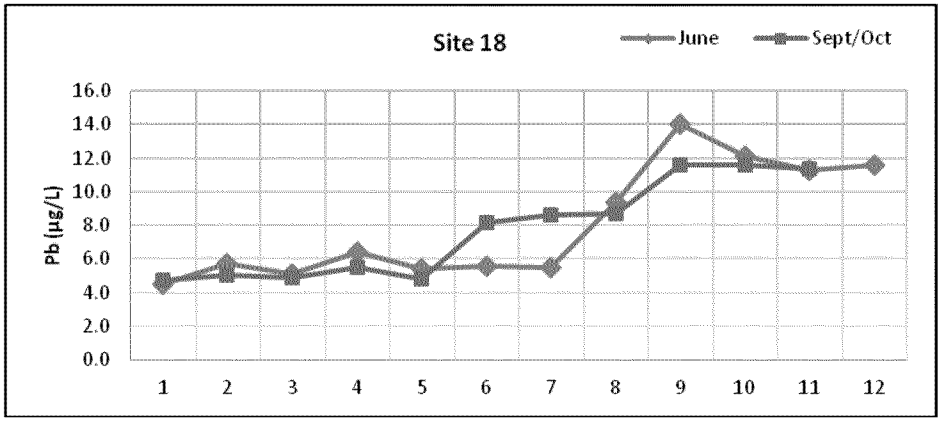
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

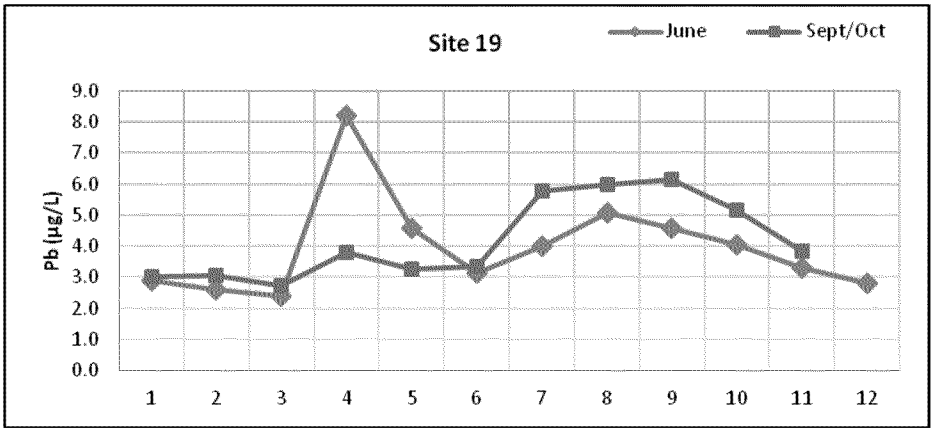
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

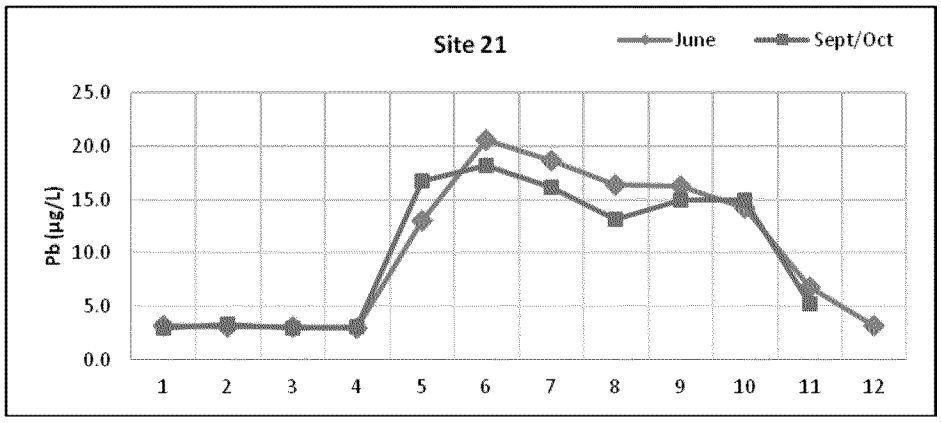
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

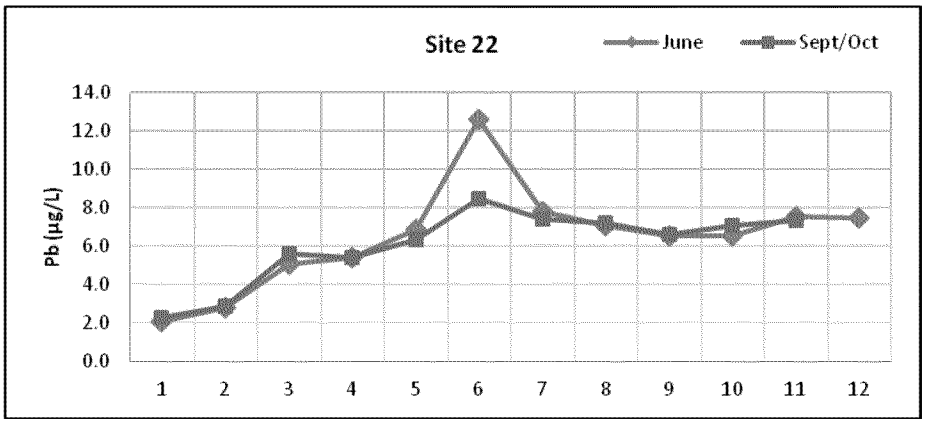
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

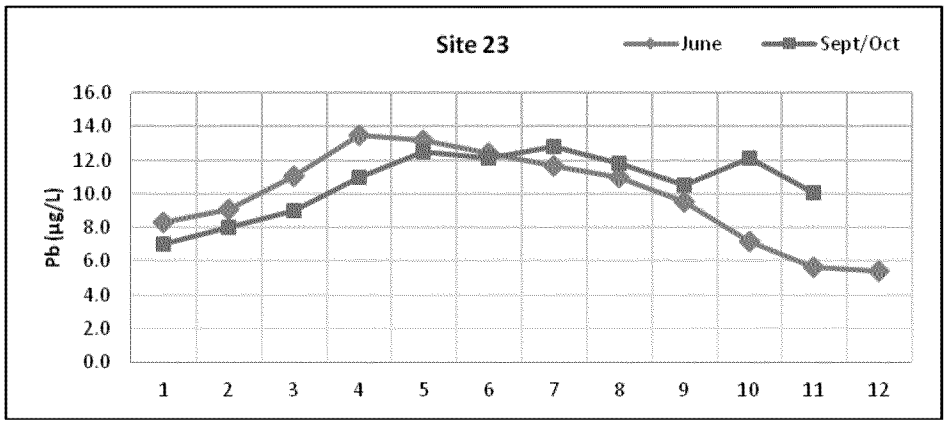
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

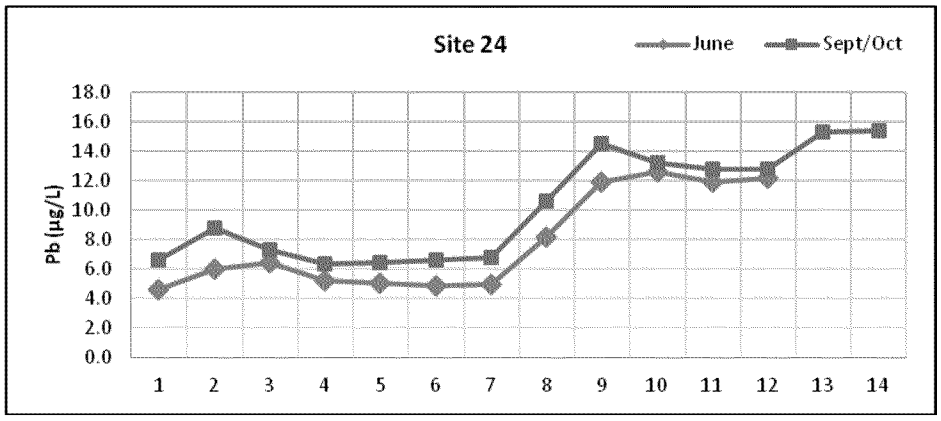
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

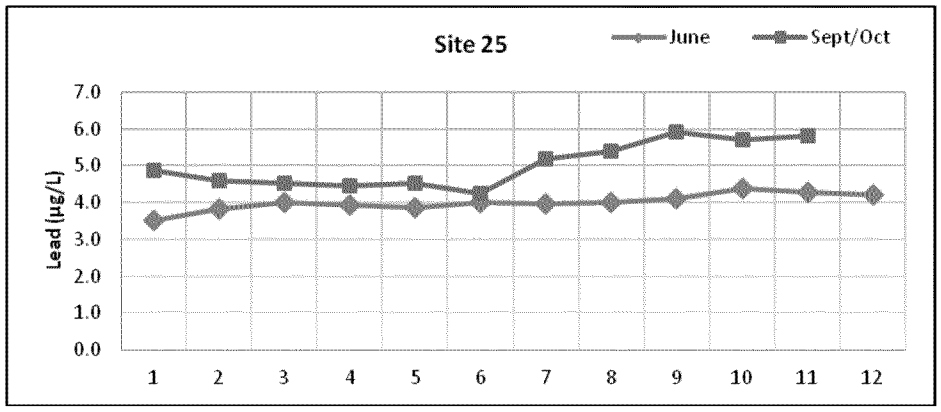
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

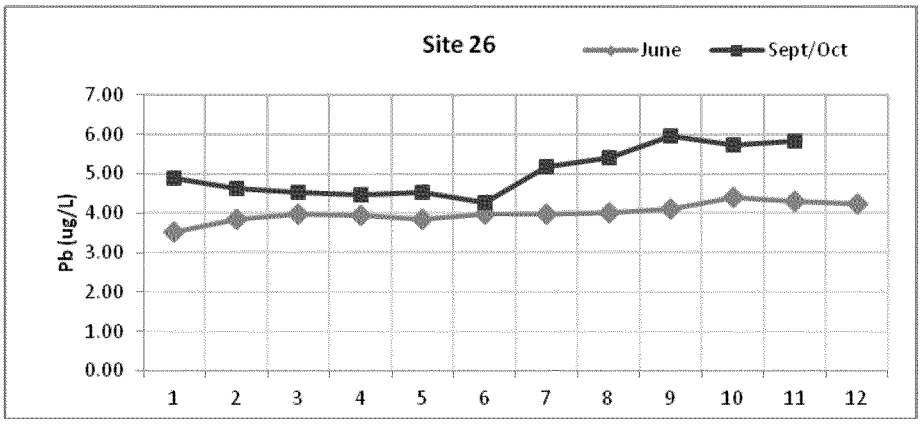
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

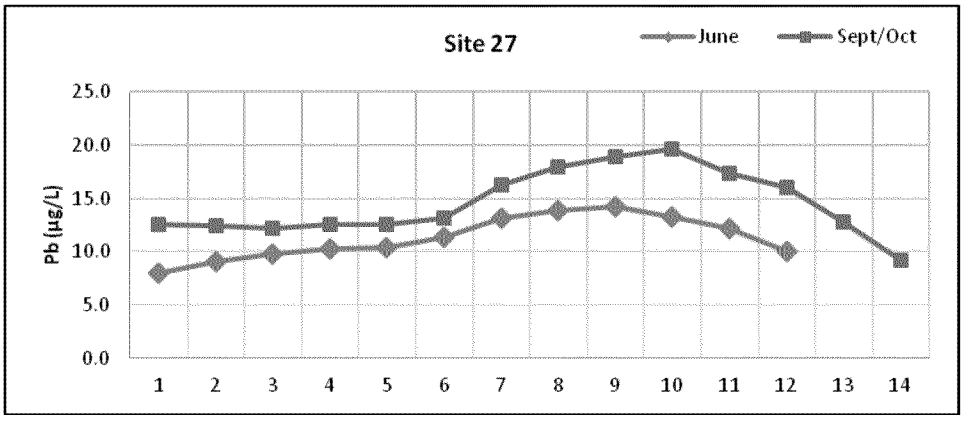
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

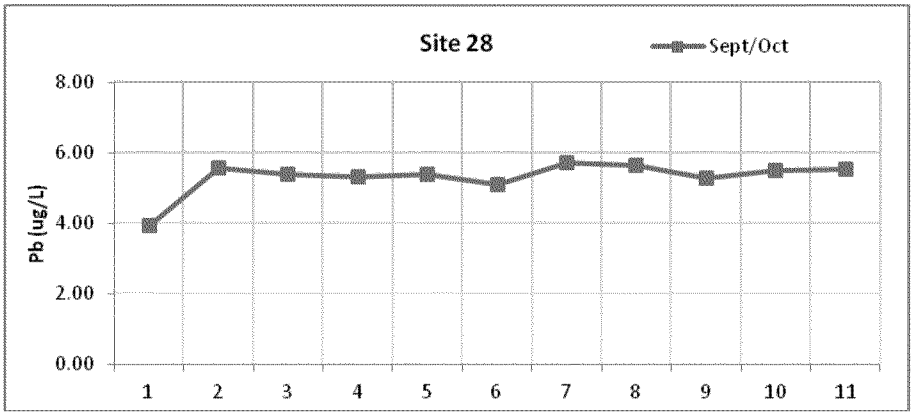
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

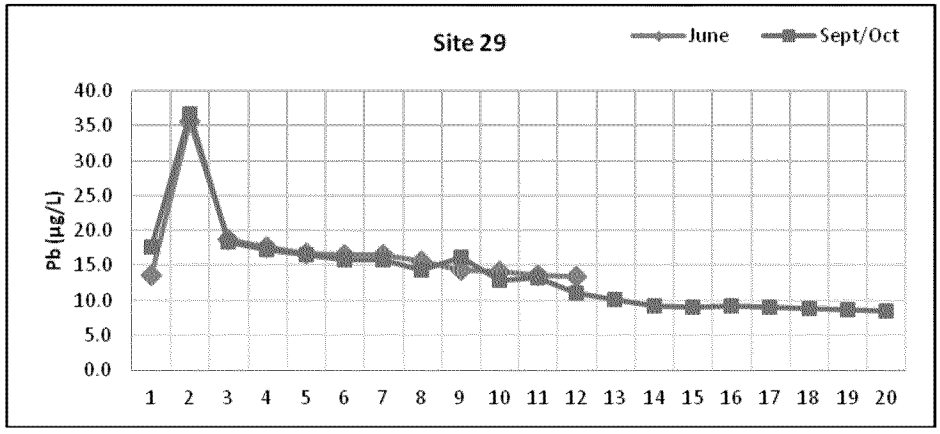
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

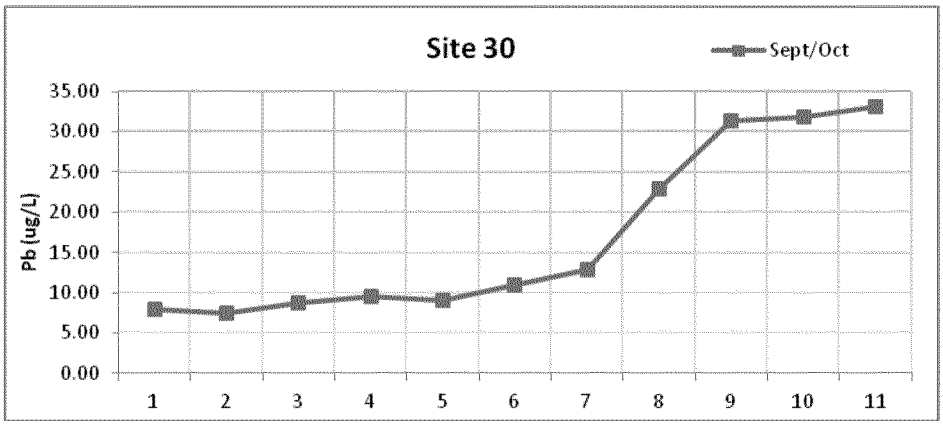
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

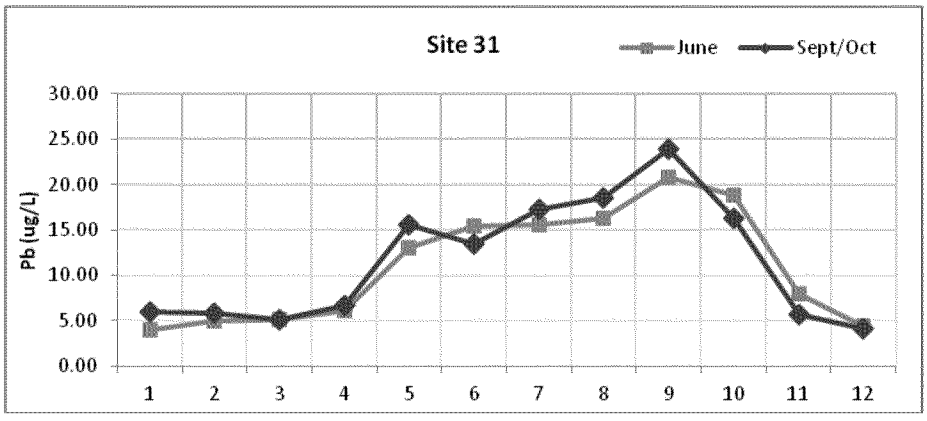
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

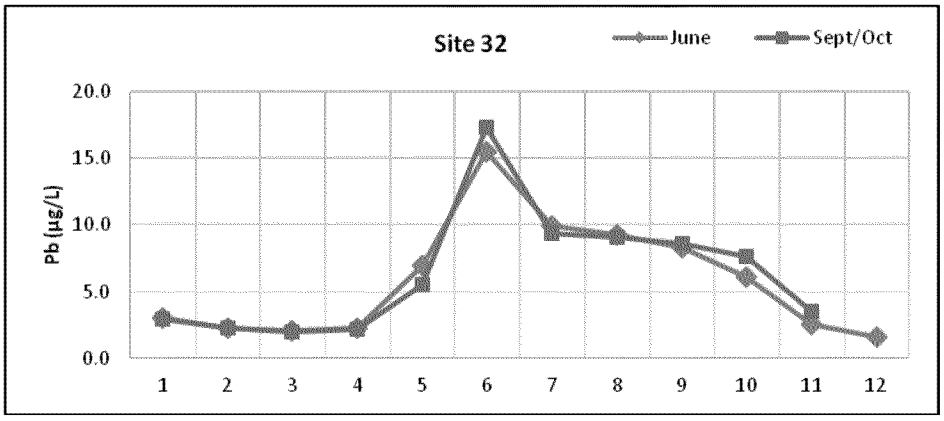
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

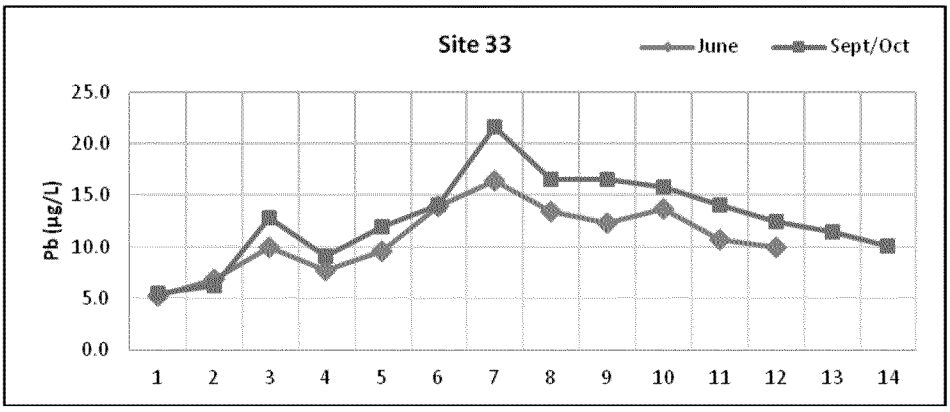
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

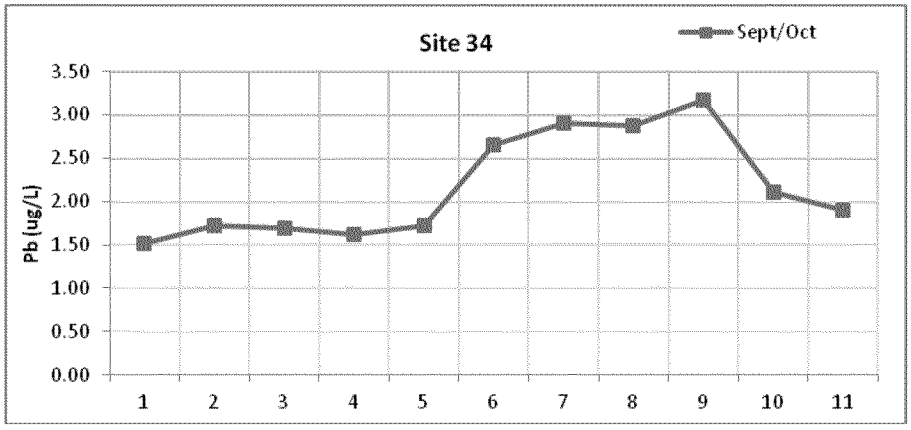
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

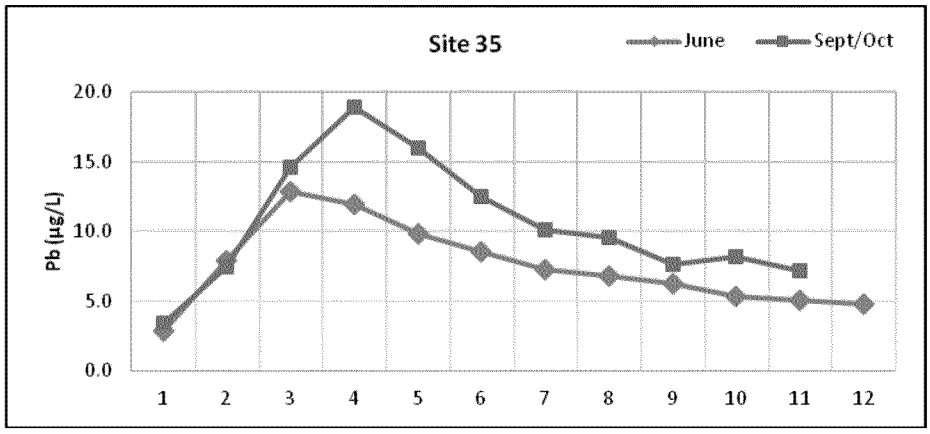
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

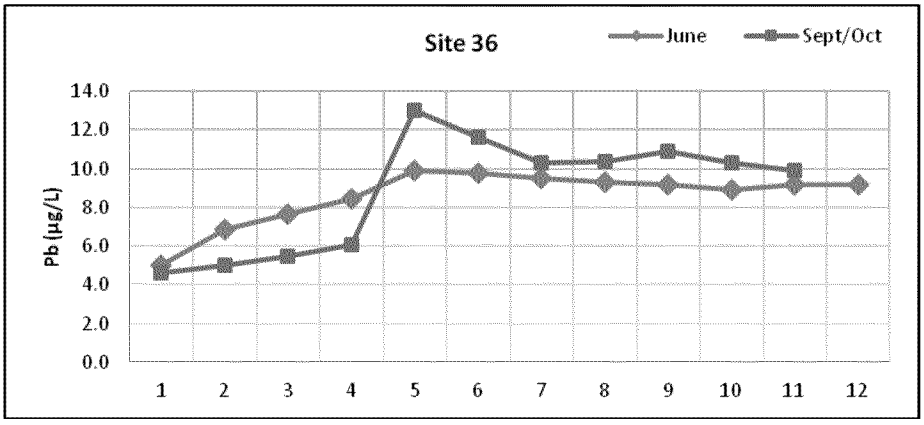
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

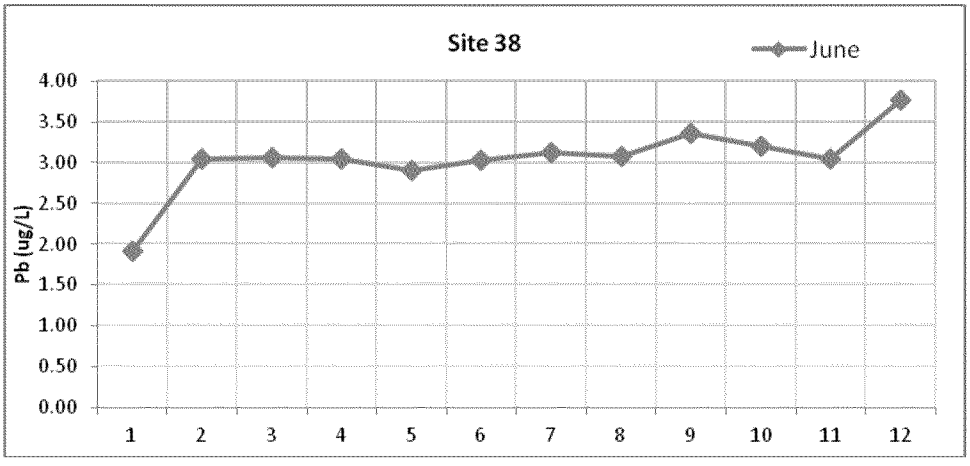
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

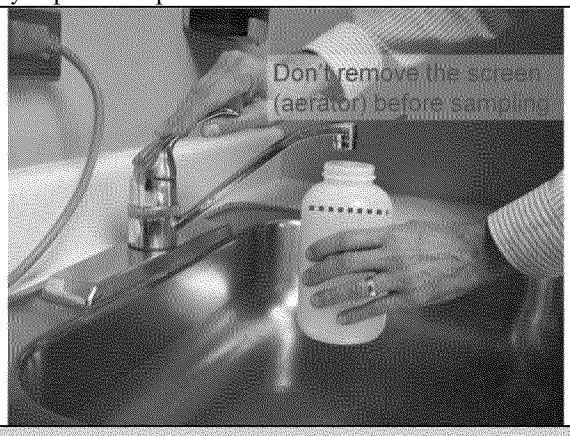
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

Literature Cited/References

1. Triantafyllidou, S.; Edwards, M., Galvanic corrosion after simulated small-scale partial lead service line replacements. *Journal American Water Works Association* **2011,** *103*, (9), 85-+.
2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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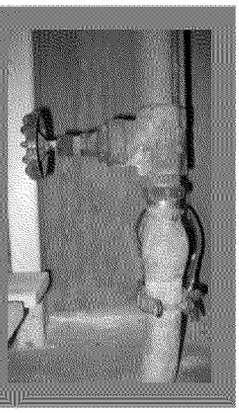
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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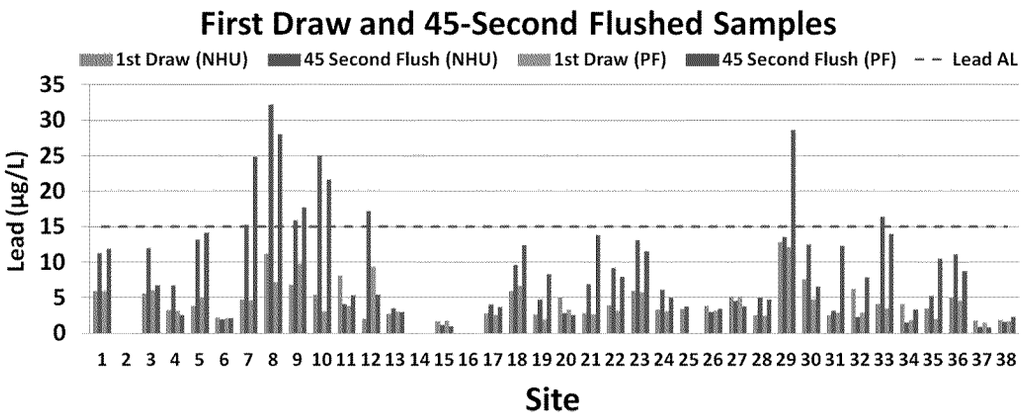

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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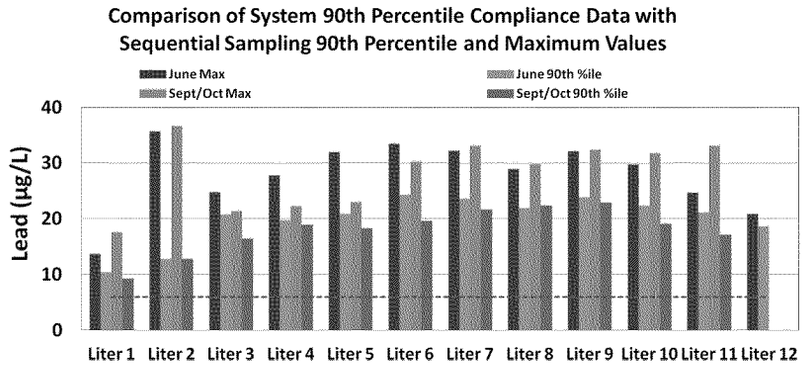

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

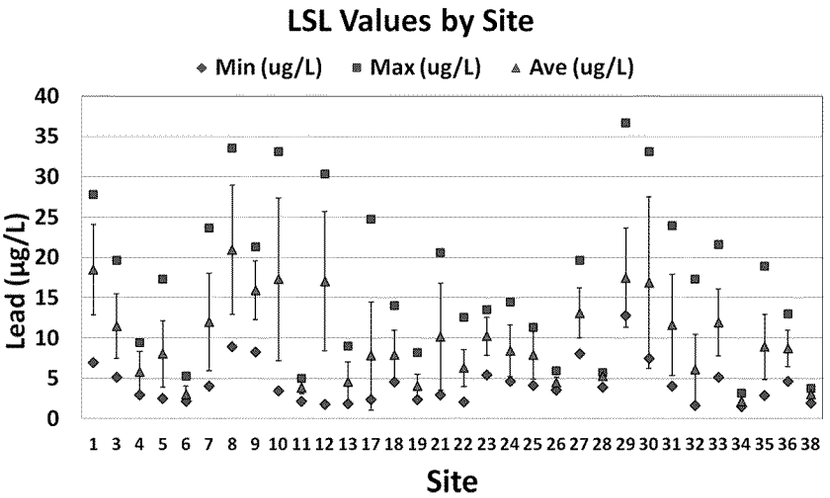

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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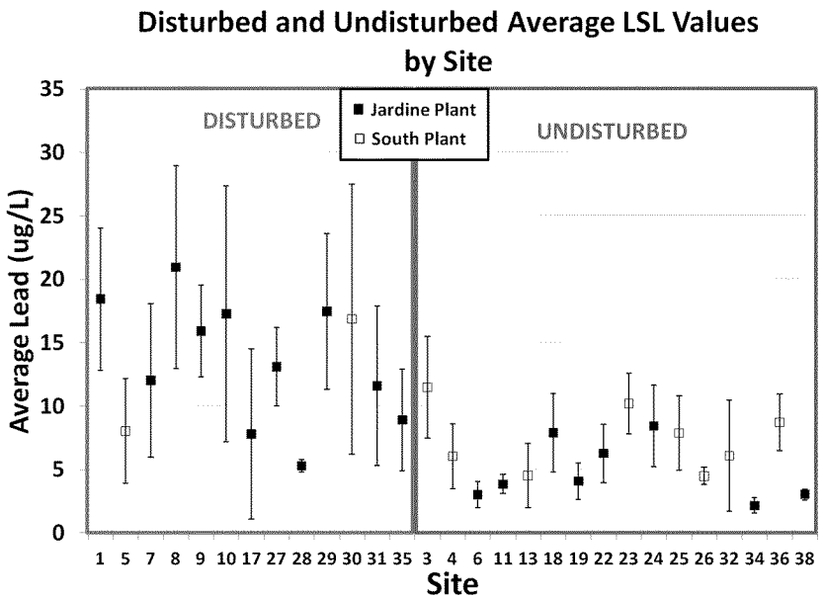

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

(Supporting Information for Manuscript ID: es-2013-003636)

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The supporting information provides additional background information, summaries and graphics for the underlying data used in the study.

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Figure S45	Sept/Oct Sample Collection and Reporting Form		

Background

The Lead and Copper Rule (LCR) is a treatment technique regulation that requires all public water systems to optimize corrosion control and utilizes tap sampling for lead and copper to determine whether additional actions need to be taken by the system. It is important to note that the sampling conducted under the LCR is not designed to evaluate individual consumers' lead exposure or risk and that the lead action level (AL) was not established as a health-based number. The lead AL is the level which EPA determined in 1991 that systems could feasibly meet, taking into account the available treatment technologies and the cost of those treatment technologies. The lead AL should not be viewed or used as a threshold value to determine whether the water is safe or unsafe to drink, and it should be reiterated that the EPA and CDC have determined that there is no safe level of lead exposure (i.e., no level at which there is not an adverse effect).

Tap sampling conducted under the LCR is intended to measure the amount of lead and copper corrosion that is occurring in public water systems using worst-case site selection and a specified sampling protocol. The sampling protocols in the current LCR were established in 1991, based on the existence of many potential sources of lead throughout the water distribution system, including lead service lines connecting the water main to the homes, leaded-solder used to join copper pipe, and leaded-brass devices, such as meters, brass connectors and shut-off valves, faucets and fixtures. The current LCR sampling requirements are prescriptive and based on the relative significance of lead sources in 1991. The sequential sampling protocol (described below, and in the accompanying paper) that resulted in capturing the highest lead, as well as the sample results themselves, are not allowed to be used in the current compliance calculation.

The LCR utilizes a combination of: worst-case site selection (sites expected to yield the highest lead results); sampling protocols used to capture the highest lead; and repeated sampling at the same sites in order to measure the level of lead corrosion that is occurring throughout the water distribution system. Utilizing this sampling structure allows U.S. EPA to keep the sampling burden on public water systems manageable, while still accomplishing the objectives of the sampling under the LCR. Absent these key components, the number of samples needed to accurately assess system-wide corrosion would necessarily need to increase substantially to accomplish the objectives of the LCR.

The action level for lead is 0.015 mg/L, but is presented here as 15 μg/L for the purpose of using consistent units for the data. An exceedance of the lead AL based on the sampling triggers specific actions that a public water system must undertake to protect public health, such as installing or adjusting corrosion control treatment and providing public education. Additionally, where the corrosion control treatment has proven ineffective at lowering lead levels below the lead AL, the removal of lead service lines is triggered. There are many different corrosion mechanisms and factors that govern lead corrosion. The selection of sampling sites, sampling protocol, and site conditions are essential components for evaluating the level of corrosion that is occurring in the distribution system, regardless of the mechanism(s) or contributing factor(s). It is therefore critically important that the sampling protocol accurately portray the level of corrosion that is occurring.

Lead Service Line and Plumbing Information

As part of the sampling protocol, residents were asked to provide a plumbing profile (figure S1), describing their internal plumbing, and identifying the location of the kitchen tap, and shut-off valve/meter.

Volunteer ID: _____

Home Plumbing and Service Line Diagrams

Below there are 4 diagrams for common household plumbing configurations and the 5th diagram is blank. Please review the diagrams and select the diagram that best matches the plumbing configuration for your home. Each of the diagrams shows where the water service line comes into the home and where the kitchen tap is located. If none of the four diagrams matches your home, use the blank diagram (number 5) to draw where the water service line comes into your home and where your kitchen tap is located. If you do not know where the service line comes into the home, you can note that in your Home Plumbing description below.

Note: Some homes have water meters and some do not. On the diagrams below, if you do not have a water meter, pick the diagram that matches where your service line comes into your home and where the kitchen tap is, and cross out the meter symbol

Home Plumbing Description: In the space below, please describe your home plumbing as best you can, from the point at which the water service line comes into your home to the location of your kitchen tap (length of pipe, diameter of pipe, pipe material, etc.):

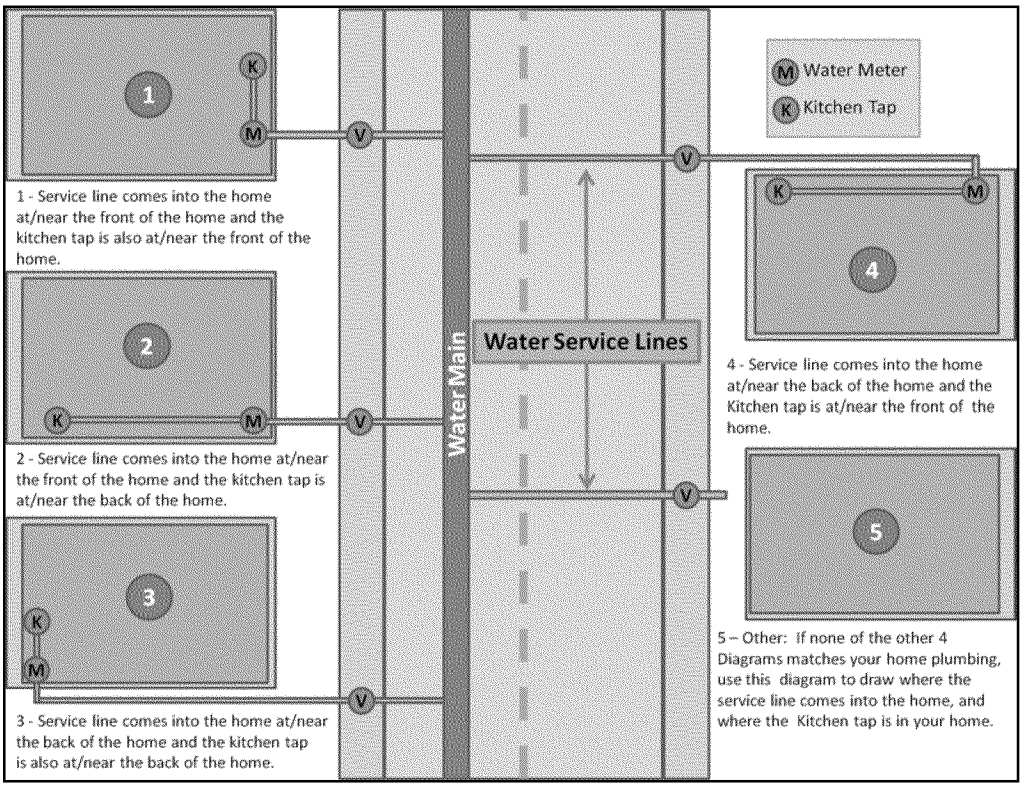


Figure S1: Plumbing Profile Diagram

Table S1 contains a summary of the LSL information for each sampling site. Due to the site-specific plumbing characteristics, the liter which first begins to capture LSL water at each site was expected to be variable, as was the liter which would begin to collect uncontaminated water from the water mains. The study findings regarding whether the current sampling protocol is capturing the corrosion that is occurring are not affected by this limitation.

Site	LSL Length ft (meters)	LSL End Point	Site	LSL Length ft (meters)	LSL End Point
1	89 (27.1)	BFW	22	65 (19.8)	IFW
3	73 (22.3)	IFW	23	66 (20.1)	IFW
4	Unknown	Unknown	24	56 (17.1)	IFW
5	80 (24.4)	IBW	25	70 (21.3)	IFW
6	60 (18.3)	IFW	26	66 (20.1)	IFW
7	59+ (18.0+)	BFW	27	47+ (14.3+)	Unknown
8	57 (17.4)	IFW	28	61+ (18.6+)	Unknown
9	102 (31.1)	BFW	29	159 (48.5)	BFW
10	48+ (14.6+)	IFW	30	49+ (14.9+)	Unknown
11	50 (15.2)	IFW	31	71+ (21.6+)	IFW
12	53 (16.2)	IFW	32	43 (13.1)	IFW
13	49+ (14.9+)	Unknown	33	43+ (13.1+)	IFW
17	58+ (17.7+)	Unknown	34	Unknown	Unknown
18	76 (23.2)	IFW	35	80 (24.4)	BFW
19	63(19.2)	IFW	36	110 (33.5)	IBW
21	46 (14.0)	IFW	38	51 (15.5)	IFW

IFW = LSL ends just inside the front wall
IBW = LSL ends just inside the back wall
BFW = LSL ends at an unknown distance beyond the front wall
+ = Indicates that the LSL was measured from the water main to the front the home, and it is not known whether the LSL extends beyond the front wall of the home.

Table S1: LSL Lengths – The length of the LSLs for most sites were measured and are presented in this table. The LSLs for two sites (site 4 and site 34) were not measured.

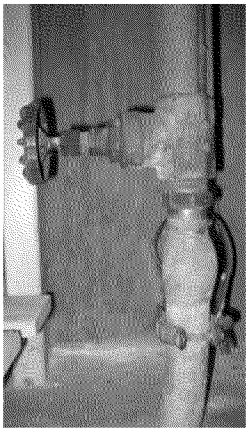


Figure S2: LSL Bulb

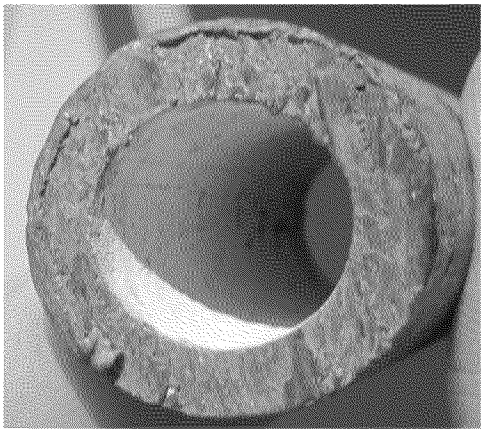


Figure S3: LSL segment (3/4 inch / 1.91 cm diameter)

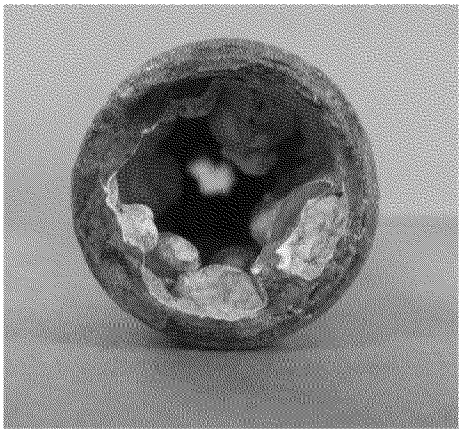


Figure S4: Severely corroded galvanized iron pipe.

Figure S2 shows a typical LSL in Chicago coming up from the foundation of the basement. The lead service line is a dull gray and easily scratched with a key. The soft LSL is typically soldered to the interior (household) plumbing, leaving a characteristic bulb. The LSL can also be connected to household pipe using a brass compression fitting.

Figure S3 is a close-up of a 3/4 inch (1.91 cm) diameter LSL, showing the thickness of a typical LSL.

Figure S4 is a cross-section of a severely corroded galvanized pipe from one of the sample sites. In this photograph the inner diameter is significantly reduced which affects the volume of water that will flow through the pipe in a set amount of time. For homes with corroded galvanized pipe, water will flow slower through the pipe and longer flushing times are generally needed to flush the lead from the plumbing.

City Information

Samples were collected from 32 single-family homes in Chicago with LSLs. Twenty-three homes were in the Jardine Plant service area and nine homes were in the South Plant service area.

Site #	Home Built	Service Area
01	1893	Jardine
03	1960	Jardine
04	1941	South
05	1901	South
06	1953	Jardine
07	1900	Jardine
08	1941	Jardine
09	1920	Jardine
10	1943	Jardine
11	1912	Jardine
12	1952	Jardine
13	1950	South
17	1907	Jardine
18	1953	Jardine
19	1912	Jardine
21	1938	Jardine
22	1924	Jardine
23	1944	South
24	1906	Jardine
25	1917	South
26	1954	South
27	1891	Jardine
28	1932	Jardine
29	1890	Jardine
30	1954	South
31	1923	Jardine
32	1923	South
33	1927	Jardine
34	1915	Jardine
35	1900	Jardine
36	1957	South
38	1927	Jardine

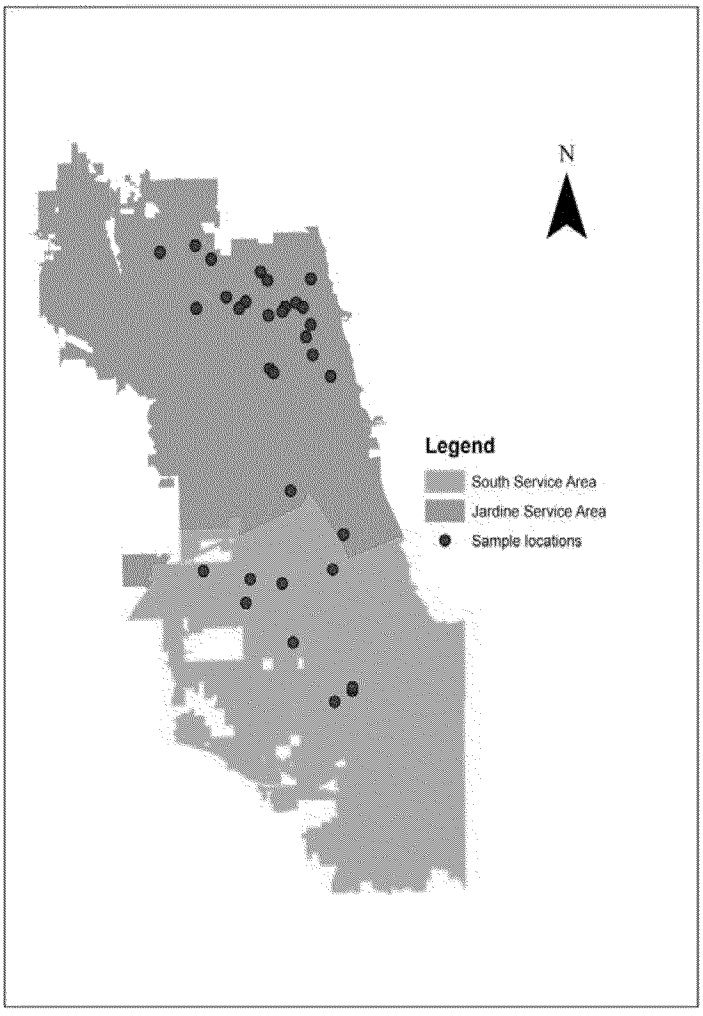


Figure S5: Home age and plant service areas for sampling locations

Table S2 contains a summary of the City's compliance monitoring data for lead. The City exceeded the lead AL only once, during the July-December 1992 compliance monitoring period.

City of Chicago (1992 – 2010) 90th Percentile Lead Values (µg/L)			
Monitoring Period Begin	**Monitoring Period End**	**Number of Samples**	**90th Percentile Value**
1/1/2008	12/31/2010	50	6
1/1/2005	12/31/2007	50	6
1/1/2002	12/31/2004	50	4
1/1/1999	12/31/2001	50	7
1/1/1999	12/31/1999	50	8
1/1/1998	12/31/1998	53	14
7/1/1997	12/31/1997	100	11
1/1/1997	6/30/1997	100	10
1/1/1993	6/30/1993	100	13
7/1/1992	12/31/1992	120	**20**
1/1/1992	6/30/1992	100	10

Table S2: City of Chicago 90th Percentile Compliance Values (1992 – 2010)

Laboratory and Analytical Information

All samples were inspected for visible particulates prior to delivery to the laboratory. In light of the significant increase in visible particulate in the final round of monitoring, the presence of fine particulates that would readily dissolve in the nitric acid preservative should not be discounted. Samples collected during the final round of monitoring coincided with the Fire Department's annual valve exercising. Colloidal lead may explain some of the variability in lead levels between the June and Sept/Oct rounds.

Laboratory blanks, laboratory fortified blanks and laboratory fortified samples were run at a frequency of at least one per twenty samples. Laboratory blanks run with the samples did not have any detections of lead above the reporting limit and all Laboratory fortified blanks and laboratory fortified samples had recoveries greater than 90%.

All laboratory instrumentation was inspected and maintained according to Chicago Regional Laboratory maintenance protocols, and calibrated daily according to Chicago Regional Laboratory standard operating procedures.

The Chicago Regional Lab Quality Assurance (QA) Contact performed a data quality assessment on the results based on laboratory blanks, laboratory fortified blanks and matrix spikes. The QA Contact identified no biases in the sample results due to these quality control measurements.

Sampling Summaries

Sample site summary table - A summary table of the types of samples collected at each site, for each sampling protocol is presented in Table S3 below. The highlighted rows for Sites 2,

14, 15, 16 & 37 were confirmed not to have LSLs and Site 20 is the same residence as Site 21 (Kitchen tap and bathroom tap). Following the first round of sampling, Site 20 (bathroom tap) was no longer sampled, to maintain consistency of using kitchen taps across all sites. Only sample results from LSL sites are presented and analyzed in the study paper. The first liter of the sequential samples in June and Sept/Oct also serve as the PF first-draw samples.

Summary of Samples Collected at Each Site							
Site #	Total # Samples	Mar/April		June	Sept/Oct		
		Day 1	Day 2	Day 1	Day 1	Day 2	Day 3
01	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
02	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
03	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
04	16	A, C	B, D	E-11 samples	DNS	DNS	DNS
05	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
06	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
07	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
08	35	A, C	B, D	E-12 samples	A	E-15 samples	F, G, H
09	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
10	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
11	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
12	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
13	16	A, C	B, D	DNS	A	E-11 samples	DNS
14	4	A, C	B, D	DNS	DNS	DNS	DNS
15	4	A, C	B, D	DNS	DNS	DNS	DNS
16	4	A, C	B, D	DNS	DNS	DNS	DNS
17	34	A, C	B, D	E-12 samples	A	E-14 samples	F, G, H
18	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
19	27	A, C	B, D	E-12 samples	DNS	E-11 samples	DNS
20	4	A, C	B, D	DNS	DNS	DNS	DNS
21	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
22	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
23	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
24	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
25	16	A, C	B, D	E-12 samples	DNS	DNS	DNS
26	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
27	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
28	30	A, C	B, D	DNS	A	E-11 samples	F, G
29	40	A, C	B, D	E-12 samples	A	E-20 samples	F, G, I
30	18	A, C	B, D	DNS	A	E-11 samples	F, G
31	31	A, C	B, D	E-12 samples	A	E-12 samples	F, G
32	28	A, C	B, D	E-12 samples	A	E-11 samples	DNS
33	33	A, C	B, D	E-12 samples	A	E-14 samples	F, G
34	18	A, C	B, D	DNS	A	E-11 samples	F, G
35	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
36	30	A, C	B, D	E-12 samples	A	E-11 samples	F, G
37	4	A, C	B, D	DNS	DNS	DNS	DNS
38	16	A, C	B, D	E-12 samples	DNS	DNS	DNS

A = NHU First-draw Sample
B = PF First-draw Sample
C = NHU 45-Second Flushed Sample
D = PF 45-Second Flushed Sample
E = Sequential Sample
F = 3-minute Flushed Sample
G = 5-minute Flushed Sample
H = 7-minute Flushed Sample
I = 10-minute Flushed Sample
DNS = Site did not sample

Table S3: Summary of samples collected at each site using each sampling protocol.

First-draw and 45-second flushed samples – Results for first-draw and 45-second flushed samples using the normal household use (NHU) and pre-flushed (PF) sampling protocols are presented in Table S4 below.

In addition to the first-draw samples, a 45-second flush sample was collected by running the water for 45 seconds immediately following the collection of the NHU first-draw and PF first-draw samples during the March/April sampling. Overall, the 45-second flush sample results were higher than the first-draw results, and yielded a higher percentage of results above the lead AL. A total of 32 NHU/45-second flushed and 32 PF/45-second flushed samples were collected, with 6 NHU 45-second flushed results above the lead AL (19%), and 5 PF/45-second flushed results above the AL (16%). The total number of 45-second flush sample results above the lead AL was 11 of 64 (17%); a percentage significantly higher than the first-draw results (2%).

First-draw and 45-second Flushed Sample Lead Results (µg/L)							
Site	A (Mar/Apr)	C (Mar/Apr)	B (Mar/Apr)	D (Mar/Apr)	B (June)	A (Sept/Oct)	B (Sept/Oct)
1	5.93	11.3	5.94	11.9	6.98	7.37	9.19
3	5.60	12.0	6.01	6.71	5.82	10.0	8.28
4	3.25	6.76	3.12	2.56	3.61	DNS	DNS
5	3.84	13.2	4.97	14.1	2.56	3.04	2.76
6	2.31	1.90	2.07	2.13	2.50	2.44	2.25
7	4.74	15.3	4.62	**24.9**	4.91	5.12	4.03
8	11.2	**32.2**	7.12	**28.0**	11.1	**17.5**	9.24
9	6.82	**15.9**	9.80	**17.7**	10.4	15.3	8.29
10	5.46	**25.0**	3.06	**21.6**	3.70	4.98	3.46
11	8.08	4.13	3.85	5.30	2.15	3.53	2.96
12	1.99	**17.2**	9.36	5.45	1.80	2.27	5.35
13	2.68	3.50	3.05	2.94	DNS	2.53	1.88
17	2.83	4.00	2.50	3.70	2.37	2.65	2.73
18	5.98	9.57	6.60	12.4	4.55	5.80	4.75
19	2.59	4.69	1.92	8.27	2.90	DNS	3.01
21	2.81	6.87	2.60	13.8	3.16	4.13	2.99
22	3.91	9.19	3.36	7.93	2.06	3.21	2.29
23	5.97	13.1	5.80	11.5	8.30	9.16	7.02
24	3.33	6.10	3.05	4.98	4.63	7.57	6.62
25	3.41	3.75	ND	ND	4.28	DNS	DNS
26	3.89	3.02	3.12	3.45	3.51	4.53	4.88
27	5.19	4.53	5.36	3.76	8.06	8.30	12.6
28	2.51	4.99	2.47	4.70	DNS	4.26	3.94
29	12.8	13.5	12.1	**28.6**	13.7	1.9	17.6
30	7.56	12.5	4.72	6.52	DNS	8.39	7.88
31	2.53	3.16	2.92	12.3	4.03	4.67	5.97
32	6.18	2.29	2.90	7.82	3.08	3.36	2.94
33	4.25	**16.4**	3.51	14.0	5.18	5.55	5.52
34	4.12	1.51	1.88	3.30	DNS	2.07	1.52
35	3.53	5.28	2.04	10.5	2.86	5.02	3.44
36	5.11	11.1	4.56	8.76	5.02	5.88	4.61
38	1.87	1.60	1.66	2.30	1.92	DNS	DNS
Ave	4.76	9.23	4.25	9.74	4.82	5.73	5.45
n	32	32	32	32	28	28	29
A = NHU First-draw Sample B = PF First-draw Sample C = NHU 45-Second Flushed Sample				D = PF 45-Second Flushed Sample DNS = Site did not sample n = number of samples collected			

Table S4a: First-Draw and 45-Second Flushed Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Summary of NHU and PF First-Draw Results					
	NHU (Mar/Apr)	PF (Mar/Apr)	PF (June)	NHU (Sept/Oct)	PF (Sept/Oct)
90th %ile Pb Value (µg/L)	8	7	8	10	9
No. of Samples	32	32	28	29	30
No. > AL	0	0	0	2	1

Table S4b: Comparison of LCR-equivalent 90th percentile results using alternative first-draw protocols.

Sequential sampling results (June 2011) – The sequential sampling approach provided a more reliable (volumetric) method for assessing corrosion as compared to a flushed (time-based) approach. Attempting to characterize the flow at each site would require an evaluation of the plumbing materials and dimensions, as well as the condition of the plumbing materials at each site, is not a feasible or reliable protocol for compliance monitoring.

The results of the each liter in the sequential sampling conducted in June are tabulated below in Table S5 by site.

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
01	6.98	10.5	**24.8**	**27.8**	**27.5**	**24.3**	**22.6**	**17.8**	**19.5**	**20.0**	**21.1**	**19.6**
03	5.82	8.91	9.18	10.2	13.1	14.6	14.4	12.9	12.1	11.6	10.7	9.34
04	3.61	5.56		7.17	8.90	9.41	8.78	8.30	5.14	3.59	3.11	2.96
05	2.56	6.73	14.0	**17.3**	**16.5**	9.85	6.72	6.29	6.01	5.73	5.65	5.60
06	2.50	2.23	2.28	2.57	2.44	2.75	2.65	2.59	3.57	5.26	4.67	4.80
07	4.91	5.45	6.28	6.73	7.03	**22.9**	**23.6**	**19.7**	**16.3**	**16.2**	**16.7**	14.6
08	11.1	12.8	**21.6**	**19.7**	**32.0**	**33.5**	**32.2**	**28.9**	**32.1**	**29.7**	**24.2**	**18.7**
09	10.4	**18.0**	**20.8**	**20.0**	**17.9**	**17.0**	**15.8**	14.7	14.3	12.9	11.5	9.48
10	3.70	5.20	5.39	6.49	14.9	**23.6**	**22.4**	**21.9**	**23.9**	**20.2**	**20.7**	**20.9**
11	2.15	2.58	2.76	2.97	3.36	3.61	3.73	3.82	4.28	4.11	4.11	4.43
12	1.80	2.95	3.55	6.69	**20.9**	**26.9**	**25.7**	**25.1**	**24.9**	**22.4**	**15.9**	7.80
17	2.37	8.46	7.12	7.20	7.27	10.5	9.91	9.56	**22.6**	**23.3**	**24.7**	6.30
18	4.55	5.73	5.12	6.43	5.41	5.62	5.5	9.38	14.0	12.1	11.3	11.6
19	2.90	2.62	2.41	8.22	4.58	3.16	4.02	5.07	4.57	4.06	3.31	2.82
21	3.16	3.12	3.08	2.97	13.0	**20.6**	**18.7**	**16.4**	**16.3**	14.2	6.78	3.21
22	2.06	2.82	5.11	5.42	6.89	12.6	7.80	7.11	6.52	6.55	7.55	7.45
23	8.30	9.06	11.1	13.5	13.2	12.4	11.7	11.0	9.55	7.16	5.69	5.41
24	4.63	6.06	6.43	5.24	5.06	4.91	5.02	8.21	11.9	12.6	11.9	12.2
25	4.28	4.28	4.15	4.23	6.82	10.9	11.3	10.9	10.1	9.68	9.17	8.82
26	3.51	3.83	3.99	3.93	3.86	3.99	4.00	4.01	4.12	4.39	4.30	4.23
27	8.06	9.13	9.84	10.3	10.4	11.4	13.10	13.9	14.2	13.3	12.2	10.1
29	13.7	**35.7**	**18.8**	**17.7**	**16.8**	**16.5**	**16.6**	**15.7**	14.4	14.1	13.7	13.4
31	4.03	5.03	5.14	6.17	13.1	**15.4**	**15.6**	**16.3**	**20.8**	**18.8**	7.91	4.48

June Sequential Sampling Results by Site/Liter (µg/L)												
	Liter											
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
32	3.08	2.29	2.07	2.28	6.95	15.5	9.91	9.27	8.30	6.12	2.60	1.65
33	5.18	6.85	10.0	7.74	9.61	13.9	**16.4**	13.5	12.3	13.7	10.7	9.95
35	2.86	7.89	12.9	11.9	9.85	8.59	7.28	6.82	6.23	5.34	5.02	4.83
36	5.02	6.90	7.68	8.46	9.90	9.81	9.51	9.34	9.19	8.93	9.20	9.19
38	1.92	3.04	3.06	3.04	2.91	3.03	3.12	3.07	3.36	3.21	3.04	3.76
Min	*1.80*	*2.23*	*2.07*	*2.28*	*2.44*	*2.75*	*2.65*	*2.59*	*3.36*	*3.11*	*2.60*	*1.65*
Max	*13.7*	***35.7***	***24.8***	***27.8***	***32.0***	***33.5***	***32.2***	***28.9***	***32.1***	***29.7***	***24.7***	***20.9***
Ave	*4.83*	*7.28*	*8.42*	*9.07*	*11.1*	*13.1*	*12.4*	*11.7*	*12.5*	*11.7*	*10.3*	*8.50*
90th %ile	*10.4*	*12.8*	***20.8***	***19.7***	***20.9***	***24.3***	***23.6***	***21.9***	***23.9***	***22.4***	***21.1***	***18.7***

Table S5: Summary of June Sequential Sampling Results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sequential Sampling Results (September and October 2011) – The results of the each liter in the sequential sampling conducted in September and October are tabulated below in Table S6 by site. Considerably more sample results contained visible particulates than in previous rounds. The presence of particulates may be a result of the Chicago Fire Department exercising valves during the time period when samples were being collected.

All sites collected at least 11 sequential samples, and some sites with high sample results in June collected additional samples. The additional sequential sample results are included here but were not included in the data analyses, since extra samples were collected only from sites with high lead. A review of the data, including and excluding these additional results was performed to ensure that a bias has not been introduced, and the review indicates that the study findings are not significantly affected by including or excluding the data. With the additional 39 samples included, a total of 80 of 358 sample results (22%) exceeded the lead AL. Using only samples 1 through 11 from each site, a total of 75 of 319 sample results (24%) exceeded the lead AL. For the purpose of the data analyses, the first liter sample from the sequential samples in June and Sept/Oct also serve as the first-draw PF sample.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)											
	Liter										
Site	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
01	9.19	12.8	**21.4**	**22.3**	**22.0**	**19.6**	**16.5**	**15.6**	14.5	14.2	13.8
03	8.28	5.58	5.17	6.43	8.46	14.9	**19.6**	**16.4**	**15.4**	14.3	**17.1**
05	2.76	10.8	12.2	10.9	12.3	7.21	5.49	5.24	4.65	5.30	5.40
06	2.25	2.18	3.43	2.37	2.30	2.28	2.81	2.32	2.20	4.16	5.03
07	4.03	4.27	5.74	5.75	9.87	15.1	15.3	15.2	12.1	14.8	13.9
08	9.24	8.95	9.45	11.8	**18.3**	**25.0**	**22.7**	**22.3**	**22.9**	**19.1**	**15.8**
09	8.29	**20.0**	**18.8**	**21.3**	**20.0**	**17.6**	**16.3**	**15.7**	14.6	14.8	**16.1**
10	3.46	6.27	6.23	5.05	14.8	**21.4**	**33.1**	**29.8**	**32.4**	**28.1**	**27.7**
11	2.96	4.05	3.90	3.91	4.30	4.44	4.35	4.71	5.02	4.75	4.47
12	5.35	**15.7**	**16.4**	**19.8**	**23.0**	**30.3**	**25.7**	**22.4**	**19.0**	**17.3**	12.2
13	1.88	7.73	9.01	3.57	2.53	3.85	2.96	2.17	2.85	7.55	5.74
17	2.73	2.38	5.45	4.41	4.07	4.09	3.72	3.42	3.35	3.42	3.17
18	4.75	5.09	4.91	5.53	4.81	8.17	8.61	8.67	11.6	11.6	11.4
19	3.01	3.07	2.75	3.80	3.25	3.37	5.80	6.01	6.15	5.18	3.83
21	2.99	3.35	3.03	3.04	**16.8**	**18.2**	**16.1**	13.2	14.9	15.0	5.24
22	2.29	2.86	5.60	5.39	6.32	8.49	7.42	7.20	6.64	7.09	7.36
23	7.02	8.00	8.99	11.0	12.5	12.1	12.8	11.8	10.5	12.1	10.1
24	6.62	8.84	7.30	6.38	6.45	6.59	6.82	10.6	14.5	13.2	12.8
26	4.88	4.61	4.52	4.46	4.52	4.26	5.18	5.40	5.94	5.72	5.82
27	12.6	12.4	12.2	12.5	12.5	13.1	**16.3**	**18.0**	**18.9**	**19.6**	**17.3**
28	3.94	5.58	5.39	5.32	5.39	5.11	5.73	5.65	5.30	5.49	5.55
29	**17.6**	**36.7**	**18.3**	**17.3**	**16.6**	**15.9**	**15.9**	14.3	**16.2**	12.8	13.2
30	7.88	7.46	8.67	9.54	9.09	11.0	12.9	**22.9**	**31.3**	**31.8**	**33.1**
31	5.97	5.82	5.20	6.72	15.6	13.4	**17.3**	**18.5**	**23.9**	**16.3**	5.70
32	2.94	2.24	2.03	2.22	5.50	**17.3**	9.42	9.07	8.63	7.64	3.50
33	5.52	6.26	12.8	9.09	12.0	14.1	**21.6**	**16.6**	**16.5**	**15.8**	14.1
34	1.52	1.72	1.69	1.62	1.73	2.66	2.91	2.87	3.17	2.10	1.90
35	3.44	7.42	14.6	**18.9**	**16.0**	12.5	10.1	9.56	7.60	8.18	7.21
36	4.61	5.01	5.51	6.11	13.0	11.6	10.3	10.4	10.9	10.3	9.93
Min	*1.52*	*1.72*	*1.69*	*1.62*	*1.73*	*2.28*	*2.81*	*2.17*	*2.20*	*2.10*	*1.90*
Max	***17.6***	***36.7***	***21.4***	***22.3***	***23.0***	***30.3***	***33.1***	***29.8***	***32.4***	***31.8***	***33.1***
Ave	*5.45*	*7.83*	*8.30*	*8.50*	*10.5*	*11.9*	*12.2*	*12.0*	*12.5*	*12.0*	*10.6*
90th %ile	*9.19*	*12.8*	***16.4***	***18.9***	***18.3***	***19.6***	***21.6***	***22.3***	***22.9***	***19.1***	***17.1***

Table S6a: Summary of September/October sequential sampling results used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Sept/Oct Sequential Sampling Results by Site/Liter (µg/L)									
	Liter								
Site	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**
01	13.9	14.1	11.7	--	--	--	--	--	--
03	--	--	--	--	--	--	--	--	--
05	--	--	--	--	--	--	--	--	--
06	--	--	--	--	--	--	--	--	--
07	12.7	9.29	6.52	6.03	--	--	--	--	--
08	12.8	9.34	7.93	6.27	--	--	--	--	--
09	--	--	--	--	--	--	--	--	--
10	--	**27.1**	**21.1**	10.7	--	--	--	--	--
11	--	--	--	--	--	--	--	--	--
12	6.98	3.28	2.04	--	--	--	--	--	--
13	--	--	--	--	--	--	--	--	--
17	2.84	2.62	2.59	--	--	--	--	--	--
18	--	--	--	--	--	--	--	--	--
19	--	--	--	--	--	--	--	--	--
21	--	--	--	--	--	--	--	--	--
22	--	--	--	--	--	--	--	--	--
23	--	--	--	--	--	--	--	--	--
24	12.8	15.3	15.4	--	--	--	--	--	--
26	--	--	--	--	--	--	--	--	--
27	**16.0**	12.8	9.24	--	--	--	--	--	--
28	--	--	--	--	--	--	--	--	--
29	11.1	10.1	9.21	9.01	9.29	8.99	8.77	8.73	8.39
30	--	--	--	--	--	--	--	--	--
31	4.17	--	--	--	--	--	--	--	--
32	--	--	--	--	--	--	--	--	--
33	12.4	11.5	10.1	--	--	--	--	--	--
34	--	--	--	--	--	--	--	--	--
35	--	--	--	--	--	--	--	--	--
36	--	--	--	--	--	--	--	--	--
Min	*2.84*	*2.62*	*2.04*	*6.03*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Max	***16.0***	***27.1***	***21.1***	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
Ave	*10.6*	*11.5*	*9.58*	*8.00*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*
90th %ile	*13.9*	*15.3*	*15.4*	*10.7*	*9.29*	*8.99*	*8.77*	*8.73*	*8.39*

Table S6b: Summary of Supplemental September/October sequential sampling results not used in data analyses. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Stagnation Times – Volunteers were asked to record the date and time water was last used, and the date and time when sampling began for each set of samples. Table S6c is a summary table which contains the stagnation times for the sequential samples, which is the amount of time the water sat motionless in the household prior to sample collection.

Sample Collection Stagnation Times			
June Sequential Sampling		Sept/Oct Sequential Sampling	
Site	Stagnation Time (hrs:mins)	Site	Stagnation Time (hrs:mins)
1	6:32	1	8:04
3	7:13	3	7:45
4	7:06	5	7:45
5	7:00	6	8:00
6	9:10	7	7:13
7	7:24	8	6:05
8	7:35	9	7:20
9	8:15	10	***
10	6:06	11	7:08
11	7:00	12	6:26
12	8:06	13	***
17	6:25	17	6:55
18	8:43	18	12:53
19	6:30	19	***
21	6:15	21	6:00
22	6:20	22	6:15
23	7:45	23	9:00
24	8:33	24	7:01
25	8:32	26	7:00
26	7:00	27	7:45
27	7:00	28	8:00
29	***	29	***
31	7:26	30	10:45
32	7:13	31	7:30
33	7:02	32	6:54
35	7:04	33	9:06
36	7:45	34	7:05
38	7:13	35	6:55
		36	8:47
***Volunteer did not record date/time the water was last used, but said it was the day before and was at least 6 hours before sampling.			

Table S6c: Summary of stagnation times for sequential sampling.

Seasonal Variability – Table S6d contains a site by site comparison of lead concentrations.

Seasonal Variability (Spring vs. Fall & Summer vs. Fall)					
First-Draw NHU	Sept/Oct > Mar/Apr	First-Draw PF	Sept/Oct > Mar/Apr	Sequential Samples	Sept/Oct > June
No. of Sample Pairs	28	No. of Sample Pairs	29	No. of Sample Pairs	285
No. Higher in Sept/Oct	19	No. Higher in Sept/Oct	20	No. Higher in Sept/Oct	156
% Higher in Sept/Oct	68%	% Higher in Sept/Oct	69%	% Higher in Sept/Oct	55%
First-Draw Samples: Mar/Apr vs. Sept/Oct (Same Site, Same First-Draw Protocol Compared)					
Sequential Samples: June vs. Sept/Oct (Same Site/Same Liter Compared)					

Table S6d: Seasonal variability effects observed.

Flushed sample results – The results of the flushed samples collected in September and October are tabulated in Table S7 by site. Most sites collected a 3 minute and 5 minute flushed sample. Some sites collected a 3, 5, and 7 minute flushed sample; and one site (site 29) collected a 3, 5, and 10 minute flushed sample, due to the length of the service line (159 ft / 48.5 m).

A flushed sample is collected by fully opening the sample tap and letting the water run for at least five minutes prior to a minimum 6 hour stagnation period. The date and time of the PF was recorded. After the minimum 6 hour stagnation period, and immediately before beginning the flushed sample collection, the date and time were again recorded and used as the start of sampling. The 3, 5, 7 and 10 minutes are measured from that start time, and water was not turned off between samples. For sequential sampling and flushed samples, the water was not turned off between samples.

EPA's current Public Notification Handbook includes instructions that advise residents to run the water between 30 and 45 seconds before collecting water for consumption if the water has not been used for an extended period of time. Running the water (flushing) for 45 seconds resulted in high lead levels at the tap for some sites. The flushed sampling results in this study indicate that EPA should develop a more appropriate flushing guidance, based on whether a home has a LSL or not, and the length of the LSL.

For homes with long LSLs, such as Site 29 (159 ft / 48.5 m), flushing may not be a practical way to reduce lead levels, as lead levels did not decline any further following 3, 5 and 10 minutes of flushing. In the case of site 29, residents would likely have a minimum of approximately 8 to 11 µg/L of lead in the drinking water for all water consumed, and should consider installing a water filter or using bottled water for drinking and cooking.

Flushed Sample Summary Table (µg/L)						
Site	Mar/Apr 2011 NHU 45sec	Mar/Apr 2011 PF 45sec	Sept/Oct 2011 3min	Sept/Oct 2011 5min	Sept/Oct 2011 7min	Sept/Oct 2011 10min
01	11.3	11.9	6.48	6.56	6.97	
03	12.0	6.71	3.78	2.93		
04	6.76	2.56				
05	13.2	14.1				
06	1.90	2.13				
07	**15.3**	**24.9**	5.49	5.46	5.32	
08	**32.2**	**28.0**	8.25	5.54	5.71	
09	**15.9**	**17.7**	14.3	7.23		
10	**25.0**	**21.6**	4.95	4.30	4.09	
11	4.13	5.30	1.75	1.69		
12	**17.2**	5.45	1.78	1.45	1.33	
13	3.50	2.94				
17	4.00	3.70	2.88	2.76	2.86	
18	9.57	12.4	4.15	3.71		
19	4.69	8.27				
20	2.80	2.54				
21	6.87	13.8				
22	9.19	7.93				
23	13.1	11.5	5.64	4.54		
24	6.10	4.98	6.38	12.4		
25	3.75	ND				
26	3.02	3.45	5.06	3.23		
27	4.53	3.76	15.0	14.1		
28	4.99	4.70	4.82	3.26		
29	13.5	**28.6**	11.9	10.9		10.8
30	12.5	6.52	5.80	4.82		
31	3.16	12.3	3.78	3.76		
32	2.29	7.82				
33	**16.4**	14.0	4.40	4.06		
34	1.51	3.30	1.83	1.75		
35	5.28	10.5	5.53	4.03		
36	11.1	8.76	7.19	5.29		
38	1.60	2.30				

NHU 45sec Samples were collected following the collection of the First-Draw NHU samples by running the water for 45 seconds following the collection of the First-Draw NHU sample.

PF 45sec Samples were collected following the collection of the First-Draw PF samples by running the water for 45 seconds following the collection of the First-Draw PF sample.

3min, 5min, 7min, and 10min flushed samples were collected after pre-flushing the tap for at least 5 minutes prior to the minimum 6 hour stagnation time during which no water was used in the home. Following the stagnation period and prior to sample collection, residents flushed the tap for 3 min to collect the 3min sample, and then an additional 2min for the 5min sample or 4min for the 7min sample. One site (site 29) had the longest lead service line so this site collected a 3 min, 5 min and 10min flushed sample (water was flushed for an additional 5 minutes following the collection of the 5min sample to collect the 10 min flushed sample). Water was not turned off in between samples to avoid the water hammer effect. Residents were instructed to have the bottles ready to insert under the faucet at the appropriate time.

Site 20 and Site 21 are the same residence. Site 20 was the upstairs bathroom and Site 21 was the kitchen sink. Note that neither the 45sec NHU nor PF samples from the upstairs tap captured any LSL water, while at least one of the kitchen tap samples did.

Table S7: Summary table of flushed sample results. Samples that were above the lead AL are in bold, and samples that contained visible particulates are shaded yellow.

Classification of Disturbed LSL Sites – A summary of the classification of each site as "disturbed", "undisturbed", or "indeterminate" is presented in Table S8, along with the number of samples collected per site and the number and percentage of sample results above the lead action level. The results from the "disturbed" and "undisturbed" sites are consistent with other research efforts showing that LSL disturbances result in higher lead levels[1-3].

Disturbed, Undisturbed and Indeterminate Site Summary								
Disturbed Sites	Total Samples Collected	# Samples Above AL (Disturbed)	Undisturbed Sites	Total Samples Collected	# Samples above AL (Undisturbed)	Indeterminate Sites	Total Samples Collected	# Samples above AL (Indeterminate)
01	27	16	03	27	4	12	27	17
05	27	2	04	14	0	21	27	7
07	27	11	06	27	0	33	27	6
08	27	19	11	27	0	---	---	---
09	27	15	13	15	0	---	---	---
10	27	15	18	27	0	---	---	---
17	27	3	19	27	0	---	---	---
27	27	5	22	27	0	---	---	---
28	15	0	23	27	0	---	---	---
29	27	15	24	27	0	---	---	---
30	15	4	25	14	0	---	---	---
31	27	10	26	27	0	---	---	---
35	27	2	32	27	2	---	---	---
---	---	---	34	15	0	---	---	---
---	---	---	36	27	0	---	---	---
---	---	---	38	16	0	---	---	---
Totals	**327**	**117**	**Totals**	**371**	**6**	**Totals**	**81**	**30**
% of samples above AL: 36%			**% of samples above AL: 2%**			**% of samples above AL: 37%**		

Table S8: Summary Table of Disturbed, Undisturbed and Indeterminate Sites, with the number and percentages of sample results above the lead AL for each site and each grouping.

Many direct LSL disturbances are localized to a specific segment of the LSL, and yet some sites have higher lead levels in sample liters over a significant portion of the LSL, not just in the immediate area of the LSL that was disturbed. A probable reason is that, except for the initial liter of water, each subsequent one-liter sample reflects both lead levels within the segment of the plumbing where the water stagnated as well as a contribution from the rest of the plumbing the water travelled through. For example, the fifth liter of water collected from a kitchen tap will not only capture the lead from the segment of LSL where the water stagnated, but it will also collect contributions from the plumbing downstream as the water passes through the remaining LSL and internal plumbing on the way to the kitchen tap. If the sample results only represented the portion of the plumbing where the water stagnated, it would be expected that a variety of metals would be found in the initial liters due to the presence of a variety of metallic plumbing materials and components, but only lead should be found in the LSL samples. In this study, a variety of metals was detected even in samples that represented LSL samples (Figure S6).

Specifically, for Site 9, information provided by the resident indicated that the internal pipe from the LSL to the kitchen tap was galvanized iron pipe. This was confirmed by the co-occurrence of higher levels of zinc and iron within the first liter of water in figure S6. There were no copper pipes in the home, so the presence of the copper is indicative of brass components (faucet, connectors, shut-off valve(s), and the water meter). Trace amounts of iron, zinc and copper are captured in the later liter samples as the water flows through the internal plumbing en route to the kitchen tap, along with traces of iron, potentially from the water main. It can reasonably be

assumed that the same phenomenon occurred for lead. Disturbed areas of the LSL have damaged scale, which can expose water passing through them to fresh lead. Therefore, lead measured in any sample upstream of the damaged area may include lead contributions from the damaged area.

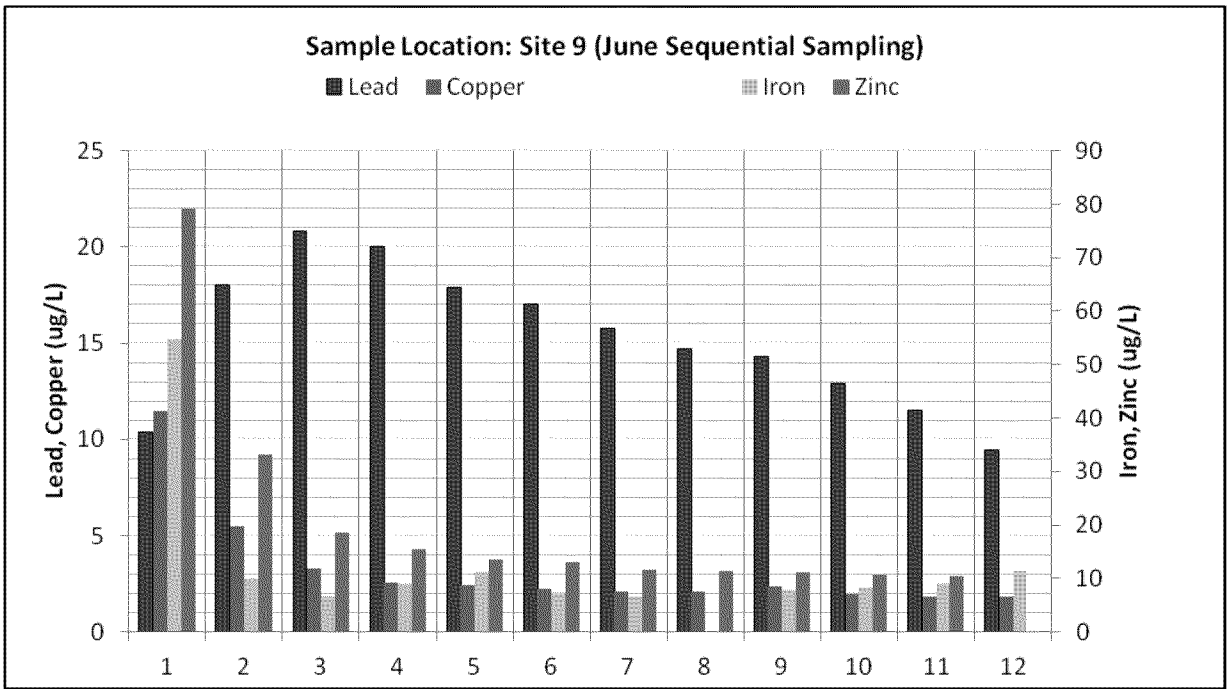


Figure S6: The LSL at Site 9 measures approximately 102 ft (31.1 m) from the water main to the meter. From the meter, there is approximately 13.5 ft (4.1 m) of 1 inch (2.54 cm) galvanized pipe to the kitchen tap.

Variability of lead levels in City B – A second city, City B, exceeded the lead AL during the July-Dec 2010 monitoring period, and was required to comply with the LSL replacement requirements in the LCR. Table S9 contains the compliance monitoring history for City B.

Monitoring Period Begin Date	Monitoring Period End Date	Number of Samples	Lead 90th Percentile Value (µg/l)
7/1/2011	12/31/2011	101	12
1/1/2011	6/30/2011	130	14
7/1/2010	12/31/2010	105	**23**
1/1/2009	12/31/2009	51	15
1/1/2008	12/31/2008	58	14
1/1/2007	12/31/2007	50	11
1/1/2006	12/31/2006	60	14
1/1/2005	12/31/2005	54	13
1/1/2004	6/30/2004	104	12
7/1/2003	12/31/2003	108	12
1/1/2002	12/31/2004	50	15
1/1/1999	12/31/1999	55	14
1/1/1998	12/31/1998	50	6
1/1/1997	12/31/1997	50	7
7/1/1996	12/31/1996	50	15
1/1/1996	6/30/1996	50	15
7/1/1992	12/31/1992	50	15
1/1/1992	6/30/1992	50	**21**

Table S9: City B 90th percentile compliance values (1992 – 2012). Samples that were above the lead AL are in bold.

The sampling instructions presented in Figure S7 are in accordance with the LCR, and were used to collect the LSL samples in City B, which has approximately 25,000 LSLs.

Instructions for Lead Sample Collection

1. Make sure the faucet used for sample collection is NOT attached to a water softener or any filtering device.
2. At bedtime, make sure the following rule is followed:
   - The water for the entire house, not just the faucet that is being used for collection, remains undisturbed for a period of at least six hours.
     - **No** faucets in the house are used, which includes the bath tub, shower and sinks.
     - The toilet is **not** flushed during this time period.
     - The water is **not** run for an ice maker.
3. When you are ready to collect the sample:
   - Make sure the sample is taken before any other water is used.
     - Open the collection container.
     - Turn on the **cold** water.
     - Allow the water to run until there is a significant change in temperature.
     - Fill the container to the shoulder.
     - Do **not** rinse the bottle out.
     - Immediately cap the sample container.
4. Fill out the enclosed chain of custody form and survey.
5. Fold and secure the chain of custody form and survey with a rubber band around the outside of the sample container.
   - Place the container outside where it was delivered.

❖ A city utilities employee will pick up the sample container. No one will enter your home. The sample must be left outside to be picked up.

Figure S7: LSL sampling instructions provided by City B to residents.

The sampling protocol used for collecting LSL samples ("allow the water to run until there is a significant change in temperature") can result in some sample results reflecting lead levels from internal plumbing rather than from within the LSLs.

The results from City B are presented below in Figure S8. Similar to the results presented for the study of Chicago, City B's results show significant variability in LSL lead levels across the system. Following the 2010 lead AL exceedance, the City B took 1,975 LSL samples, with a total of 1,762 results (89%) below the lead AL and 213 results (11%) above the lead AL. LSL results above the AL were significantly variable, ranging from 16 μg/L to 580 μg/L with a large number of sample results in exceedance of 50 μg/L.

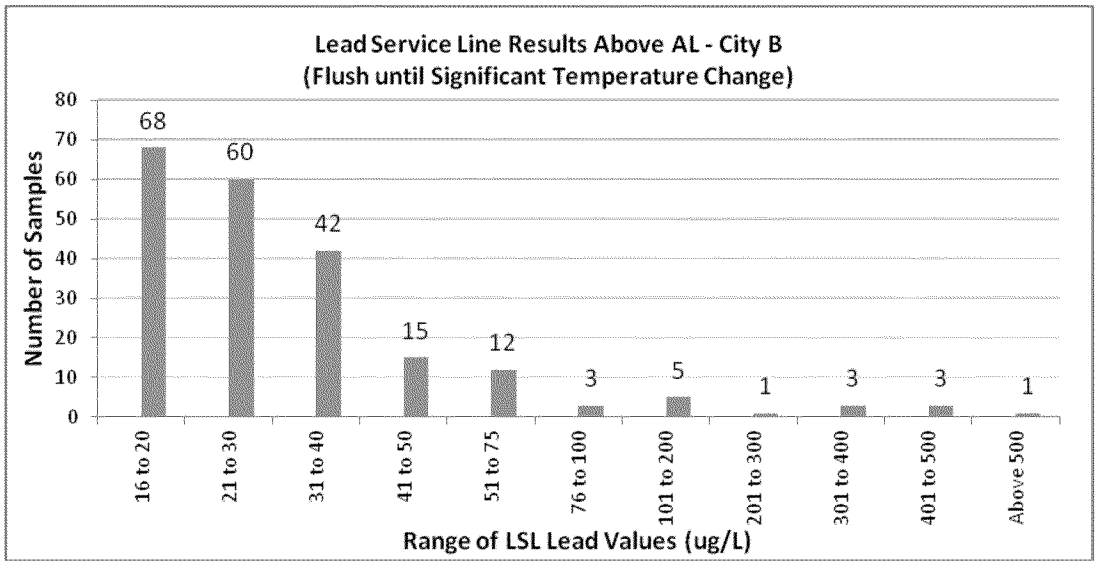
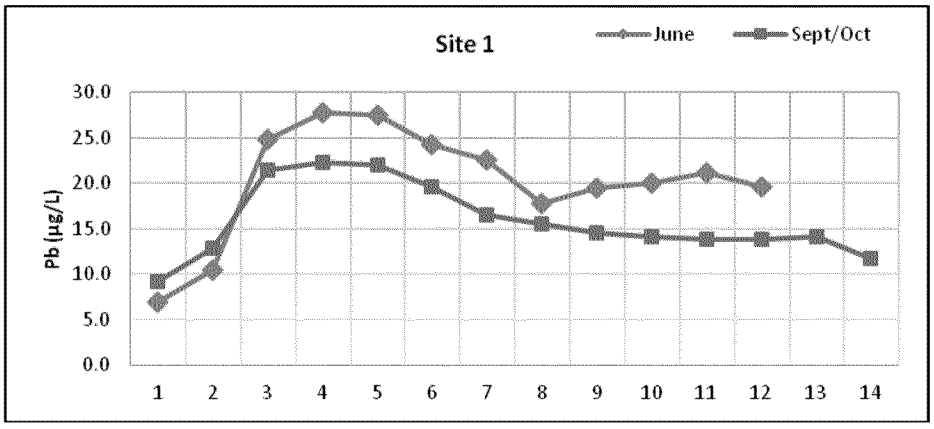


Figure S8: Range of lead values for City B LSL sampling results

Sequential Sampling Summary Graphs –The headers are color-coded based on whether the site has a disturbed LSL (red) or an undisturbed LSL (green). Sites for which this could not be determined (indeterminate sites) are color-coded orange. Water usage information is listed for each site. The samples which contained visible particulates are highlighted yellow, and the results that are above the lead AL are in bold text in the data tables. For sites that conducted sequential sampling in both June and Sept/Oct, the sequential sampling profiles were generally consistent during both sampling periods (see Figures S9 – S40).

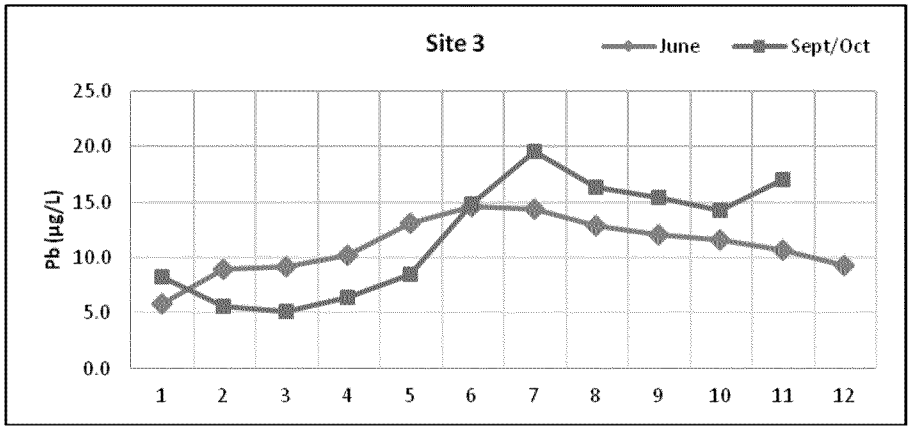
Site 1		
Liter	June	Sept/Oct
1	7.0	9.2
2	11	13
3	**25**	**21**
4	**28**	**22**
5	**28**	**22**
6	**24**	**20**
7	**23**	**17**
8	**18**	**16**
9	**20**	15
10	**20**	14
11	**21**	14
12	**20**	14
13		14
14		12



Disturbance(s): Water meter installed in 2010
Approximate LSL Length: 89 ft (27.1 m)
Ave Monthly Water Use: 3,444 gal. (13,037 L)

Figure S9: Sequential Lead Results - Sample Site #1 (June and Sept/Oct)

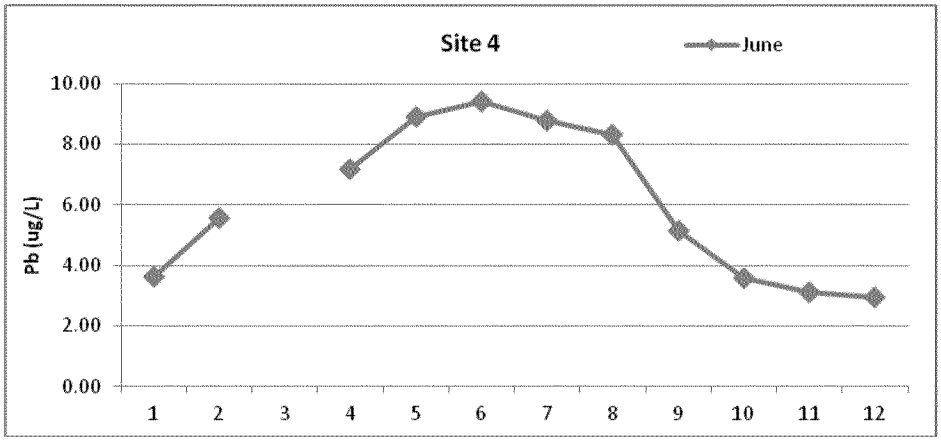
Site 3		
Liter	June	Sept/Oct
1	5.8	8.3
2	8.9	5.6
3	9.2	5.2
4	10	6.4
5	13	8.5
6	15	15
7	14	**20**
8	13	**16**
9	12	15
10	12	14
11	11	**17**
12	9.3	



Disturbance(s): No known disturbance
Approximate LSL Length: 73 ft (22.3 m)
Ave Monthly Water Use: Not metered

Figure S10: Sequential Lead Results - Sample Site #3 (June and Sept/Oct)

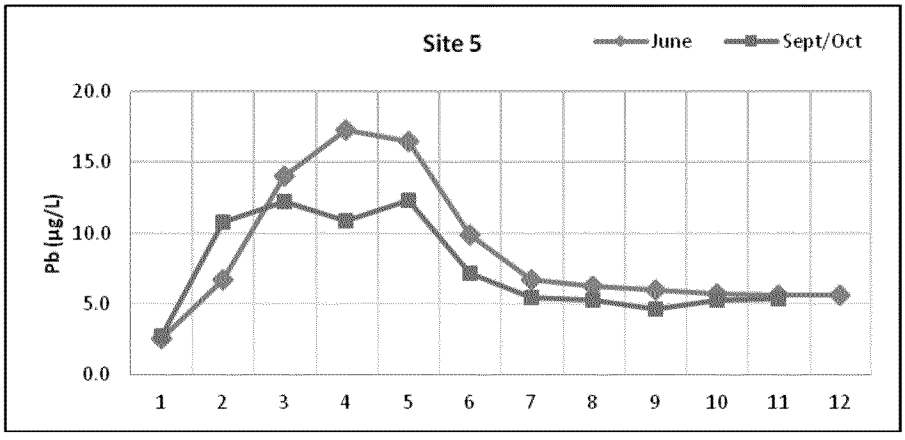
Site 4	
Liter	June
1	3.61
2	5.56
3	
4	7.17
5	8.90
6	9.41
7	8.78
8	8.30
9	5.14
10	3.59
11	3.11
12	2.96



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S11: Sequential Lead Results - Sample Site #4 (June)

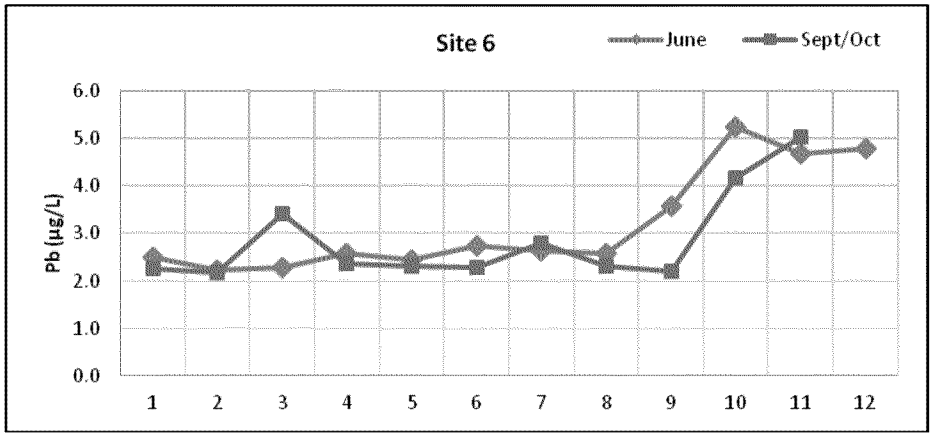
Site 5		
Liter	June	Sept/Oct
1	2.6	2.8
2	6.7	11
3	14	12
4	**17**	11
5	**17**	12
6	9.9	7.2
7	6.7	5.5
8	6.3	5.2
9	6.0	4.7
10	5.7	5.3
11	5.7	5.4
12	5.6	



Disturbance(s): Water meter installed in 2011
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 10,400 gal. (39,368 L)

Figure S12: Sequential Lead Results - Sample Site #5 (June and Sept/Oct)

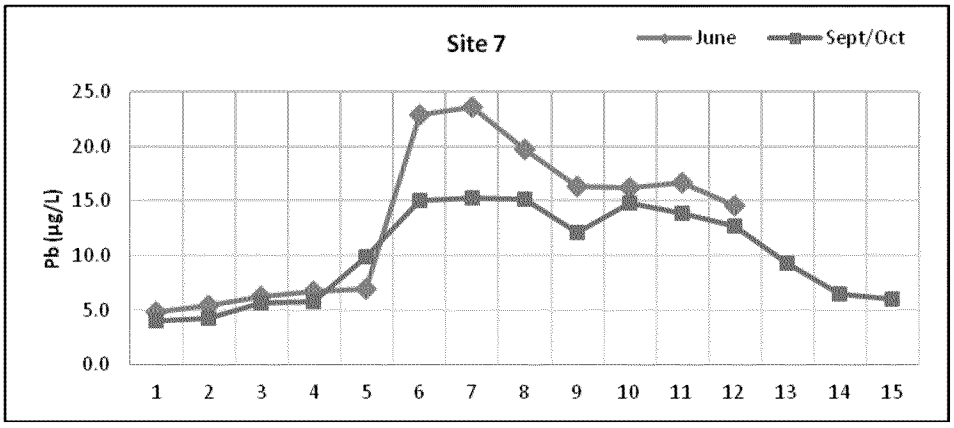
Site 6		
Liter	June	Sept/Oct
1	2.5	2.3
2	2.2	2.2
3	2.3	3.4
4	2.6	2.4
5	2.4	2.3
6	2.8	2.3
7	2.7	2.8
8	2.6	2.3
9	3.6	2.2
10	5.3	4.2
11	4.7	5.0
12	4.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 60 ft (18.3 m)
Ave Monthly Water Use: Not metered

Figure S13: Sequential Lead Results - Sample Site #6 (June and Sept/Oct)

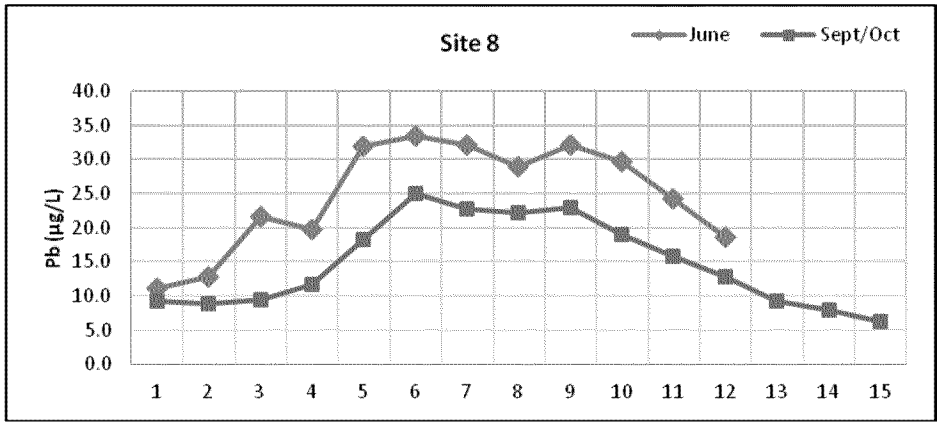
Site 7		
Liter	June	Sept/Oct
1	4.9	4.0
2	5.5	4.3
3	6.3	5.7
4	6.7	5.8
5	7.0	9.9
6	**23**	15
7	**24**	15
8	**20**	15
9	**16**	12
10	**16**	15
11	**17**	14
12	15	13
13		9.3
14		6.5
15		6.0



Disturbance(s): Street excavation, potential installation of Cu whip at service connection in 2008
Approximate LSL Length: 59+ ft (18.0+ m)
Ave Monthly Water Use: Not metered

Figure S14: Sequential Lead Results - Sample Site #7 (June and Sept/Oct)

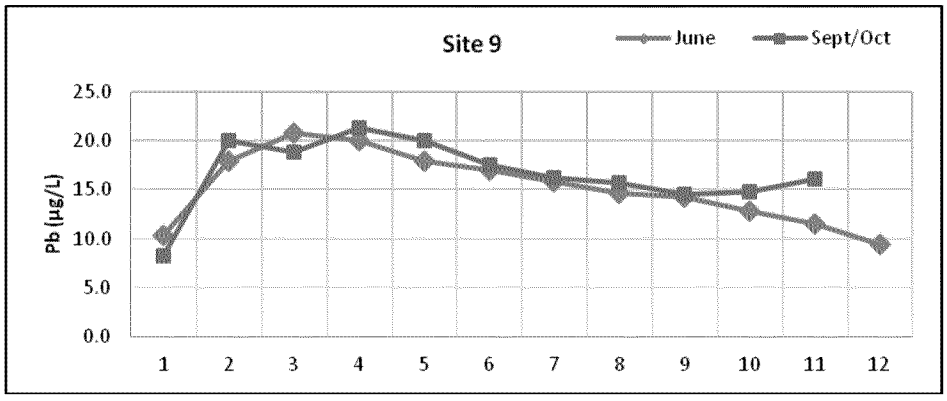
Site 8		
Liter	June	Sept/Oct
1	11	9.2
2	13	9.0
3	**22**	10
4	**20**	12
5	**32**	**18**
6	**34**	**25**
7	**32**	**23**
8	**29**	**22**
9	**32**	**23**
10	**30**	**19**
11	**24**	**16**
12	**19**	13
13		9.3
14		7.9
15		6.3



Disturbance(s): Leak in parkway, repaired roundway in 2005.
Approximate LSL Length: 57 ft (17.4 m)
Ave Monthly Water Use: Not metered

Figure S15: Sequential Lead Results - Sample Site #8 (June and Sept/Oct)

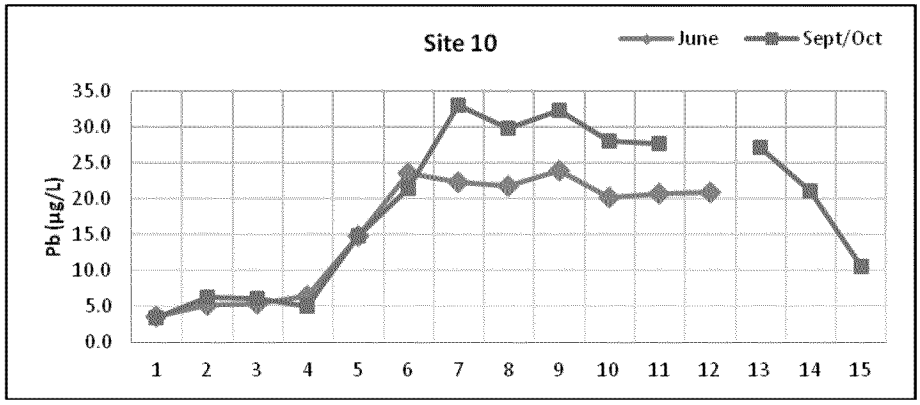
Site 9		
Liter	June	Sept/Oct
1	10	8.3
2	**18**	**20**
3	**21**	**19**
4	**20**	**21**
5	**18**	**20**
6	**17**	**18**
7	**16**	**16**
8	15	**16**
9	14	15
10	13	15
11	12	**16**
12	10	



Disturbance(s): Water meter installed in 2008.
Approximate LSL Length: 102 ft (31.1 m)
Ave Monthly Water Use: 3,190 (12,075 L) – In Sept 2011, usage was 24,000 gal. (90,850 L) due to hose left running for one or more days. In calculating the overall average, the Sept 2010 value of 8,000 gal. (30,283 L) was also used for Sept 2011 instead of the 24,000 gal. (90,850 L) value.

Figure S16: Sequential Lead Results - Sample Site #9 (June and Sept/Oct)

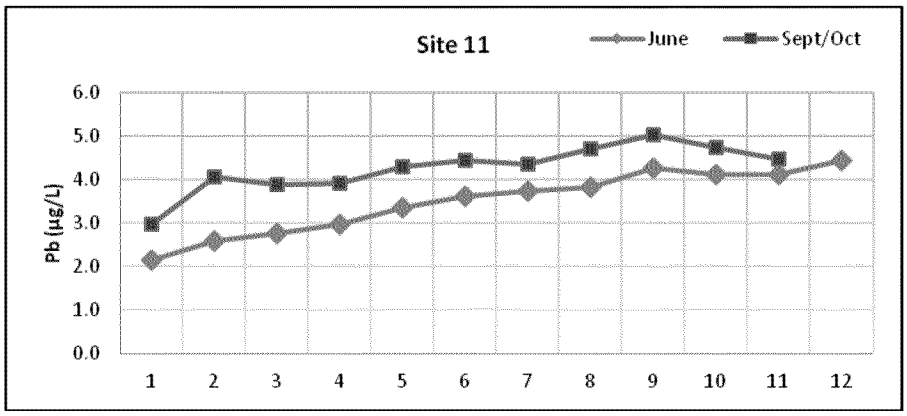
Site 10		
Liter	June	Sept/Oct
1	3.7	3.5
2	5.2	6.3
3	5.4	6.2
4	6.5	5.1
5	15	15
6	**24**	**21**
7	**22**	**33**
8	**22**	**30**
9	**24**	**32**
10	**20**	**28**
11	**21**	**28**
12	**21**	
13		**27**
14		**21**
15		11



Disturbance(s): Service leak repair, water meter installed in 2009.
Approximate LSL Length: 48+ ft (14.6 m)
Ave Monthly Water Use: 1,826 gal. (6,912 L)

Figure S17: Sequential Lead Results - Sample Site #10 (June and Sept/Oct)

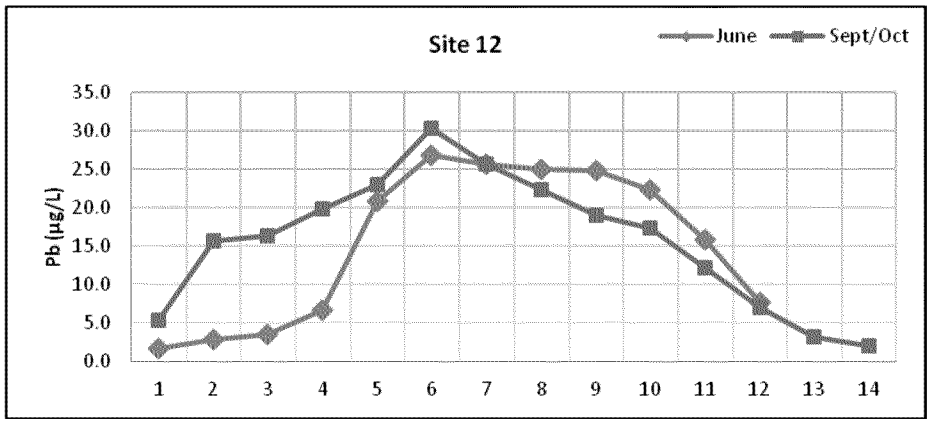
Site 11		
Liter	June	Sept/Oct
1	2.2	3.0
2	2.6	4.1
3	2.8	3.9
4	3.0	3.9
5	3.4	4.3
6	3.6	4.4
7	3.7	4.4
8	3.8	4.7
9	4.3	5.0
10	4.1	4.8
11	4.1	4.5
12	4.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 50 ft (15.2 m)
Ave Monthly Water Use: Not metered

Figure S18: Sequential Lead Results - Sample Site #11 (June and Sept/Oct)

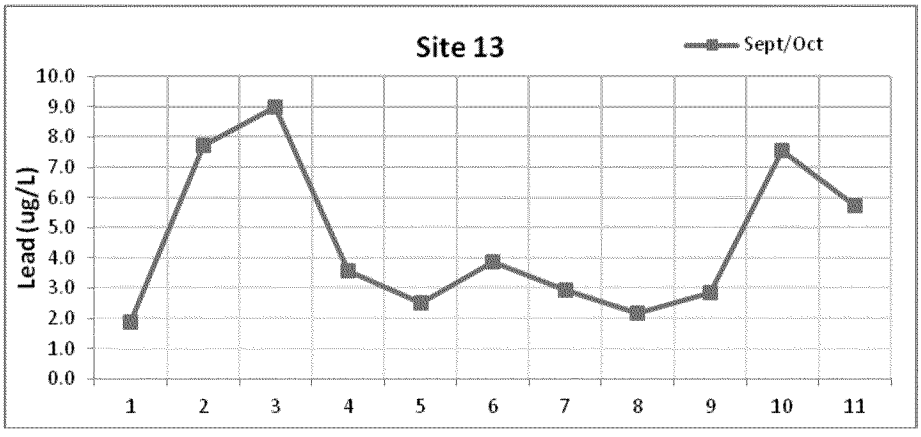
Site 12		
Liter	June	Sept/Oct
1	1.8	5.4
2	3.0	**16**
3	3.6	**16**
4	6.7	**20**
5	**21**	**23**
6	**27**	**30**
7	**26**	**26**
8	**25**	**22**
9	**25**	**19**
10	**22**	**17**
11	**16**	12
12	7.8	7.0
13		3.3
14		2.0



Disturbance(s): Indeterminate
Approximate LSL Length: 53 (16.2 m)
Ave Monthly Water Use: Not metered

Figure S19: Sequential Lead Results - Sample Site #12 (June and Sept/Oct)

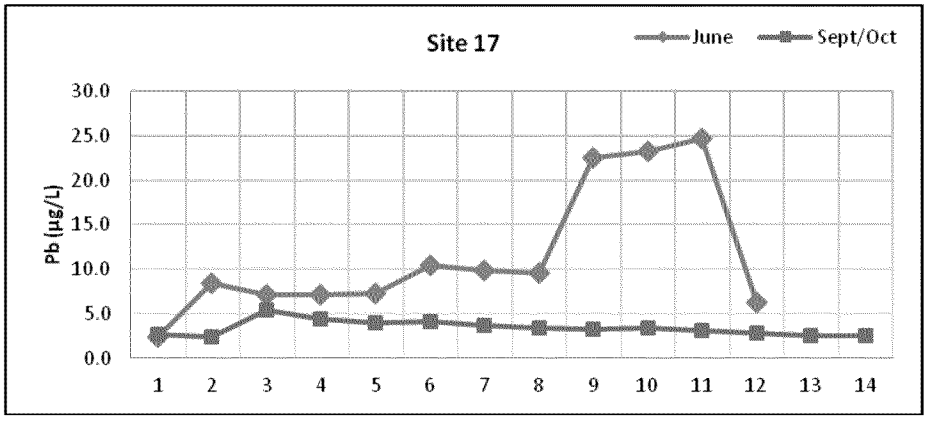
Site 13	
Liter	Sept/Oct
1	1.9
2	7.7
3	9.0
4	3.6
5	2.5
6	3.9
7	3.0
8	2.2
9	2.9
10	7.6
11	5.7



Disturbance(s): No known disturbance
Approximate LSL Length: 49+ ft (4.9 m)
Ave Monthly Water Use: Not metered

Figure S20: Sequential Lead Results - Sample Site #13 (Sept/Oct)

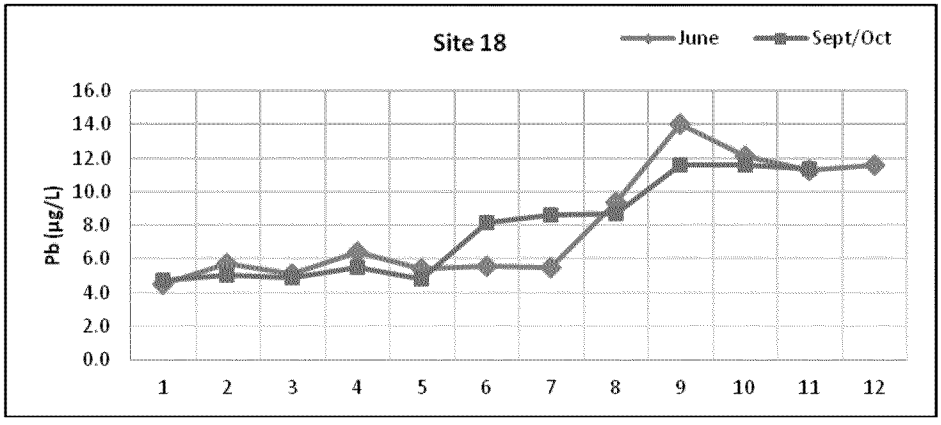
Site 17		
Liter	June	Sept/Oct
1	2.4	2.7
2	8.5	2.4
3	7.1	5.5
4	7.2	4.4
5	7.3	4.1
6	11	4.1
7	9.9	3.7
8	9.6	3.4
9	**23**	3.4
10	**23**	3.4
11	**25**	3.2
12	6.3	2.8
13		2.6
14		2.6



Disturbance(s): Meter replacement in 2008.
Approximate LSL Length: 58+ ft (17.7+ m)
Ave Monthly Water Use: 9,772 gal. (36,991 m)

Figure S21: Sequential Lead Results - Sample Site #17 (June and Sept/Oct)

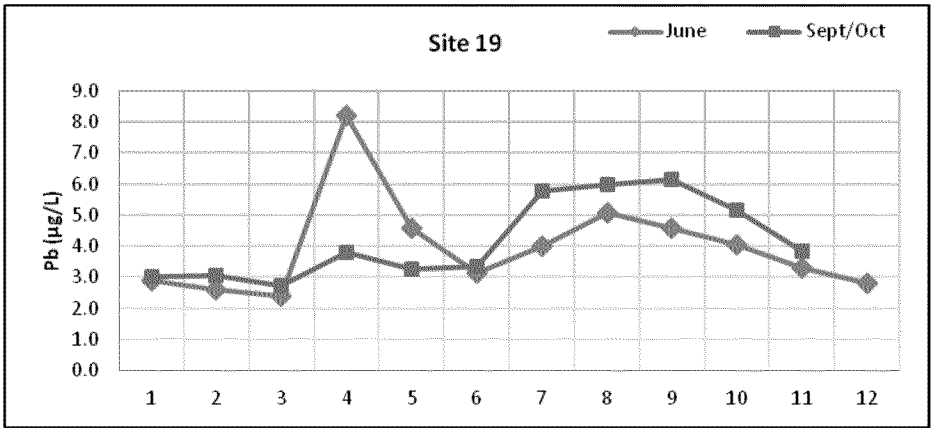
Site 18		
Liter	June	Sept/Oct
1	4.6	4.8
2	5.7	5.1
3	5.1	4.9
4	6.4	5.5
5	5.4	4.8
6	5.6	8.2
7	5.5	8.6
8	9.4	8.7
9	14	12
10	12	12
11	11	11
12	12	



Disturbance(s): No known disturbance
Approximate LSL Length: 76 ft (23.2 m)
Ave Monthly Water Use: Not metered

Figure S22: Sequential Lead Results - Sample Site #18 (June and Sept/Oct)

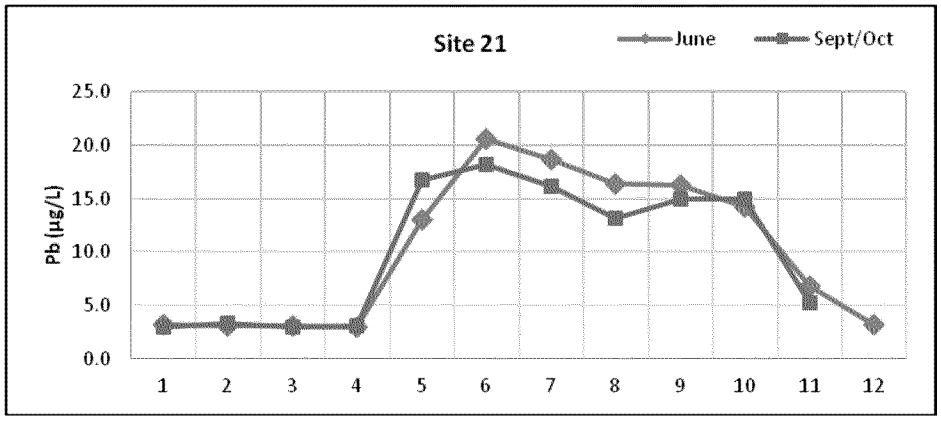
Site 19		
Liter	June	Sept/Oct
1	2.9	3.0
2	2.6	3.1
3	2.4	2.8
4	8.2	3.8
5	4.6	3.3
6	3.2	3.4
7	4.0	5.8
8	5.1	6.0
9	4.6	6.2
10	4.1	5.2
11	3.3	3.8
12	2.8	



Disturbance(s): No known disturbance
Approximate LSL Length: 63 ft (19.2 m)
Ave Monthly Water Use: Not metered

Figure S23: Sequential Lead Results - Sample Site #19 (June and Sept/Oct)

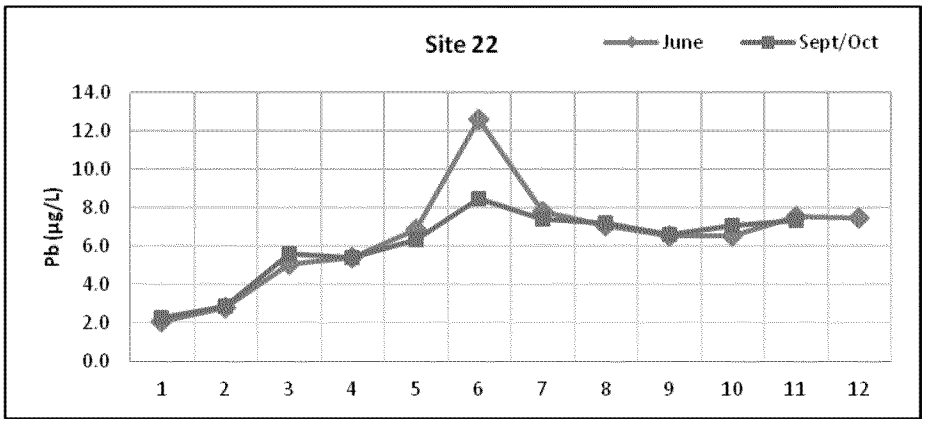
Site 21		
Liter	June	Sept/Oct
1	3.2	3.0
2	3.1	3.4
3	3.1	3.0
4	3.0	3.0
5	13	**17**
6	**21**	**18**
7	**19**	**16**
8	**16**	13
9	**16**	15
10	14	15
11	7.0	5.2
12	3.2	



Disturbance(s): Indeterminate
Approximate LSL Length: 46 ft (14.0 m)
Ave Monthly Water Use: Not metered

Figure S24: Sequential Lead Results - Sample Site #21 (June and Sept/Oct)

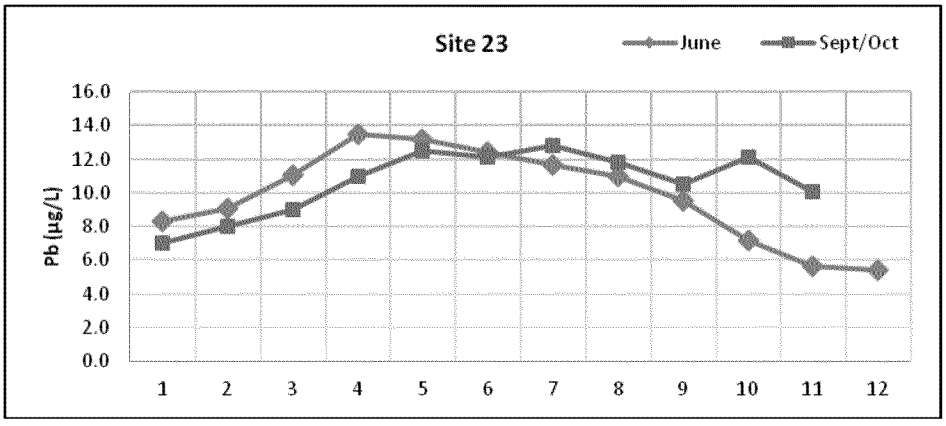
Site 22		
Liter	June	Sept/Oct
1	2.1	2.3
2	2.8	2.9
3	5.1	5.6
4	5.4	5.4
5	6.9	6.3
6	13	8.5
7	7.8	7.4
8	7.1	7.2
9	6.5	6.6
10	6.6	7.1
11	7.6	7.4
12	7.5	



Disturbance(s): No known disturbance
Approximate LSL Length: 65 ft (19.8 m)
Ave Monthly Water Use: Not metered

Figure S25: Sequential Lead Results - Sample Site #22 (June and Sept/Oct)

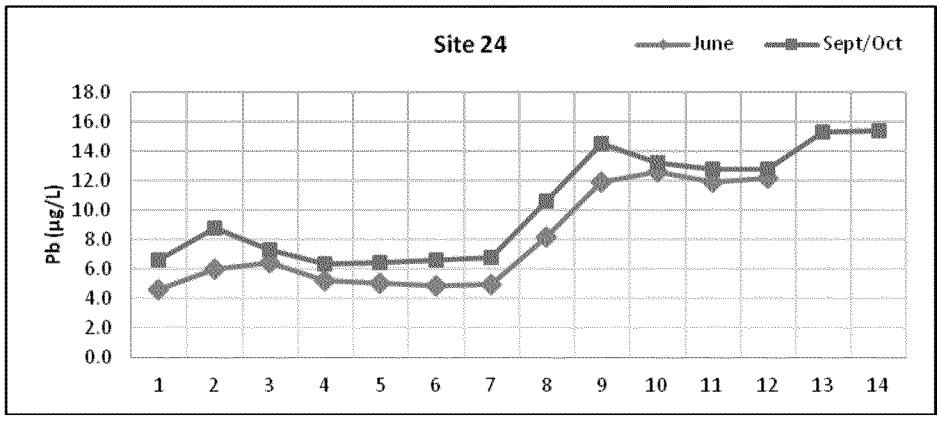
Site 23		
Liter	June	Sept/Oct
1	8.3	7.0
2	9.1	8.0
3	11	9.0
4	14	11
5	13	13
6	12	12
7	12	13
8	11	12
9	9.6	11
10	7.2	12
11	5.7	10
12	5.4	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S26: Sequential Lead Results - Sample Site #23 (June and Sept/Oct)

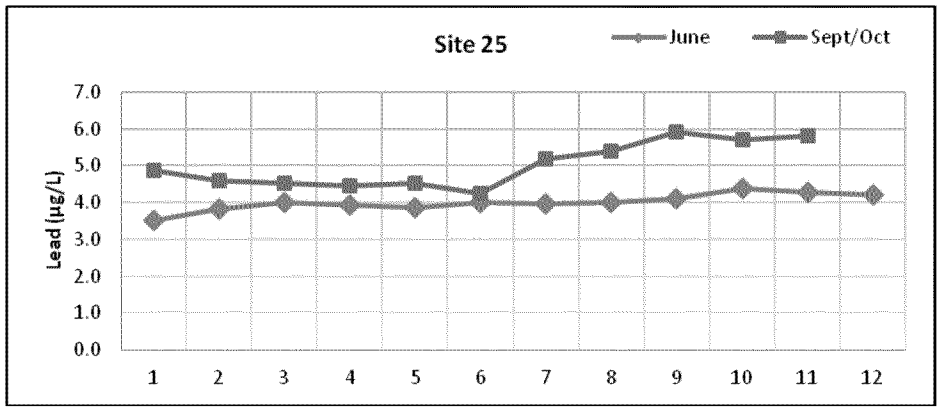
Site 24		
Liter	June	Sept/Oct
1	4.6	6.6
2	6.1	8.8
3	6.4	7.3
4	5.2	6.4
5	5.1	6.5
6	4.9	6.6
7	5.0	6.8
8	8.2	11
9	12	15
10	13	13
11	12	13
12	12	13
13		15
14		15



Disturbance(s): No known disturbance
Approximate LSL Length: 56 ft (17.1 m)
Ave Monthly Water Use: Not metered

Figure S27: Sequential Lead Results - Sample Site #24 (June and Sept/Oct)

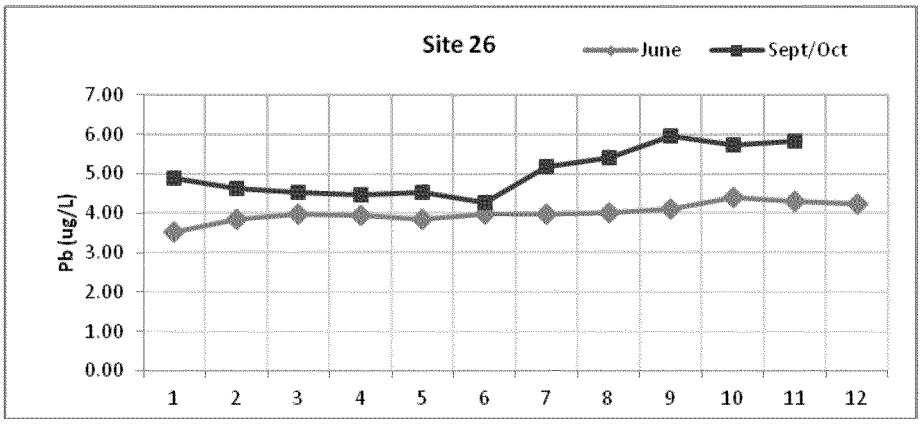
Site 25		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 70 ft (21.3 m)
Ave Monthly Water Use: Not metered

Figure S28: Sequential Lead Results - Sample Site #25 (June and Sept/Oct)

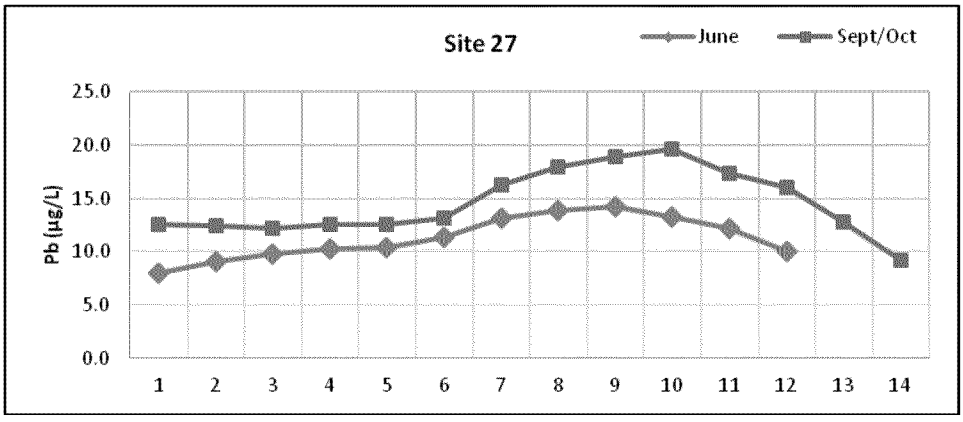
Site 26		
Liter	June	Sept/Oct
1	3.5	4.9
2	3.8	4.6
3	4.0	4.5
4	3.9	4.5
5	3.9	4.5
6	4.0	4.3
7	4.0	5.2
8	4.0	5.4
9	4.1	5.9
10	4.4	5.7
11	4.3	5.8
12	4.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 66 ft (20.1 m)
Ave Monthly Water Use: Not metered

Figure S29: Sequential Lead Results - Sample Site #26 (June and Sept/Oct)

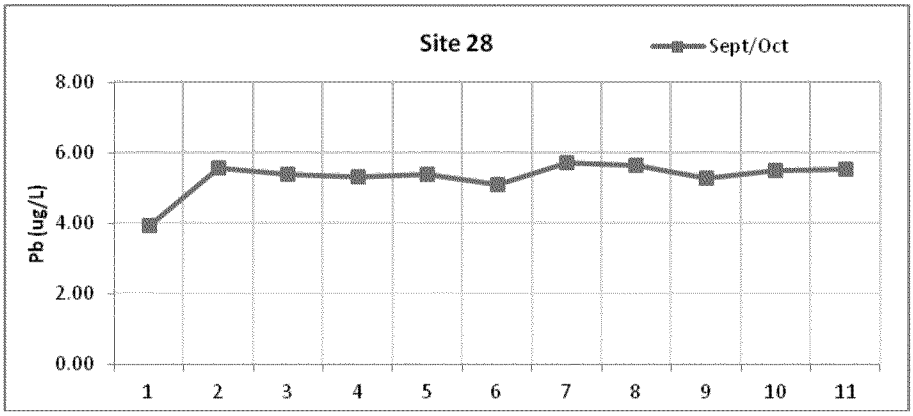
Site 27		
Liter	June	Sept/Oct
1	8.1	13
2	9.1	12
3	9.8	12
4	10	13
5	10	13
6	11	13
7	13	**16**
8	14	**18**
9	14	**19**
10	13	**20**
11	12	**17**
12	10	**16**
13		13
14		9.2



Disturbance(s): Meter replacement in 2010.
Approximate LSL Length: 47+ ft (14.3 m)
Ave Monthly Water Use: 4267 gal. (16,152 L)

Figure S30: Sequential Lead Results - Sample Site #27 (June and Sept/Oct)

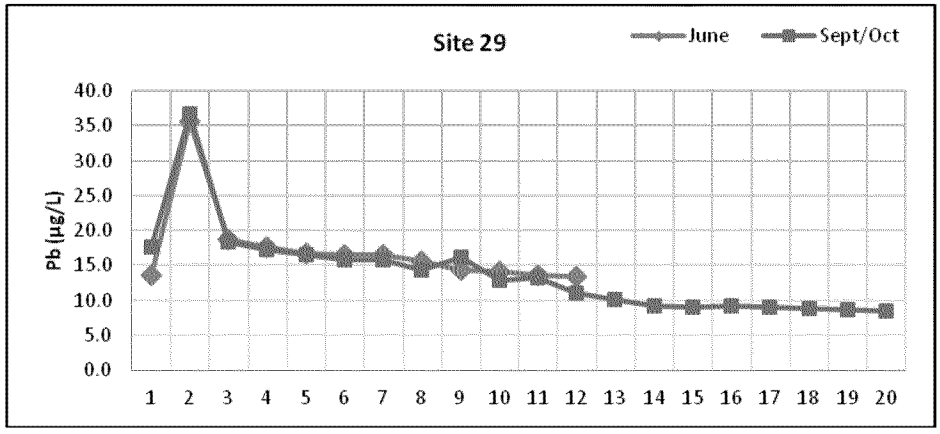
Site 28	
Liter	Sept/Oct
1	3.9
2	5.6
3	5.4
4	5.3
5	5.4
6	5.1
7	5.7
8	5.7
9	5.3
10	5.5
11	5.6



Disturbance(s): Meter replacement in 2009.
Approximate LSL Length: 61+ ft (18.6+ m)
Ave Monthly Water Use: 4273 gal. (16,175 L)

Figure S31: Sequential Lead Results - Sample Site #28 (Sept/Oct)

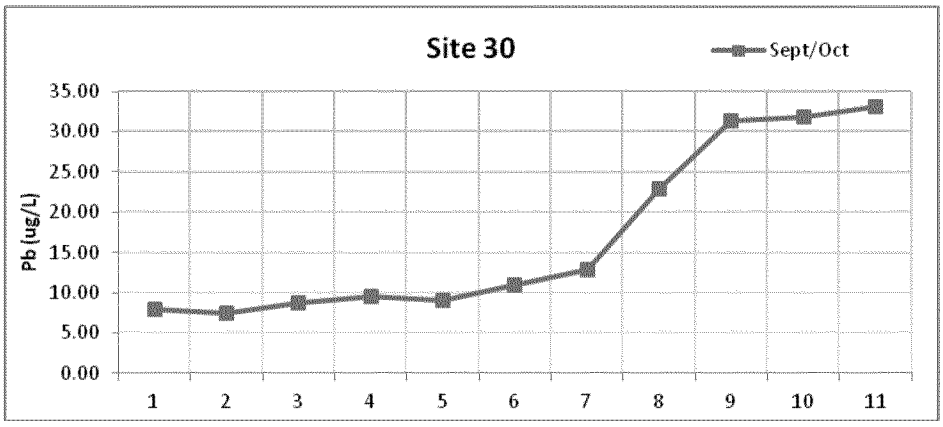
Site 29		
Liter	June	Sept/Oct
1	14	**18**
2	**36**	**37**
3	**19**	**18**
4	**18**	**17**
5	**17**	**17**
6	**17**	**16**
7	**17**	**16**
8	**16**	14
9	14	**16**
10	14	13
11	14	13
12	13	11
13		10
14		9.2
15		9.0
16		9.3
17		9.0
18		8.8
19		8.7
20		8.4



Disturbance(s): Probable Approximate LSL leak repair, meter installed in 2010.
Approximate LSL Length: 159 ft (48.5 m)
Ave Monthly Water Use: 1,438 gal. (5,443 L)

Figure S32: Sequential Lead Results - Sample Site #29 (June and Sept/Oct)

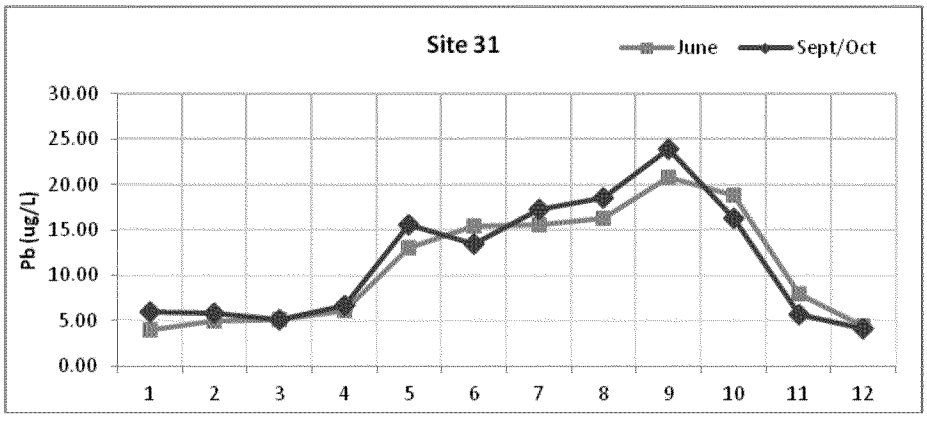
Site 30	
Liter	Sept/Oct
1	7.9
2	7.5
3	8.7
4	9.5
5	9.1
6	11
7	13
8	**23**
9	**31**
10	**32**
11	**33**



Disturbance(s): Broken water main in 2000, sidewalk replaced & street re-surfacing.
Approximate LSL Length: 49+ ft (14.9 m)
Ave Monthly Water Use: Not metered

Figure S33: Sequential Lead Results - Sample Site #30 (Sept/Oct)

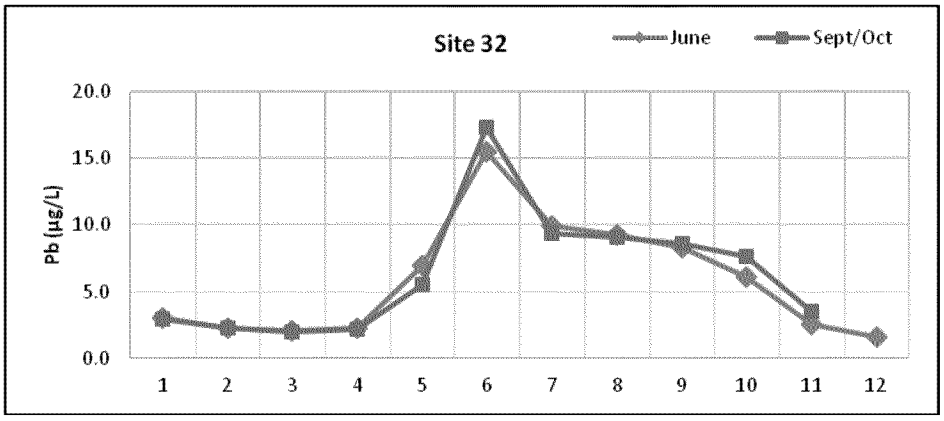
Site 31		
Liter	June	Sept/Oct
1	4.0	6.0
2	5.0	5.8
3	5.1	5.2
4	6.2	6.7
5	13	**16**
6	15	13
7	**16**	**17**
8	**16**	**19**
9	**21**	**24**
10	**19**	**16**
11	8	5.7
12	4.5	4.2



Disturbance(s): Approximate LSL leak repair in 2010.
Approximate LSL Length: 71+ ft (21.6+ m)
Ave Monthly Water Use: Not metered

Figure S34: Sequential Lead Results - Sample Site #31 (June and Sept/Oct)

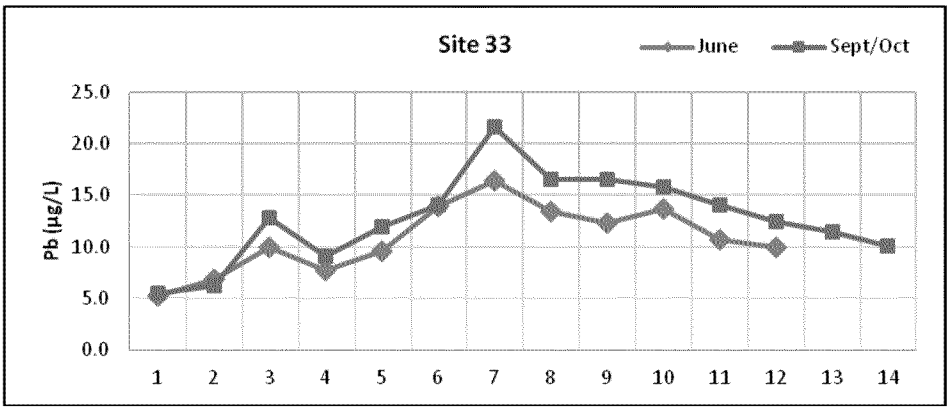
Site 32		
Liter	June	Sept/Oct
1	3.1	2.9
2	2.3	2.2
3	2.1	2.0
4	2.3	2.2
5	7.0	5.5
6	**16**	**17**
7	9.9	9.4
8	9.3	9.1
9	8.3	8.6
10	6.1	7.6
11	2.6	3.5
12	1.7	



Disturbance(s): No known disturbance
Approximate LSL Length: 43 ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S35: Sequential Lead Results - Sample Site #32 (June and Sept/Oct)

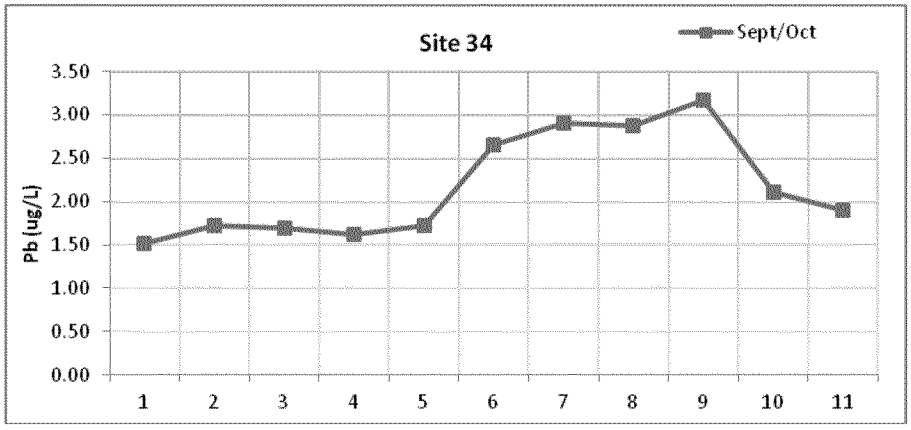
Site 33		
Liter	June	Sept/Oct
1	5.2	5.5
2	6.9	6.3
3	10	13
4	7.7	9.1
5	9.6	12
6	14	14
7	**16**	**22**
8	14	**17**
9	12	**17**
10	14	**16**
11	11	14
12	10	12
11		12
12		10



Disturbance(s): Indeterminate
Approximate LSL Length: 43+ ft (13.1 m)
Ave Monthly Water Use: Not metered

Figure S36: Sequential Lead Results - Sample Site #33 (June and Sept/Oct)

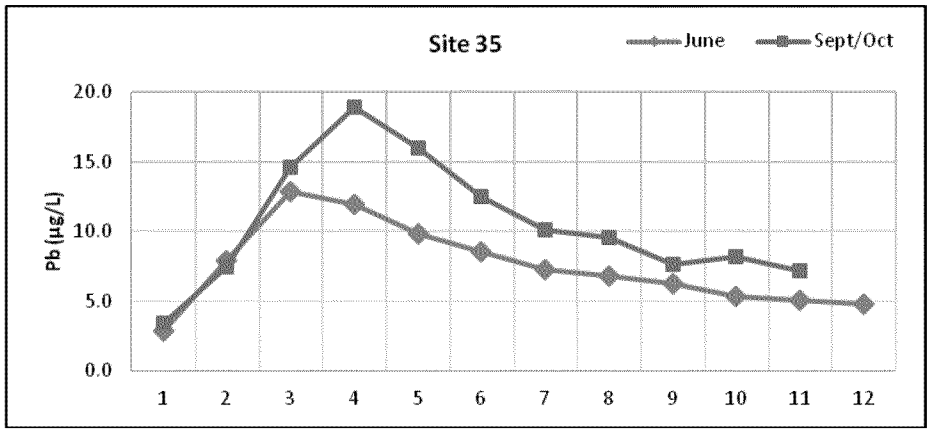
Site 34	
Liter	Sept/Oct
1	1.5
2	1.7
3	1.7
4	1.6
5	1.7
6	2.7
7	2.9
8	2.9
9	3.2
10	2.1
11	1.9



Disturbance(s): No known disturbance
Approximate LSL Length: Unknown
Ave Monthly Water Use: Not metered

Figure S37: Sequential Lead Results - Sample Site #34 (Sept/Oct)

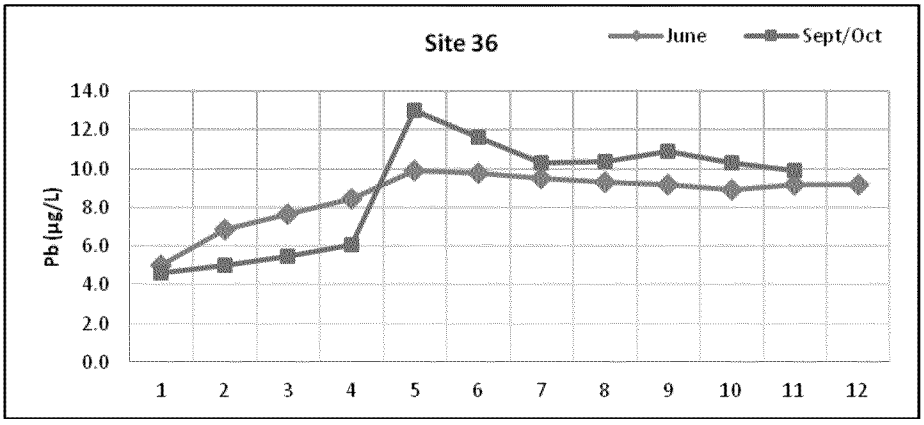
Site 35		
Liter	June	Sept/Oct
1	2.9	3.4
2	7.9	7.4
3	13	15
4	12	**19**
5	9.9	**16**
6	8.6	13
7	7.3	10
8	6.8	9.6
9	6.2	7.6
10	5.3	8.2
11	5.0	7.2
12	4.8	



Disturbance(s): Meter installed in Aug 2011 (between June and Sept/Oct sampling).
Approximate LSL Length: 80 ft (24.4 m)
Ave Monthly Water Use: 4,667 gal. (17,667 L) – Data available only for Aug-Oct 2011

Figure S38: Sequential Lead Results - Sample Site #35 (June and Sept/Oct)

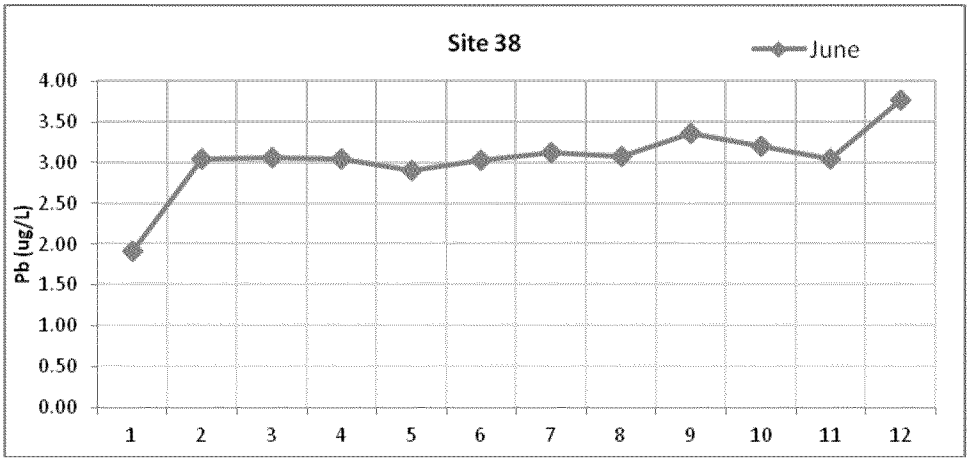
Site 36		
Liter	June	Sept/Oct
1	5.0	4.6
2	6.9	5.0
3	7.7	5.5
4	8.5	6.1
5	9.9	13
6	9.8	12
7	9.5	10
8	9.3	10
9	9.2	11
10	8.9	10
11	9.2	9.9
12	9.2	



Disturbance(s): No known disturbance
Approximate LSL Length: 83+ ft (25.3 m)
Ave Monthly Water Use: Not metered

Figure S39: Sequential Lead Results - Sample Site #36 (June and Sept/Oct)

Site 38	
Liter	June
1	1.9
2	3.0
3	3.1
4	3.0
5	2.9
6	3.0
7	3.1
8	3.1
9	3.4
10	3.2
11	3.0
12	3.8



Disturbance(s): No known disturbance
Approximate LSL Length: 51 ft (15.5 m)
Ave Monthly Water Use: Not metered

Figure S40: Sequential Lead Results - Sample Site #38 (June)

Sampling collection and reporting instructions and forms

March/April sampling – The sampling instructions and forms below were used in the March/April sampling. Sampling was scheduled to conclude in March, but the sampling ran into April. As a result of the instructions below, some volunteers sampled one day at the kitchen tap and one day at the bathroom tap. The intent was to have all samples collected from the same tap, so volunteers that split the samples were asked to collect replacement samples so that a complete set of four samples was collected at the same tap. We chose the kitchen tap, and all samples collected thereafter were also collected at the kitchen tap. In addition, the 45-second flushed

sampling protocol was not used after the March/April sampling due to the complication with corroded galvanized pipe.

General Sampling Instructions

You will be taking a total of 8 samples for this study. One set of 4 samples will be taken in March 2011 and one set of 4 samples (using the same instructions) will be taken in August 2011.

General Instructions for all four samples of a set
Sample #1 and Sample #2 must be collected one after another on the same day.
Sample #3 and Sample #4 must also be collected one after another on the same day, and within the same week as Sample #1 and Sample #2.

All samples should be collected from taps that are generally used by your household for drinking water. Do not collect samples from a taps that have not been used within the last 24 hours. Use a kitchen or bathroom cold-water faucet for your sampling.

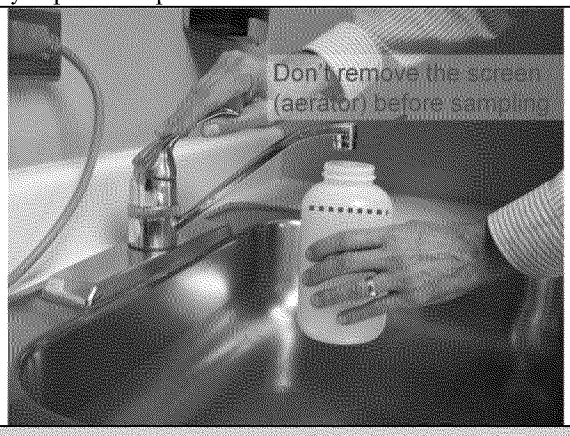
Do not collect samples from a tap that has a water filter or is connected to a water softener. If you have a water softener or water filter on your kitchen tap, collect your sample from a bathroom tap that is not attached to the water softener or water filter, if possible.

Instructions for Collecting Sample #1

Important: Please make sure you use the bottle labeled 'Sample #1' for your first sample!

Collecting Sample #1: The first sample is to be collected after water throughout the household *has not been used for a minimum of 6 hours* (example: midnight to 6am). During these 6 hours, do not flush toilets, shower, or run water from other faucets. The best time to collect samples is either:
1) First thing in the morning, before any water is used in the household; or 2) Immediately upon returning from work, and prior to using any water, as long as water has not been used in the household during the day.

1. When you are ready to collect your first sample, use the sample bottle labeled 'Sample #1'.
2. Do not run any water from the tap before collecting the first sample.
3. Place the opened sample bottle below the faucet and gently open the cold water tap.
4. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs below) and turn off the water. Tightly cap the sample bottle.



Instructions for Collecting Sample #2

Important: Please make sure you use the bottle labeled 'Sample #2' for your second sample!

Collecting Sample #2: This sample is to be collected from the same faucet as Sample #1, immediately after collecting Sample #1.

1. Immediately after collecting Sample #1, run the water for 45 seconds. Shut off the water, and place the opened

sample bottle (labeled Sample #2) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #3

Important: Please make sure you use the bottle labeled 'Sample #3' for your third sample!

Collecting Sample #3: Collect on a different day in the same week as Samples #1 & #2.

1. *Before* letting the water sit for a minimum of 6 hours, run the water from the faucet for 5 minutes at a high rate, and then do not use any water in the household for at least 6 hours after that (Example: Run the water for 5 minutes at midnight before going to bed, and then do not use any water in the household until collecting the third sample at 6 am the following morning).
2. Do not run any more water from the tap before collecting the third sample. Place the opened sample bottle below the faucet and gently open the cold water tap.
3. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Instructions for Collecting Sample #4

Important: Please make sure you use the bottle labeled 'Sample #4' for your fourth sample!

Collecting Sample #4: This sample is to be collected from the same faucet as Sample #3.

1. Immediately after collecting Sample #3, run the water for 45 seconds. Shut off the water, and place the opened sample bottle (labeled Sample #4) below the faucet and gently open the cold water tap.
2. Fill the sample bottle as you would normally fill a glass of water for drinking, up to the neck of the bottle (see photographs on first page) and turn off the water. Tightly cap the sample bottle.

Figure S41: March/April sampling instructions.

Sample Collection and Reporting Page

Sample Reporting – Sample #1 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #1): _____ **Date/time Sample #1 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #1: _____

Was sample #1 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #2 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #2): _____ **Date/time Sample #2 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the water was last used in household before collecting Sample #2: _____

Was Sample #2 collected from the same faucet as Sample #1: Yes ☐ No ☐

Sample Reporting – Sample #3 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #3): _____ **Date/time Sample #3 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #3: _____

Was sample #3 collected from a faucet that has a water softener or water filter? Yes ☐ No ☐

Sample Reporting – Sample #4 | **EPA Use: Visible Particulate? Yes ☐ No ☐**

Sample ID (from Sample Bottle #4): _____ **Date/time Sample #4 was collected:** _____

Volunteer ID: _____ **Sampling Location: Kitchen Faucet ☐ Bathroom Faucet ☐**

Date/time the faucet was flushed before collecting Sample #4: _____

Was Sample #4 collected from the same faucet as Sample #3: Yes ☐ No ☐

Have there been any plumbing repairs or plumbing work done within the household during the last six months (including installation of new faucets)? Yes ☐ No ☐

If yes, explain briefly (*Example – 'New faucet installed one week ago'*):

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S42: March/April sample collection and reporting form.

Sequential Sampling Instructions for June – The sampling instructions and forms below were used in the June sequential sampling.

Sequential Sampling Instructions

Please read all instructions before beginning your sampling

General Information

•Use only the kitchen faucet for all of these samples.

•Use only **cold water** and open the cold water tap all the way when filling the bottles.

•Fill each bottle to the top of the label on the sample bottle.

Sampling Instructions

•The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form on the back side of this page.

•The water must sit motionless in the home plumbing for at least 6 hours before collecting the samples so do not use water in the home after you finished running the water and until all samples are collected the following morning. Showering, flushing toilets, or other water use will affect the sampling results. It may help to tape a sign in the kitchen and bathrooms with a reminder not to use the water, in case people forget.

• The bottles are numbered, and it is very important to collect them in order (Sample 1 first, Sample 2 second, etc.).

•In the morning, when you are ready to sample, place the open bottles in order by sample number. You will be collecting the samples without shutting off the water in between samples, so you should remove the caps from all bottles so that you have all of the bottles ready to fill. You can put the caps on after all samples have been collected. Try not to let any water spill in between samples.

•Write down the date/time right before you sample on the form on the back side of this page.

•Begin by placing the Sample 1 bottle under the faucet and open the **cold water** slowly until the faucet is fully open. While one bottle is filling, grab the next bottle so that you are ready to move it under the faucet quickly.

•Once the bottle is filled to the top of the label, quickly place the Sample 2 bottle under the faucet, and continue until you have filled all sample bottles.

Sequential Sampling – Sample Collection and Reporting Form

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S43: June sampling instructions and sample collection and reporting form.

Sampling instructions for September/October – In the final round of sampling, the number and type of samples was customized to each site and sites collected 3 days of sampling. The instructions below were for a site collecting one NHU First-draw sample, 11 sequential samples and a 2 flushed samples. Some sites collected additional sequential samples and some collected 3 flushed samples instead of two.

Sampling Instructions
Please read all instructions before you start sampling.
General Information ☐ Use **only the kitchen faucet** for all of these samples. ☐ Use **only cold water**. ☐ Open the cold water tap all the way when filling the bottles. ☐ Fill each bottle to the top of the label on the sample bottle.
Sampling Instructions ☐ There are three different sets of samples for you to collect (Sample Set #1, #2 and #3). ☐ Each set will be taken on a different day. (The three sampling sets do not have to be taken on three days in a row.) ☐ A section of the reporting form (attached) needs to be filled in for each day of sampling.
A) Sample Set #1 (1 bottle, Blue Label) 1. The water must sit motionless in the home plumbing for at least 6 hours before collecting the sample. Typically, the night before taking the sample, make sure that no one uses water in the home until you collect the sample from the kitchen the following morning. 2. In the morning, when you are ready to sample, write down the date/time on the attached form. 3. Fill up the bottle with the BLUE LABEL. That's it for collecting the first sample set.
B) Sample Set #2 "Sequential Sampling" (11 bottles, WHITE LABELS) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, your first water usage should be collecting eleven samples in a row (one after another). Use the bottles with the WHITE LABELS. The samples should be collected without shutting off the water in between samples. To do this, remove the caps from all eleven bottles before you turn on the water. 3. Place the eleven open bottles in order by sample number before you start collecting the samples Try not to waste water in between the samples. You can put the caps on after all 11 samples have been collected. The bottles are numbered Seq 01, to Seq 11. **It is very important to collect the samples in order** (Seq 01 first, Seq 02 second, etc.). 4. Use the attached reporting form to note the date and time that you started taking the sample set.
C) Sample Set #3 (2 Bottles, GREEN LABEL and YELLOW LABEL) 1. The night before sampling (right before everyone goes to bed) run the water from the kitchen tap for at least 5 minutes. Write down the date/time you finished running the water on the form. After running the water for 5 minutes, it should sit motionless in the home plumbing for at least 6 hours. 2. In the morning, when you are ready to sample, write down the date/time on the attached reporting form. 3. Run the water for 3 minutes, then collect a sample in the jar with the GREEN LABEL. Continue to let the water run for an additional 2 minutes (for a total of 5 minutes), and collect the final sample in the bottle with the YELLOW LABEL.

Figure S44: Sept/Oct sampling instructions.

Sample Collection and Reporting – Sampling set # 1 (Blue label)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____ _

Date/Time Volunteer Began Collecting Samples: _____ _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____ ____

Sample Collection and Reporting - Sampling set # 2 (11 samples, White labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ _ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Sample Collection and Reporting - Sampling set # 3 (Green and Yellow labels)

Volunteer ID: _____

Sampling Information

Date/time the water was last used in household (the night before collecting the samples): _____

Date/Time Volunteer Began Collecting Samples: _____

Were All Samples Collected from the Kitchen Tap? Yes ☐ No ☐

FOR EPA USE: Samples received by _____ **Date/Time:** _____

Samples transferred to Region 5 Laboratory by _____ **Date/Time:** _____

EPA Use: Visible Particulate in any samples? Yes ☐ No ☐ **If Yes – List Samples With Particulate** _____

Volunteer Certification: I have read the sampling instructions and have collected the samples in accordance with the instructions provided.

_____ ***OR*** _____

Signature/Date ***Volunteer ID/Date***

Figure S45: Sept/Oct sample collection and reporting form.

Literature Cited/References

1. Triantafyllidou, S.; Edwards, M., Galvanic corrosion after simulated small-scale partial lead service line replacements. *Journal American Water Works Association* **2011,** *103*, (9), 85-+.
2. Renner, R., Reaction to the Solution: Lead Exposure Following Partial Service Line Replacement. *Environmental health perspectives* **2010,** *118*, (5).
3. Cartier, C.; Arnold Jr, R. B.; Triantafyllidou, S.; Prévost, M.; Edwards, M., Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research* **2012,** *46*, (13), 4142-4152.

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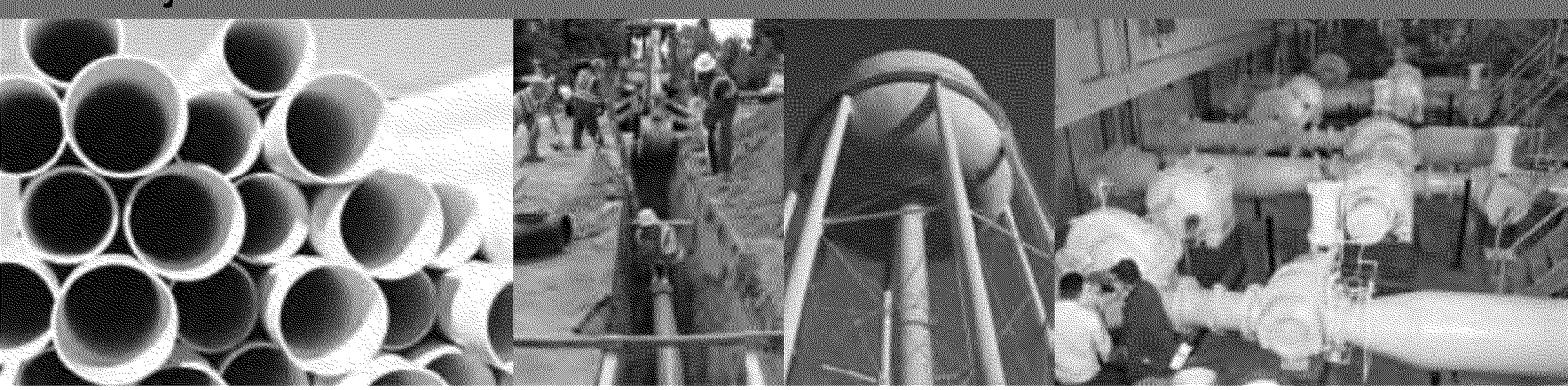
Web Report #4088b

Subject Area: Infrastructure



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Contribution of Galvanic Corrosion to Lead in Water After Partial Lead Service Line Replacements

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ABSTRACT

Partially replacing lead service lines with copper is a common practice for some water utilities, and it can be required by law if the federal limit for lead in tap water is exceeded. The goal of this practice is to reduce lead levels in tap water, by reducing the amount of lead pipe that contacts the water. However, field data have indicated that lead concentrations may increase for a short duration after partial replacements, and their potential longer-term effects have never been subjected to a rigorous scientific study. Furthermore, a recent announcement by the Centers for Disease Control (CDC) revealed that partial lead service line replacements may have actually increased the incidence of elevated blood lead in children. The preliminary results of the CDC epidemiological study prompted an important update to lead poisoning prevention program managers on potential dangers of the practice.

The work presented herein was supported by a Water Research Foundation Project Continuation Reserve (PCR) fund, and is an extension to Project 4088 entitled "Impact of Chloride:Sulfate Mass Ratio (CSMR) Changes on Lead Leaching in Potable Water." The Water Research Foundation extended that project to examine effects of CSMR and galvanic corrosion on lead leaching to potable water after partial lead service line replacements.

Due to property ownership issues, partial lead service line replacements (and not full replacements) are widely implemented in the US, with a purported goal of reducing lead-in-water problems. During a partial-pipe replacement, a portion of the lead service line is typically replaced with copper pipe, and the dissimilar metallic pipe materials are then connected to restore drinking water service. This condition creates an electrochemical or galvanic cell, which can accelerate corrosion of the lead pipe via galvanic connection to copper. In this work, the adverse effects of such connections in the context of lead leaching were confirmed in experiments of simulated lead service line replacements. Under stagnant water conditions, galvanic connections between lead pipe (either new or old) and copper pipe increased lead release into the water, compared to a full length of lead pipe alone.

The extent of galvanic corrosion observed was dependent on drinking water quality. Exposure to synthetic water of high CSMR (i.e., CSMR of 16) increased lead release from the Pb:Cu test rigs by 3-12 times, compared to low CSMR water (i.e., CSMR of 0.2). Higher galvanic currents between lead and copper were measured when the CSMR was high, mechanistically explaining the trends in lead release. Even though more work is needed in order to quantify the relevant contribution of galvanic corrosion to lead release, consideration of its long-term impact after partial lead service line replacements with copper is very important.

A conference presentation is available on this work:

- Triantafyllidou, S.; Nguyen, C.; Edwards, M. Contribution of Galvanic Corrosion to Lead in Water after Partial Lead Service Line Replacements. 2009 AWWA Water Quality Technology Conference. Seattle, WA. Presentation WED7. November 2009.

Key aspects of this study are also available in the form of a webcast, accessible through the Water Research Foundation website:

- Nguyen, C., Triantafyllidou, S., Stone, K., Clark, B., Edwards, M., Gagnon, G, Knowles, A.. Impact of Chloride:Sulfate Mass Ratio (CSMR) Changes on Lead Leaching in Potable Water. March 16, 2010.

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Contribution of Galvanic Corrosion to Lead in Water After Partial Lead Service Line Replacements

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Detection and Evaluation of Elevated Lead Release from Service Lines: A Field Study

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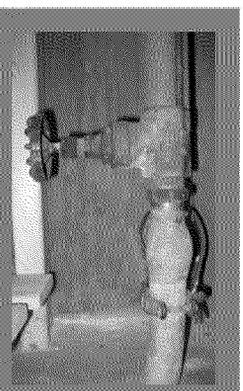
* Supporting Information

Comparative stagnation sampling conducted in 32 homes in Chicago, Illinois with lead service lines demonstrated that the existing regulatory sampling protocol under the U.S. Lead and Copper Rule systematically misses the high lead levels and potential human exposure. Lead levels measured with sequential sampling were highest within the lead service lines, with maximum values more than four times higher than Chicago's regulatory compliance results using a first-draw sampling protocol. There was significant variability in lead values from different points within individual lead service lines and among different lead service line sites across the city. Although other factors could also influence lead levels, the highest lead results most often were associated with sites having known disturbances to the lead service lines. This study underscores the importance and interdependence of sample site selection, sampling protocol, and other factors in assessing lead levels in a public water system.



INTRODUCTION

Background. Most lead in drinking water comes from premise plumbing materials and lead service lines (LSLs). LSLs are generally the largest source of lead in drinking water when they are present in public water systems.[1] The 1986 Safe Drinking Water Act Amendments banned new lead pipes in the potable water network, but a legacy of millions of partial or whole LSLs remains in many public water systems.[2] Where the term "lead corrosion" is used, it refers to the corrosion of lead plumbing materials that result in the transfer of dissolved or particulate lead into the drinking water.

The Lead and Copper Rule (LCR) sampling is intended to measure the lead levels in drinking water to assess the effectiveness of corrosion control treatment utilized by public water systems (PWSs) to minimize lead in drinking water. PWSs are required to use sampling sites that are presumed to be the highest-risk sites for lead release, and to optimize corrosion control to minimize lead levels at consumers' taps. Most published sampling studies typically focus on systems having high lead levels or systems that have experienced challenges in attempting to balance LCR compliance with various other treatment or water quality objectives. Except for LCR compliance data, little published data exists or is available for systems that are considered to be operating with optimal corrosion control and meeting the lead action level (AL) in the LCR. This study focuses on a system that is considered to have optimized corrosion control using a blended phosphate, with a relatively stable water quality, and compliance results historically well below the lead AL. This situation is representative of a large percentage of systems serving 100,000 or more people that utilize orthophosphate or blended phosphates for corrosion control and the vast majority of systems are meeting the lead AL based on the current sampling protocol in the LCR. Additional information on the LCR and study is available in the Supporting Information (SI). This study focused on whether (1) the current LCR compliance sampling protocol adequately captures the peak lead levels in a water system; (2) "preflushing" (PF) results in capturing lower lead levels in samples compared to samples collected under normal household usage (NHU) conditions; (3) a first-draw sampling protocol appropriately determines the adequacy of optimal lead corrosion control in water systems with LSLs; and (4) there is seasonal variability in the sampling results using the different sampling protocols.

System Information. The Chicago Department of Water Management (CDWM) operates two similar conventional surface water filtration treatment plants serving approximately 5.4 million residents, including those in 125 suburbs. Lake Michigan is the sole water source, with relatively stable water quality leaving the treatment plants and in the distribution system (Table 1). Before the LCR, CDWM utilized pH/alkalinity adjustment for corrosion control. CDWM switched to a proprietary blended phosphate at both plants between 1993 and 1994 which is still used as the primary corrosion control treatment.

The LCR requires public water systems to collect lead samples using a first-draw (FD) sampling protocol, and samples were collected almost exclusively from single-family homes with LSLs as required by the LCR sample site selection require-

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Table 1. Water Quality Data 2011

parameter	outlets min	outlets max	distribution min	distribution max
temp (°C)	4	24	5	23
turbidity (NTU)	0.1	0.2	0.1	0.4
pH	7.5	7.8	7.7	7.8
Cl_2 residual (mg/L)	1.0	1.2	0.7	0.9
total alkalinity (mg/L as $CaCO_3$)	103	108	98	108
chloride (Cl, mg/L)	16	20	17	20
sulfate (mg/L)	29	31	29	30
Ca (mg/L)	34	39	34	39
PO_4 (mg/L)	0.4	0.6	0.5	0.5
total PO_4 (mg/L)	0.8	1.1	0.8	1.2
Al (μg/L)	34	126	29	113
Fe (μg/L)	<5	<5	<5	34
Mn (μg/L)	<3	<3	<3	<3

ments.[3] Since the initial LCR monitoring, Chicago has exceeded the lead AL only once, during July-December 1992, with an average 90th percentile compliance monitoring value between 1999 and 2010 of 6 μg/L (SI Table S2).[3]

The LCR requires 1-L, FD tap samples of water that has stood motionless in the plumbing system (i.e., has stagnated within the plumbing) for at least 6 h. The two variants of the FD sampling protocol currently used by public water systems are defined herein as the NHU first-draw sample, where water is used in a normal household manner, and then allowed to sit motionless in the plumbing for at least 6 h before the sample is collected; and the PF first-draw sample, where the water is run from the sampling tap for a specified amount of time immediately prior to the stagnation period. However, the LCR does not provide specific details on water use during the stagnation period.

Almost all PWSs in the U.S. rely on residents to collect compliance samples under the LCR and there are differences across the U.S. in how systems instruct residents not to use the water during the stagnation period prior to collecting the sample. A review of example sets of sampling instructions provided to residents by large PWSs in the U.S. found that some are instructed not to use any water from the tap to be sampled during the stagnation period. Others are instructed not to use any water in the household. Prior to 2009, CDWM used the PF first-draw sampling protocol, with a 5-min preflush preceding stagnation. Recent instructions to residents included not using water from the sampling tap or from any nearby tap until the (poststagnation) samples were collected, and to collect samples as soon as possible after the minimum required 6-h stagnation period. Regardless of the sampling protocol, resident-collected samples necessitate the use of simple instructions and make it difficult to ensure strict adherence to any sampling protocol. In addition, the diverse premise plumbing materials and configurations (SI Table S1) represent varying effects of flow rates, hydraulic flow characteristics, and possible lead sorption/particle release effects on the shapes of the lead profiles, particularly with corroded galvanized pipe locations.[4,5]

MATERIALS AND METHODS

Sampling Objectives and Protocol. Since the promulgation of the LCR, new research on lead corrosion has shown that there are many mechanisms and water quality factors involved.[1,4,6-11] Specifically, the sampling protocols used in this study were evaluated to determine if

- preflushing biases results;
- first-draw samples, with or without preflushing, capture the "worst-case" level of lead corrosion under normal use conditions; and
- seasonal variability affects lead concentrations (in this water system).

Consistent with the LCR requirements and CDWM compliance sampling, samples for this study were collected by volunteer residents from 32 single-family residences, built between 1890 and 1960, with LSLs. An additional 5 homes were sampled and determined not to have LSLs, and were therefore excluded from further sampling. All results are included in the Supporting Information, but the non-LSL sites were not used in the data analysis (SI Tables S4a, S5, S6a, S6b, and S7).

Information was requested on the specific plumbing configurations of each sampling site to a much greater extent than the regulatory requirements which simply require the plumbing material to be identified. This information, along with analyses conducted for lead, copper, iron, and zinc for each sample, facilitated a better understanding of the observed water lead levels. Residents were asked to (1) complete a plumbing profile identifying the kitchen tap and meter or internal shut-off valve, and (2) describe the internal plumbing, including any recent plumbing work (SI Figure S1). The information provided by residents along with the results of the four metals provided additional information on the sequences of plumbing materials, and the presence of in-line brass plumbing components. CDWM provided the locations of water mains, service line materials, work conducted by the city at each residence (meter installation or repair, shut-off valve repair/replacement, service line leak repair, street excavation), and monthly water use data for residences with water meters. The information provided by CDWM on water main locations was used to measure the distance from the water main to each residence, and internal plumbing information provided by residents was used along with the measured length from the water main to the residence to approximate the LSL length (SI Table S1).

Residents were provided with written sampling and reporting instructions for each sampling event (SI Figures S41-S45). One-liter, high-density polyethylene (HDPE), wide-mouth (5.5 cm, 2.2 in.) sample bottles were used to collect all samples. Residents were instructed not to remove aerators prior to sampling and not to collect samples after point-of-use or point-of-entry treatment devices.

Several prior studies have suggested that significant contributions of particulate-associated lead can be mobilized as a function of flow rate and turbulence in certain water chemistries, though studies have not developed predictive relationships to premise plumbing material, scale composition, and hydraulic flow characteristics.[6,10-15] To try to achieve the most aggressive high flow conditions under realistic field conditions, residents were instructed to collect all samples by slowly opening the cold water kitchen tap until fully open. Upon receipt, the samples were inspected by EPA for visible particulate matter prior to delivery to the laboratory.

For all first-draw samples, residents were instructed not to use any water throughout the household (i.e., no showering, washing clothes/dishes, flushing toilets, etc.) during the

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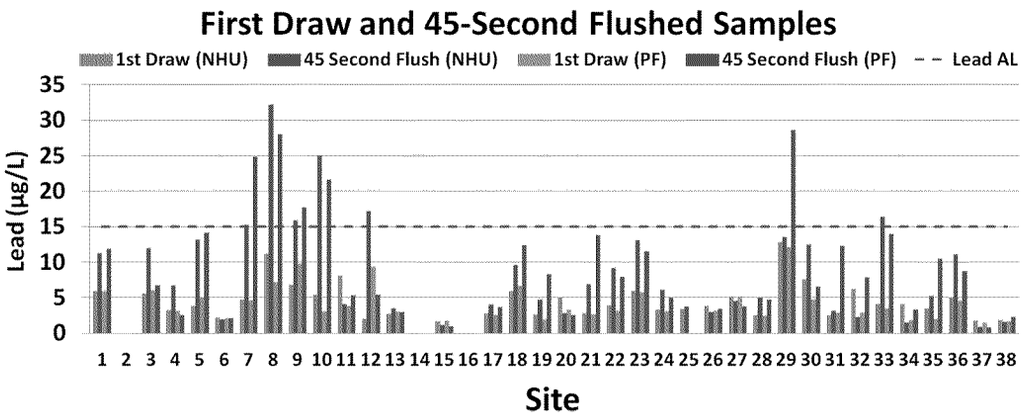

Figure 1. First round lead results for all sites.

minimum mandatory 6-h stagnation period. In this study, PF samples include a flush of at least 5 min prior to the mandatory minimum 6-h stagnation period. A NHU sample had no preflushing prior to the mandatory minimum stagnation period. Residents were instructed to allow the water to sit motionless in the household plumbing a minimum of 6 h, but not more than 24 h, and to record the dates/times the taps were flushed prior to the stagnation period, and the dates/times samples were collected following the stagnation period. First-draw samples using both variants (NHU and PF) were collected in the first and third rounds of monitoring in March/April and September/October, respectively. Additionally, 45-s flushed samples were collected in the first round to evaluate whether a second-draw sample more accurately captured the level of corrosion. Three-min, 5-min, and 7-min flushed samples were collected in the third round of sampling to provide guidance to volunteers when high lead levels were found (SI Table S7). This information can also be used to provide site-specific guidance on minimum flushing times necessary to reduce consumer exposure to lead in drinking water.

In the first round of sampling, each resident collected a NHU first-draw sample and then a second-draw (45-s flushed) sample after allowing the water to run for 45 s. On the second day, residents collected a PF first-draw sample and then a second 45-s flushed sample. EPA's current Public Notification Handbook advises[16] residents to run the water 30 s or until it turns cold before consuming, if the water has not been used for an unspecified "extended period of time", which can result in higher lead levels at the tap for consumers. It has also been previously demonstrated that in some situations, this advice can cause residents to consume the worst-case water sitting stagnant in the LSL.[17] (Figure 1)

Sites 14, 15, 16, and 37 were verified as not having LSLs and were excluded from further sampling. Site 2 was verified as not having a LSL following the June sequential sampling and was excluded from the final round of monitoring. The 45-s flushed sampling was discontinued following the March/April sampling first round due to the presence of severely corroded galvanized pipe in some of the residences (SI Figure S4) which reduced the inner pipe diameter, restricting water flow and resulting in varying volumes of water flowing through the plumbing for the same flush time.

In June 2011, each resident collected a total of twelve PF sequential samples in one day of sampling. The first PF sequential sample was also the PF first-draw sample for the data analysis. All samples were analyzed for lead, copper, zinc, and iron. The co-occurrence of the metals, along with plumbing details, was used in qualitative assessments to correlate lead results with potential sources of lead in the plumbing network (SI Figure S6).[4,10]

In September/October 2011, each resident collected a NHU first-draw sample, and a minimum of 11 PF sequential 1-L samples. Sites with high lead levels in the previous rounds collected an additional 3 or 4 PF sequential samples, and one site with a very long LSL (159 ft, 48 m) collected an additional 9 PF sequential samples. The additional PF sequential samples were collected to determine the point at which lead levels consistently dropped below the AL. All samples collected are included in the sampling summary with the numbers and types of samples collected at each site (SI Table S3).

Most stagnation times were relatively consistent across most sites at between 6 and 8.5 h, and all but two sites had stagnation times between 6 and 9 h 10 min, which facilitated unadjusted comparisons (SI Table S6c).

Additional flushed samples were collected in September/October for high lead sites in order to provide residents with guidance on minimizing lead levels in their drinking water. Recommended minimum flushing times were then estimated based on the lead levels and LSL lengths. These results are included in the Supporting Information, but not discussed here.

Sample Analyses. All samples were visually inspected for particulate matter prior to delivery to the EPA Chicago Regional Laboratory. Samples were preserved upon receipt by the laboratory using concentrated nitric acid to pH <2 and held for a minimum of 24 h prior to analysis.[18] The laboratory's Reporting Limits (RL) for lead, copper, and zinc in drinking water samples, using EPA Method 200.8, are 0.5, 1, and 10 µg/L, respectively. The laboratory's RL for iron in drinking water samples, using EPA Method 200.7, is 80 µg/L. Additional laboratory information is included in the Supporting Information.

RESULTS AND DISCUSSION

Both Variants of the First-Draw Protocol Significantly Underestimated Peak Lead Levels, and the NHU First-Draw Protocol Yielded Higher Results Overall than the PF First-Draw Protocol. The 90th percentile lead values for all three rounds of first-draw sampling using both variants were slightly higher than Chicago's historical compliance results, but still fell well below the lead AL (SI Table S4b). Only 2% of the total number of first-draw samples (3 of 151) exceeded the AL despite the presence of lead levels well above the lead action

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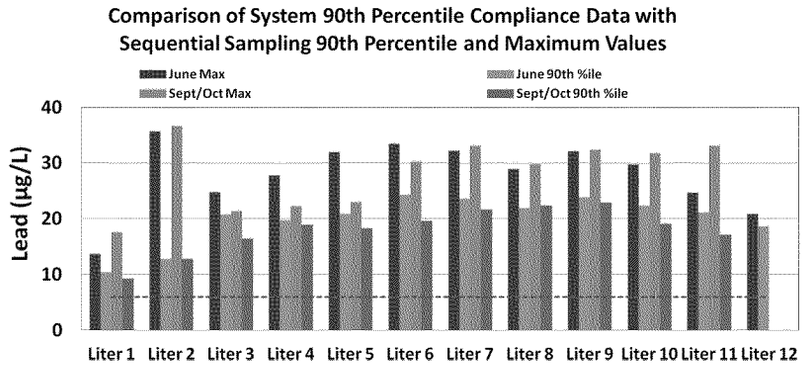

Figure 2. Comparison of 90th percentile LCR compliance data to 90th percentile values from LSL samples (across sites by liter) and maximum values from LSLs. The green dashed line indicates the average 90th percentile compliance monitoring value for Chicago between 1999 and 2010 of 6 µg/L.

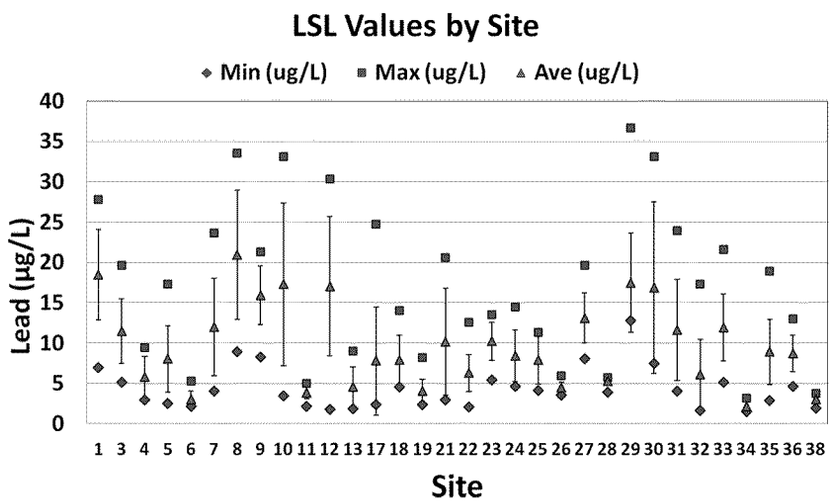

Figure 3. LSL results were highly variable within each LSL and from site to site. Error bars represent 1 standard deviation.

level within the service lines as indicated by the 45-s flushed results in the first round of monitoring and sequential sampling results in the second and third rounds.

In contrast, if the 90th percentile value of each of the successive sequential liter samples from the LSLs is computed across all sampling sites, the lead levels were up to four times higher than Chicago's average 90th percentile value using FD samples. Some peak values for each sequential liter calculated across all sampling sites were over twice the lead AL and up to six times higher than the regulatory compliance data (Figure 2). In summary, 69 of 336 (21%) of the individual sequential samples collected in June and 75 of 319 (24%) of sequential samples in September/October exceeded the lead AL, indicating that current sampling protocols will often considerably underestimate the peak lead levels and overall mobilized mass of waterborne lead in a system with lead service lines.

The NHU results were numerically higher overall than the corresponding PF values for most sites, but the differences were not statistically significant. The PF first-draw protocol produced lower individual results than NHU first-draw protocol in 23 of 32 sample pairs in March/April, and 20 of 27 sample pairs in Sept/Oct (SI Table S4a). Although NHU first-draw samples were collected without directing the residents to flush the tap prior to the stagnation period, NHU can involve showering, washing dishes, or doing laundry a short time prior to the stagnation period, which could clear the lead from the pipes similar to preflushing the tap. Thus a NHU sample can be effectively the same as a PF sample and yield similar results. Since the sequential sampling results from these same sites show that there is much higher lead present within the LSL at the same time that the NHU and PF first-draw samples were collected, it stands to reason that if the NHU activities were not undertaken, and a larger sample set were used, the NHU samples would yield results that were statistically higher than the corresponding PF samples. The distance from the kitchen tap to the beginning of the LSL was highly variable, ranging from approximately 3 to 87 feet (0.9 to 27 m), and the measured LSL lengths ranged from 43 to 159 feet (13 to 48 m). Consequently, for sites with shorter total plumbing lengths, the initial and final sequential samples would include relatively uncontaminated water from the water main following the 5-min tap preflushing. These samples would contain little to no LSL lead contribution, consistent with plumbosolvency and radial diffusion/flow principles.[5,19,20] A targeted LSL sampling protocol isolating only LSL contact water would likely yield a higher percentage of results above the lead AL for systems with Pb(II) pipe scale chemistry, but the specific location of the peak lead levels will necessarily vary with premise plumbing configurations.

Seasonal Variability. In a site-by-site comparison, lead concentrations were higher in Sept/Oct than in Mar/Apr or June, with the starkest statistical difference between first-draw

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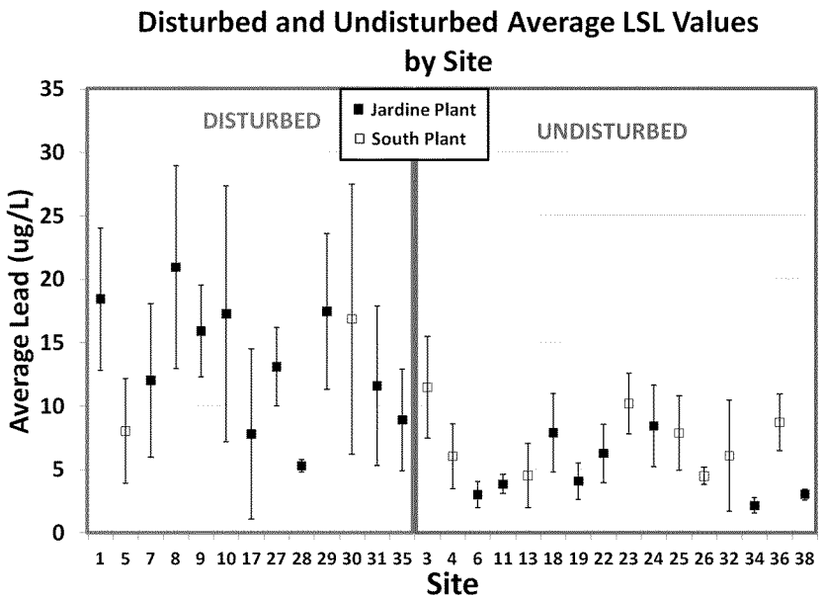

Figure 4. Average lead levels at disturbed and undisturbed sites. Error bars represent 1 standard deviation.

NHU samples collected in Mar/April and Sept/Oct ($p = 0.03$ for two-tailed paired Student's t-test). Overall, 68% and 69% of NHU and PF first-draw samples, respectively, were higher in Sept/Oct than in Mar/Apr, while 55% of paired sequential samples were higher in Sept/Oct than in June. Seasonal variation in lead levels consists of multiple contributing factors from the source water through the premise plumbing which could not be precisely isolated in this study, but the results in this study are consistent with other findings on seasonal variability (SI Table S6d).[21] Factors include (1) water temperature, (2) water chemistry variation, and (3) fluctuations in water usage for Sept/Oct versus June, which could increase or decrease lead levels.[22,23]

Lead Concentrations Vary Throughout Each Individual LSL and among Different LSLs Across the System. There was a high degree of variability in sequential sample results at most sites, some of which could include a particulate-bound component as reflected in spikes in some sequential sampling results (SI Figures S9-S40). For most sites, no individual sample result from within the LSL can characterize the lead concentrations at the site. Within the complete sampling profile results, lead levels at most sites ranged from well below to well above the AL (Figure 3). Under the LCR, this would mean that a system would meet the action level and have no additional regulatory requirements or would exceed the AL and be required to implement additional requirements, depending on which sample result is selected as the compliance sample. The variability within sites and between sites is similar in trend to that found in several other studies reporting sequential sampling conducted in water systems with different corrosion control strategies and chemistries from CDWM.[1,4,10,12,14,15,24-27]

Additional compliance data from a second large utility (City B) which exceeded the lead AL and conducted sampling using the temperature change LSL sampling protocol in the LCR,[3] yielded similar variability across the system (SI Figure S8 and Table S9). A total of 1975 LSL sites were sampled, with 1762 results (89%) below the lead AL; 128 results (6.5%) from 16 to 30 µg/L; 57 results (2.8%) from 31 to 50 µg/L; and 28 results (1.4%) between 51 and 580 µg/L. This LSL sampling protocol is similarly vulnerable to low biases, although many results were considerably higher than the AL (SI Figure S8).

Factors Affecting Lead Levels. The majority of high lead results occurred at sites with a documented physical disturbance of the LSL between 2005 and 2011 (Figure 4). The actual extent to which the LSL was physically disturbed is unknown for all sites, and the records of disturbances are based on information provided by CDWM and by the sampling volunteers (SI Figures S9-S40).

For the purpose of this study a physical LSL disturbance is defined as a meter installation or replacement, autometer-reader (AMR) installation, service line leak repair, external service shut-off valve repair or replacement, or significant street excavation directly in front of the home that could disturb the LSL. An "undisturbed" site is an unmetered site where neither the CDWM nor resident have a record or recollection of any disturbance, as defined above. A third category, "indeterminate", is used for three sites where CDWM has no record of any LSL disturbance, and the resident did not provide a response as to whether there has been any LSL disturbance. Cross-checking was important because information provided by volunteers in some cases contradicted CDWM records, and upon further investigation, the records were found to be incomplete and were corrected, which resulted in reclassification of the site.

Of the 13 disturbed sites, 11 sites had 3 or more sequential sampling results above the lead AL, two sites had 2 results each above the AL, and one site had no results above the AL. Of the 16 sites with no known disturbance, only three sites had any results above the lead AL. In the remaining 3 "indeterminate" sites, 30 of 81 sample results (37%) were above EPA's lead AL (Table 2).

A recent AWWA publication on the state of water infrastructure highlights the need for major infrastructure work.[28] This necessary infrastructure work will potentially increase the incidence of damage to the protective scales within LSLs as this work is performed. Inevitably, these physical LSL disturbances will continue to occur with increased frequency as part of daily routine water system maintenance and nonwater related community infrastructure work.

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Table 2. Lead Results for Disturbed, Undisturbed, and Indeterminate Sites[a]

disturbed sites			undisturbed sites			indeterminate sites		
no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL	no. sites	no. samples	no. above AL
13	327	117	16	372	6	3	81	30
% samples over AL: 36%			% samples over AL: 2%			% samples over AL: 37%		

[a]Most lead results above the AL were found at sites with LSL disturbances. Additional results above the AL were also found at sites where the status of the LSL (disturbed or undisturbed) could not be confirmed. Sites without LSL disturbances had few if any results above the AL.

Possible Implications of Water Conservation and Use. Information provided by CDWM and volunteers anecdotally suggests that low water usage may also play a role in high lead levels at some sites. Of the four locations with the highest average lead levels, three (Sites 1, 29, and 10) had documented low water usage. Site 1 had average monthly water usage of 3444 gallons (13 037 L) which does not appear to be low usage. However, information provided by the resident indicates that the majority of the monthly water usage occurs during a relatively small number of days during the month when there is a high volume of water usage. Site 29 had average monthly usage of 1826 gallons (6912 L), and Site 10 had an average usage of 1438 gallons/month (5443 L/month). For comparison, the mean single-family household water usage is approximately 8582 gallons/month (32 486 L/month), with a sizable standard deviation.[29]

In two locations (Sites 17 and 5), lead levels decreased with an increase in water usage. As water usage approximately doubled at Sites 17 and 5, maximum lead levels from sequential sampling decreased from 25 to 5.5 μg/L and from 17 to 12 μg/L, respectively. Although this represents a small set of samples, these observations support the idea that higher lead levels can be associated with low water usage.[30]

Extrapolating from prior research suggests the necessity of consistent flow to deliver corrosion inhibitor effectively into passivating films,[31] and correlates increased inhibitor dosages with reduced lead release.[10,32-35] Low water usage may inhibit healing of the damaged scales, and influence the rate of galvanic corrosion. Water usage effects cannot be separated from other seasonal effects in this study, but prior literature and the combined sequential graphs showing entire profiles shifted up or down from the June to Sept/Oct sampling suggest further investigation is warranted (SI Figures S9-S40). As conservation efforts increase, it will become increasingly important to conduct further research on the relationship between water usage and increases in lead levels.

The results in this study also indicate that more appropriate flushing guidance must be developed, based on neighborhood and premise plumbing characteristics, and whether a home has a LSL or not. Much of the current published and web-based flushing guidance inadvertently increases the risk of exposure to elevated lead levels by clearing an insufficient amount of water volume.[17] Even fully flushing LSLs may only lower lead levels to a limiting, measurable lead level, that relates to the plumbosolvency of the water, the flow rate, the length and internal diameter of the pipe,[5-7,10,19,20] and possibly effects of prior disturbances (SI Table S7).

Risk Identification and Management. Recently, CDC issued a health alert associating higher elevated blood lead levels with partial LSL replacement,[36] and also concluded that LSLs were an independent risk factor for elevated blood lead levels even when lead levels in drinking water met the LCR lead AL of 0.015 mg/L.[37] As highlighted in this study, LSLs can contribute high lead when they are disturbed in many different ways, not just due to partial LSL replacement, and water usage may also play a role in the resultant high lead levels and potential increased human exposure. In an August 2012 update on lead in drinking water and blood lead levels, the CDC notes that "The recent recommendations from the CDC Advisory Committee on Childhood Lead Poisoning Prevention to reduce or eliminate lead sources for children before they are exposed underscore the need to reduce lead concentrations in drinking water as much as possible".[38]

As the ultimate human and environmental health goal, LSLs should be completely removed where possible. The stability of the protective scales within LSLs depends on many factors which can change over time. For example, changes to water quality or treatment have resulted in high lead levels over a sustained period of time (years).[10,39-41] Under the current regulatory framework, elevated lead levels from disturbances, water quality, treatment, or water usage changes can potentially go undetected for up to 3 years between LCR compliance monitoring periods, which can result in increased public exposure over a significant period of time.

Proper selection of sampling sites, sampling protocol, and other site conditions is critical for evaluating the amount of lead corrosion and release that is occurring in the distribution system. Successful optimization of the plumbosolvency treatment depends on an accurate understanding of the corrosion mechanisms, pipe scale mineralogy and structure, and the consequences of LSL disturbances and water conservation efforts. No published studies could be found that systematically investigated the time and inhibitor doses/water quality adjustments necessary to overcome the disturbances and damage to the lead pipe scales that will be routinely occurring throughout cities across the U.S., as long as full or partial lead service lines remain in service.

Analyses of the Chicago LSL scales by EPA (to be reported elsewhere) reveal that the surface coatings on both lead service line and galvanized interior pipes from CDWM are primarily composed of amorphous aluminum, calcium, and phosphorus-rich deposits, and not crystalline lead(II) (or zinc)-orthophosphate phases that are predicted by conventional divalent lead plumbosolvency theory for orthophosphate dosing.[10,33,42] An understanding of the scales is essential to study and implement procedures and strategies for effective and timely repair of the protective scales damaged by LSL disturbances, and to minimize the public's exposure to high lead levels that can result from damaging the scales. Experimental evaluations are critical when scale compositions fall outside the scope of well-understood predictive corrosion control practices.

ASSOCIATED CONTENT

* Supporting Information

Additional background information, tabular summaries of sampling results, and graphics. This material is available free of charge via the Internet at http://pubs.acs.org.

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The authors declare no competing financial interest.

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The occurrence of particulate lead in drinking water deserves increased scrutiny. This is especially true because models of human exposure to lead, sampling protocols, analytical methods, and environmental assessments are often based on the presumed dominance of soluble lead in drinking water. Recent cases of childhood lead poisoning were tied to solder particles that detached from the plumbing and contaminated the potable water supply. In cases such as these, common sample-handling procedures can "miss" particulate lead present in water samples. In some instances, the actual amount of lead present in drinking water samples may be five times higher than that obtained using approved protocols. The presence of chloride, warmer temperature, and lower pH in the human stomach may render a significant fraction of this "missed" particulate lead as bioavailable when ingested.

Lead Particles in Potable Water

BY SIMONI TRIANTAFYLLIDOU,
JEFFREY PARKS, AND MARC EDWARDS

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Reaction to the Solution

Lead Exposure Following Partial Service Line Replacement

The safety of a commonly used construction technique for getting the lead out of drinking water—digging up old lead water pipes ("service lines") and replacing a portion with new copper pipe—has been debated for many years. At best, critics charge, this technique may waste millions of dollars by failing to reduce levels of lead in drinking water. At worst, the partial replacement technique can backfire and substantially increase lead levels for months or longer. The Centers for Disease Control and Prevention (CDC) recently warned public health officials that new agency findings suggest partial replacement of lead service lines may be linked to an increased incidence of high blood lead levels in children. Some drinking water experts are now saying the CDC and the U.S. Environmental Protection Agency (EPA) should consider jointly recommending a moratorium on partial service line replacement based on this new information.

To understand the potential impact of partial lead service

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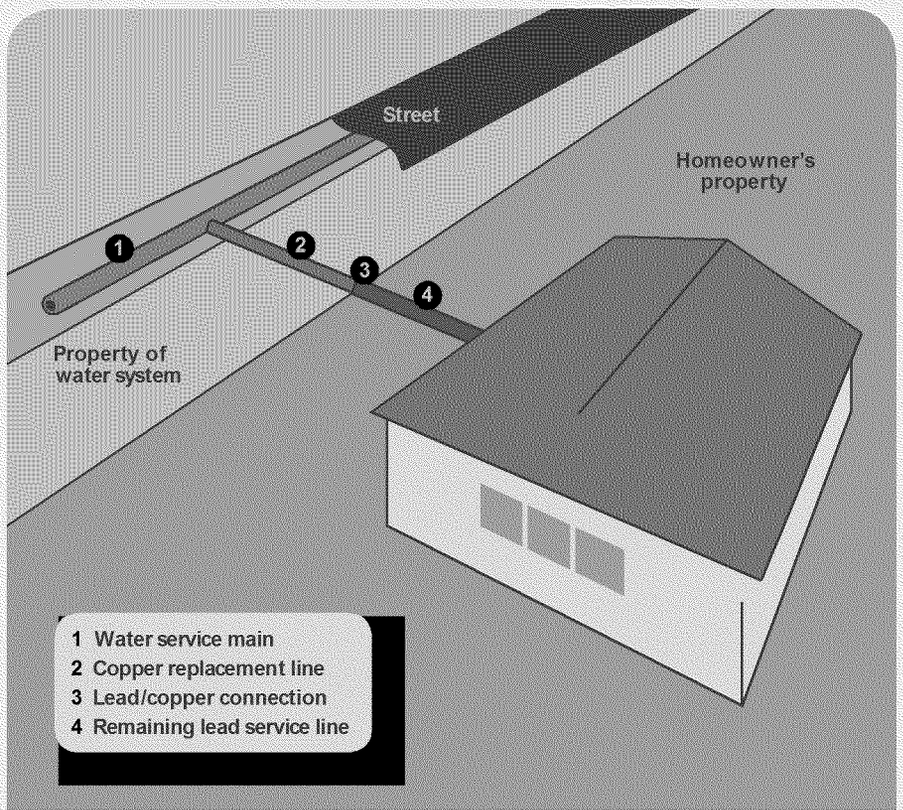

Initially the Lead and Copper Rule required water utilities to replace entire lead service lines, but constitutional and legal concerns about this stipulation led to a revision of the law in 2000 to allow for partial service line replacement. Utilities may offer homeowners the option of replacing the privately owned portion of the line at the homeowner's cost, but few exercise this option.

Matthew Roy/EHP

line replacement, Virginia Tech environmental engineer Marc Edwards suggests an analogy with lead paint. "We all understand that lead paint becomes a hazard if it's disturbed," he says. "Our research indicates that doing a partial [lead] service line replacement is analogous to creating a source of lead dust right in your home, where it is very accessible by children. The 'dust' in this case, which is in the form of lead rust on the pipe, builds up continuously over time. If and when it comes off [the pipe], the amount of lead [in drinking water] can vastly exceed health standards." He adds, "But instead of realizing this is a potential hazard, homeowners have a false sense of security, because they are told that partial service line replacements are being conducted to improve the situation."

"There is no doubt that partial lead service line replacements can result in significantly elevated levels of lead in tap water and that this contamination can continue for weeks and months, particularly in situations where [drinking water] corrosion control is not optimized," says EPA chemist Michael Schock. "Why and where these high levels occur is still the topic of research, but their occurrence is fact."

One Thing Leads to Another

The EPA started regulating lead in tap water in 1991 in the wake of numerous health studies that linked lead in drinking water to cases of severely elevated blood lead [see "Out of Plumb: When Water Treatment Causes Lead Contamination," *EHP* 117:A542–A547 (2009) and "Exposure on Tap: Drinking Water as an Overlooked Source of Lead," *EHP* 118:A68–A74 (2010)]. Water companies generally try to keep lead levels low by controlling water chemistry; water that is too corrosive can liberate more lead from pipes and solder. But if tap water lead levels continue to exceed the action level of 15 ppb after corrosion control is implemented, the federal Lead and Copper Rule (LCR) requires water utility companies to begin replacing lead service lines.

Initially the LCR required the replacement of the entire lead pipe, both the publicly and privately owned sections. But requiring water utilities to remove privately owned lead service lines raised constitutional and legal issues in terms of private property and eminent domain. A 1994 challenge in the DC Circuit Court by the American Water Works Association (AWWA) limited the EPA's jurisdiction to just the public portion of the service line. As a result the LCR was revised in 2000 to allow for partial service line replacement, although utilities may offer homeowners the option of replacing their portion of the line at the homeowner's cost.

This change was backed by the EPA's interpretation of field studies that examined the impact of partial replacement, according to EPA spokeswoman Enesta Jones. She explains that the limited number of available studies all eventually found that lead levels declined after the new pipes were installed.

Jeff Kempic, treatment technology and cost team leader with the EPA Office of Ground Water and Drinking Water, notes that weighing the relative benefits or disadvantages associated with partial service line replacement is a task that extends beyond the EPA's control. "The vast majority of partial lead service line replacements occur on a voluntary basis and are not covered by the LCR requirements," he says.

In the United States lead service lines are most prevalent in the older cities of the Northeast and Midwest. Their installation was actually required by plumbing codes in U.S. cities through the 1950s, and in some places they were installed up until Congress imposed restrictions on the lead content of plumbing in 1986. The 1990 report *Lead Service Line Replacement: A Benefit-to-Cost Analysis*, prepared for the AWWA, provides the most recent estimates for the amount of lead plumbing in the United States: approximately 3.3 million lead service lines and 6.4 million lead connections (curved pieces that join one pipe to another). Today, there could still be millions of U.S. homes with lead service lines.

Although numbers vary dramatically from city to city or even from home to home, a national survey conducted as part of a Water Research Foundation (WaterRF) project and published in the 2008 report *Contribution of Service Line and Plumbing Fixtures to Lead and Copper Rule Compliance Issues* found the average total length of service lines was 55–68 feet, with 25–27 feet of that length controlled by the water utility. The cost of replacing the customer portion can add up to several thousand dollars, and few customers do so voluntarily, according to the AWWA.

Providence Water in Rhode Island is currently required to replace 7% of its 26,000 lead service lines each year to comply with the LCR. Water companies also may voluntarily replace large numbers of lead service lines as a proactive measure to ensure water quality. Louisville, Kentucky, is replacing lead service lines voluntarily at

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an annual cost of $1.5–2 million, according to 2008 figures from the Louisville Water Company.

Partial Replacement and Elevated Water Lead

But Washington, DC abandoned an extensive and expensive lead service line replacement program in 2008 in part due to data indicating partial replacement caused higher levels of lead in drinking water for at least several months, according to George Hawkins, general manager of the District of Columbia Water and Sewer Authority, Washington, DC's water utility. The district now does partial service line replacements only when road repair or a broken water main necessitates such work, says Hawkins. In these cases the water company provides filters and monitors homeowners' drinking water for 5 months.

A 1992 report for the UK Department of the Environment titled *Economics of Lead Pipe Replacement* noted that in theory, when a lead pipe is partially replaced, the amount of lead in water should be reduced because there is less expanse of lead in contact with the water. But practical experience "shows an unexpected rise in measured lead levels after the replacement has been carried out," stated the report. "The estimates for the duration of the observed rise vary from 4 up to 18 months."

One explanation for the spike is that the replacement work disturbs the pipes and knocks off lead-bearing pipe scale (built-up minerals that coat the inside of the pipe). In addition, "partial replacements using copper piping can result in the creation of a galvanic cell," states the UK report. "This chemical cell can exacerbate the problem of plumbosolvency [lead release] and give rise to increased and erratic levels of lead observed at the tap. The effect can be persistent and may well annul any beneficial effects of reducing the length of lead pipe in the system."

Galvanic corrosion is an electrochemical process in which a metal corrodes when it is in contact with a different metal and both are immersed in an electrolyte. Galvanic corrosion creates the voltage in car batteries. This potentially powerful electrochemical phenomenon is also used to extend the life of steel water heaters through the use of sacrificial anode rods, so named because they are designed to dissolve and thereby protect other metallic parts; the sacrificial rod corrodes, not the water tank.

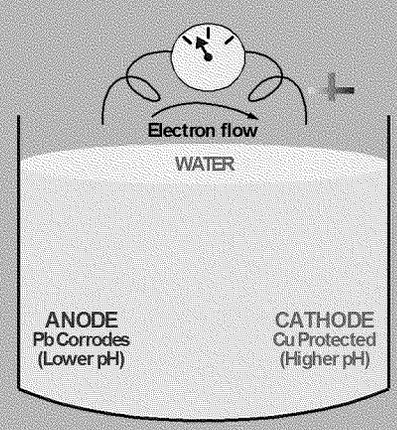
When copper water pipe is connected to lead water pipe, standard electrochemistry indicates the lead pipe should be more susceptible to galvanic corrosion. If corrosion is significant and long-lasting, it would significantly add to lead release. "The possible role of galvanic corrosion in the release of lead to water was recognized over 150 years ago, although we don't fully understand the conditions that promote or deter it," says Simoni Triantafyllidou, a graduate student in Edwards' lab at Virginia Tech.

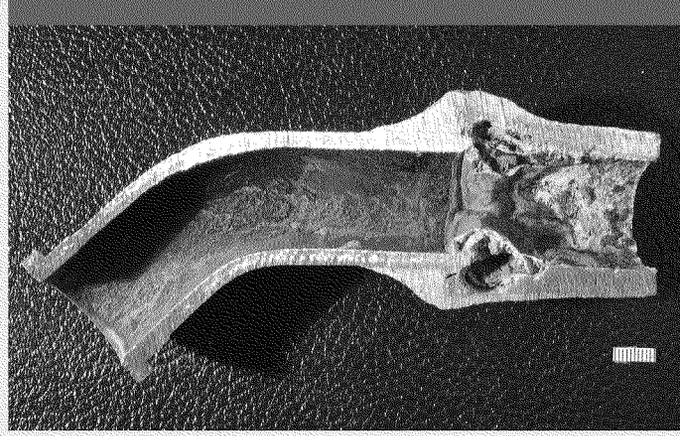
In volume 35, issue 5 (1981) of the *Journal of the Institution of Water Engineers and Scientists*, A. Britton and W.N. Richards described a number of examples taken from Scotland, where high prevalence of lead water pipes, lead water tanks, and naturally corrosive water caused long-standing problems with high levels of lead in drinking water. They studied 195 households in Glasgow, 186 of which had some lead

Galvanic Corrosion

When lead and copper plumbing pipes are connected, a galvanic cell is created. Metallic lead serves as the anode and is oxidized (corroded). The copper pipe serves as the cathode, and the drinking water flowing through the joined pipes serves as the electrolyte.

Brass contains varying percentages of copper, zinc, lead, and other metals. Some brass alloys also can be cathodic to lead; the makeup of the given alloy determines if the brass or the lead will corrode, and the pH chemistry of the water also makes a difference. In some waters, use of a short brass connector may alleviate the very high corrosion rate that can occur when lead is connected directly to copper. However, that is not always the case—as seen in these photographs of a curved brass "gooseneck" soldered to a piece of lead pipe that has corroded badly.





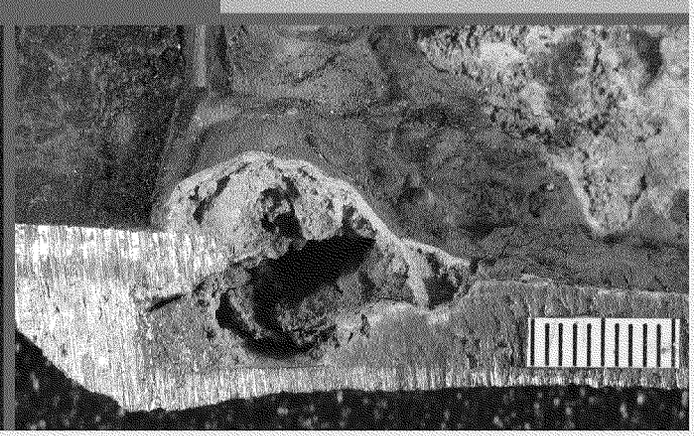


figure: Matthew Ray/EHP, adapted from Triantafyllidou et al.; photographs: Michael DeSanti/Pegasus Technical Services

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water pipes. The 69 households with mixed lead and copper piping were more likely to have high levels of lead in their tap water. "Occasionally the insertion of copper pipe can produce particularly bad results and despite satisfactory pH control it may be impossible to obtain any satisfactory samples," they wrote. Ironically, this is one of the studies used by the EPA to support partial service line replacements, notes Edwards, who adds that other researchers in England, most prominently Oliphant and Gregory, noted that in many waters serious lead contamination was caused and influenced by galvanic corrosion.

More recently, motivated by elevated levels of drinking water in Greenville and Durham, North Carolina, which appear to be related to water treatment changes that increased the ratio of chloride to sulfate in drinking water, Edwards and Triantafyllidou have been conducting experiments that evaluate galvanic corrosion by measuring electrical current between joined copper and lead pipes and also by measuring release of dissolved and particulate lead. "We have confirmed that a high chloride-to-sulfate mass ratio can trigger galvanic corrosion of lead solder and release hazardous levels of lead into drinking water, consistent with early English studies," says Triantafyllidou. "We believe this explains the elevated levels of lead in Greenville and Durham."

The chloride-to-sulfate mass ratio can change in treated drinking water due to a change in coagulant chemicals used to remove organic matter, as occurred prior to spikes in water lead in Greenville and Durham. Other factors that could increase this ratio include road salt entering a water supply from runoff, desalination, anion exchange treatment, or brine from sodium hypochlorite generators (used to disinfect drinking water) leaking into a water supply.

In contrast to Edwards and Triantafyllidou's galvanic corrosion study, which was funded by WaterRF, another WaterRF-funded experimental study conducted by consultancy HDR and published in March 2010 found that lead release due to galvanic corrosion was short-lived and transient. "Both projects recommended that more research be done in this area to better understand the short- and long-term effects of galvanic corrosion, as well as water quality effects that both exacerbate and mitigate galvanic corrosion," says WaterRF project manager Traci Case. "For now, we know that galvanic corrosion leads to lead release after partial [replacements]."

Schock and Michael DeSantis, a mineralogist with EPA contractor Pegasus Technical Services, have been examining old connections between lead and copper or lead and brass to look for signs of galvanic corrosion. Their preliminary examination of more than a dozen decades-old pipes was presented at the AWWA Water Quality Technology Conference in November 2009 and has revealed clear examples of galvanic corrosion. "We see corrosion very locally at lead junctions," says Schock. "The minerals formed on some of the lead sides of the connections show that locally the lead can be hundreds of milligrams per liter, but we can't tell how long it persists or how much of the high lead generated there [gets] into the water." But they have also found pipes where corrosion is minor as well as joints where the lead appears stable and the copper has corroded.

"There are many waters in which we are sure that galvanic corrosion is not a problem," says Edwards. "But our lab data show that the worst case can be quite bad. When problems occur, they can be very hard to detect due to erratic release of lead scale at the [lead–copper] joint. More research is needed to understand the issues associated with sloughing off of lead scale and galvanic corrosion," he says.

Signs of Health Effects

In January 2010 the CDC announced the findings of an unpublished epidemiologic study suggesting a relationship between elevated blood lead levels in children and partial lead service line replacements. A notice published on the agency's website (http://www.cdc.gov/nceh/lead/waterlines.htm) advises public health managers that customers should be informed when partial lead service line replacement occurs so they can take steps such as flushing taps and cleaning aerators after service line disruption. The CDC notice does not address how long after the service line replacement taps should be flushed or how many minutes the tap should be flushed at each use before using the water.

The notice, written by Howard Frumkin, CDC's former director of the National Center for Environmental Health, says the

Timing Is Everything

A 2005 conference proceeding document prepared by John Joseph Wujek for the District of Columbia Water and Sewer Authority, Minimizing Peak Lead Concentrations after Pwartial Lead Service Replacements, has often been cited as showing how partial lead service line replacement can improve water quality if the replacement is followed by vigorously flushing water through the tap. However, this study suffers from a flaw, according to EPA Region 3 environmental scientist Jennie Saxe.

In Washington, DC, high levels of lead occurred in part due to a November 2000 change in the disinfectant used to treat water from chlorine to chloramine. In the spring of 2004, the city switched back to chlorine for a month to control bacteria, and lead levels plummeted citywide. For the project described by Wujek, the initial, pre-replacement measurements of lead in water were made close to the time when chloramine was in the water, and lead levels were high. The post-replacement sampling was impacted by benefits from chlorine in the water when lead levels were low.

The factors that marred this study illustrate how difficult it can be to conduct field studies on the effects of partial service line replacement on tap water, says Edwards. "Water chemistry, temperature, whether the pipes have been flushed, even how fast the tap is running when samples are collected—all these factors affect field study results," he says. "With no standard protocol, these studies are difficult to compare and may provide misleading results."

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One City's Experience with Full Lead Service Line Replacement

Although there is general agreement that replacing the entire lead service line is preferable to partial service line replacement, few utilities in the United States or elsewhere have found ways to conquer the legal and financial hurdles that block full replacement. One city that has succeeded is Madison, Wisconsin.

In 1994 Madison Water Utility was faced with drinking water lead levels that exceeded EPA requirements, but studies found that most standard chemical corrosion treatments were ineffective at reducing lead levels. The exception was orthophosphate; however, orthophosphate dosing was unpalatable because the area's wastewater treatment plant had just built a biological removal system for phosphate to manage nutrient loading to the Great Lakes. In a 2006 case study, consulting engineer Abigail Cantor of Process Research Solutions wrote, "If phosphorus was to be added to the drinking water, the removal system would not work properly and a chemical phosphorus removal system would need to be added. In addition, the water that would runoff directly to the lakes would carry phosphorus with it." Already, she observed, there was talk in the city council of banning phosphorus lawn fertilizers in Madison.

Madison determined that the long-term costs of orthophosphate would be greater than the cost of total lead service line replacement and that replacing lead service lines would reduce lead levels more than dosing with orthophosphate would. The state regulator, the Wisconsin Department of Natural Resources, agreed that removing lead service lines was the only reasonable means of corrosion control available—but this meant removing the entire lead service line, both the public and private portion.

When the program began on 1 January 2001, there were approximately 6,000 lead service lines on the publicly owned portion and 5,000 on the homeowner side. The state set a goal of replacing all service lines by 2011. In 2000 the city passed an ordinance that prioritized replacements in schools and day care facilities.

But there was disagreement about who should pay for replacing privately owned service lines, says utility finance manager Robin Piper. First the utility tried to add a charge to the bills of its more than 60,000 customers, but this was vetoed by Wisconsin's utilities commissioners. "The sewer authority was able to add a surcharge, and this makes sense because choosing service line replacements over orthophosphate dosing saved water treatment costs," he says. Additionally monies came from cell phones—the utility receives rent for allowing cell phone antennas to sit atop its water towers. "At the height of replacement efforts we were spending $500,000 to $600,000, and the antenna rental fees contributed several hundred thousand," Piper explains.

The threat of lake nutrient loading made orthophosphate a no-go for Madison, Wisconsin.

Alamy

The city uses this money to help reimburse customers for half the cost of replacing their lead service line, up to $1,000. For low-income customers, the city provides a loan for the other half of the cost, with repayment deferred until the property is sold. Costs for the utility portion have averaged about $2,000, and for the customer portion about $1,400, Piper says. To date Madison is on target with 5,394 pipes replaced.

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study's preliminary results suggest that when the public portion of a lead service line is replaced, children are more likely to have blood lead levels of 10 µg/dL or higher, compared with children living in housing with undisturbed lead service lines or nonlead service lines. The epidemiologic study of children living in Washington, DC, is undergoing peer review, and publication in a scientific journal is anticipated.

"It is important to share these preliminary findings with the nation's childhood lead poisoning prevention managers to provide guidance about leadsafe water practices in homes with lead-based water lines or lead solder following plumbing work including water service line replacement," says CDC spokeswoman Bernadette Burden in explaining why the agency issued the notice in advance of the paper's publication.

"There have always been doubts about the benefits of partial service line replacement," says Alan Roberson, director of security and regulatory affairs for the AWWA. "The CDC study is the first I know of that links partial replacement to adverse health effects, but these new findings appear to confirm the existing, long-standing doubts."

Jim Elder, who headed the EPA drinking water program from 1991 to 1995, says, "Given these data, CDC and EPA should jointly recommend a moratorium on partial service line replacements."

Roberson agrees that a moratorium makes sense. Following an October 2008 EPA meeting that discussed possible long-term revisions to the LCR, AWWA representatives submitted comments to the agency stating that "aggressive and expensive [lead service line] replacement programs which do not result in exposure reductions and may in fact result in exposure increases over the short-term seem to be an inappropriate allocation of limited funds. The primary role of [lead service line] replacement in the LCR appears to be as a regulatory 'hammer,' and based on available research, it appears to be a hammer with previously unanticipated risks that are inappropriate for the LCR."

The EPA has committed to re-evaluating its regulations that cover partial lead service line replacement. In testimony before the Senate Environment and Public Works Committee in late 2009, EPA Assistant Administrator for Water Peter Silva committed to finalizing long-term revisions to the LCR by 2012. "But to meet that timetable with meaningful proposals, EPA should be doing research and fact-finding to sum up the current situation and look into options," says Roberson. "There are no indications as far as I'm aware that they are doing this."

> **This issue definitely cuts across public health, scientific, technical, and legal issues. It would be great to see collaboration among these disciplines and across agencies to figure this out.**
>
> —Michael Schock
> U.S. Environmental Protection Agency

Possible Solutions

Everyone contacted by *EHP* for this story, including EPA sources, agrees that full lead service line replacement is preferable to partial replacement. However most believe the financial and legal impediments to full replacement are difficult to surmount. When partial lead service line replacements occur, the tap should be flushed; utility recommendations vary from 15 minutes to 1 hour of full-flow flushing immedately after partial service line replacement. Afterwards, residents should use "point-of-use" home water filters that are certified under National Sanitation Foundation/American National Standards Institute (NSF/ANSI) Standard 53 for both particulate and dissolved lead (or NSF/ANSI Standard 58 for reverse osmosis systems), according to Schock. Kempic says one of the long-term LCR revisions currently under consideration at the EPA is a requirement that water filters be supplied to homes where partial service line replacement occurs.

Since galvanic corrosion requires contact between metals, there may be a fairly simple engineering solution—the use of a longer brass connector might put enough distance between the lead and copper to reduce the corrosion, says Edwards. However, the effectiveness of this solution would depend on the composition of the brass alloy used and the pH of the water. Edwards also says the use of some sort of nonconducting dielectric might be beneficial. Existing pipes can also be lined with epoxy resin, although such lining has not been tried on a large scale in the United States.

In the United Kingdom, where approximately 40% of homes have lead pipes, partial service line replacement is not widespread, according to corrosion expert Colin Hayes of Swansea University. "The emphasis in the UK has been on the optimization of corrective water treatment, most of which includes dosing orthophosphate," Hayes says. He says the success of corrective water treatment has much reduced the need for lead pipe replacement in the short term; in England and Wales, 99.77% of samples in 2008 complied with the current European lead standard.

Hayes says UK systems use relatively high levels of orthophosphate—about 3 times the levels used in the United States. However, in the United States, concerns about increasing levels of phosphate loading to the environment might limit this option. Orthophosphate dosing has not been implicated in such problems in the United Kingdom, Hayes says, although he acknowledges that "elsewhere in Europe there have been environmental concerns about using orthophosphate—mostly unfounded—and many water companies have been replacing their lead connection pipes but not those lead pipes owned by consumers."

A potential legal solution might be to stop partial service line replacements and instead require full replacement when property changes hands. Analogous steps are currently taken to correct problems with underground fuel oil tanks in some states such as Washington.

"Taken as a whole, the data suggest that galvanic corrosion can occur, but we don't know enough about the specific conditions, the scale on which it occurs, or what causes worst-case effects," says Schock. "Having said that, it is hard to see how any public agency could justify to the public purposely leaving even part of the lead pipe in the ground. Advising people to use a filter and flush their taps following partial replacement should help. This issue definitely cuts across public health, scientific, technical, and legal issues. It would be great to see collaboration among these disciplines and across agencies to figure this out."

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