

Long-Term Behavior of Simulated Partial Lead Service Line Replacements

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Abstract

In this 48-month pilot study, long-term impacts of copper:lead galvanic connections on lead release to water were assessed without confounding differences in pipe exposure prehistory or disturbances arising from cutting lead pipe. Lead release was tracked from three lead service line configurations, including (1) 100% lead, (2) traditional partial replacement with 50% copper upstream of 50% lead, and (3) 50% lead upstream of 50% copper as a function of flow rate, connection types, and sampling methodologies. Elevated lead from galvanic corrosion worsened with time, with 140% more lead release from configurations representing traditional partial replacement configurations at 14 months compared to earlier data in the first 8 months. Even when sampled consistently at moderate flow rate (8 LPM) and collecting all water passing through service lines, conditions representing traditional partial service line configurations were significantly worse ($\approx 40\%$) when compared to 100% lead pipe. If sampled at a high flow rate (32 LPM) and collecting 2 L samples from service lines, 100% of samples collected from traditional partial replacement configurations exceeded thresholds posing an acute health risk versus a 0% risk for samples from 100% lead pipe. Temporary removal of lead accumulations near Pb:Cu junctions and lead deposits from other downstream plastic pipes reduced risk of partial replacements relative to that observed for 100% lead. When typical brass compression couplings were used to connect prepassivated lead pipes, lead release spiked up to 10 times higher, confirming prior concerns raised at bench and field scale regarding adverse impacts of crevices and service line disturbances on lead release. To quantify semirandom particulate lead release from service lines in future research, whole-house filters have many advantages compared to other approaches.

Key words: corrosion; drinking water; galvanic corrosion; lead; partial lead service line replacement

Introduction

THE ENVIRONMENTAL PROTECTION AGENCY's (EPA) Lead and Copper Rule (LCR) was implemented to minimize lead and copper exposure from drinking water by various interventions, including replacement of lead service pipes. However, there is growing concern that the action of replacing a portion of lead service line with a copper pipe, to reduce consumer lead exposure, may not always be effective and in some cases actually increases exposure (Britton and Richards, 1981; US EPA, 1991, 2011; Swertfeger *et al.*, 2008, 2011; Muylwyk *et al.*, 2011). Specifically, when utilities partially

replace lead service lines with a copper pipe, either in response to requirements of the LCR or voluntarily, galvanic corrosion between the lead and newly installed copper and brass connectors can increase lead corrosion rates, which can either cancel or reverse anticipated benefits of having less lead pipe (Triantafyllidou and Edwards, 2010, 2011; Cartier *et al.*, 2012a; Giammar *et al.* 2012; Wang *et al.*, 2013).

A range of factors are suspected to influence the extent of galvanic corrosion between copper and lead, including flow rate, connector type, deposition corrosion, lead surface passivation, relative position of lead versus copper pipe, presence of crevices, and corrosivity of water (Triantafyllidou and Edwards, 2010; US EPA, 2011; Xie and Giammar, 2011; Cartier *et al.*, 2012a, 2013; Hu *et al.*, 2012; St. Clair *et al.*, 2012; Wang *et al.*, 2012, 2013; Clark *et al.*, 2013; Zhou *et al.*, 2015). Better understanding the duration of the adverse galvanic effects is extremely important, as early practical work suggested that such impacts could be sustained for years or decades (Britton and Richards, 1981). A few recent reports by EPA and others speculated that such effects would be insignificant and sustained only for days or a few weeks (Reiber and Dufresne, 2006; National Drinking Water

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Advisory Council, 2011; US EPA, 2011; Boyd *et al.*, 2012), although an “expression of concern” is now associated with that work as detailed elsewhere (Edwards, 2012; Retraction Watch, 2015). All recent studies that rigorously examined the issue at bench or pilot scale (up to 8 months duration) concluded that problems with elevated lead from galvanic corrosion and partial replacements could be sustained over the entire duration of the experiments (Triantafyllidou and Edwards, 2011; Cartier *et al.*, 2012a, 2013; Doré *et al.*, 2012; Wang *et al.*, 2012). Moreover, a recent study in Chicago by US EPA also reported longer term adverse consequences of partial lead service line replacement (PLSLR) (i.e., disturbed lead service lines) in terms of elevated lead in water risks (Del Toral *et al.*, 2013).

Detection of elevated lead problems from galvanic corrosion has been complicated by differing results dependent on flow rate and sampling methodologies, and recommended sampling methods for compliance monitoring are still under review (US EPA, 2012). If water samples are collected at low flow, lead particulates and sediments are not mobilized, which can hinder detection of visible deposits of lead scale created at the lead:copper pipe connection due to galvanic corrosion (Deshommes *et al.*, 2010; Triantafyllidou and Edwards, 2011; Cartier *et al.*, 2012a). Samples collected at moderate (8 LPM) and higher flow rates (32 LPM) from partially replaced lead pipe configurations revealed sporadic detachment of lead particulates at levels posing an acute health risk (Cartier *et al.*, 2012a). The latter observation could explain why a higher incidence of elevated blood lead in children was reported in homes with partial pipe replacements versus undisturbed lead service lines (Brown and Margolis, 2012), and higher rates of fetal death were noted to occur during periods of partial pipe replacements (Edwards, 2014), even when routine sampling of such homes at low flow rates did not detect serious problems (Giani *et al.*, 2004; Giani, 2008). Practical sampling methods that are capable of better detecting semi-random release of lead particulates that can pose an acute health risk to consumers (McNeill and Edwards, 2004; Triantafyllidou *et al.*, 2007) are needed.

Other factors have been speculated to play a role in service line corrosion and galvanic impacts arising from lead:copper connections. For instance, copper and lead pipe sections are cathodic to iron water mains, which imply that lead connections to unlined iron might be cathodically protected from corrosion (Clark *et al.*, 2013), consistent with other anecdotal field evidence suggesting that iron mains cathodically protect copper service lines when they are directly connected in the distribution system (Gehring *et al.*, 2003; Rajani and Kleiner, 2003). It is hypothetically possible that an iron main coupled to a lead service line could provide cathodic protection to the lead or, possibly, eliminate galvanic corrosion of the lead in a partial pipe replacement if unlined iron mains are present. If this protection was significant, replacement of unlined iron mains with either new lined pipes or plastic, as occurs routinely with infrastructure upgrades, might also produce increased lead contamination of water. There are no laboratory studies refuting or supporting these hypothesized effects. Prior bench scale research also suggested that partially replacing lead pipes with copper can create much more serious problems with lead contamination of water during prolonged stagnation events when compared to a situation with a 100%

lead pipe (Triantafyllidou and Edwards, 2010; Arnold and Edwards, 2012; Cartier *et al.*, 2012a).

The goal of the current work is to (1) extend data collection in the 8-month pilot study described in Cartier *et al.* (2012a) to a time period of >2.5 years, (2) better define the nature of elevated lead release after partial pipe replacements, (3) examine galvanic impacts on prepassivated lead pipes using “real” brass connectors, (4) evaluate the strengths/weaknesses of sampling protocols that could detect public health concerns arising from partial pipe replacements, (5) conduct preliminary analysis of unlined iron main impacts on corrosion of service lines, and (6) determine how prolonged stagnation periods influence lead release.

Materials and Methods

The pilot is that described by Cartier *et al.* (2012a) using Blacksburg tap water, but extending the previously reported results from 8 months to a total time of 48 months of operation. Blacksburg tap water is relatively noncorrosive due to pH adjustment and dosing of a zinc orthophosphate corrosion inhibitor. Three configurations representing a full-lead service line (100% Pb), a PLSLR with lead downstream of copper (Pb-D; 50% Cu and 50% Pb), and a PLSLR with lead upstream of copper (Pb-U; 50% Pb and 50% Cu) were tested in triplicate. Each 50:50% pipe section was galvanically coupled with an external wire. A 100% copper control was also tested. Four phases of research tested three different sampling methods (Table 1). During all test phases, galvanic corrosion currents were continually measured by a zero-resistance ammeter using a GAMRY Potentiostat. During months 8–13, the pilot operated at low flow and was not sampled.

Phase A: The pilot was operated and sampled like that described by Cartier *et al.* (2012a) during months 13–14, including low, moderate, and high flow rates. A first draw, 2 L, sample was collected after 16-h stagnation periods for all three flow rates. After the flushing events, filtered samples were immediately collected from the 2 L sample to quantify soluble lead using a 0.45 μm filter. Thereafter, the 2 L first draw and filtered sample were acidified with nitric acid to a concentration of 2% v/v and allowed to digest for a minimum of 5 days before analysis through an inductively coupled plasma mass spectrometer according to the Standard Method 3125B (APHA, 1998).

Phase B: From months 15–22, consistent flow rates emulating a household faucet (8–9 LPM) were tested, in contrast to the earlier work (months 0–15) in which flow rates were 1.3 LPM, except during sampling events. Daily composites (five times per week) of all water flowing through the pipes from three flow events per day (2 min of flow after 8 h of stagnation) were collected in separate bins for each replicate (48 L total per rig; Phase B-1). On Mondays, the composite included a flow event after a 54 h weekend stagnation period. Sample aliquots from each unacidified bin were collected after rigorously stirring the bins to mobilize any particulates; QA/QC by decanting the top fraction and acidification of the remaining 2 L demonstrated that this approach did not systematically underquantify particulate lead (<10% error).

To evaluate how accumulations of lead deposits (i.e., reservoirs in the plumbing, including lead scale on pipe at the copper junction or walls of copper and downstream plastic plumbing that might have been coated with lead) were

TABLE 1. EXPERIMENTAL PHASES AND SAMPLING METHODS

Phase	Time period (months during experiment)	Experimental change from Cartier <i>et al.</i> (2012a)	Sampling method
A	13–14	No change	Same as Cartier <i>et al.</i> (2012a), (first 2 L sample and grab samples)
B-1	15–17	Consistent 8–9 LPM flow rate	Collection of all water passing through pipes (bins)
B-2	17–18	Removed 4" coupons	
B-3	19–22	Replaced copper piping	
C	22–25	Installed connectors	Collection of all water passing through pipes (bins)
D	26–27	Prolonged stagnation events	First 2 L sample
E	28–48	Increased flow rate (~18–20 LPM)	Whole-house filters

contributing to lead release, reservoirs of lead that could accumulate in the plumbing system were systematically removed and replaced. Specifically, after 17 months (492 days), the lead coupons with heavy galvanic corrosion products at the copper junction were removed, and the rigs were then sampled during months 17–18 (Phase B-2; days 493–524). Thereafter, for both the Pb-U and Pb-D triplicates, the old copper pipes were replaced with new copper during month 19 (day 560) and sampled for lead release during months 21–22 (Phase B-3; days 623–643) and the rig was sampled for lead release.

The mass of lead deposits in each of the above sections was quantified. Easily removed lead rust was cleaned from the lead coupons, and total lead mass and weight loss were calculated. Copper pipe sections were filled with deaerated 10% hydrochloric acid to dissolve most of the lead that remained on the copper pipe sections. Finally, the plastic plumbing downstream of the metal pipe sections was acid cleaned and the mass of lead was quantified.

Phase C: This portion of the research explored how new low lead (<1%) brass connectors might enhance lead leaching from passivated lead service lines through galvanic corrosion without any pipe cutting. Side experiments demonstrated very low lead leaching (<5 ppb) from the brass used in this research. Three types of connections representing extremes encountered in practice were tested, including a brass compression fitting, copper sleeve (which has crevices similar to compression fitting, but made of copper only and thus presents no potential for lead contamination, identical to Clark *et al.*, 2013), and the existing clear plastic tubing connection with a small dielectric spacer. A baseline for lead release was established for each of the pipe rigs during the final stage of Phase B. On the basis of the baseline results, the pipe with the highest lead release remained connected as before with plastic tubing. The pipe with the lowest baseline lead release was connected using a copper sleeve, and the pipe with the middle level of lead release was connected using a brass compression fitting (same corporation valve as Clark *et al.*, 2013). From months 22–25, all water flowing from each rig was collected in bins from each condition and sampled as per Phase B.

Phase D: During months 26–27 the rigs underwent two 1-month-long stagnation events. The prolonged stagnation events represented conditions of prolonged homeowner absences with long periods of little to no flow. After each stagnation event, a 2 L first draw was collected from each rig at a flow rate of 15 LPM during a 5-min flow event. The 2 L samples were acidified and analyzed as described previously.

Phase E: After extensive bin sampling during Phase C, whole-house filter cartridges (polypropylene, 1 μ m), used to filter water at the point of entry, were installed downstream of the various rigs to collect and quantify lead release. During months 28–34, rigs were flushed at 18–20 LPM for 5 min, three times per day, to achieve a total daily volume of ~300 L per rig; during this phase, flow events continued over weekend periods with no 54-hr stagnation event. Filters were allowed to collect lead for periods of 38 days. Thereafter, filter housings and cartridges were removed and acidified with nitric acid to a concentration of 5% v/v. Sample aliquots were taken after 120 h digestion and analyzed through ICP-MS as described previously. From months 35–48, rigs operated under the same conditions and filters were collected and analyzed every 3–4 months following the procedure outlined above. During these periods, 2 L first draws were also collected at the beginning of flushing events.

Phase F: Concerns related to unlined iron main connections to services lines were explored using a small 10.2 cm lead pipe section that was coupled to a 305 cm galvanized iron pipe section through varying lengths of copper pipe ranging from 2.5 to 305 cm copper using clear plastic tubing; a control condition consisted of the lead and galvanized iron (no copper in between) connected by the plastic tubing. The apparatus was filled with Blacksburg water, and the sacrificial galvanic currents to the lead were measured immediately using a handheld multimeter. Then, the experiment was repeated with varied lengths of copper pipe. Testing demonstrated that a zero resistance ammeter and the multimeter gave equivalent results to within $\pm 5\%$.

Another short-term experiment briefly connected 3 m of galvanized iron to the first section of the pilot in the flow sequence. Each rig was filled with water, and the galvanic current sacrificing the lead was quantified.

Statistical analyses were performed using Student's *t*-test. All confidence testing reported herein was conducted at the 95% confidence level ($p < 0.05$), unless otherwise stated.

Results and Discussion

Six experimental objectives of the work are addressed sequentially in the sections that follow.

Phase A: low, moderate, high flow sampling

Trends in lead release through 9–14 months mirrored those obtained during the 8-month pilot reported by Cartier *et al.* (2012a), confirming that serious water contamination problems arising from galvanic connections can certainly persist

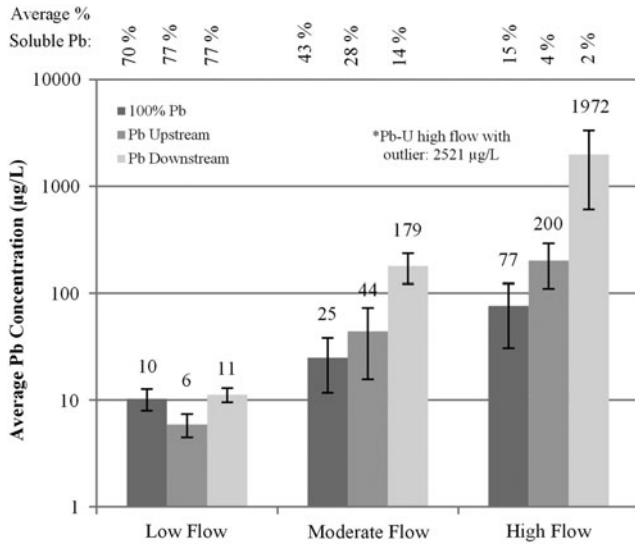


FIG. 1. Average lead concentration in water after 14 months from three sampling periods as described by Cartier *et al.* (2012a) with average soluble lead percentage in samples. Error bars represent 95% confidence interval. *High flow rate, Pb-U condition excludes high outlier of 20,090 µg/L. (*N*=9, 3 sampling events of triplicates).

for years, as suggested in earlier field sampling (Britton and Richards, 1981; Del Toral *et al.*, 2013). Lead release increased for samples collected at a higher flow rate for all three conditions (100% Pb, Pb-U, and Pb-D), but the worst case was the configuration created by partial replacement, with lead pipe after copper pipe in the flow sequence (Pb-D rigs; Fig. 1). Specifically, at low flow rates over this time period, average lead concentrations were not statistically different among the tested service line configurations, but at moderate and high flow rate sampling, the condition with Pb-D vs 100% full-lead pipe was 7 and 26 times higher, respectively ($p < 0.05$). Filtered samples were analyzed to determine the soluble fraction of lead in the first 2 L flush (Fig. 1). During low flow, a major fraction of lead was soluble (70–80%) for all conditions. As flow rate increased particulate lead became the dominant fraction of lead released (e.g., at high flow,

soluble lead <5% of the total lead in both the Pb-U and Pb-D conditions).

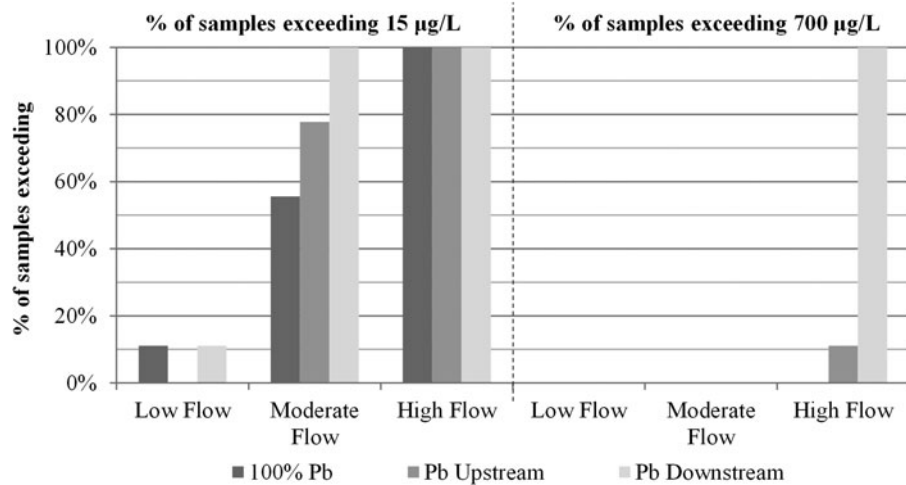
Comparison of these results to the 8-month study of Cartier *et al.* (2012a) obtained using the same apparatus; at high flow rate, lead release decreased 68% (240 µg/L to 77 µg/L) for the 100% Pb and 52% for Pb-U (416 µg/L to 200 µg/L) configuration after the initial study period. Conversely, for Pb-D, lead release *increased* 135% (839 µg/L to 1972 µg/L) during months 13–14 compared to the first 8 months of the pilot. These are unambiguous data demonstrating that the elevated lead originating from configurations of partial replacements, in which a copper pipe is placed upstream of the lead pipe, does not necessarily ameliorate with time but can actually worsen.

At low flow, only one sample (11%) from the service line for the condition with 100% Pb and Pb-D exceeded the LCR action level of 15 µg/L; while at high flow, 100% of samples exceeded the EPA action level (Fig. 2). Moreover, lead concentrations in 100% of samples collected from the Pb-D service line at high flow even exceeded acute health risk standards of >700 µg/L as per the analysis of Cartier *et al.* (2012a), whereas only 11% of Pb-U and 0% of full-lead pipe samples exceeded this threshold (Triantafyllidou, 2014; Fig. 2). This result provides a basis for heeding prior warnings to avoid placing copper in front of lead in the flow path sequence (Britton and Richards, 1981; Breach *et al.*, 1991; Copper Development Association, 1999; Triantafyllidou and Edwards, 2010, 2011; Clark *et al.*, 2011; Cartier *et al.*, 2012a) and health warnings especially for young children and developing fetus (Edwards, 2014). Moreover, the results indicate that collecting samples at low flow rates, or forcing consumers to collect samples at low flows by providing bottles with very small openings as is now commonplace in many cities, can dramatically underestimate water lead levels that can be present during normal use.

Phase B: consistent moderate flow sampling

It was hypothesized that detriments of partial replacements and galvanic corrosion might disappear if flow rates were maintained at very consistent levels and all water from the rig was collected. During the first 8 weeks of sampling with flow at 8 LPM and collecting all the water (Phase B-1), the

FIG. 2. Percentage of first 2 L service line samples exceeding 15 µg/L (LCR action level) and 700 µg/L (acute health risk level) during Phase A—low, moderate, and high flow sampling (*N*=9, 3 sampling events of triplicates).



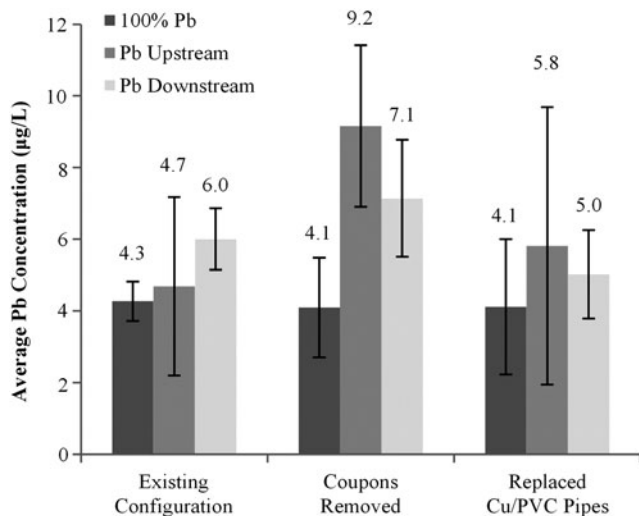


FIG. 3. Average Pb release during various stages of Phase B. Error bars represent 95% confidence interval (data from replicated pooled for each individual condition). (Phase B-1, $N=69$; Phase B-2, $N=27$; Phase B-3, $N=45$).

concentration of lead was not statistically different for conditions representing 100% Pb pipe or if a lead service line was followed by copper (Pb-U) (Fig. 3). However, the Pb-D was 40% worse than the 100% Pb condition (100% Pb: $4.3 \mu\text{g/L}$, Pb-D: $6.0 \mu\text{g/L}$, $p < 0.05$). In other words, even if the flow rate was maintained at stable and normal levels, the average lead release in 48 L sampled from a partial pipe replacement configuration is statistically worse than a full-lead service line with twice the lead pipe surface area.

Lead reservoirs

Elevated galvanic currents are expected to contribute to a reservoir of lead in the pipe rigs and especially at the junction between lead and copper (Triantafyllidou and Edwards, 2011; Cartier *et al.*, 2012a). The mounds of lead corrosion products at the lead:copper junction became more prominent as the study progressed (Fig. 4, 492 days compared to 281 days, fig. 7-Cartier *et al.*, 2012a). However, lead might also accumulate on other surfaces in the rigs (copper, plastic pipe downstream of the metal pipe), similar to prior observations made for lead accumulations in plumbing of homes with galvanized iron installed downstream of lead service lines (HDR Engineering, Inc., 2009). The potential contribution from each reservoir was examined sequentially.

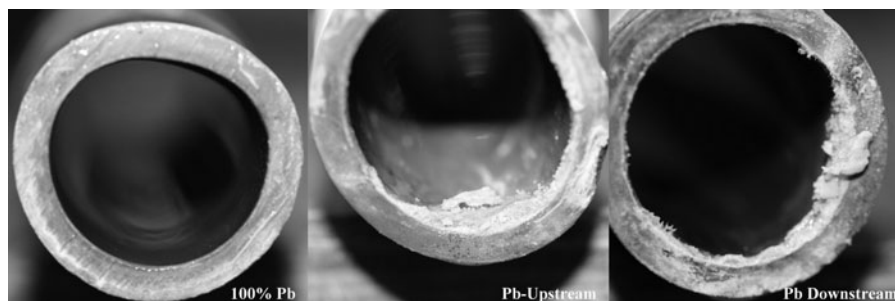


FIG. 4. Visual comparison of lead rust buildup on pipe coupons with and without galvanic corrosion after 17 months (492 days) of experiment. Large mounds of rust from partials were localized at the end closest to the Cu:Pb junction.

To assess the lead released from the previously formed mounded lead rust (Phase B-2), the 10 cm lead coupons immediately adjacent to the copper pipe were completely removed from the rigs without replacement. Thereafter, the Pb-D condition remained 73% higher than the 100% Pb condition (Fig. 3; $p > 0.05$), and the Pb-U condition was 125% higher than the 100% Pb condition ($p < 0.05$). After the existing copper and plastic piping downstream of the lead pipe was also replaced (Phase B-3), lead release was not statistically different among the three conditions (Fig. 3). Hence, removal of all three possible reservoirs of particulate lead (lead junction, copper pipe, plastic pipe) finally produced a situation in which conditions with 50% lead pipe were at least temporarily not worse than the 100% lead pipe. This restored the rig to relative performances noted in the earliest phases (months 1–3) of the study (Cartier *et al.*, 2012a), before reservoirs of lead particulates had accumulated throughout the plumbing system. In addition, replacing the old passivated copper pipe with a new copper pipe did not worsen the galvanic corrosion currents between lead and the copper pipe (comparing coupons removed to Cu replaced in Fig. 5).

Quantification of the mass of lead rust accumulation within the different parts of the plumbing system was revelatory and consistent with previously defined trends for lead release. Visually, a larger accumulation of lead rust was present on the lead surface nearest to the galvanic junction in the PLSLR conditions, whereas there was little or no accumulation on the lead pipe surface of the 100% Pb rigs (Fig. 4). When surface deposits (i.e., rusts) were later gently removed from all the removed pipe surfaces and weighed (Fig. 6), the mass of lead recovered in the conditions with galvanic corrosion was markedly greater than the 100% Pb replicates (Pb-U: 2.4 times higher, Pb-D: 3.5 times higher). This verifies the concerns originally expressed by Triantafyllidou and Edwards (2011) regarding galvanically induced accumulations of lead deposits and provides mechanistic support for the data indicating that pipes with galvanic Pb:Cu sections had a greater mass of lead mobilized at higher flow.

Galvanic corrosion current

In theory, as long as galvanic corrosion currents persist between lead and copper, problems with sporadic and elevated lead can also persist, and expected benefits of having a 50% lower lead surface area may never be realized. Galvanic currents remained elevated throughout the entire 48 months of the study, but eventually decreased somewhat relative to levels observed during the first year (Fig. 5). Removing the lead coupons or replacing the old copper with a new copper pipe did not result in an increase of galvanic current. From

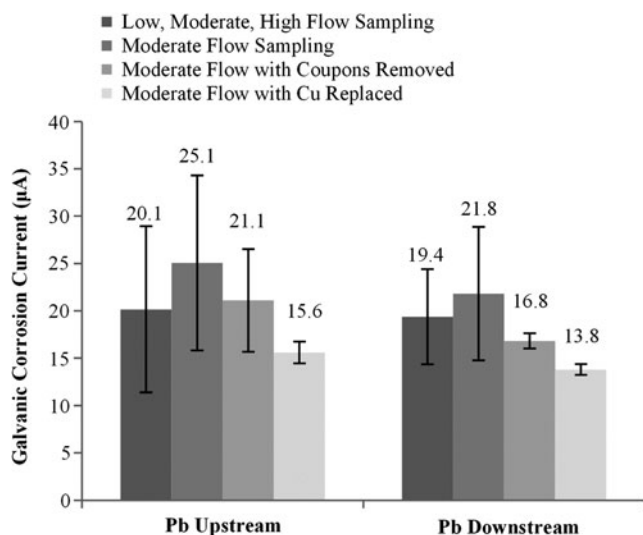


FIG. 5. Galvanic corrosion currents during Phase A (low, moderate, and high flow rate sampling per Cartier *et al.* 2012a) and Phase B (moderate flow rate sampling). Pb-U and Pb-D averages and 95% confidence intervals calculated from average current of individual triplicates from each condition.

initial experimental conditions to the final stage of Phase B (months 20–22), galvanic currents decreased 35–50% over the nearly 2-year period. Pb-U galvanic corrosion current decreased from 25 μA to 20 μA after the first year and decreased further to 16 μA after 22 months. The Pb-D galvanic current decreased from 29 μA to 19 μA in the first year and then to 14 μA after 22 months.

The expected weight loss of the short solid Pb pipe sections should be at least that calculated by Faraday's Law and the integrated sacrificial current (Supplementary Fig. S1). In this work, Faraday's Law overestimated actual weight loss by a factor of 4–5; however, it is clear that there was a greater weight loss with higher galvanic current (Fig. 6) compared to the condition with 100% lead and no galvanic current. In addition, galvanic current for Pb-U was generally higher than Pb-D ($p > 0.05$), but average weight loss was greatest for Pb-D (Fig. 6). The discrepancy between weight loss predicted by Fara-

day's law versus that observed might be due to deposition (plating) of copper ions onto lead pipe surfaces causing a weight gain (Hu *et al.*, 2012; Clark *et al.*, 2015), which is hinted at by an increase in the ratio of copper to lead in the scale for cases with partial replacements (Supplementary Fig. S2). Alternatively, the discrepancy could also be explained by difficulties in removing all the lead deposits/scales with the gentle abrasion used herein or possible presence of reactions other than lead oxidation contributing to galvanic corrosion currents.

Phase C: influence of connectors

Many different types of brass devices can be used to connect lead to a copper pipe, and it has recently been demonstrated that certain brass devices with crevices have high propensity to cause much worse elevated lead in water than direct connections between lead and copper pipe (Clark *et al.*, 2013). Others have speculated that connection of well passivated lead to copper or brass will not create significant problems with elevated lead in water (Reiber and Dufresne, 2006). However, recent work in Chicago demonstrated a significant problem with elevated lead if lead pipes were disturbed and a brass connector was used (Del Toral *et al.*, 2013). These issues were tested for the lead pipe, which had been prepassivated for 2 years in this research, with no freshly cut lead surfaces present.

Before the brass or copper connections were made, there was no statistical difference in the average concentration of total lead in water between each replicate during the initial 3-week baseline experiment without brass connectors (last 3 weeks of Phase B; 100% Pb = 4.1 $\mu\text{g/L}$, Pb-U = 5.8 $\mu\text{g/L}$, Pb-D = 5.0 $\mu\text{g/L}$). After installation of a commercially available brass device with crevices or a copper sleeve (to simulate brass without lead), lead release for the control conditions (existing plastic connector) decreased, but the lead concentrations for the copper sleeve increased significantly by 3.7 times (Fig. 7, $p < 0.05$). Lead release from the 100% Pb rig coupled with a brass compression connector increased 10-fold ($p < 0.05$), and the Pb-U and Pb-D rigs with brass connectors increased significantly by 6.2 times and 3 times ($p < 0.05$), respectively.

Total galvanic corrosion current sacrificing the lead pipe increased by 215 and 357% for the conditions connected with the copper sleeve or brass compression fitting, respectively

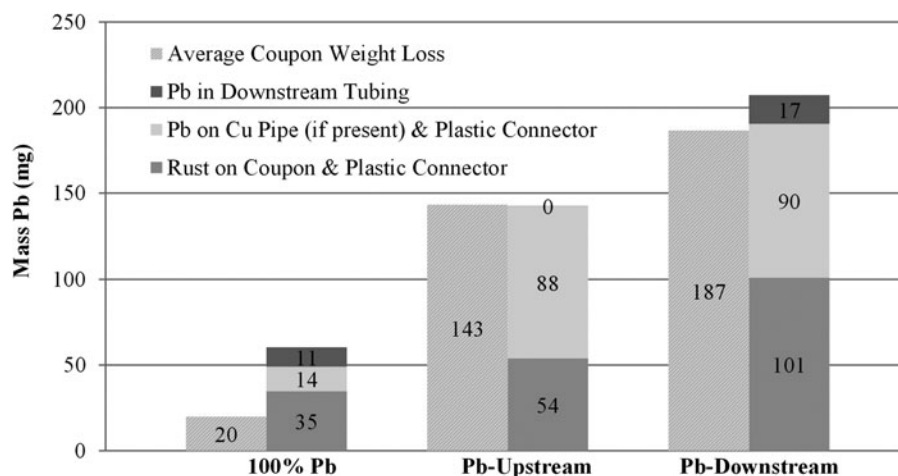


FIG. 6. Average weight loss from 10 cm lead sections (coupons), and average mass of lead recovered from individual reservoirs for each condition.

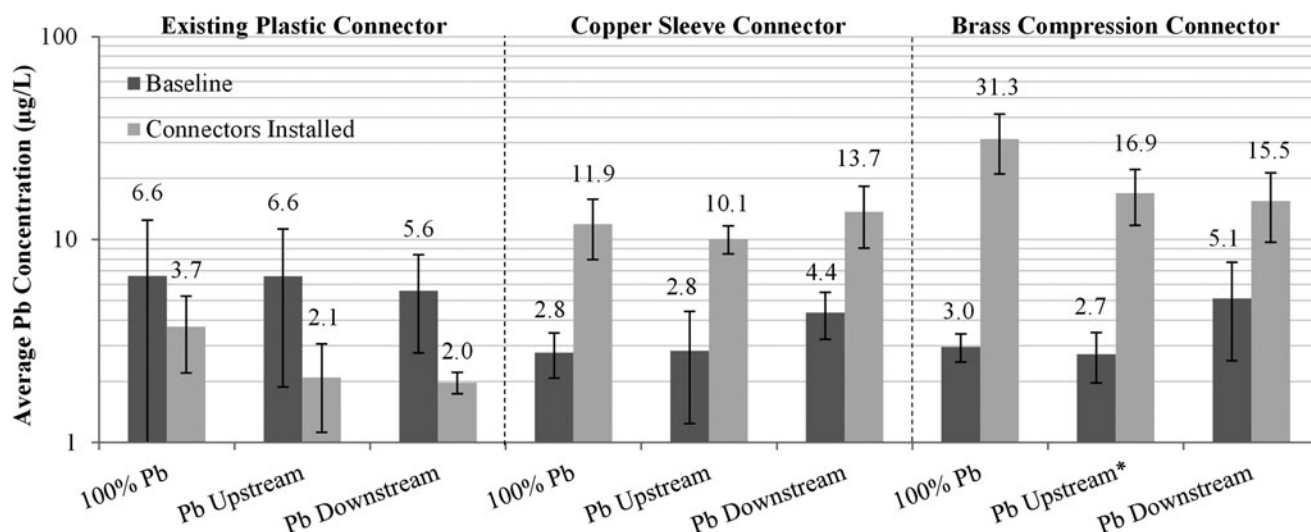


FIG. 7. Average Pb concentration in water during baseline period and after installing connectors. Error bars represent 95% confidence interval. (Baseline $N = 15$ per triplicate, Connectors installed $N = 55$ per triplicate) *High outlier excluded from Pb-U3 baseline.

(Fig. 8; note that galvanic current is zero for 100% Pb conditions when connected with the plastic tubing). The higher galvanic current compared to the prior situation where the lead pipe was connected to copper with a plastic connector and 0.3 cm distance is likely due to (1) changing the separation distance between lead and copper, (2) adding a third metal such as brass, and (3) creating crevices as described by Clark *et al.* (2013). Across all conditions, the sacrificial galvanic current of the lead pipe (through connection to brass or copper) strongly correlated to lead release (Fig. 9).

Contrary to expectations, the “worst case condition” of lead release and galvanic corrosion occurred when two lead pipes were connected through a brass connector (Fig. 7). While the magnitude of this effect was initially surprising, in retrospect it is explained by the fact that this condition creates two lead:copper alloy junctions and crevices. Lead release increased sharply (14–19 times higher) for the two

100% Pb conditions with brass or copper connectors immediately after connection (Fig. 10). Lead in water for the copper sleeve connection returned to levels slightly above the plastic connector at the end of the sampling period while lead release from the brass compression connector remained elevated (Fig. 10). This finding further supports that prior research results in experiments using lead pipe connected to the copper pipe are conservative in terms of galvanic impacts on lead release relative to “real world” connectors (Edwards, 2012). Clearly, well-passivated lead pipes connected to the copper pipe through commercial brass connectors can sometimes create severe lead contamination from galvanic corrosion, consistent with results of other recent work using a harvested lead pipe that had been exposed for decades in distribution systems or field studies (Cartier *et al.*, 2013; Wang *et al.*, 2012, 2013; Del Toral *et al.*, 2013; Welter *et al.*, 2013).

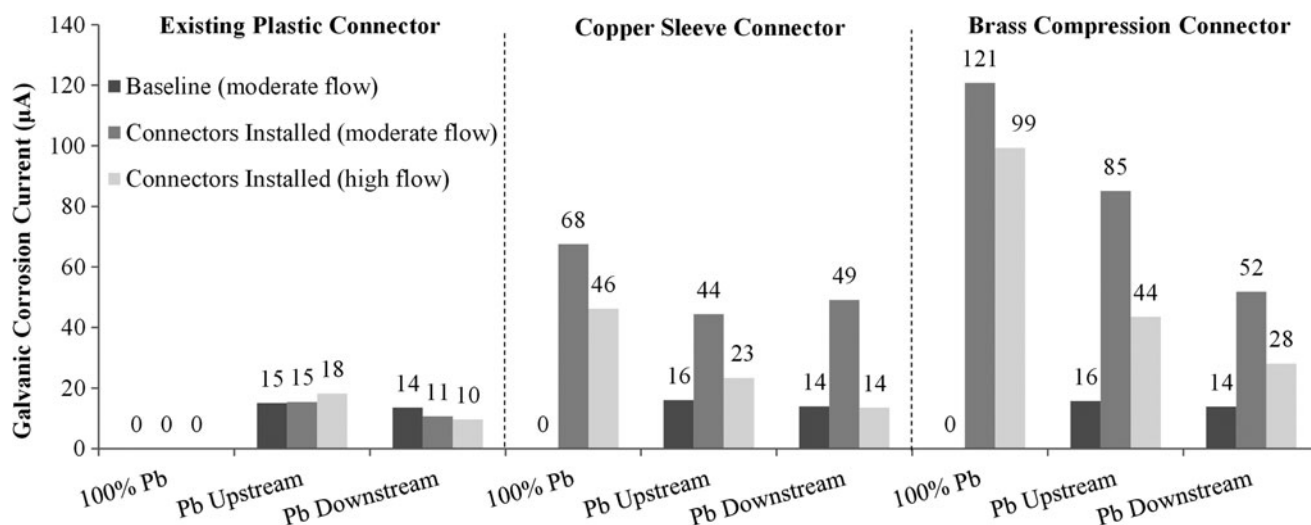


FIG. 8. Galvanic corrosion currents before and after installing connectors and Phase E.

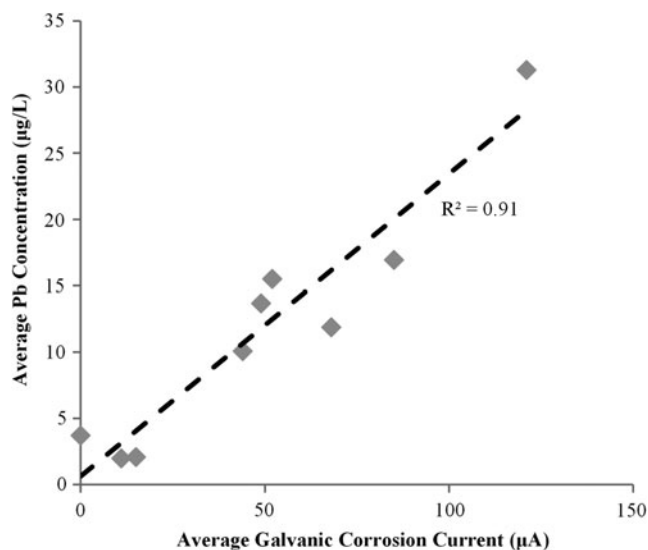


FIG. 9. Average Pb in water versus average galvanic current with connectors installed.

Phase D: prolonged stagnation event

During a 1-month stagnation event, the mass of lead released to water from either a 100% lead pipe or simulated direct Pb:Cu pipe connections was 3000–4000 ppb in a 2 L sample (Supplementary Fig. S3). The use of copper sleeves and brass connectors had between three and seven times higher lead release. Over the month, long stagnation event galvanic corrosion currents dropped only 42–82% compared to that measured after an 8 h stagnation time. Clearly, prolonged stagnation raises health concerns in systems with partial replacements.

Phase E: detecting problems with particulate lead using filters and synthesis of sampling methods

Filters were installed downstream of the various conditions and allowed to collect lead for two periods of 38 days while operating the rig at a relatively high flow rate (18–20 LPM) for a short duration of time (separated by long periods of stagnation). Additional 2 L first flush samples were collected without the filters at the end of both time periods. For both

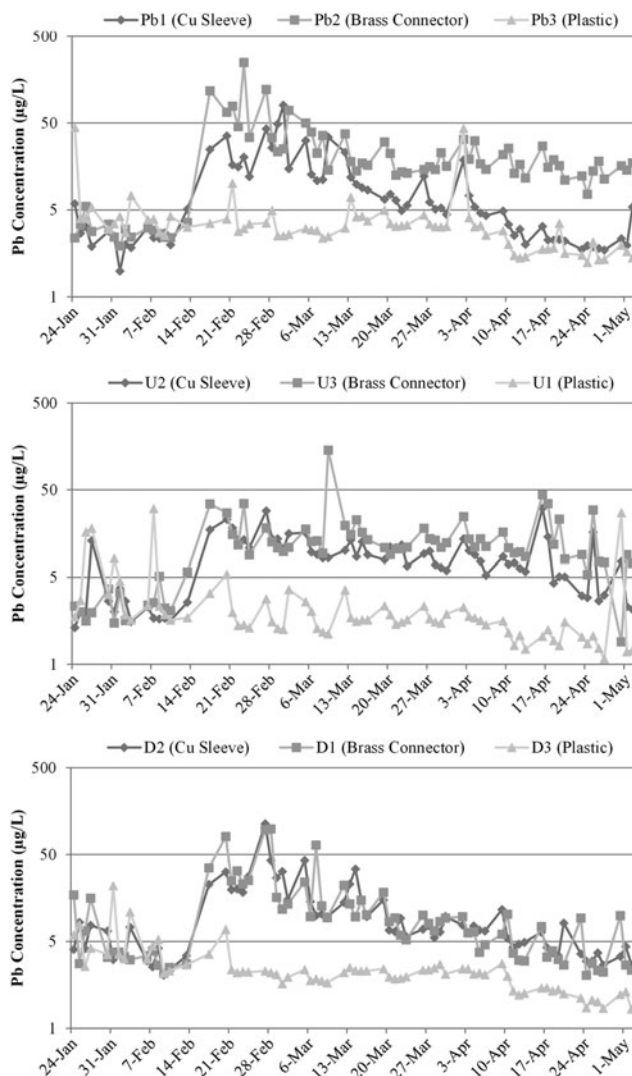


FIG. 10. Lead in water for 100% Pb conditions with connectors installed for duration of Phase C ($N=70$ for each condition).

TABLE 2. SAMPLING METHODS FOR ONCE THROUGH FLOW EVENTS

Method	Description	Volume sampled	Pros/Cons	Likelihood to detect soluble and particulate lead
First draw & profiling	Collect first amount of water during flushing and subsequent samples	1–2 L or extent of profile	Quantifies all lead in samples collected, very sensitive to flow rate, and likelihood of missing spikes	Detects all Pb in water (particulate and soluble) collected but can miss sporadic spikes
Grab samples	Collect small samples periodically	10 mL	Provides profile of soluble lead, does not sample large portion of flow event, and sensitive to flow	Detects soluble lead, may miss particulate spikes
Proportional sampler	Splits flow such that a portion is collected for analysis	A fraction of daily flow	Conventional designs “miss” particulates. Other designs could concentrate particulates.	Depends on design
Bins to collect all water from pipe rigs	Collects all water during a daily flush	Entire daily	Collects all water and quantifies suspended fraction, including spikes but not practical	Confined to laboratory. Likely to detect all Pb (particulate and soluble)
Whole-house filters	Filters lead from all water for an extended period of time	Entire	Samples large volumes with minimal effort. Misses soluble and small colloidal lead.	Potentially quantifies a large fraction of particulates Pb > 1 μ m

sampling methods, Pb in water increased with increasing galvanic corrosion current (Supplementary Fig. S4). Pb recovered using the filters was better correlated with galvanic current than the first 2 L samples ($R^2=0.41$ and $R^2=0.23$, respectively). One time sampling events, such as the first 2 L samples, are useful but imperfect since high variability is expected from semirandom release of particulates during the high flow events. However, the representative sampling from the service line correlated strongly ($R^2=0.85$) with the cumulative Pb captured by filters over the 1-month experiment with only one exception ($R^2=0.72$, including exceptional point).

Filters were installed downstream of the various conditions and allowed to collect lead for 37 days while operating the rig at a relatively high flow rate (18–20 LPM) for short periods of time (separated by long periods of stagnation). The daily mass collected on the filters correlated strongly with prior data in which all the flow was collected in bins the prior 5 weeks and at a lower flow rate. In summary, whole-house filters have promise in detecting problems arising from particulate lead release from service lines in real systems. Galvanic currents decreased slightly ($\sim 20\%$) during this phase of testing, presumably due to passivation of the lead pipe from the initial increased galvanic corrosion rates (Supplementary Fig. S4).

During months 35–48, rigs were continuously sampled using the filters as described previously, and there was no difference between configurations that were partially replaced compared to that of the 100% lead pipe section. Thus, in this water, 4 years of exposure and removal of lead reservoirs were required, before a lead release from a partially replaced lead pipe was not worse than a full-lead pipe. The Blacksburg water did contain chloramine, which has been reported to cause more persistent and serious elevations of lead with galvanic connections than free chlorine (Arnold and Edwards, 2012; Zhou *et al.*, 2015).

The array of sampling methods used in this work to detect health risks to consumers arising from sporadic particulate lead release from service lines each had potential benefits and drawbacks (Table 2). Profiling at high or low flow is a useful tool to quantify lead release, but large numbers of samples must be collected at a range of flow rates to quantify the nature of particulate lead release, and it is always possible that a propensity for very large spikes in water lead might be missed (Cartier *et al.*, 2012a; Clark *et al.*, 2012). Collecting a few grab samples (e.g., second draw) is even more prone to missing semirandom release of particulate lead. Proportional samplers can be used to collect fractions of total daily flow (van den Hoven and Slaats, 2006); however, our experiences demonstrate that these conventional designs may systematically “miss” larger lead particulates because the momentum of particles carries them past small side stream samplers and causes the lead to be “missed.” Conversely, other designs may concentrate lead particulates in water samples.

The least ambiguous approach used in this research, through collection and sampling all the water passing through the service line over a period of weeks, is obviously not practical in consumer homes. However, if the whole-house filters captured a large fraction of the particulate lead (the dominant fraction of lead released), filters can detect problems from service lines in a single sample (the filter), improve the quality of consumed water in the home through removal of some lead particulates, eliminate the need for required

stagnation events before collection, create a composite sample (for large captured particulates) at representative flow rates, and use patterns without interfering with the consumers' daily routine. These advantages of using filters in field work on partial lead service lines may outweigh the disadvantages of missing soluble lead.

Phase F: exploring concerns related to unlined iron main connections to service line

If there was no copper pipe installed between the iron main and lead pipe, the iron cathodically protected the lead pipe with an initial current of 92 μA . However, in all cases when the length of copper installed between the two pipes was >2.5 cm, the lead was sacrificed by the connection to copper. Thus, in a conventional partial replacement, where copper pipe lengths of >2.5 cm are used, a connection of the service line to unlined iron is not expected to influence galvanic corrosion problems.

This was further confirmed by temporarily connecting a 3 m galvanized iron pipe section to the individual pipe rigs in this work. The iron pipe section did sacrificially reduce lead corrosion rates (measured by galvanic current, 12–15 μA) in cases where the iron was directly connected to lead, as occurs in the field before partial replacement. However, for the case in which 1.5 m of copper was placed between the lead and iron as per a traditional partial replacement, the galvanic current between copper:lead was not affected by the upstream connection between copper:iron. In summary, this short-term experiment suggests that there could be some very small benefits to lead release if a lead pipe is connected to an unlined iron main, but sacrificial currents from the unlined iron main will not reduce galvanic impacts of copper on lead pipe corrosion in a traditional partial service line replacement. Clearly, there are also other detriments from lead mobilization by iron particulates through a sorption mechanism as illustrated elsewhere (Hulsmann, 1990; Lytle *et al.*, 1993; Triantafyllidou *et al.*, 2007; Deshommes *et al.*, 2010; Cartier *et al.*, 2012b; Schock *et al.*, 2014; Masters *et al.*, 2015).

Conclusions

- After 14 months of exposure, starting with new lead pipe and no scale and without any disturbances from cutting pipes, configurations representative of partial pipe replacements continued to release much more lead than the full-lead service pipe at moderate and high flow rates (as observed in Cartier *et al.*, 2012a). Under low flow rate sampling, lead release is comparable between the three conditions tested. However, at moderate and high flow rates, galvanic corrosion increased lead in water for Pb-D by a factor of 26 compared to 100% Pb at highest flow.
- When a consistent moderate flow rate was used, galvanic corrosion increased lead release from a partial service line configuration, and these effects were significant for a period of at least 48 months. By the end of this experiment, the particulate lead release from the partial pipe (Pb-U and Pb-D) was comparable to the full-Pb pipe, even though the surface of lead in contact with water is 50% lower.

- Galvanic current contributed to reservoirs of lead rust/scale accumulation in the downstream plumbing that created spikes of particulate lead. Galvanic corrosion currents had decreased 35–50% from initial rates after 2 years of operation, but still persisted at rates greater than 10 μA .
- The addition of brass and copper connectors to the passivated lead pipe dramatically increased lead corrosion and subsequent lead in water, apparently by the formation of crevices and galvanic corrosion. Lead leaching was markedly higher in conditions with brass compression fittings compared to plastic connectors with a dielectric spacer. Negative effects from galvanic corrosion and brass connectors, during exceptionally long stagnation events, created serious issues with elevated lead. Galvanic corrosion from newly installed brass connectors coupled to the lead pipe increased total lead mass release 5–7 times.
- Cathodic protection from iron mains coupled to lead does not appear to be capable of stopping problems arising from galvanic copper:lead connections.
- Collecting all water from the service pipes identified some important trends in total lead release that first draw and grab samples missed. While further research is required, whole-house filters can be utilized as an experimental collection method, capable of quantifying a fraction of the total particulate lead release while also partly mitigating lead in water risks for consumers.

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