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Evaluating the Effects of Full and Partial Lead Service Line Replacement on Lead Levels in Drinking Water

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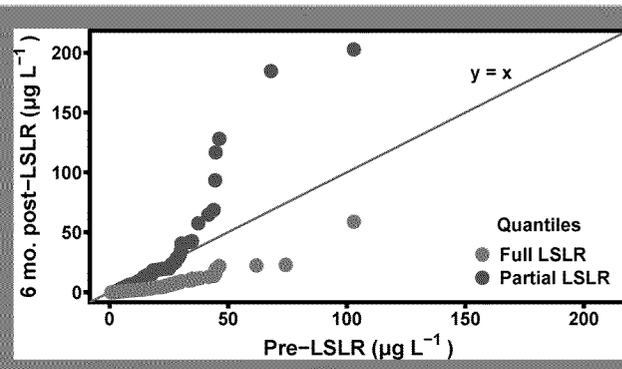
* Supporting Information

Lead service line replacement (LSLR) is an important strategy for reducing lead exposure via drinking water, but jurisdictional issues can sometimes interfere with full replacement of the lead line. The effects of full and partial LSLR on lead levels were assessed using 5 × 1-L sample profiles collected at more than 100 single-unit residences. Profiles comprised four sequential standing samples (L1−L4) and a free-flowing sample (L5) drawn after a 5 min flush of the outlet. At 45 sites with full lead service lines, 90th percentile lead levels in standing samples ranged from 16.4 to 44.5 μg L^{-1} (L1 and L4, respectively). In the free-flowing sample (L5), 90th percentile lead was 9.8 μg L^{-1}. Within 3 days, full LSLR had reduced L3−L5 lead levels by more than 50%, and within 1 month, lead levels were significantly lower in every liter of the sample profile. Conversely, partial LSLR more than doubled premises plumbing (L1, L2) lead release in the short term and did not reduce L1, L2 lead release in the long term. Even 6 months after partial LSLR, 27% of first-draw lead levels were greater than 15 μg L^{-1} (the U.S. EPA action level), compared with 13% pre-replacement.



INTRODUCTION

Lead service lines (LSLs) · pipes connecting distribution mains to premises plumbing · were installed widely throughout the first half of the 20th century in North America and even occasionally up until the U.S. congressional ban in 1986.[1] In Canada, the National Plumbing Code permitted installation of LSLs until 1975.[2] At sites where they were installed, 50−75% of drinking water lead may be attributable to the LSL.[3]

Elevated lead in drinking water is a significant public health concern because water lead levels have been shown to correlate positively with blood lead levels.[4−7] Childhood blood lead levels below 10 μg d L^{-1} · and early childhood levels as low as 2 μg dL^{-1} · are linked with deficits in cognitive and academic skills.[8−10] Chronic lead exposure in adults is associated with renal dysfunction[11] and hypertension,[12] and lead is a known abortifacient.[13] Lead in U.S. drinking water is regulated under the Lead and Copper Rule,[14] which specifies an action level of 15 μg L^{-1} for the 90th percentile first-draw lead level. In Canada, the maximum acceptable concentration in a free-flowing sample is 10 μg L^{-1}, a benchmark that serves as the current Nova Scotia regulation.[15] Health Canada also recommends corrective action when the 90th percentile first-draw lead level exceeds 15 μg L^{-1}.[2,16]

Lead service line replacement (LSLR) is an important strategy for reducing lead exposure via drinking water, but joint (public−private) ownership can interfere with full replacement of the LSL. Typically, partial LSLR occurs when the public LSL is replaced with copper and joined to the private LSL left in place.[1] Partial LSLR can cause elevated lead in drinking water; disturbance of LSL corrosion scale during replacement may release high levels of lead for an extended period post-replacement. The lead−copper junction is a specific concern due to the potential for galvanic corrosion.[3,17] Elevated lead release owing to a galvanic lead−copper couple has been demonstrated in laboratory and pilot studies and is a likely factor in persistent high lead levels following partial LSLR.[18−21] Mineralogical evidence of galvanic corrosion in lead−copper and lead−brass joints excavated from several distribution systems has also been reported.[22]

Limited residential data suggest that partial LSLR may be associated · at best · with insignificant decreases in lead exposure risk. Partial LSLR did not reduce the risk of elevated blood lead among children in Washington, DC, and children living in homes with partial LSLs were more than three times as likely to have elevated blood lead (≥10 μg dL^{-1}) compared to those in homes constructed without LSLs.[23] Other residential studies · on five or fewer sites · have reported lead spikes following partial LSLR and modest or minimal subsequent reductions in lead release.[3,24,25] Camara et al.[26] reported greater lead release following partial LSLR relative to full LSLR but did not provide detailed pre- and post-replacement comparisons.

In 2011, an advisory board to the U.S. EPA concluded that existing data were inadequate to fully assess the effect of partial

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LSLR on drinking water lead levels.[27] The board found that the few studies available had limitations · including small sample sizes and limited follow-up sampling · but did conclude that partial LSLR could not be relied upon to reduce lead levels in the short term.[27]

The objective of this work was to estimate changes in lead exposure due to full and partial LSLR via a standardized profile sampling protocol. This protocol was implemented within a water system where Pb(II) compounds · in contrast to highly insoluble Pb(IV) oxides[28] · were presumed to dominate on the basis of distributed water quality and consistent observation of peak lead levels in samples representative of LSLs. Water distribution infrastructure was considered typical of older North American municipalities. Water system characteristics featured several risk factors for elevated lead release, including (1) a significant number of unlined cast iron distribution mains,[26] (2) distributed water with low alkalinity (20 mg L^{-1} as $CaCO_3$) and pH (7.3),[29] (3) a low orthophosphate residual (0.5 mg L^{-1} as PO_4^{3-}),[29] and (4) a chloride-to-sulfate mass ratio above the critical threshold (0.5−0.77) identified in previous work as a driver of galvanic corrosion.[30,31] This study contributes to the literature by helping to address limitations identified in the EPA advisory board report: small sample sizes and limited follow-up sampling.[27] This paper also expands on previous work,[26] with analysis of a much greater volume of data: 74 and 61 full and partial LSL replacements · including paired before-and-after comparisons of 18 partial replacements · and 13 additional sites with LSLs.

MATERIALS AND METHODS

Study Area. The study area comprised single-unit residences in Halifax, NS, Canada, that underwent LSLR between 2011 and 2015. In the event of partial LSLR, electrical continuity between lead and copper was expected but not verified; following open-trench replacement, lead and copper were typically joined via a brass union as described in Clark et al.[32] Participating residences were predominantly older homes; in areas of widespread pre-1950 construction, thousands of LSLs are still in use.[26]

Sample sites received distributed water from a treatment facility employing free chlorine disinfection, and this facility is described in detail elsewhere.[33] Table 1 lists 2013−14 typical values for key treated water quality parameters, and no relevant changes in treatment were made over the study period.[34] Beginning in 2002, a blended zinc ortho/polyphosphate corrosion inhibitor (75% orthophosphate, 25% polyphosphate) was added at a treated water residual of 0.5 mg L^{-1} (as PO_4^{3-}).

Table 1. Typical Values for Treated Water Quality Parameters, Pre-Distribution

parameter	typical value
zinc ortho/polyphosphate (as PO_4^{3-})	0.5 mg L^{-1}
alkalinity (as $CaCO_3$)	20.0 mg L^{-1}
free chlorine	1.2 mg L^{-1}
hardness (as $CaCO_3$)	12.0 mg L^{-1}
total organic carbon	1.5 mg L^{-1}
pH	7.3
chloride	9 mg L^{-1}
sulfate	8.5 mg L^{-1}
turbidity	0.06 NTU
iron	<0.05 mg L^{-1}
lead	<0.5 μg L^{-1}

Sample Collection. Residents, with direction from utility staff, collected profiles of four sequential 1 L standing samples from kitchen cold-water taps, beginning with the first draw following a minimum 6 h standing period. The 4 × 1-L sample profile (L1−L4) was followed by a 5 min flush of the outlet and subsequent collection of a fifth 1 L sample (L5). Profile sampling was carried out before and at four follow-up rounds (3 days, 1 month, 3 months, and 6 months) after LSLR. Residents were instructed to record exact stagnation times (median, 7 h, 40 min; minimum, 6 h; maximum, 23 h) and to sample at a constant flow rate, but they were not instructed to remove faucet aerators prior to sample collection. Sampling instructions given to residents are provided as Supporting Information, and data were excluded from analysis when these instructions were not followed.

A complete series of pre- and post-replacement sample profiles was not available for every residential site, owing to incomplete resident participation. For this reason, sample sizes for before-and-after comparisons differ by follow-up round. However, lead levels at a given follow-up round were not significantly different (two-tailed rank-sum tests, $\alpha = 0.05$) at sites where residents participated at the next round compared to sites where they did not; reporting lead levels to residents did not appear to influence subsequent participation.

In order to assess the effect of water temperature on lead release, 13 × 1-L sample profiles were collected from kitchen cold-water taps at two other sites with LSLs (denoted sites A and B), following a 5 min flush of the outlet and subsequent 30 min stagnation. The final liter of the 13 × 1-L profile was a free-flowing sample collected after a second 5 min flush of the outlet. Sample collection by the authors at these two sites, as opposed to residents, necessitated use of an alternate sampling protocol; however, differences in collection methods limit the comparisons that can be made between these two sites and the rest of the data. Samples were collected weekly over a period of 7 or 8 weeks (sites A and B, respectively). Water temperature was measured, using a glass thermometer, in the first and last liter of each profile from the second week on.

Analytical Methods. Total lead, copper, iron, and aluminum were measured by ICP-MS (Thermofisher X Series II) according to Standard Methods 3125 and 3030.[35] Reporting limits for Pb, Cu, Fe, and Al were 0.4, 0.7, 6.0, and 4.0 μg L^{-1}, respectively. Lead was also quantified in 0.45 μm filtrate for a subset of 386 sample profiles. Filtration via cellulose nitrate membrane filters was generally performed within 2 days of sample collection (but prior to acid preservation), so results should be interpreted with care, as changes in speciation between collection and filtration cannot be ruled out. Loss of lead to sample bottles was estimated at 1.2% (SD 17.3%) by comparing total lead in 10 mL aliquots drawn from well-mixed 1 L samples before and 24 h after acid preservation of the entire sample (N = 82 sample profiles of 5 × 1-L).[36] Losses due to filtration were estimated at 21.1% (SD 0.4%) by comparing lead in 10 mL aliquots filtered once with lead in 10 mL aliquots filtered twice, using a new filter each time.[36] Polyethylene (HDPE) bottles and caps were immersed in ~2 M reagent-grade HNO_3 for a minimum of 24 h and rinsed three times with ultrapure water prior to use, and method blanks were prepared by holding ultrapure water preserved with trace-metal-grade HNO_3 in acid-washed bottles for 24 h at 4 °C.

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Table 2. Estimated Fraction of Pre-Replacement Lead Levels Remaining after Full and Partial LSLR[a]

	follow-up round	liter 1	liter 2	liter 3	liter 4	flushed	no. of sites
partial LSLR (paired)	3 d	2.89*	2.45**	1.55	1.04	0.65	15
	1 mo	1.65**	1.26	1.15	1.22	0.61**	18
	3 mo	1.24	0.86	0.54[b]	0.28[b]	0.24[b]	16
	6 mo	1.04	0.57	0.31[b]	0.25[b]	0.17[b]	16
full LSLR (unpaired)[c]	3 d	1.04	0.85	0.38***	0.29***	0.22***	48
	1 mo	0.65*	0.43***	0.20***	0.13***	0.13***	56
	3 mo	0.62*	0.41***	0.16[b]	0.08[b]	0.09[b]	45
	6 mo	0.60*	0.33***	0.10[b]	0.06[b]	0.07[b]	45

[a]Statistical significance: *$p < 0.05$, **$p < 0.01$, ***$p < 0.001$. [b]Statistically significant, but likely to have been influenced by variation in water temperature. [c]Compared against an independent set of 45 sites with full LSLs.

Data Analysis. Lead levels at each of the four follow-up rounds after LSLR were compared with pre-replacement lead levels. Sample profiles collected after full LSLR (Table 2) were compared, using two-tailed rank-sum tests,[37] with an independent set of 45 profiles collected at sites with full LSLs. Differences between groups were multiplicative (i.e., best described as ratios), but a natural log transformation yielded additive differences (i.e., best described as constants). Changes in lead level were quantified using a Hodges−Lehmann estimator, c, where $c = \text{median}(y_i/x_j)$ for all i = 1, ..., n and j = 1, ..., m. Variables x and y denote lead levels observed at m and n sites with full and fully replaced LSLs, respectively. The quantity c estimates the ratio of lead levels between the two groups, where $y = cx$.[38]

Sample profiles collected before and after partial LSLR were paired by address and compared using two-tailed signed rank tests by profile liter and follow-up round.[37] Natural log transformations were applied to the paired data to achieve symmetry in the distribution of differences. Multiplicative differences in before-and-after replacement lead levels were also quantified using a Hodges−Lehmann estimator; details, though similar to those provided above, may be found elsewhere.[38] No control of the familywise error rate for multiple comparisons was employed.

RESULTS AND DISCUSSION

Pre-Replacement Lead Levels. Lead and copper levels representing 5 × 1-L sample profiles collected at 45 sites with full LSLs are provided in Figure 1. Peak copper levels occurred in L1 (90th percentile: 151 μg L^{-1}) and peak lead levels in L4 (90th percentile: 44 μg L^{-1}). For single-unit residences, peak lead levels are often observed by L4 of the sample profile.[39] The higher median (11 μg L^{-1}) and increased variability in L3 lead levels (90th percentile: 29 μg L^{-1}) suggest that L3 stagnated at least partially within the LSL at some of the 45 sites. This is consistent with previous work, where Cartier et al.[40] estimated median (mean) premises plumbing volumes in 88 pre-1970 homes at 2.0 L (2.3 L). At sites with full LSLs, L1 significantly underestimated peak lead levels; in systems where lead(II) compounds form preferentially, lead in water that contacted the LSL during stagnation may be considerably higher than lead in the first-draw sample.[26,28,41]

Longer sample profiles (13 × 1-L) collected at two residential sites with LSLs (sites A and B, Figure 2) provide insight into the ability of the 5 × 1-L profile to estimate lead exposure. Plumbing configuration can be inferred by comparing lead and copper levels over each profile, although mixing among profile liters and the 5 min flush prior to stagnation (sites A and B only) may have influenced apparent plumbing volumes. Site A is a typical example of the single-unit residences that underwent LSLR (the large apparent premises plumbing volume of 8−9 L at site B is explained by sample collection from a second-level kitchen). The apparent premises plumbing volume at site A was 2−3 L, and copper declined sharply after the first 2 L and quickly approached the level of the flushed sample (L13, 10 μg L^{-1}). Peak lead levels were observed by L4, although L3−L9, or parts thereof, appear to have stagnated within the LSL. In light of LSL lead release observed at sites A and B following 30 min of stagnation (maximum 28 and 14 μg L^{-1}, respectively), the apparent LSL lead release at the 45 sites represented in Figure 1 was lower than expected (minimum 6 h stagnation, 90th percentile of 44 μg L^{-1} in L4). This suggests that L4 may sometimes have fallen short of the LSL. Furthermore, while L4 does appear to have reached the LSL in at least some cases, it may not have reached the lead−copper junction at sites with partial LSLs, as sample profiles reported in previous work have shown.[42] Thus, 4 × 1-L standing sample profiles may only provide an indirect assessment of the effect of galvanic corrosion following partial LSLR.

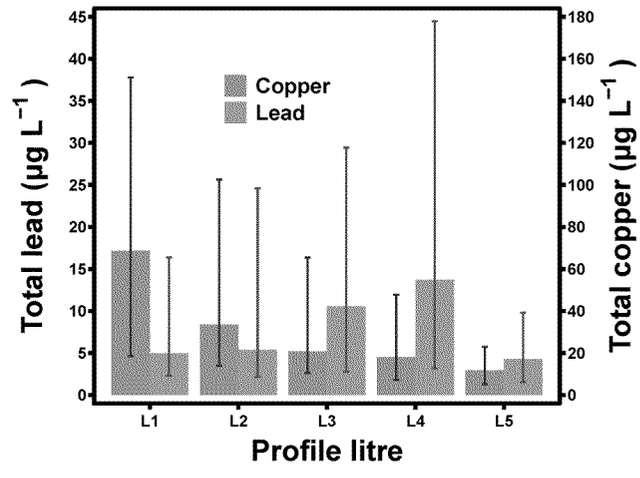

Figure 1. Median lead and copper levels in 5 × 1-L sample profiles collected at 45 sites with full LSLs. L5 is a 5 min flushed sample, and error bars represent the 10th and 90th percentiles.

Influence of Water Temperature. Pre-replacement profiles were collected in summer, and collection dates of the initial round had a July median. This introduces water temperature as a potential confounding variable. One-month follow-up collection dates had a September median, and at this interval the variation in water temperature is likely to have been low; the typical 5 min flushed sample temperature was 17.9 °C in July (this study) and 17.8 °C in October.[43] Comparisons between full and partial LSLR for the same follow-up round are

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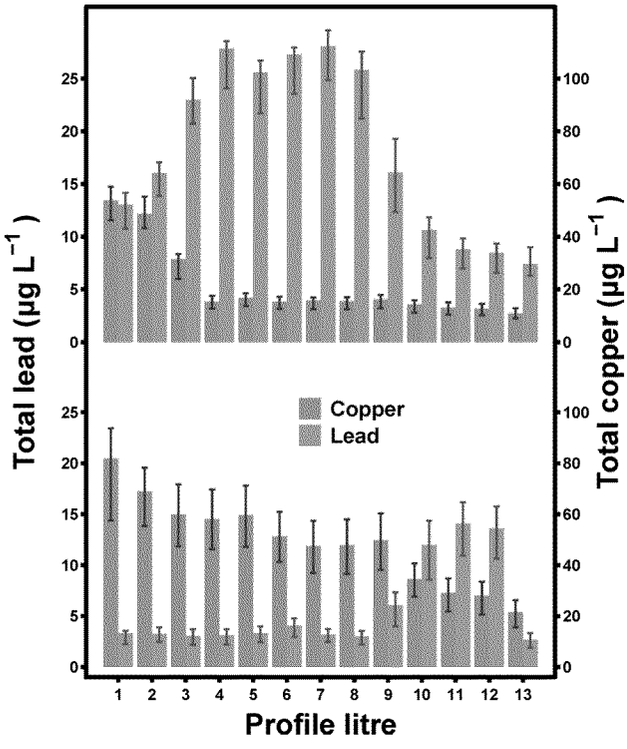

Figure 2. Median lead and copper in 13 × 1-L sample profiles collected at two sites. Site A (top) is a typical site with a full LSL, while the large apparent premises plumbing volume (8−9 L) at site B (bottom) is explained by sample collection from a second-level kitchen. Liter 13 is a 5 min flushed sample, and error bars represent the 10th and 90th percentiles.

also unlikely to have been strongly influenced by temperature. However, partial LSL profile collection dates for the 3- and 6-month follow-up rounds had December and April medians, respectively. Pre- and post-replacement comparisons of these intervals are subject to decreases in water temperature of approximately 10 °C: the typical 5 min flushed sample temperature in February was 7.2 °C.[43]

The effect of water temperature on lead release is complex: the positive effect of temperature on the rate of electrochemical reactions may be counterbalanced by the reduced solubility of lead minerals at higher temperatures, depending on the composition of corrosion scale.[44] Temperature effects also depend on whether the source of lead is premises plumbing, where seasonal variation is expected to be minimal, or LSLs, where variation is greater.[17] Previous work has shown that lead release can be temperature-dependent,[40,45] and data from 13 × 1-L profiles collected at sites A and B show that lead release was moderately to highly correlated with water temperature (R^2 = 0.46−0.98, average of 0.79). Between 13 and 19 °C, a 1 °C increase in 5 min flushed sample temperature accompanied an average 1.1 µg L^{-1} increase in lead release from LSLs [Figure S1, Supporting Information (SI)], although other seasonally varying water quality parameters, such as free chlorine residual, could have influenced observed lead levels.

Water temperature was correlated with lead release from premises plumbing at sites A and B as well, likely a consequence of the short (30 min) stagnation time. More generally, lead release from premises plumbing does not often exhibit strong temperature dependence, provided that stagnation time is sufficient for standing water to reach building temperatures (e.g., 6 h).[17] In a survey of 365 U.S. drinking water utilities, 90th percentile first-draw lead levels · collected following a minimum 6-h stagnation · were not a function of season.[46] Within the present study area, first-draw lead levels (minimum 6 h stagnation, 34 residential sites) were no higher in October than in February.[43]

Owing to the effect of water temperature on lead release from LSLs, long-term (3 and 6 month) pre- and post-replacement comparisons were only interpreted for the portion of the sample profile least likely to have been influenced by temperature, L1 and L2. It is possible that, for sites with very small premises plumbing volumes, L2 lead levels were affected by temperature variation, but no significant drop in L2 lead levels from summer to winter was observed following partial LSLR (Table 2). For L3 and L4, however, decreasing water temperature likely contributed to · and may have been entirely responsible for · observed reductions in lead release.

Effect of Full LSL Replacement. For each profile liter and follow-up round, the fraction of pre-replacement lead remaining after LSLR was estimated (Table 2), and full LSLR reduced lead levels in every liter of the sample profile within 1 month. Before and after differences were multiplicative (not additive), meaning that sites with high lead levels pre-replacement tended to see greater reductions in lead post-replacement. For a given profile liter, a ratio of less than 1 signifies a reduction in lead over pre-replacement levels, and a ratio greater than 1 signifies an increase.

At sites that underwent full LSLR, public and private LSL sections were not often replaced on the same day, and a delay of several months was not uncommon. In many cases, the pre-replacement profile represented the partial LSL configuration. In order to properly evaluate the effect of full LSLR, lead levels after full replacement were compared, without pairing, against 45 sites with full LSLs; 41 of these 45 underwent partial LSLR only. Unpaired comparisons have the advantage of larger sample sizes, but they are expected to be less accurate when other sources of lead are present (e.g., in L1 and L2). A key benefit of pairwise comparisons is that they tend to account for variation due to uncontrolled factors (e.g., other sources of lead, variations in flow rate).[47]

Reductions in lead following full LSLR were immediate · lead levels in L3−L5 were less than half that of their pre-replacement counterparts at the 3-day follow-up (Table 2). One month after full replacement, lead release from premises plumbing (L1, L2) had dropped significantly as well. Distribution quantiles, representing before and after full LSLR comparisons at the 1-month follow-up, are provided in Figure 3. Quantiles adhered well to the ratio estimates listed in Table 2, except in the upper extremes, where outliers occasionally deviated. One month post-replacement, 90th percentile lead levels ranged from 2 to 12 µg L^{-1} (L5 and L1, respectively), while pre-replacement 90th percentiles ranged from 10 to 44 µg L^{-1} (L5 and L4, respectively). Reductions in lead release from premises plumbing (L1, L2) may be attributed to gradual flushing of lead that had accumulated pre-replacement, as previous work has suggested.[3] Since premises plumbing upgrades were not performed in conjunction with LSLR, leaded solder and brass were not expected to have contributed to changes in lead release post-replacement. Accumulation of lead in premises plumbing may have been driven in part by adsorption to iron deposits or surfaces; as described elsewhere, colloidal lead (<0.45 µm) was strongly associated with colloidal iron at residential sites within

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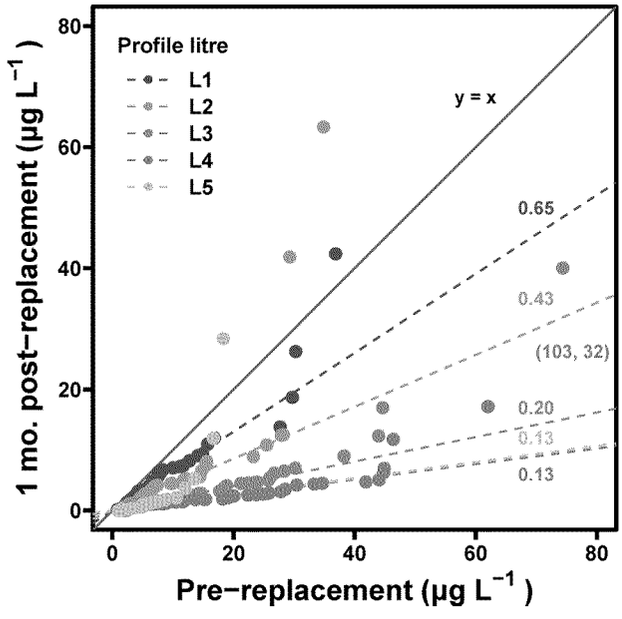

Figure 3. Lead levels (as distribution quantiles) before (45 sites) and 1 month after full LSLR (56 sites). Dashed lines (representing y = cx) are labeled by the corresponding c, where c is the estimated fraction of pre-replacement lead levels remaining at the 1-month follow-up. Points beyond the plot limits are represented as (x,y) coordinates.

the same water system.[36] Previous work has accounted for a correlation between iron and lead at the point of use with reference to the strong tendency for lead to adsorb to iron oxide deposits or galvanized iron plumbing.[42,48,49] Manganese deposits in premises plumbing have also been implicated as a sink for • and subsequent source of • lead in drinking water.[50]

Effect of Partial LSL Replacement. Partial LSLR more than doubled premises plumbing (L1, L2) lead levels at the 3-day follow-up (Table 2). One month post-replacement, L1 was still elevated by more than 60%, while subsequent standing sample lead levels (L2−L4) had not changed significantly relative to their pre-replacement counterparts. Even 6 months after partial LSLR, no significant reductions in L1 or L2 lead levels were observed. Reductions in L3 and L4 lead release at the 3- and 6-month follow-up rounds were expected to have been enhanced by • and could have been entirely due to • decreasing water temperature. (Increases, relative to pre-replacement, in LSL lead release at 3 and 6 months were sometimes observed despite temperature differences; see Figure S2, SI.) Applying a 1.1 µg L^{-1} °C^{-1} correction (described in the Supporting Information), based on expected water temperature differences, eliminated statistically significant reductions in L3 and L4 lead release that would otherwise have been attributed to partial LSLR.

In contrast to standing samples, L5 lead levels were not significantly different from their pre-replacement counterparts at the 3-day follow-up (90th percentile of 14 µg L^{-1}) and were significantly lower at the 1-month follow-up (90th percentile of 6 µg L^{-1}). However, data from this study do not support 5 min of flushing as a strategy for protecting against the short-term effects of partial LSLR: 9% of L5 samples were greater than 15 µg L^{-1} at the 3-day follow-up compared with just 4% pre-replacement. Moreover, an L5 sample collected 3 days post-replacement measured 230 µg L^{-1}.

Changes in lead release due to partial LSLR are illustrated in Figure 4 (bottom), which displays the distribution of pairwise

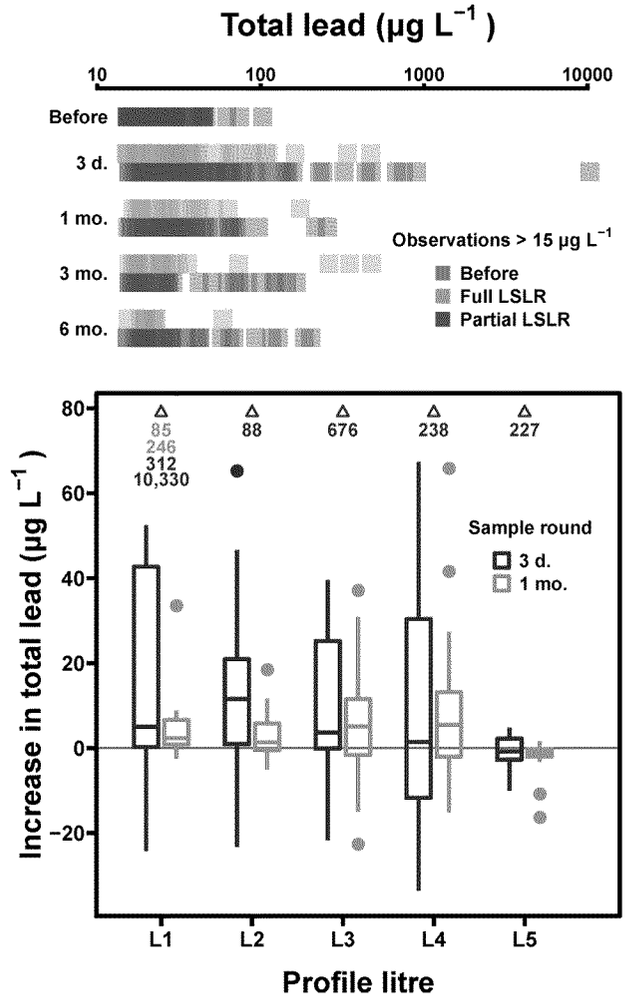

Figure 4. (Top) Lead levels greater than 15 µg L^{-1} over the 5 × 1-L sample profile, pre-LSLR, and at four follow-up rounds after full and partial LSLR. (Bottom) Box-and-whisker plots of the increase in total lead (pairwise differences) at the 3-day and 1-month follow-up rounds after partial LSLR, relative to pre-replacement. Boxes enclose the interquartile range (IQR), medians divide the boxes, and whiskers extend from the upper and lower quartile to the most extreme value within 1.5 times the IQR. Increases in lead beyond the plot limits are annotated.

differences in lead release (after − before), grouped by profile liter for the first two follow-up rounds (3 days and 1 month). Positive differences correspond to an increase in lead release post-replacement, and negative differences correspond to a decrease (sample sizes are provided in Table 2). Increased lead release following partial LSLR is evident, especially at the 3-day follow-up; at this interval, more than a quarter of sites saw increases of 20 µg L^{-1} in at least one standing sample (L1−L4). At the 1-month interval, L3 and/or L4 lead increased by 10 µg L^{-1} at more than a quarter of sites.

Increased lead release to L1 and L2 can likely be attributed to accumulation of particulate lead in premises plumbing following replacement-induced destabilization of LSL corrosion scale.[3] Galvanic corrosion at the lead−copper junction has been linked with elevated particulate lead release as well.[18−21] Occurrence of particulate lead (>0.45 µm) was more frequent following partial (relative to full) LSLR (Figure 5). At the 3-day follow-up, 11 and 26% of samples collected following full and partial LSLR, respectively, had more than 10 µg L^{-1} of

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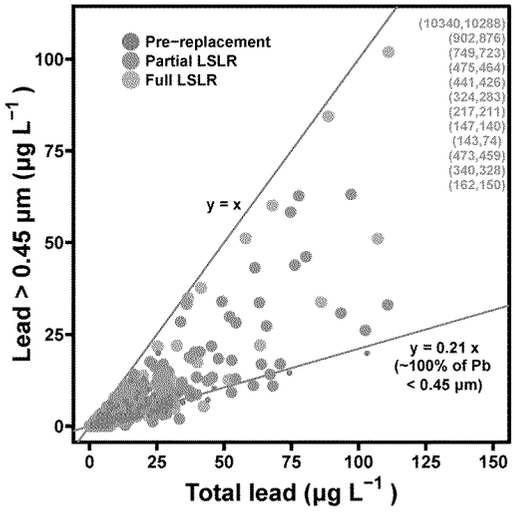

Figure 5. Particulate (>0.45 µm) lead release as a function of total lead, pre-replacement and at the first two follow-up rounds (72 h and 1 mo post-replacement). Points beyond the plot limits are represented as (x,y) coordinates and the line y = 0.21x represents the estimated loss due to 0.45 µm filtration (~100% of lead <0.45 µm).

particulate lead (compared to 3% pre-replacement). Elevated lead release in general was dominated by particles, and at higher total lead levels, the particulate fraction was larger, approaching unity at lead levels higher than approximately 100 µg L^{-1} (Figure 5). Available data suggest that lead in 0.45 µm filtrate was dominated by colloidal particles (0.05−0.45 µm).[36]

Post-Replacement Lead Exposure. Serious spikes in lead sometimes followed LSLR and were much more frequent following partial LSLR. Elevated lead levels after partial replacement have been reported in previous work as well.[3,24,25,51] Trends in post-replacement lead release diverged immediately according to replacement type. At sites with full LSLs (pre-replacement), 29% of standing sample (L1−L4) lead levels were greater than 15 µg L^{-1} (45 sites). Three days after partial LSLR, 45% of L1−L4 lead levels were greater than 15 µg L^{-1} (34 sites), while 3 days after full LSLR, just 14% were greater (48 sites). Lead levels exceeding 15 µg L^{-1}, by follow-up round and LSL configuration, are displayed in Figure 4 (top). This threshold represents a concentration above which the Centers for Disease Control and Prevention considers drinking water unsuitable for consumption by children and pregnant women.[52] At the 3-day follow-up, three observations also exceeded the U.S. Consumer Product Safety Commission's acute exposure level for children (700 µg L^{-1}, based on a 250 mL intake)[53] · all at sites with partial LSLs. These extreme lead levels, including a sample with 10 340 µg L^{-1}, were associated with premises plumbing (L1 or L2). High-velocity, multiple outlet flushing post-replacement is a possible strategy for protecting against these short-term spikes in lead.[51]

Unusually high lead levels were also observed at several sites following full LSLR (Figure 4, top). The highest observations at 1 and 6 months were both first-draw samples, and the highest four observations at 3 months represent standing samples (L1−L4) collected at a single site. Lead in these four samples was more than 90% particulate (>0.45 µm). These samples were also unusually rich in particulate iron, copper, and aluminum, suggesting that the source was corrosion scale within premises plumbing that had accumulated multiple contaminants over time. In the case of full LSLR, public and private LSL replacements were not often performed simultaneously, and disturbances associated with two replacements · as well as possible galvanic corrosion in the interim · may have contributed to elevated lead release and accumulation of particulate lead within premises plumbing. While full LSLR was associated with substantial reductions in lead levels, staggered replacements may have caused the true benefit of full replacement to be underestimated.

In the long term, elevated lead was observed more often at sites with partial LSLs than at sites with either full LSLs or with full copper service lines (post-LSLR). At the 6-month follow-up after partial LSLR, 22% of premises plumbing (L1, L2) lead levels · and 30% of service line lead levels (L3, L4) · were greater than 15 µg L^{-1} (30 sites). At sites with copper service lines, 7% of L1 and L2 samples · and none of the L3 or L4 samples · were greater than 15 µg L^{-1} (45 sites). The frequency of high (>15 µg L^{-1}) L1, L2 lead levels was substantially greater · even 6 months post-replacement · at sites with partial LSLs relative to sites with full LSLs (22 vs 16% respectively). Moreover, the fraction of first-draw samples greater than 15 µg L^{-1} at 6 months was double the pre-replacement fraction (27 vs 13%). Despite the possibility that the sample profile did not reach the lead−copper junction · and that the flow regime did not represent a worst-case scenario · these data captured the greater tendency, identified in previous work,[19,20] for elevated lead at sites with partial LSLs compared to sites with full LSLs.

Implications for Controlling Drinking Water Lead. This study used a standardized profile sampling protocol to assess the effect of full and partial LSLR on lead release to drinking water. The strength of this work was the comparatively large volume of data collected pre- and post-replacement, and the principal limitations were the inevitable uncertainties associated with sample collection by residents and the lack of information on plumbing volumes and configurations that limits a mechanistic understanding of the results.

Full LSLR reduced service line (L3, L4) and 5 min flushed sample (L5) lead levels within 3 days. At 1 month, full replacement had caused lead level reductions in every liter of the sample profile. Partial LSLR, on the other hand, caused substantial short-term increases in premises plumbing (L1, L2) lead levels and did not significantly reduce L1 and L2 lead levels within 6 months. Furthermore, first-draw lead levels were greater than 15 µg L^{-1} at a considerably higher frequency than at sites with full LSLs, even 6 months post-replacement. This finding could have important implications in jurisdictions where drinking water lead is regulated based on the first-draw sample.

This study generated a considerable volume of data corroborating previous work that showed (1) full LSLR · in addition to removing the primary source of lead · is effective for reducing lead release from premises plumbing, (2) partial LSLR dramatically increases lead at the point of use in the short term, (3) partial LSLR may be worse than leaving the LSL intact due to the potential for elevated lead release in the long term, (4) in Pb(II)-dominated water systems, first-draw lead levels are likely to underestimate lead exposure at residences with LSLs, and (5) lead release from LSLs is sometimes strongly influenced by water temperature.

The short-term elevated lead levels that sometimes followed partial LSLR are a serious concern · some were high enough to pose acute health risks. High-velocity flushing of outlets,[51] use of pipe-cutting methods that minimize disturbances to LSLs,[3] and point-of-use lead removal[54] are potential strategies that

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could be implemented to reduce health risks associated with this mode of exposure. In assessing the effects of full and partial LSLR, the potential for dramatically elevated lead levels post-replacement is an important consideration. While the rapid reductions in lead that typically follow full LSLR outweigh the risk of short-term disturbance-induced spikes, the modest long-term benefits from partial LSLR described in some previous work[24,25] may be overshadowed by the greater risk of elevated lead in both the short- and long-term.

ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b01912.

Supplementary figures, a description of temperature correction methods, and a transcription of the sampling instructions given to residents (PDF)

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Notes

The authors declare no competing financial interest.

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