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Contribution of Galvanic Corrosion to Lead in Water After Partial Lead Service Line Replacements

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Subject Area: Infrastructure

Contribution of Galvanic Corrosion to Lead in Water After Partial Lead Service Line Replacements

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Contribution of Galvanic Corrosion to Lead in Water After Partial Lead Service Line Replacements

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FOREWORD

The Water Research Foundation (Foundation) is a nonprofit corporation that is dedicated to the implementation of a research effort to help utilities respond to regulatory requirements and traditional high-priority concerns of the industry. The research agenda is developed through a process of consultation with subscribers and drinking water professionals. Under the umbrella of a Strategic Research Plan, the Research Advisory Council prioritizes the suggested projects based upon current and future needs, applicability, and past work; the recommendations are forwarded to the Board of Trustees for final selection. The Foundation also sponsors research projects through the unsolicited proposal process; the Collaborative Research, Research Applications, and Tailored Collaboration programs; and various joint research efforts with organizations such as the U.S. Environmental Protection Agency, the U.S. Bureau of Reclamation, and the Association of California Water Agencies.

This publication is a result of one of these sponsored studies, and it is hoped that its findings will be applied in communities throughout the world. The following report serves not only as a means of communicating the results of the water industry's centralized research program but also as a tool to enlist the further support of the nonmember utilities and individuals.

Projects are managed closely from their inception to the final report by the Foundation's staff and large cadre of volunteers who willingly contribute their time and expertise. The Foundation serves a planning and management function and awards contracts to other institutions such as water utilities, universities, and engineering firms. The funding for this research effort comes primarily from the Subscription Program, through which water utilities subscribe to the research program and make an annual payment proportionate to the volume of water they deliver and consultants and manufacturers subscribe based on their annual billings. The program offers a cost-effective and fair method for funding research in the public interest.

A broad spectrum of water supply issues is addressed by the Foundation's research agenda: resources, treatment and operations, distribution and storage, water quality and analysis, toxicology, economics, and management. The ultimate purpose of the coordinated effort is to assist water suppliers to provide the highest possible quality of water economically and reliably. The true benefits are realized when the results are implemented at the utility level. The Foundation's trustees are pleased to offer this publication as a contribution toward that end.

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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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INTRODUCTION

Lead (Pb) is widely recognized as one of the most pervasive environmental health threats in the United States (US). Water consumption contributes to an estimated 10-20% of the general population's total lead exposure (Levin et al. 2008), but can occasionally be the dominant source of exposure (Edwards et al. 2009, Levin et al. 2008). The harmful health effects from lead exposure through drinking water have been historically recognized since the 1850s. In that era drinking water contamination by lead pipes was thought to be the main source of human-ingested lead, causing infant mortality, neurological effects, and digestion problems (Troesken 2006).

PARTIAL LEAD SERVICE LINE REPLACEMENTS

Lead service lines were the standard in many US cities through the 1950s, and were even occasionally installed up to the ban of lead pipe in 1986 (Renner 2010). As of 1990, 3.3 million lead service lines were estimated to be present across the country, while 6.4 million lead connections (e.g., goosenecks) were also acknowledged (Weston and EES 1990). The actual number of old lead service lines in the US today is unknown. Old lead service lines can still be significant contributors to lead-in-water hazards. Depending on their length and diameter, the water corrosivity, water use patterns as well as hydraulic patterns, these old lead service lines can account for 50-75% of lead contamination observed at the tap, when they are present in front of US homes (Sandvig et al. 2009). Lead in US drinking water is regulated under the Lead and Copper Rule (LCR), which may require replacement of utility-owned lead service lines, if the LCR lead action level of 15 parts per billion (ppb) is exceeded (US EPA 1991).

If the lead service line extends onto the homeowner's property, the utility is only required to replace the portion of pipe that it owns, leaving behind the customer-owned portion of lead pipe (Figure 1, left). Although numbers vary dramatically from city to city or even from home to home, a national survey (Sandvig et al. 2009) indicated that the typical service line length in the US averages 55-68 feet, with 25-27 feet (i.e., 40-45%) being under the utility's jurisdiction. Partially replacing a single lead service line can cost from $1,000 to more than $3,000 (AWWA 2005a), and thus constitute considerable financial burden for drinking water utilities. The cost of replacing the customer portion can add up to several thousand dollars, and few customers voluntarily replace their portion of the lead service line (Swertfeger et al. 2006). Different methods may be used in the field, in order to connect the remaining lead pipe to new copper pipe. For example, utilities may use a brass corporation valve, or a brass compression fitting between the lead and the copper.

The practice of only replacing the utility portion of lead pipe, referred to as a "partial lead service line replacement," is fraught with controversy because it has been known to increase the concentration of lead in drinking water. The increased lead can arise from a variety of mechanisms and can possibly be short-term (days to weeks) or even long-term (months to years) in duration. Short-term problems occur from disturbing the lead rust (i.e., scale) that has accumulated on the pipe over decades of use, and/or from creating metallic lead particles when the pipe is cut (Schock et al. 1996). In the US these short-term mechanisms from cutting and scale disturbance were documented in laboratory experiments (Boyd et al. 2004) and in field studies undertaken by several utilities (Sandvig et al. 2009).

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Longer-term problems might arise from creation of a new electrochemical or galvanic cell, between the old lead pipe and the newly installed copper pipe (Figure 1, right). Britton and Richards (1981) documented a case in Glasgow where more than four months were required for lead levels to drop. In the US Swertfeger et al. (2006) measured lead levels in water of homes after partial lead service line replacements, and reported that even after one year of sampling, replacement did not show an improvement over keeping a complete line in place. Practical experiences in the UK showed an unexpected rise in measured lead levels, four months to eighteen months after partial replacements (Chambers and Hitchmough 1992). The UK report partly attributed this problem to galvanic corrosion, which "can be persistent and may well annul any beneficial effects of reducing the length of lead pipe in the system" (Chambers and Hitchmough 1992). In the US, a project conducted for the Water Research Foundation (Sandvig et al. 2009) evaluated, among other issues, the effectiveness of full versus partial lead service line replacements, with the collaboration of several water utilities. The report provided tremendous information regarding those practices, but it did not address the issue of galvanic corrosion as a potential contributor to lead in water after partial lead service line replacements.

In addition, some research work on galvanic corrosion did not confirm adverse consequences. Reiber and Dufresne (2006) concluded that galvanic impacts were short-lived on aged/passivated lead service lines, based on surface potential measurements between lead and copper under continuous flow. Lead in water was not measured in that work. Another experimental study, which also employed continuous flow, also concluded that galvanic coupling had little relevance to accelerating metal release on the lead service line (HDR Engineering 2010). In both studies the water never stagnated inside the pipes, a situation which is known to occur regularly in homes and to promote galvanic corrosion problems in practice (Dudi 2004). Based on data collected under continuous flow experimental conditions, potential long-term problems were described as likely inconsequential, and were believed to provide a strong basis for discounting claims and concerns relative to accelerated metal release after partial replacements (Reiber and Dufresne 2006).

Unfortunately, partial lead service line replacement was recently linked to adverse health effects in Washington, D.C. A 2010 announcement by the Centers of Disease Control and Prevention (CDC) warned that partial replacement of lead pipes was linked to increased incidence of high blood lead levels in children. The epidemiological study found that "when lead service lines are partially replaced, children are more likely to have blood lead levels greater than or equal to 10 μg/dL, compared to children living in housing with either undisturbed lead service lines or service lines that are not made of lead" (Frumkin 2010). The CDC study is the first to link partial replacement to adverse health effects, although utilities have long warned of increased lead-in-water levels for an indeterminate length of time when partial replacements are conducted (AWWA 1996, AWWA and AMWA 2008).

The US Environmental Protection Agency (EPA) has indicated a willingness to re-evaluate its regulations that cover partial lead service line replacement, as part of the 2012 long-term revisions to the LCR (Renner 2010). Considering the financial burden on utilities to conduct partial lead service line replacements, the new concerns about adverse health effects associated with the practice, and the opportunity to generate data and insights to inform revision of the LCR, additional research on this topic is very timely.

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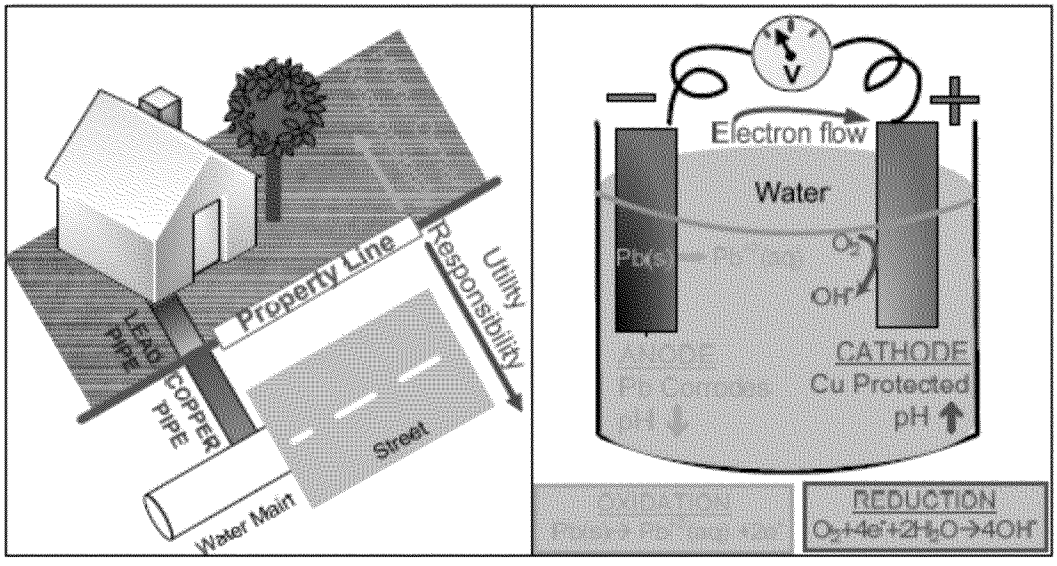

Figure 1: Typical plumbing configuration after partial lead service line replacement, where new copper pipe is directly connected to old lead pipe (left). Conceptualization of galvanic corrosion due to direct electrical connection of copper pipe to lead pipe (right).

GALVANIC CORROSION OF LEAD FROM LEAD/COPPER CONNECTIONS

When copper is electrically connected to lead pipe, it can accelerate corrosion of the lead pipe by galvanic action, above and beyond corrosion that would normally occur for lead pipe alone (Figure 1, right). The drinking water in contact with these dissimilar metals serves as the electrolyte. Based on the galvanic series, metallic lead typically serves as the anode of this galvanic cell and is therefore oxidized (i.e., corroded) to form Pb^{+2}. The production of Pb^{+2}, which is a Lewis acid, causes a local pH drop at the anode (i.e., the lead pipe surface) (Figure 1, right). The copper pipe serves as the cathode, where the cathodic reaction (such as oxygen reduction) occurs over its surface (Figure 1, right). In this situation, lead leaching to water could increase due to higher corrosion rate and/or a lower pH at the surface of the lead material. Since lower pH tends to prevent the formation of a passivating film on lead surfaces, high galvanic currents could tend to be self-perpetuating (Dudi 2004). After a partial lead service line replacement is conducted, it is the connection of lead pipe to a dissimilar metal, as is copper, which creates a galvanic cell. If part of the lead service line was replaced with a non-metallic (and thus non-conductive) pipe material, such as plastic, galvanic corrosion of lead would not be possible.

The contribution of galvanic corrosion to elevated lead in water was first recognized in England. As early as 1859, it was noted that galvanic corrosion is a very powerful agent in promoting lead release, dependent on the corrosive action of certain waters upon lead (Ingleson 1934). Recently, DeSantis et al. (2009) found mineralogical evidence of galvanic corrosion in domestic lead pipes in the US, which were excavated after 70-114 years in service. Their work provided clear visual and mineralogical documentation of galvanic corrosion in harvested lead-copper junctions. Consistent with galvanic theory, the study found that lead pipe sometimes behaved as the anode and was corroded. Interestingly, in some of the junctions, the lead pipe behaved as the cathode and was protected from corrosion, even though the mechanism for such a shift was unclear (DeSantis et al. 2009). The mineralogical observations of DeSantis et al. (2009)

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also verified areas of radically altered water chemistry near the galvanic joints, consistent with prior hypothesis forwarded by Edwards and Triantafyllidou (2007) and recently confirmed by Nguyen et al. (2010).

Other work in the United Kingdom (Britton and Richards 1981) and in the US (Nguyen et al. 2009, Nguyen et al. 2010) suggests that sometimes serious long-term problems can occur from this mechanism. The UK study described an instance in Glasgow, where water samples from homes with Pb:Cu pipe connections had abnormally erratic or much higher than normal lead concentrations. These authors went further to state that "occasionally the insertion of copper pipe can produce particularly bad results and despite satisfactory pH control it may be impossible to obtain satisfactory samples." Laboratory experiments at Virginia tech (Nguyen et al. 2009) illustrated that the water chemistry controls the magnitude of galvanic current between lead and copper. One critical water quality factor that was found by researchers at Virginia Tech to influence the magnitude of galvanic corrosion is the chloride-to-sulfate mass ratio (CSMR). Specifically, waters with high CSMR resulted in strong galvanic currents when lead pipe (both new and passivated) was connected to copper pipe, whereas waters with low CSMR resulted in lower galvanic currents. Consistent with the higher currents, exposure to high CSMR water released 2 to 5 times more lead than did exposure to low CSMR water, under the stagnant water conditions of the Virginia Tech study.

THE EFFECT OF CHLORIDE-TO-SULFATE MASS RATIO (CSMR)

The influence of chloride and sulfate levels on the corrosivity of water relative to lead release is not a new concept. In an English study where various synthetic waters were examined, Oliphant (1983) noted that chloride stimulated attack on lead when it was galvanically connected to copper. Sulfate inhibited corrosion of lead-bearing materials both in isolation and in galvanic connections to copper. Lead release was never measured directly in the Oliphant study, and conclusions were based on galvanic current measurements under continuous flow conditions.

Follow-up studies of Gregory (1985), who compared thirty eight real waters in England, reinforced the importance of the relative concentration of chloride to sulfate in exacerbating galvanic corrosion of lead. Gregory developed the concept of chloride-to-sulfate mass ratio to explain this tendency. To illustrate, for water containing 15 mg/L Cl^- and 30 mg/L SO_4^{-2}, the resulting CSMR is 0.5 (Equation 1). Gregory (1985) determined that CSMRs above 0.5 increased galvanic corrosion of lead solder connected to copper pipe, as evidenced by increased galvanic voltage measurements.

$$\text{Chloride to Sulfate Mass Ratio (CSMR)} = \frac{[Cl^-]}{[SO_4^{-2}]} = \frac{15\,\text{mg/L}\,Cl^-}{30\,\text{mg/L}\,SO_4^{-2}} = 0.5 \quad \text{Equation 1}$$

As part of a US utilities' survey, Edwards et al. (1999) later determined that as the relative concentration of chloride to sulfate increased in a water supply, a utility was more likely to exceed the LCR action level for lead. Specifically, for a sub-set of utilities studied in-depth, 100 percent of utilities with CSMR below 0.58 met the LCR lead action level of 15 ppb, whereas only 36 percent of utilities with CSMR above 0.58 were in compliance. The identified critical CSMR of 0.58 cited for causing lead compliance problems was remarkably similar to the 0.5 threshold identified as causing galvanic corrosion of lead in the preceding English studies. Other laboratory experiments (Nguyen et al. 2009, Edwards and Triantafyllidou 2007, Dudi 2004) as well as anecdotal evidence from specific US utilities (Edwards and Triantafyllidou 2007,

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AWWA 2005b, Kelkar et al., 1998) supported the notion that lead release was impacted by higher CSMR (ranging from 1.0 to as high as 19.8 depending on the study).

Clark and Edwards (2007) provided a mechanistic explanation for the success of the empirical CSMR in explaining lead contamination of potable water. They examined the solution chemistry of Pb^{+2} in the presence of chloride and sulfate. They conducted experiments at relatively low pH (≈ 3.0-5.0), since previous microelectrode measurements had shown that local pH at the lead surface of galvanic connections to copper can drop substantially (e.g. Edwards and Triantafyllidou 2007). Pb^{+2} formed sulfate solids which were relatively insoluble even at pH of 3.0. On the contrary, Pb^{+2} formed soluble complexes with chloride, which could significantly increase the solubility of lead. Since increased lead solubility can translate to lead contamination of drinking water, these data illustrated how a high CSMR can indeed worsen lead problems at low pH.

RESEARCH OBJECTIVES

Previous research and observations highlight the potential impacts of galvanic corrosion on lead release after partial lead service line replacements with copper, but experimental studies documenting these effects are limited. In order to provide more insights, long-term experiments of simulated partial lead service line replacements were conducted, with emphasis on:

a) Rigorously testing the longevity of galvanic corrosion with respect to lead contamination of drinking water,

b) Assessing the relative effects of chloride and sulfate by measuring lead release into drinking water at two different CSMRs, and

c) Exploring mechanisms by which lead leaching is increased.

MATERIALS AND METHODS

EXPERIMENTAL APPARATUS

The experimental apparatus was constructed to track lead leaching from simulated small-scale partial lead service line replacement with copper. The test rigs consisted of a copper pipe section (type M, 3/4 inch ID, 7/8 inch OD) that was electrically connected to lead pipe (3/4 inch ID, 1 inch OD), with a total rig length of three feet (Figure 2). The lead portion and the copper portion of each rig were separated by an insulating spacer, and could be externally connected via grounding strap wires (Figure 2). If the wires were disconnected, direct galvanic corrosion between lead and copper was not possible due to their spatial separation by the insulating spacer, and if the wires were connected the galvanic current flowed as usual (Figure 2).

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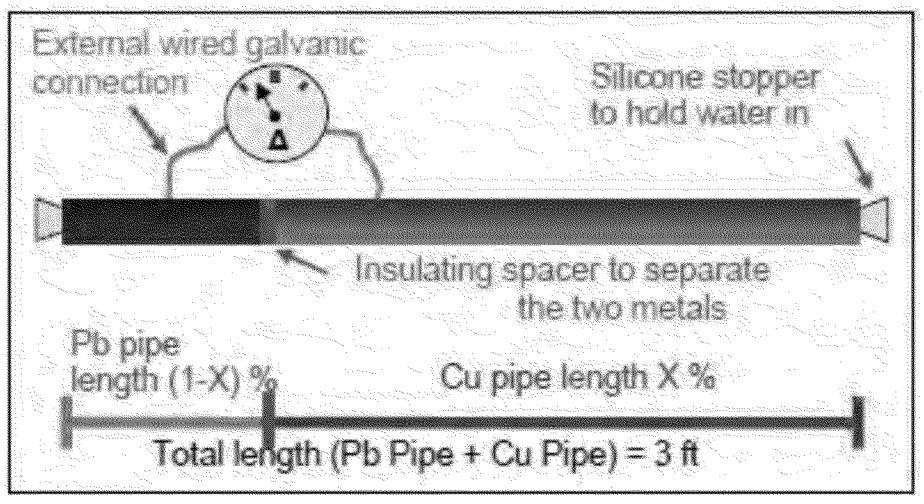

Figure 2: Generalized schematic of experimental setup. This design assessed the contribution of galvanic corrosion to lead in water, as it would occur after partial lead service line replacement.

The portion of the pipe that is lead and the portion that is copper were systematically varied - as would occur in partial replacements with different percentage of consumer ownership of the service line - to examine the effect of anode (lead) to cathode (copper) surface area. Specifically, 100% lead pipe (simulating a lead service line before replacement), 100% copper pipe (simulating full replacement), and four increments in between (17% copper pipe, 50% copper pipe, 67% copper pipe, and 83% copper pipe to simulate partial replacements) were tested (Figure 3).

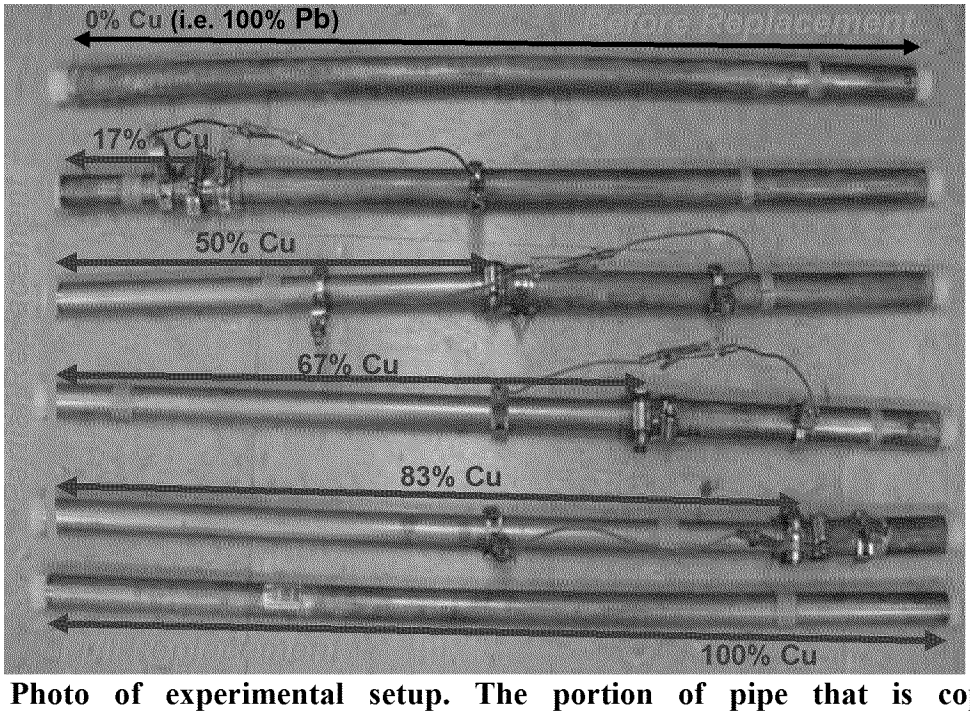

Figure 3: Photo of experimental setup. The portion of pipe that is copper was systematically varied, in order to examine the effect of anode (lead) to cathode (copper) surface area.

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Three sets of rigs were constructed, utilizing three different types of lead pipe:

- **New Pb pipe**, i.e., lead pipe which had not yet been used in experiments
- **Old Pb pipe A**, i.e., lead pipe which was initially new but had been previously used in other short-term experiments of four months,
- **Old Pb pipe B,** i.e., older Pb pipe which was initially new but had been previously used in other longer-term experiments of one year. Each of these older lead pipes was 0.5 feet long. For the needs of this experiment, the third set of rigs was constructed by connecting in series as many short Pb pipes together as needed, in order to add up to the desired Pb length.

The three sets of Pb pipes will be referred to as "new Pb pipe," "old Pb pipe A" and "old Pb Pipe B" respectively, in subsequent discussion. All three sets of lead pipe were pre-treated in the same way prior to construction of the rigs: They were exposed to re-circulating pH 2.0 water (de-ionized water with the addition of H_2SO_4) for three hours, in order to remove surface rust and deposits. They were subsequently rinsed with de-ionized water for fifteen minutes, exposed to pH 10.0 water (de-ionized water with the addition of NaOH) for another three hours, and finally rinsed with de-ionized water for another fifteen minutes.

At the beginning of the experiment (i.e., after pre-treatment), visual differences were apparent in the internal scale of the three sets of Pb pipes. Specifically, the older lead pipes had developed a uniform internal scale that was more white in color, compared to the uniform scale of the new lead pipe (Figure 4). This color was visual confirmation of corrosion scale deposits, and suggested that the older pipes had a somewhat more passivated internal surface. In practice, during partial lead service line replacements, new copper pipe is electrically connected to old lead pipe. Therefore, examination of more passivated lead pipe (old Pb Pipe A, old Pb pipe B), in addition to the new Pb pipe, gives further insights on the practical implications following partial lead service line replacements.

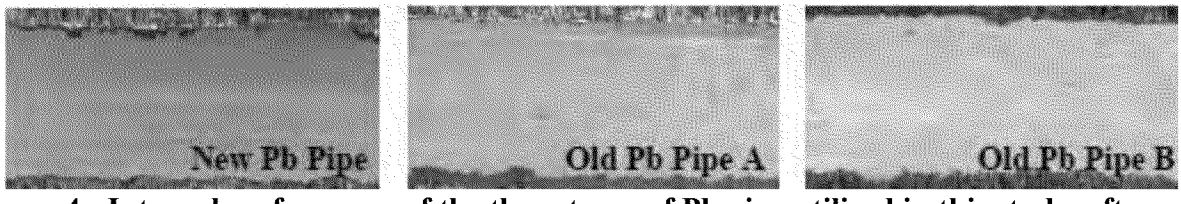

Figure 4: Internal surface area of the three types of Pb pipe utilized in this study, after pre-treatment.

EXPERIMENTAL PROTOCOL AND WATER CHEMISTRY

Three distinct phases of experimentation were undertaken for all three sets of rigs (new Pb pipe, old Pb pipe A, old Pb pipe B):

- **Phase 1.** During weeks 1-11, all rigs were exposed to synthetic tap water with a low chloride-to-sulfate mass ratio (CSMR) of 0.2 ("low CSMR water"). This water also had an alkalinity of 15 mg/L as $CaCO_3$, monochloramine disinfectant dosed at 4.0 mg/L as Cl_2, ionic strength of 4.6 mmol/L (by addition of salts to mimic other tap water constituents) and pH of 8.0 (Table 1).

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- **Phase 2.** After baseline results were established in the non-aggressive water, the test water was switched to an aggressive synthetic tap water with a high CSMR of 16 during weeks 12-25 ("high CSMR water"). All other water parameters such as alkalinity, monochloramine, ionic strength and pH were kept the same as in the "low CSMR water" of Phase 1 (Table 1).
- **Phase 3.** For weeks 26-31 the rigs continued to be exposed to the "high CSMR water" as in Phase 2, but without direct galvanic corrosion between the lead and copper pipe due to removal of the connecting strap wires.

Table 1: Key characteristics of the two synthetic waters utilized in the experiment.

	Cl^- (mg/L)	SO_4^{-2} mg/L)	CSMR (-)	Alkalinity (mg/L as $CaCO_3$)	NH_2Cl (mg/L as Cl_2)	Ionic Strength (mmol/L)	pH
Low CSMR Water	22	112	0.2	15	4.0	4.6	8.0
High CSMR Water	129	8.0	16	15	4.0	4.4	8.0

Throughout the experiment (i.e., in all three phases), water was completely changed inside the pipes three times per week, using a "dump and fill" protocol. Lead release from the rigs was therefore evaluated under "worst case" stagnation conditions, which are known to promote galvanic corrosion problems in practice (e.g., Dudi 2004). These worst-case stagnation conditions represent extremes in water usage that may be encountered at schools/ municipal-type buildings during weekends/breaks and at homes during prolonged absences. In such situations, tap water is not being consumed but rather sits stagnant inside the pipes for extended periods of time.

The contribution of galvanic connection (or lack thereof) to lead release was assessed by measuring total lead concentration in water and galvanic current magnitude:

- One composite water sample was collected at the end of each week from each rig, by pouring the three water samples of that week into the same container. Total lead was quantified in these unfiltered composite water samples using a Thermo Electron X-Series inductively coupled plasma mass spectrometer (ICP-MS) per Standard Method 3125-B (APHA, AWWA, and WEF, 1998). Water samples and instrument calibration standards were prepared in a matrix of 2% nitric acid by volume.
- Galvanic current between the dissimilar metals (for Phases 1 and 2 when the external wires were connected) was measured with a digital multi-meter purchased at a retail electronics store (RadioSack model 22-812, internal resistance determined at 100 Ω). The current flowing in each rig was measured by connecting the multi-meter in-line for 15 seconds after disconnecting the wire between the two metals.

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RESULTS AND DISCUSSION

TEMPORAL TRENDS ON LEAD RELEASE

New Pb pipe connections to Cu pipe

After an initial stabilization period of three weeks, lead release under exposure to low CSMR water (weeks 4-11) became relatively low for each rig (Figure 5). The rig with pure lead pipe (i.e. 0% Cu) released 120-320 ppb of lead during that time frame. As expected, the rig with no lead pipe (i.e. 100% Cu) released no lead, providing assurance that sample contamination with lead was not taking place. In rigs with copper and lead galvanic connections, lead leaching was always increased when compared to pure lead pipe, during exposure to low CSMR water (Figure 4).

When the water was switched to high CSMR in Phase 2 (weeks 12-25), lead release from the galvanic connections dramatically increased, and remained elevated throughout that time period (Figure 5). High CSMR water also increased lead release from pure lead pipe (i.e. 0% Cu pipe) (Figure 5). Taking into account that lead release is generally expected to decrease with time, since protective layers begin forming on the inside surface area of the lead pipe, this general trend of sustained elevated water lead levels further highlights the detrimental effect of high CSMR, consistent with prior published research.

During weeks 26-31 (Phase 3), when galvanic corrosion was inactivated by disconnecting the wires, lead levels dropped even though the aggressive high CSMR water was still fed to the rigs. This demonstrates the direct role of galvanic corrosion in sustaining high lead concentrations in water, when lead and copper pipe are electrically connected.

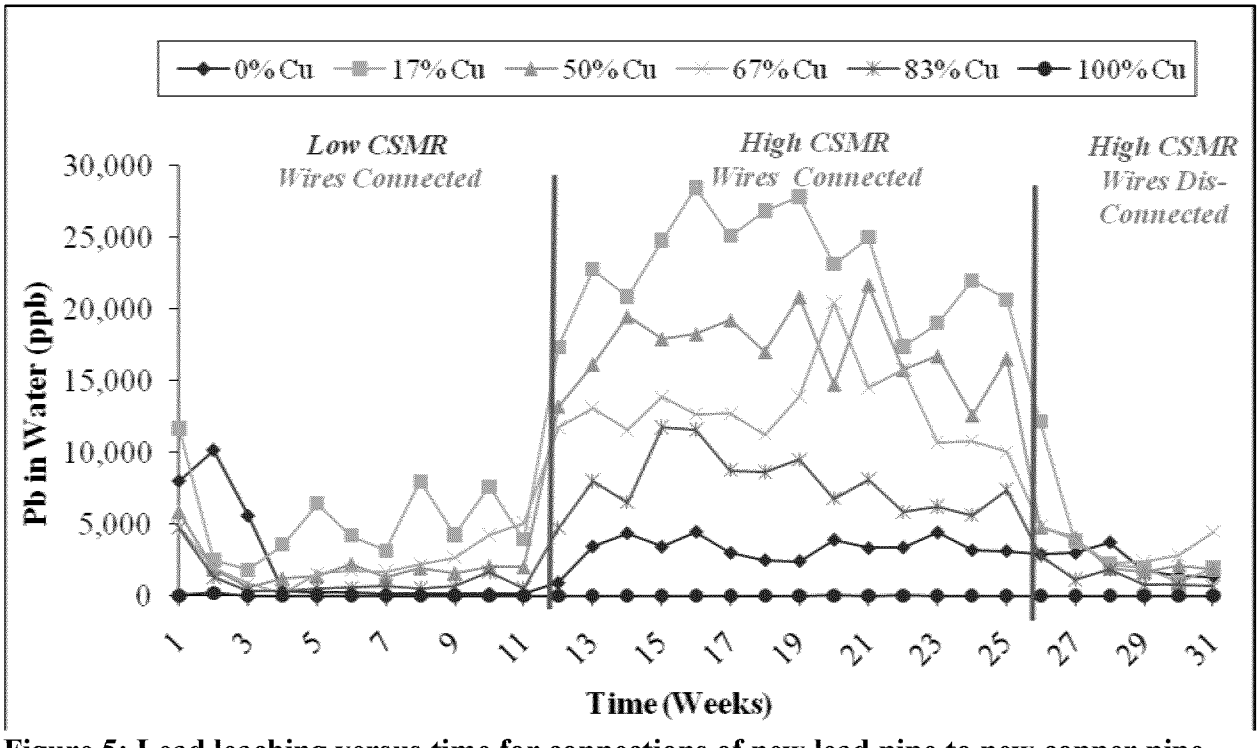

Figure 5: Lead leaching versus time for connections of new lead pipe to new copper pipe.

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Old Pb pipe connections to Cu pipe

Similar trends were observed for two types of old Pb pipe, as with the new Pb pipe. That is, an initial stabilization period of three weeks was required in order for lead release to become relatively stable (Figure 6, Figure 7). When the wires between lead and copper were connected (weeks 1-26), the galvanic rigs (i.e., 17% Cu, 50% Cu, 67% Cu and 83% Cu) typically released more lead to the water, compared to the pure lead pipe (i.e., 0% Cu).

Exposure to low CSMR water (Phase 1, weeks 1-11) released much lower lead to the water than did exposure to high CSMR (Phase 2, weeks 12-25) for all rigs. Exposure to high CSMR water for 14 weeks resulted in sustained grave lead contamination of the water, for the galvanic rigs. In other words, under the stagnant water conditions of this study, galvanic corrosion was sustained for a prolonged period of time, even in the old (and thus more passivated) Pb:Cu rigs. Disconnecting the wires (Phase 3, weeks 26-31) typically released much lower lead to the water, compared to the galvanic connections of Phase 2 (Figure 6, Figure 7). Throughout the duration of the study, the extent of lead release was higher for old Pb pipe B (Figure 7), compared to old Pb pipe A (Figure 6).

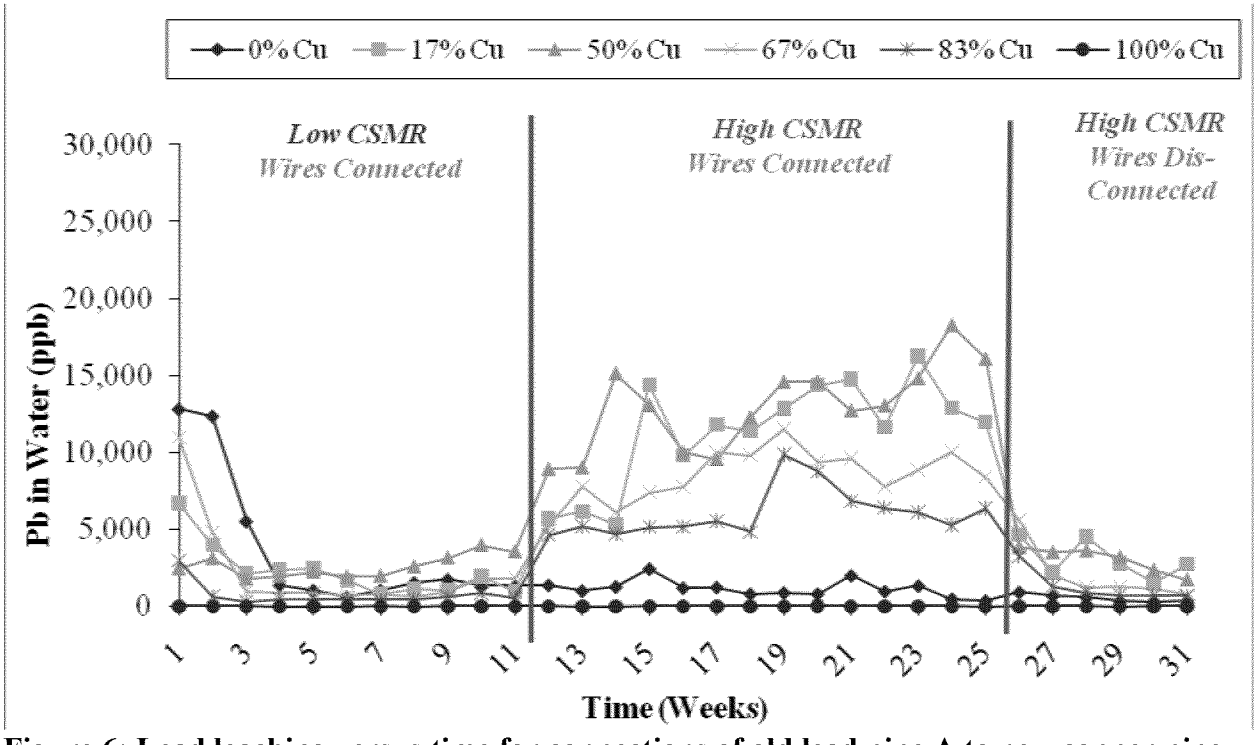

Figure 6: Lead leaching versus time for connections of old lead pipe A to new copper pipe.

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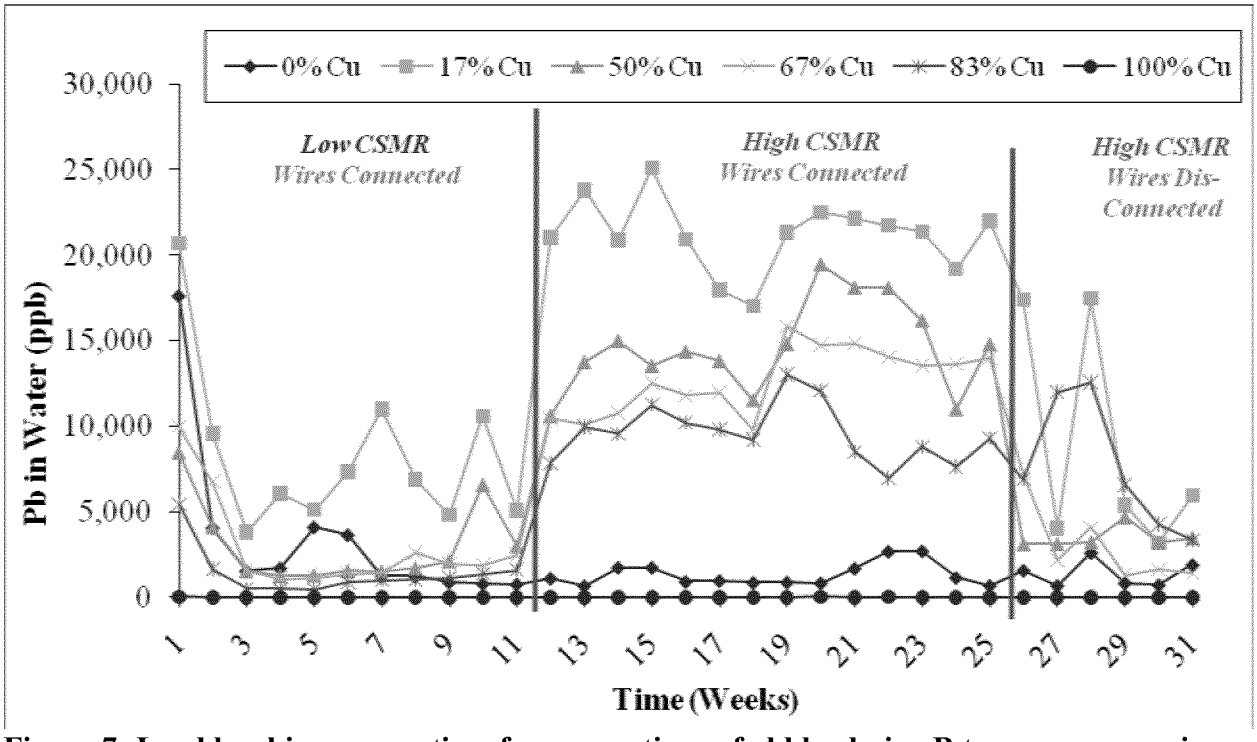

Figure 7: Lead leaching versus time for connections of old lead pipe B to new copper pipe.

SYNTHESIS: EFFECT OF CSMR ON LEAD RELEASE

With the exception of weeks 1-3, when lead release had not yet stabilized, results were synthesized by averaging the lead data for each experimental phase.

New Pb pipe connections to Cu pipe

All simulated partial replacements (17%, 50%, 67% and 83% of Pb replaced by Cu) released the same or more lead to the water than did the rig consisting of pure lead (i.e. 0% Cu) (Figure 8). This was true for all three experimental phases, and results were statistically significant at the 95% confidence level (error bars plotted, Figure 8). Even though these results appear to be counter-intuitive, they are not surprising considering early knowledge on galvanic corrosion. That is, the 0% Cu rig, consisting only of lead pipe, has a higher Pb surface area in contact with the water compared to all other conditions (see Figure 3). This condition would therefore be expected to release more lead to the water. However, when lead pipe is connected to copper pipe, the effect of galvanic corrosion in enhancing lead release can be so strong, that even a smaller lead surface area exposed to the water results in increased lead contamination of the water. This experimental observation is consistent with the notion of Chambers and Hitchmough (1992) that the galvanic effect "may well annul any beneficial effects of reducing the length of lead pipe in the system."

High CSMR released much more lead to the water compared to low CSMR, when the wires were connected. In fact, when comparing high CSMR water (Phase 2) to low CSMR water (Phase 1), lead release increased by 5 times (in the case of 17% Cu) to as much as 12 times (in the case of 83% Cu) (Figure 8). These differences were statistically significant at the 95%

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confidence level (Error bars plotted, Figure 8). Disconnecting the wires under high CSMR water (Phase 3) decreased lead release by 4-6 times in all Cu:Pb galvanic couples (Figure 8).

The extent of lead contamination was dependent on the extent of partial replacement, for the new Pb pipe. For instance, for the worst-case experimental condition of High CSMR and connected wires (Phase 2), the rig consisting of 17% Cu released the highest lead, followed by the 50% Cu, 67% Cu, and 83% Cu rigs (Figure 8). In other words, less copper relative to lead worsened lead corrosion, and these results were statistically significant at the 95% confidence level (Error bars plotted, Figure 8).

Similarly, when the rigs were exposed to low CSMR water with connected wires (Phase 1), the rig consisting of 17% Cu (with the remaining 83% being lead) released the highest lead. The 50% Cu and 67% Cu were not statistically different, and the 83% Cu rig released the lowest lead. On the contrary, when the rigs were exposed to High CSMR water with disconnected wires (Phase 3), there were not statistically significant differences in lead release between the 17%, 50%, 67%, and 83% Cu rigs (Error bars plotted, Figure 8). During Phase 3, lead release from all the de-activated galvanic rigs was not statistically higher, when compared to lead release from pure lead pipe. Lead release from those rigs was not lower either, as would be expected due to their smaller lead surface area, compared to the pure lead pipe. This could be because the rigs had not yet "recovered" from the previous Phase (Phase 2) of galvanic connection due to some form of memory effect. Alternatively, other additional corrosion mechanisms could be present in the Pb:Cu rigs during Phase 3 compared to the pure Pb pipe, even in the absence of galvanic connection to copper (e.g., copper deposition corrosion onto the lead pipe).

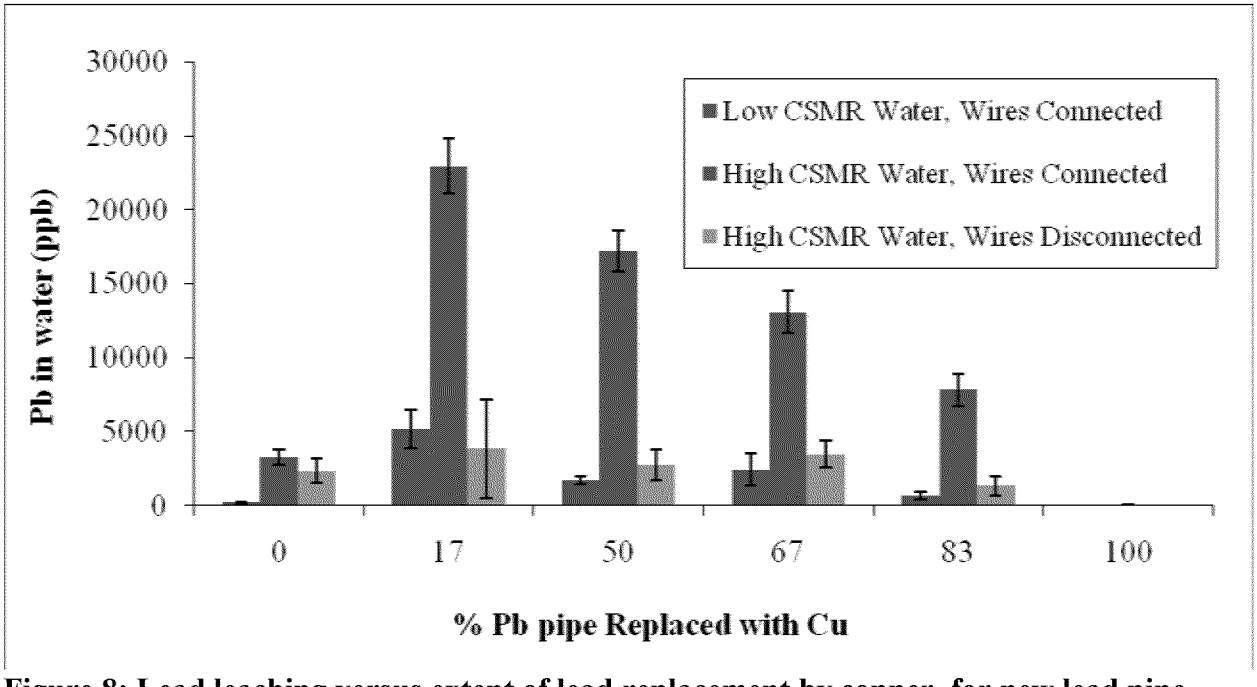

Figure 8: Lead leaching versus extent of lead replacement by copper, for new lead pipe. Data were averaged for each experimental phase. The error bars denote 95% confidence intervals.

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Old Pb pipe connections to Cu pipe

Overall, similar trends were observed for the two types of old Pb pipe as with the new lead pipe, with a few exceptions. Similarly to the new Pb pipe results, high CSMR water released much more lead to the water compared to low CSMR water, when the wires were connected and thus galvanic corrosion was activated. More specifically, when comparing high CSMR water (Phase 2) to low CSMR water (Phase 1), lead release increased by 5-11 times for old Pb pipe A (Figure 9). These differences were statistically significant at the 95% confidence level (Error bars plotted, Figure 9). For old Pb pipe B, lead release increased by 3-10 times, and these results were also statistically significant (error bars plotted, Figure 10).

For old Pb pipe A, disconnecting the wires under high CSMR water (Phase 3) decreased lead release by 3-4 times, when compared to the previous phase (Phase 2) of same water quality but active galvanic corrosion (Figure 9). For old Pb pipe B, disconnecting the wires in Phase 3 decreased lead release by 2-4 times (Figure 10). These reductions are smaller than those observed in the case of new Pb pipe (where the corresponding lead release decreased by 4-6 times). This is consistent with expectations, since old Pb pipe is more passivated, "weakening" the galvanic battery effect when connected to new copper pipe. However, the reductions in lead release for old lead pipe were still significant. That is, under an aggressive water condition of high CSMR, connections of old (and thus more passivated) lead pipe to copper still significantly contributed to galvanic corrosion of lead.

The extent of lead release from old Pb pipe was dependent on the extent of partial replacement, but these trends were not the same as with new Pb pipe. For old Pb pipe A, under the worst-case experimental condition of High CSMR and connected wires (Phase 2), the rig consisting of 50% Cu released the highest lead, followed by the 17% Cu, 67% Cu, and 83% Cu rigs (Figure 9). For old Pb pipe B, under Phase 2 conditions, the rig consisting of 17% released the highest lead, followed by the 83% Cu, 50% Cu, and 67% Cu rigs (Figure 10).

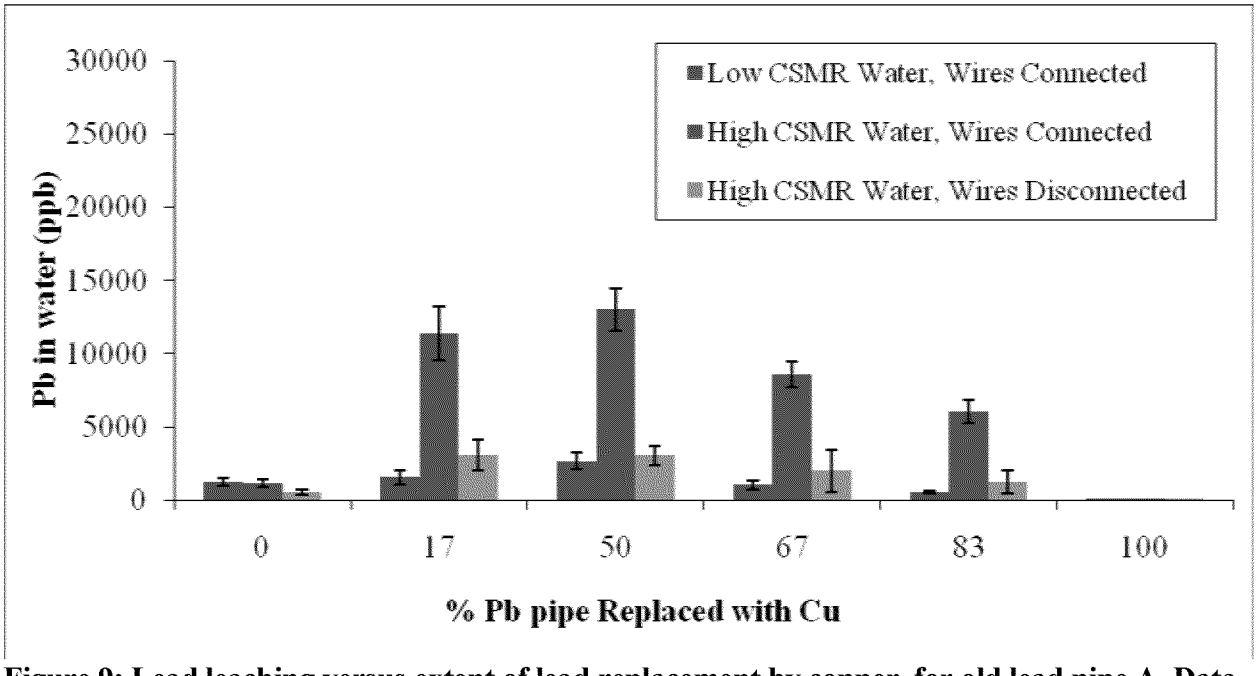

Figure 9: Lead leaching versus extent of lead replacement by copper, for old lead pipe A. Data were averaged for each experimental phase. The error bars denote 95% confidence intervals.

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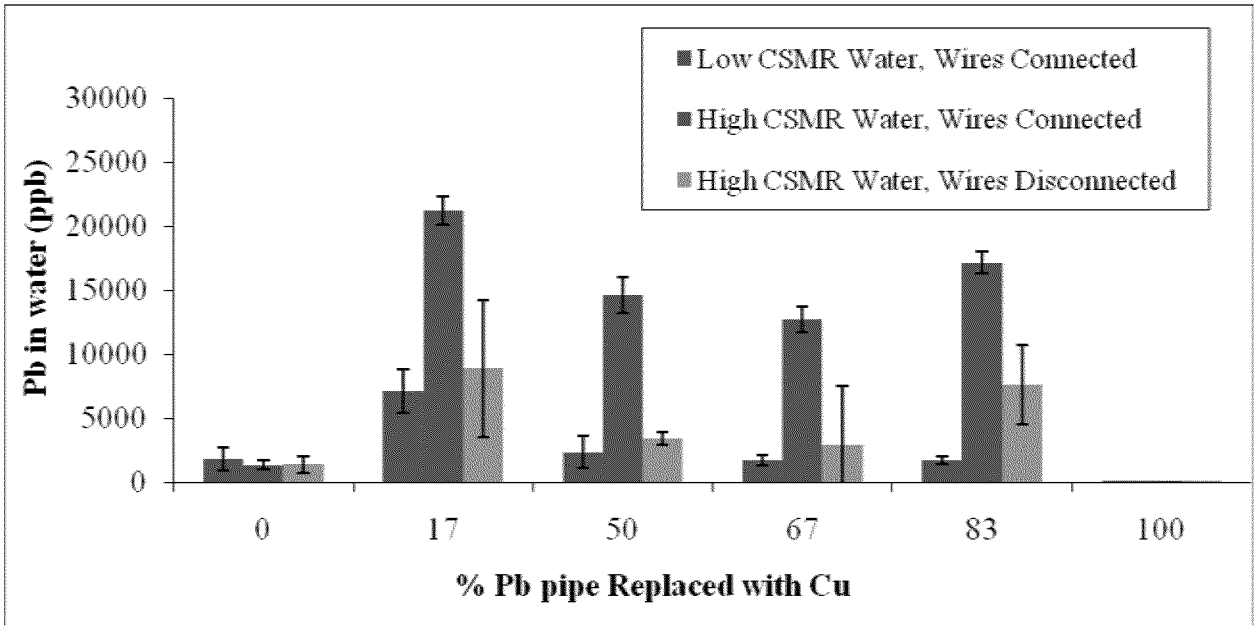

Figure 10: Lead leaching versus extent of lead replacement by copper, for old lead pipe B. Data were averaged for each experimental phase. The error bars denote 95% confidence intervals.

When compared to new Pb pipe, the extent of lead contamination of water was either lower (for old Pb pipe A) or relatively similar (for old Pb pipe B). For instance, the highest amount of lead released to the water was 23,000 ppb for new Pb pipe (Figure 8), whereas the highest lead released to the water was 13,000 ppb for old Pb pipe A (Figure 9), and 21,000 ppb for old Pb pipe B (Figure 10).

MECHANISTIC INSIGHTS VIA GALVANIC CURRENT MEASUREMENTS

Measurement of the galvanic current between the lead and copper portion of the rigs can provide mechanistic insights on the observed lead leaching trends. The galvanic current is a direct measure of the instantaneous rate of galvanic corrosion between the lead solder and the copper pipe. A higher magnitude of current indicates a higher rate of galvanic corrosion.

Galvanic current measurements were taken during Phase 1 and Phase 2 of the experiment, when galvanic corrosion was activated. Since the wires were disconnected during Phase 3, thereby blocking electron flow between the two dissimilar metals, no current was flowing and thus no such measurements were conducted during that phase. All of the current values measured in this study were positive. This means that the actual direction of current was the same as that of the reference multi-meter direction. The reference multi-meter direction had been chosen based on the expectation that lead would be the anode and copper would be the cathode. Thus, the positive currents suggest that lead was indeed being sacrificed (i.e., corroded), and that its corrosion was accelerated by connection to the copper.

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New Pb pipe connections to Cu pipe

Higher currents were measured when high CSMR water was fed to the rigs (Phase 2), compared to when low CSMR water was fed to the rigs (Phase 1) (Figure 11). This trend is consistent with the lead leaching results (Figure 8).

Under the high CSMR water condition, the highest current of 87 uA was measured for the 17% Cu rig, followed by the 50% Cu, 67% Cu, and 83% Cu rigs (Figure 11). These differences were statistically significant at the 95% confidence level (Error bars plotted, Figure 11). Thus, the overall galvanic reaction in this water was anodically limited, since it increased as the relative surface area of Pb:Cu (i.e. anode:cathode) increased. The ranking of the rigs with respect to the magnitude of the measured galvanic currents is consistent with that based on the lead leaching results. For instance, the 17% Cu rig had the highest measured current (Figure 11), and it also resulted in the highest lead-in-water concentrations (Figure 8).

Under the low CSMR condition, the highest current of 52 uA was measured for the 17% Cu rig, followed by the 50% Cu, 67%, and 83% Cu rig. These differences were statistically significant at the 95% confidence level (Error bars plotted, Figure 11). This ranking in terms of galvanic current magnitude under the low CSMR condition did not always agree with the ranking based on the lead leaching results (Figure 8). A possible explanation for this discrepancy is that galvanic current between lead and copper is a measure of galvanic corrosion only, and does not account for lead release due to normal lead dissolution or deposition corrosion. Total lead measured in the water accounts for all lead release mechanisms. In addition, galvanic current magnitude determines the rate of lead oxidation, but not necessarily the rate of lead release. It does not give information about how much lead is actually released into the water, because the oxidized lead can either be deposited onto the pipe surface as corrosion scale, or can be released into the water. For all these reasons, even though galvanic current magnitude is a direct indicator of galvanic corrosion of the lead pipe, it cannot be expected to perfectly predict lead levels in water.

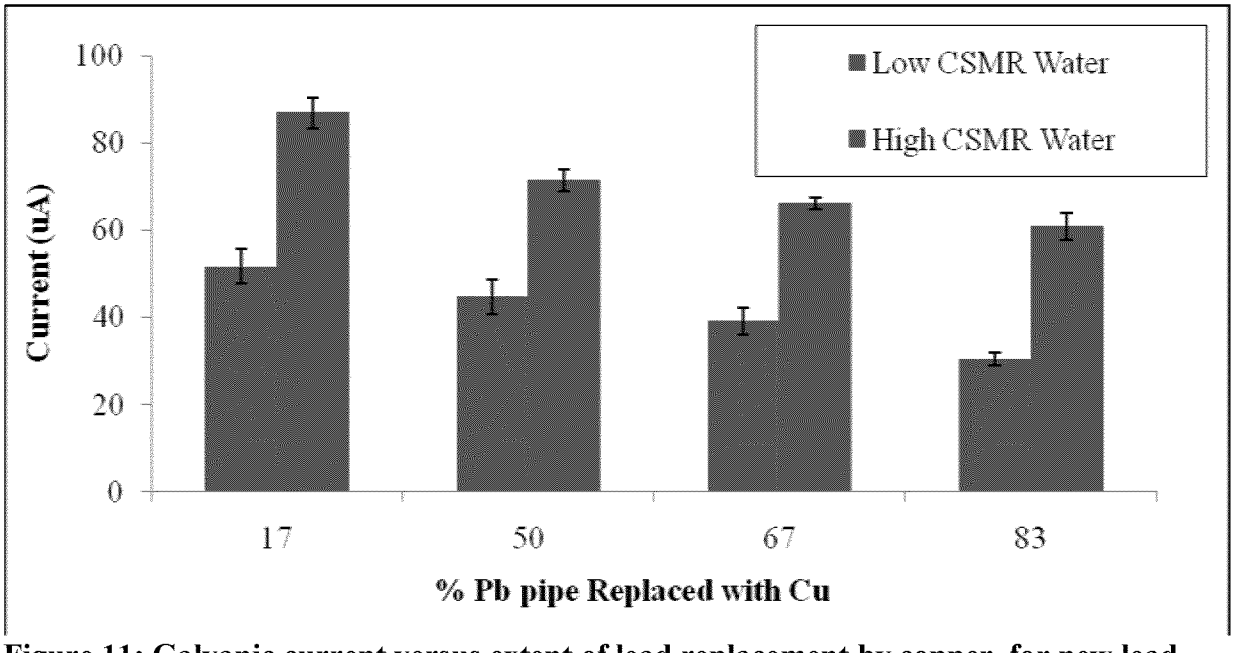

Figure 11: Galvanic current versus extent of lead replacement by copper, for new lead pipe. The error bars denote 95% confidence intervals.

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Old Pb pipe connections to Cu pipe

Higher currents were measured when high CSMR water was fed to the rigs (Phase 2), compared to when low CSMR water was fed to the rigs (Phase 1). This was true for old Pb Pipe A (Figure 12), consistent with the lead leaching results (Figure 9), as well as for old Pb Pipe B (Figure 13), consistent with the corresponding lead leaching results (Figure 10).

For old Pb Pipe A under the high CSMR water condition (Phase 2), the highest current of 67 uA was measured for the 17% Cu rig, followed by the 50% Cu, 67% Cu and 83% Cu rig (Figure 12). Under the low CSMR condition (Phase 1), the highest current of 37 uA was measured for the 17% Cu rig, followed by the 50% Cu, 83% Cu, and 67% Cu rig. For old Pb Pipe A, the ranking of the rigs in terms of galvanic current measurements under both the low CSMR and the high CSMR water condition (Figure 12) did not correspond to their ranking based on the lead leaching results (Figure 9).

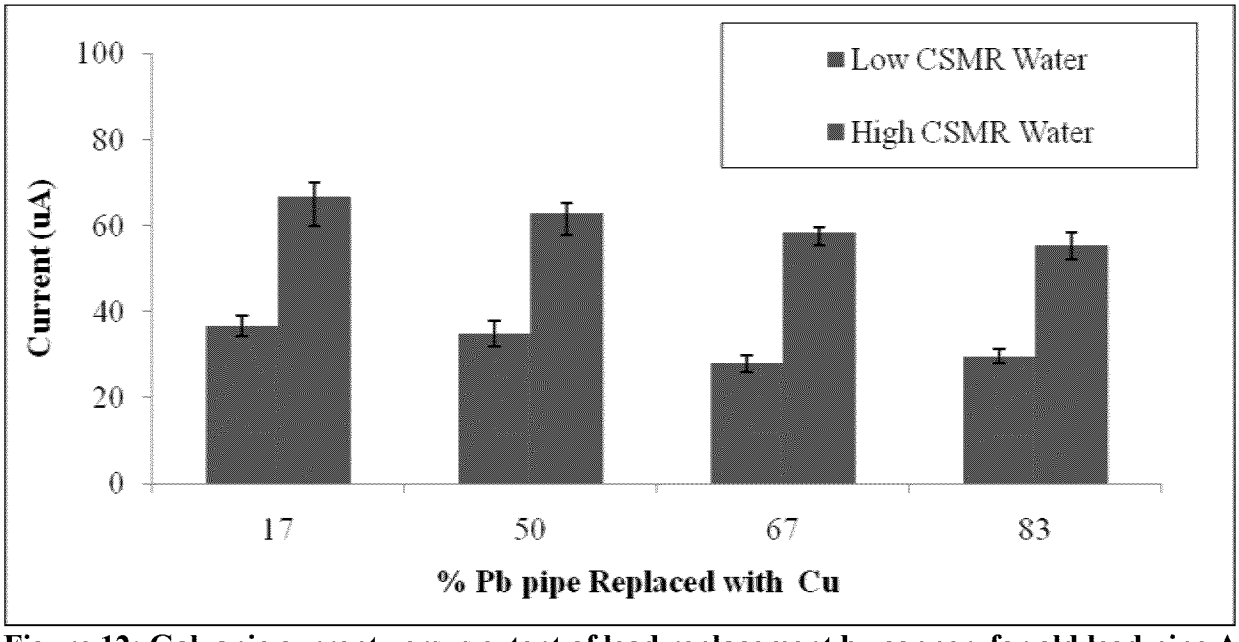

Figure 12: Galvanic current versus extent of lead replacement by copper, for old lead pipe A. The error bars denote 95% confidence intervals.

For old Pb Pipe B under the high CSMR water condition (Phase 2), the highest current of 73 uA was measured for the 17% Cu rig, followed by the 50% Cu, 67% Cu and 83% Cu rig (Figure 13). Under the low CSMR condition (Phase 1), the highest current of 50 uA was measured for the 17% Cu rig, followed by the 67% Cu 83% Cu, and then the 50% Cu rig. For old Pb Pipe B, the ranking of the rigs in terms of galvanic current measurements under either the low CSMR or the high CSMR water condition (Figure 13) did not correspond to their ranking based on the lead leaching results (Figure 10).

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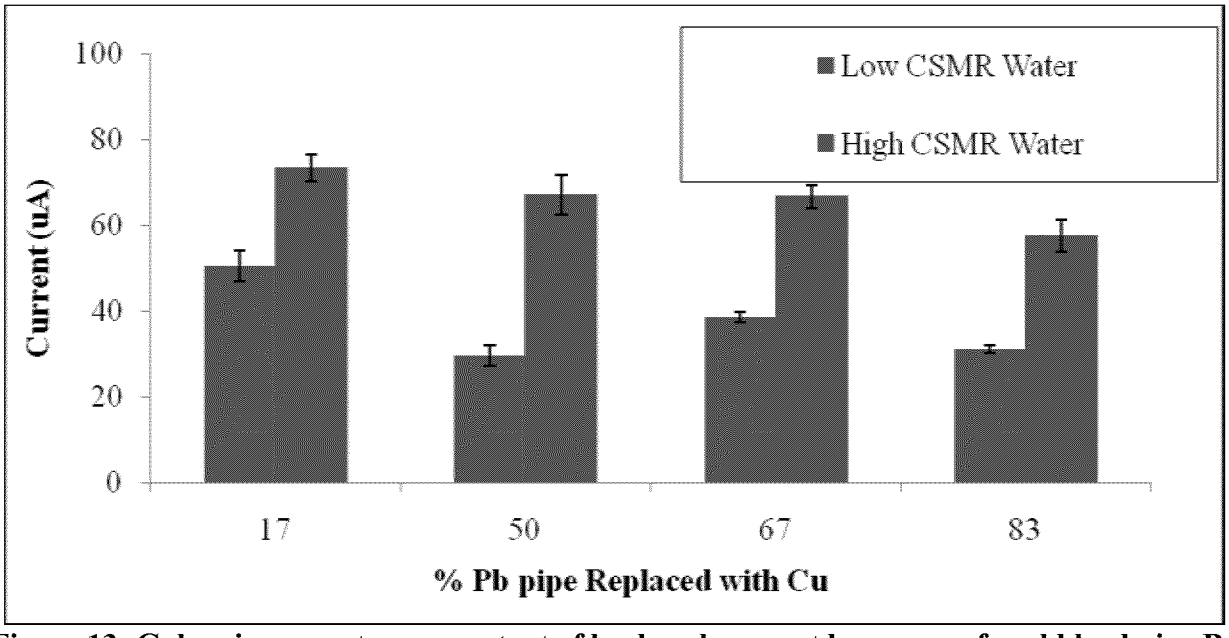

Figure 13: Galvanic current versus extent of lead replacement by copper, for old lead pipe B. The error bars denote 95% confidence intervals.

The galvanic currents in the three sets of Pb:Cu rigs were consistent with their corresponding lead leaching results. Overall, the ranking of Pb pipes, with respect to lead release into the water and galvanic current magnitude, was:

New Pb pipe > Old Pb pipe B > Old Pb pipe A

In other words, new Pb pipe connections to Cu pipe produced the highest galvanic currents (up to 87 uA, see Figure 11), and they resulted in the highest lead-in-water contamination (up to 23,000 ppb, see Figure 8). Old Pb pipe B connections to Cu pipe produced the second highest galvanic currents (up to 73 uA, see Figure 13) and resulted in the second highest lead-in-water contamination (up to 21,000 ppb, see Figure 10). Finally, old Pb pipe A connections to Cu pipe had the lowest measured galvanic currents (up to 67 uA, see Figure 12) and resulted in the lowest lead-in-water contamination (up to 13,000 ppb, see Figure 9).

Area of galvanic influence and potential implications

The third set of rigs (Old Pb pipe B) was constructed by connecting in series many short Pb pipes together, in order to add up to the desired Pb length. For example, the 17% Cu rig consisted of one 0.5ft Cu section, connected to five sequential 0.5 ft Pb sections (Figure 14, bottom). This setup allowed measuring the galvanic current between the Cu and each of the Pb sections. The total current between the Cu and Pb pipe was the sum of the individual currents between the Cu and each Pb section.

Throughout Phases 1 and 2 of the experiment (when the wires were connected between lead and copper), the galvanic current was concentrated to the Pb segment that was closest to the galvanic junction. During Phase 1 of Low CSMR Water, the current flowing between the Cu and the first Pb segment constituted on average 90% of the total current between Pb and Cu in the rig

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(Figure 14, top). The second Pb segment received 7% of the total current. The remaining three Pb segments, those furthest away from the connection to Cu pipe, accounted for only 3% of the total current (Figure 14, top). Similarly, during Phase 2 of High CSMR water, the first Pb segment was impacted by 95% of the total galvanic current, the second Pb segment by 3% of the galvanic current, and the remaining three Pb segments by 2% of the current (Figure 14, top). Since galvanic current is a direct measure of galvanic corrosion, these data suggest that the Pb segment that is closest to the galvanic junction is the one that gets impacted the most (i.e., corroded), due to galvanic connection to copper.

These results are consistent with the observations of other researchers. Based on surface potential measurements, Reiber and Dufresne (2006) determined the area of galvanic influence to be in the first few inches of the lead pipe, in the vicinity of the coupling to copper. Based on mineralogical and visual observations, DeSantis et al. (2009) determined the zone of galvanic influence to be a few milimeters wide, immediately adjacent to the joint.

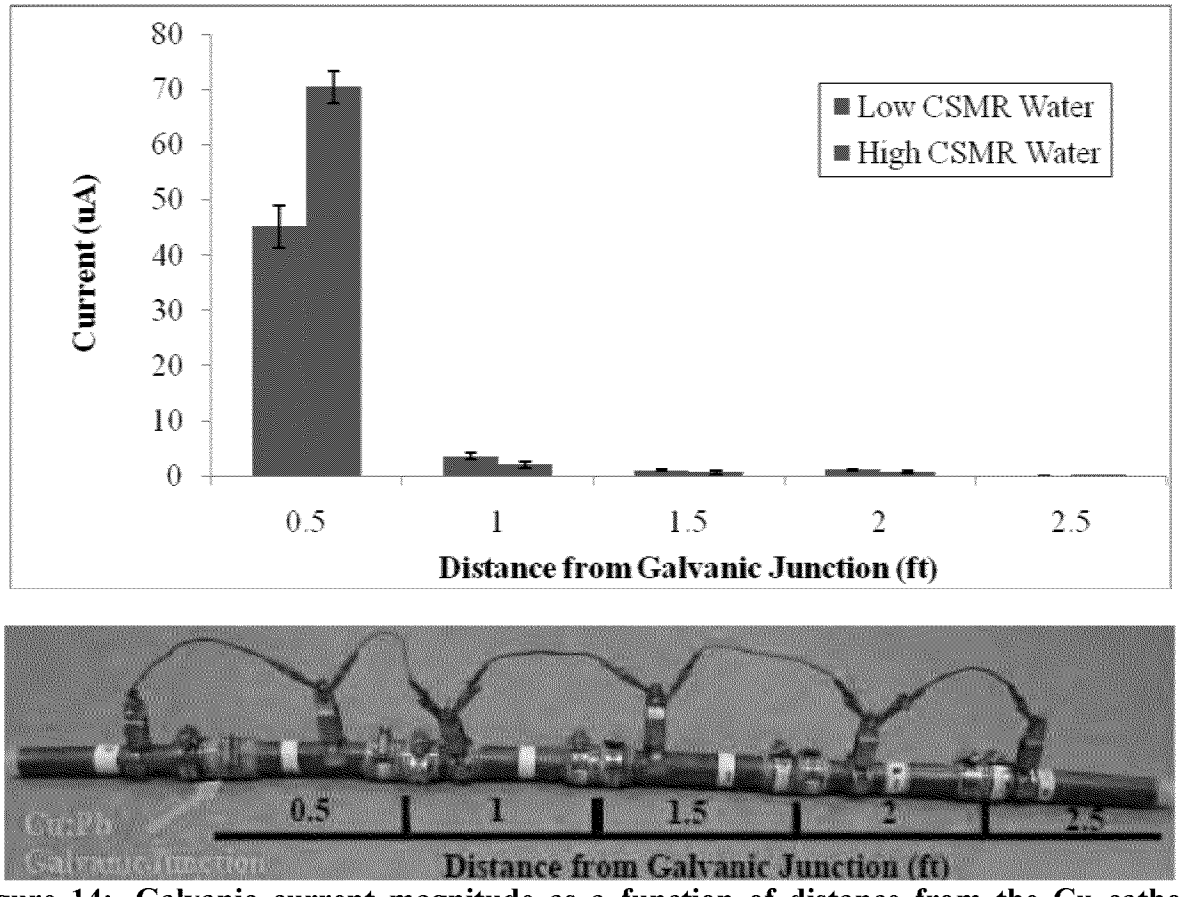

Figure 14: Galvanic current magnitude as a function of distance from the Cu cathode (top). Small segments of Pb pipe (Old Pb pipe B) were placed in series, allowing current measurements as a function of distance from the junction (bottom).

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The limited zone of galvanic impact provided a basis upon which researchers have dismissed the effect as inconsequential (Reiber and Dufresne 2006). However, translating galvanic effects to lead release and human exposure can offer a different perspective. DeSantis et al. (2009) identified failures at joints (i.e., lead pipe wall completely eaten away), due to the depth of galvanic corrosion in the pipe wall. Simple calculations suggest that even if only 1 inch length of lead pipe is half-eaten away at the pipe wall due to galvanic corrosion, the released lead mass equals 25 grams (calculation based on lead density of 11.3 g/cm^3, lead pipe internal diameter of ¾ inches, and lead pipe external diameter of 1 inch). This mass of lead is sufficient to contaminate every drop of water used by a family of four for 4 years, with lead levels above the LCR action limit of 15 ppb (calculation based on 300 gal/day, or else 1135 L/day water usage for the whole family). Thus, the limited area of attack does not translate to limited impacts on public health.

EFFECT OF ALKALINITY

Alkalinity is a measure of buffering capacity: high alkalinity waters are thought to be well buffered, and able to resist pH changes such as the detrimental pH drop close to the lead surface at galvanic junctions (see Figure 1, right). Throughout the three phases of this study, the alkalinity of the water entering the rigs was maintained at 15 mg/L as $CaCO_3$ (see Table 1). Waters with this low alkalinity level have been described as alkalinity deficient in terms of lead corrosion control (Edwards et al., 1999). Edwards et al. (1999) identified alkalinity of 100 mg/L as $CaCO_3$ as an approximate upper bound to obtaining substantial improvements in lead release for waters with a pH below 8.5, as is the synthetic water utilized in this experiment (pH was maintained at 8.0 throughout this study, see Table 1).

In order to examine the impact of higher alkalinity levels on lead release due to galvanic corrosion, additional tests were performed. After Phase 3, galvanic corrosion was re-activated in the rigs (by re-connecting the wires between lead and copper). The rigs were then exposed to High CSMR water of gradually increased alkalinity. Specifically, alkalinity was increased from 15 to 50 mg/L $CaCO_3$ for four weeks, and from 50 to 100 mg/L $CaCO_3$ for 10 weeks. Alkalinity levels throughout the experiment were adjusted by adding the appropriate amount of sodium bicarbonate ($NaHCO_3$) from a fresh stock solution to the synthetic water.

Surprisingly, increasing the alkalinity was not able to alleviate lead corrosion for any of the three types of lead pipe, as evidenced by lead release to the water, as well as galvanic current magnitude. For example, for old Pb pipe A, increasing the alkalinity from 15 to 50 and then to 100 mg/L as $CaCO_3$ resulted in either similar, or even higher, lead-in-water levels for all rigs (Figure 15).

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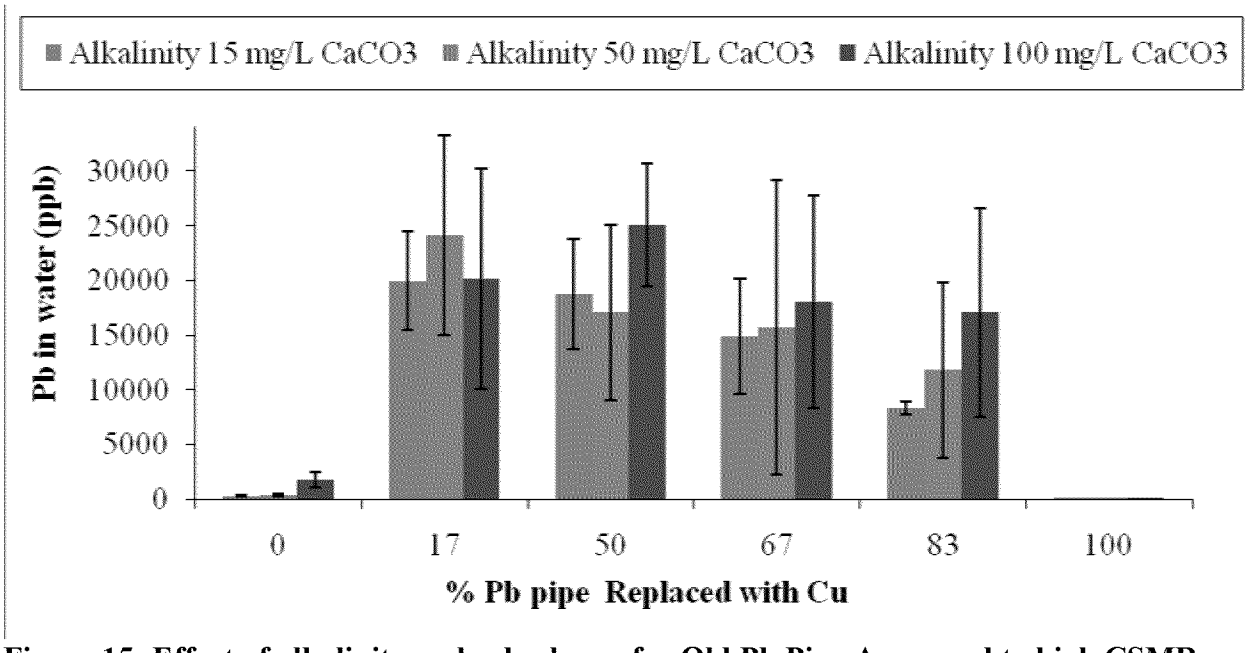

Figure 15: Effect of alkalinity on lead release, for Old Pb Pipe A exposed to high CSMR water. The error bars denote 96% confidence intervals.

Likewise, the galvanic currents between lead and copper increased, when the alkalinity increased (Figure 16). Similar trends in lead release and galvanic current were observed for the other two types of lead pipe examined (results not presented herein).

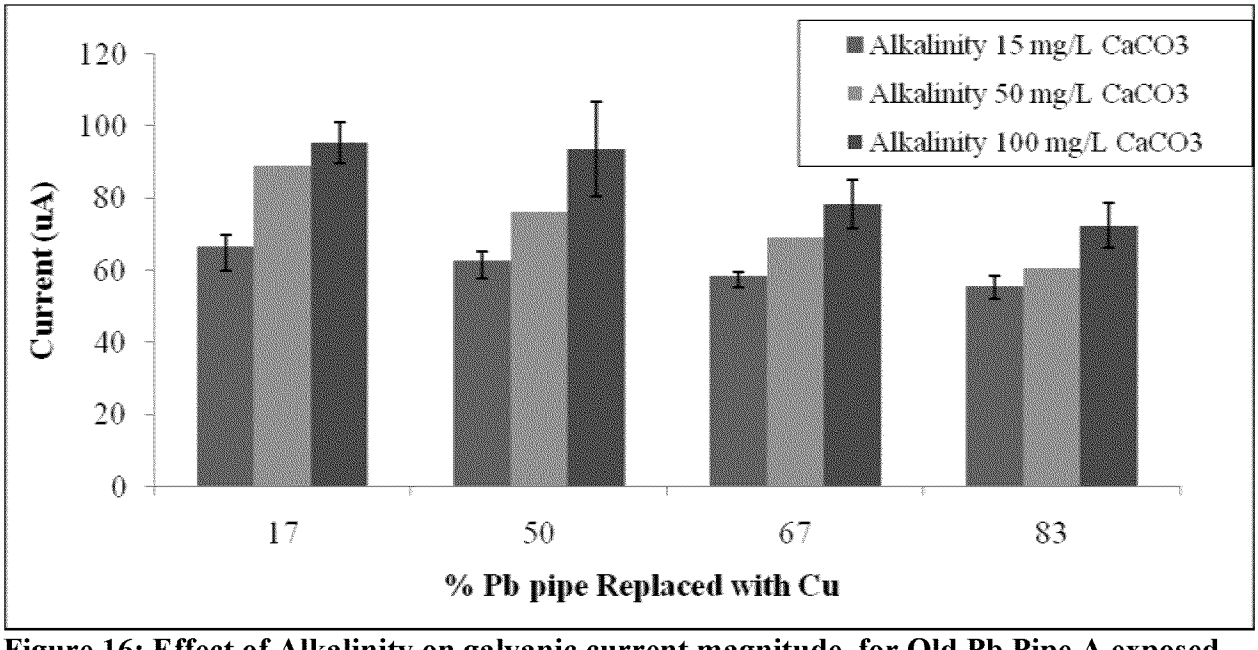

Figure 16: Effect of Alkalinity on galvanic current magnitude, for Old Pb Pipe A exposed to high CSMR water. The error bars denote 95% confidence intervals.

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CONCLUSIONS

Under controlled experiments of simulated partial lead service line replacements that lasted for more than seven months:

- Galvanic connections between copper pipe and lead pipe (either new or old) increased lead release compared to lead pipe alone, under stagnant water conditions.
- Removal of the galvanic connection between copper pipe and lead pipe decreased lead release by 2-6 times (depending on the age of lead pipe), under a high CSMR water condition (i.e., CSMR of 16).
- Water with a high CSMR of 16 released 3-12 times more lead to the water than did low CSMR water of 0.2, for both new and old lead pipes connected to copper.
- High sustained galvanic currents between copper and lead pipe (up to 87 uA for new lead pipe) were measured when the CSMR was high, resulting in galvanic corrosion of the lead. When the CSMR was low, galvanic currents were lower (up to 52 uA for new lead pipe), consistent with corresponding lower lead leaching results.
- Increasing the alkalinity from 15 mg/L to 50 and then to 100 mg/L as $CaCO_3$ was not able to overcome the corrosive effects to the lead pipe from high CSMR water in galvanic connections.
- Based on galvanic current measurements, the area of lead pipe adjacent to the copper joint (<0.5 ft) is most affected by galvanic corrosion. The lead pipe sections that are furthest away from the copper junction (>0.5 ft) are the least affected by the galvanic connection to the copper.

Based on these initial results and an in-depth literature review, any decision to conduct partial lead service line replacements with copper pipe should be carefully considered. These preliminary results indicate that galvanic corrosion might significantly contribute to lead leaching even after more than seven months, depending on drinking water chemistry. As a result, the practice of partial lead service line replacements with copper may actually increase lead contamination of potable water, under worst-case conditions. If this effect was shown to persist under more realistic exposure scenarios, remedial strategies might be necessary to protect consumers from exposure to lead due to galvanic corrosion.

The work presented herein is a first step in understanding the implications of galvanic corrosion to lead release into drinking water, relative to partial lead service line replacements with copper pipe. More research is needed, in order to:

- Quantify the relevant contribution of galvanic corrosion to lead release, compared to other mechanisms such as normal dissolution, deposition corrosion, particle detachment, and lead retention in pipe scale
- Examine galvanic corrosion under realistic water flow regimes (i.e., intermittent flow which is more representative of typical water usage at homes). This work examined stagnant water conditions, while other research examined continuous flow conditions
- Assess the effect of couplings between lead pipe and copper pipe on galvanic corrosion (e.g., brass corporation valve or brass compression fitting)

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- Test lead service lines harvested from the field after decades of use or conduct additional testing on partially replaced lead pipes in practice. The pipes used in this study were designed to examine consequences of galvanic corrosion on lead leaching under worst-case scenarios, and may not reflect the behavior of pipes with decades of accumulated scale
- Evaluate the effects of other water qualities on galvanic corrosion (e.g., corrosion inhibitor added to the water)

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ABBREVIATIONS

CDC	Centers for Disease Control and Prevention
CSMR	Chloride-to-Sulfate Mass Ratio
cm	centimeter
Cu	copper
EPA	Environmental Protection Agency
ft	feet
gal	gallon
g	gram
ICP-MS	Inductively Coupled Plasma - Mass Spectrometry
ID	Internal Diameter
LCR	Lead and Copper Rule
L	liter
uA	microampere
OD	Outside Diameter
PAC	Project Advisory Committee

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Pb	lead
ppb	parts per billion
UK	United Kingdom
US	United States of America
Ω	Ohm

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