

Effect of connection methods on lead release from galvanic corrosion

YIN WANG,¹ VRAJESH MEHTA,² GREGORY J. WELTER,³ AND DANIEL E. GIAMMAR²

¹Department of Civil and Environmental Engineering, University of Illinois, Urbana-Champaign

²Department of Energy, Environmental, and Chemical Engineering, Washington University in St. Louis, St. Louis, Mo.

³O'Brien and Gere, Bowie, Md.

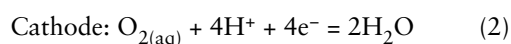
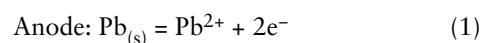
Laboratory experiments were performed to evaluate the effect of galvanic corrosion on lead release following simulated partial lead service line replacements. Experiments were conducted in a recirculating flow mode with intermittent stagnation periods using aged lead pipes harvested from Washington, D.C. Lead pipe and copper tubing were connected by brass, brass dielectric, and plastic couplings; additional experiments were performed with plastic couplings and an external wire to connect the lead

and copper. Lead release increased in the order of systems connected with plastic, plastic with external wires, brass dielectric, and brass couplings. The trends are consistent with galvanic reactions between lead and copper and between lead and brass. For systems with galvanic corrosion, the increased lead release was primarily in a particulate form. Increasing the chloride-to-sulfate mass ratio from 0.7 to 7 did not increase the dissolved or total lead release.

Keywords: *drinking water, galvanic corrosion, lead, partial lead service line replacement*

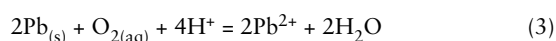
Lead service line replacement (LSLR) may be required when the tap water lead concentrations exceed the action level, which has been set as 15 µg/L for a 1-L first-draw sample of tap water, and cannot be lowered by corrosion control methods (USEPA, 1991). LSLR may also be necessitated as incidental to programmed water infrastructure renewal projects or unplanned emergency repair operations. In many instances, only partial lead service line replacements (PLSLRs) are conducted because utilities can typically replace only the publicly owned portion of the service line; the private side of the service line is not replaced (Swertfeger et al, 2006). Generally, PLSLRs are implemented by replacing the old lead pipe with new copper tubing and connecting the remaining lead pipe with a brass fitting.

In a PLSLR, a galvanic couple can be created if the lead pipe and copper tubing are electrically connected either by direct contact or by a metal coupling. Lead serves as the anode and copper serves as the cathode as shown in Eqs 1 and 2:



in which $\text{Pb}_{(s)}$ is solid elemental lead, $\text{O}_{2(aq)}$ is dissolved oxygen, and e^{-} is an electron.

This can result in an acceleration of the corrosion and release of lead, and the net reaction is shown in Eq 3:



The disinfectants in drinking water, including free chlorine and chloramines, may also serve as electron acceptors at the cathode. Laboratory examination of field-exhumed lead/copper connections has indicated significant corrosion on the lead pipe surface in the immediate vicinity of the connection, which was probably the result of galvanic corrosion (DeSantis et al, 2009). Several recent studies found that lead concentrations did not decrease soon after PLSLRs; instead, higher lead concentrations were observed for some utilities even several months after PLSLRs (Muylwyk et al, 2011; USEPA, 2011; Sandvig et al, 2009; Swertfeger et al, 2006). Muylwyk et al (2011) measured lead concentrations in tap water that persisted for at least 18 months following some PLSLRs in Guelph, Ont. Swertfeger et al (2006) found that lead concentrations were only slightly lower for connections with partial replacement than for connections that retained the lead service lines even one year after replacement. Galvanic corrosion has been proposed as a mechanism that may result in the long-term persistence of lead concentrations (Triantafyllidou & Edwards, 2011; USEPA, 2011; Edwards & Triantafyllidou, 2007).

The effect of galvanic corrosion on lead release following PLSLRs can be affected by the methods used to connect the lead pipes and copper tubing. Connecting lead pipes and copper tubing joined by plastic tubing but with external electrical wires resulted in measurable galvanic currents and increases of 1.5–75 times the amount of lead release (Cartier et al, 2012; Triantafyllidou & Edwards, 2011). Although this connection method allows direct measurement of galvanic currents, actual connections in PLSLRs use couplings such as brass compression fittings, brass dielectric compression fittings, and brass valves. Recent

studies have found that lead release was significantly increased by connecting lead pipes and copper tubing with brass compression fittings—with enhancement primarily attributed to galvanic corrosion (Cartier, 2012; Wang et al, 2012). Clark et al (2011) measured lead release after connecting lead pipes and copper tubing using different methods and found that the lead concentrations increased in the order of systems connected by direct end-to-end connection, brass compression fittings, and brass valves.

Water chemistry can affect galvanic corrosion. Several studies have reported that increasing the chloride-to-sulfate mass ratio (CSMR) can accelerate lead release because of galvanic corrosion (Triantafyllidou & Edwards, 2011; Nguyen et al, 2010a; Edwards & Triantafyllidou, 2007; Gregory, 1990). Triantafyllidou and Edwards (2011) quantified lead release in externally wired pipe assemblies and found that 3–12 times more lead was released from a system with a CSMR of 16 than from one with a CSMR of 0.2. Cartier (2012) observed that lowering the CSMR from 0.9 to 0.3 significantly decreased lead release from lead pipes connected to copper tubing with brass fittings. Decreasing the pH level (Gregory, 1990) or increasing the nitrate concentration (Nguyen et al, 2011a) was also found to increase lead release resulting from galvanic corrosion. In contrast, increasing bicarbonate, zinc, or silicate concentrations could inhibit galvanic corrosion (Nguyen et al, 2011b; Gregory, 1990). Orthophosphate is often added to inhibit lead corrosion (McNeill & Edwards, 2002), but it was not found to decrease lead release from galvanic corrosion (Cartier, 2012; Arnold et al, 2011).

The corrosion products that constitute pipe scales can potentially passivate the pipe surface and have an effect on galvanic corrosion. By measuring the surface potential, Reiber and Dufresne (2006) found that although galvanic corrosion was significant on new lead pipe surfaces, it was minimal and highly transient on aged lead pipes. For a system in which new lead pipe and copper tubing were galvanically connected, Arnold et al (2011) observed that Pb(IV) oxides formed on lead pipes after approximately two months of continuous recirculation with chlorinated water and that the presence of the Pb(IV) oxides protected the lead pipes from galvanic corrosion. Although corrosion products may passivate the pipe surface, they may also

break off and be released as particulate lead at high flow rates (Cartier et al, 2012). Although new or cleaned lead pipes were commonly used in previous work to simulate PLSLRs (Boyd et al, 2012; Cartier et al, 2012; Clark et al, 2011; Triantafyllidou & Edwards, 2011), only a few studies have used lead pipes that were harvested from actual distribution systems (Cartier, 2012; Wang et al, 2012).

The extent and duration of galvanic corrosion can be affected by the flow regime. Triantafyllidou and Edwards (2011) observed that lead concentrations were significantly increased in stagnant water as a result of galvanic corrosion and suggested that stagnation represented a worst-case scenario for lead release. In determining lead release from galvanically connected lead–copper pipe assemblies, Arnold et al (2011) found that greater amounts of lead were released in intermittent flow than during continuous flow. Flow rates also affected lead release from galvanic corrosion with medium (8 L/min) and high (32 L/min) flow rates, resulting in much higher lead release than at a low flow rate (1.3 L/min; Cartier et al, 2012).

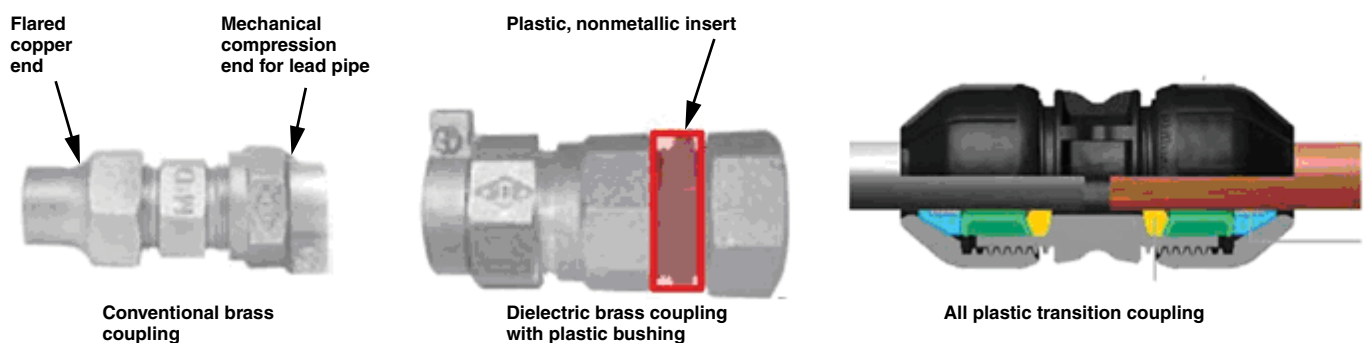
The primary objective of the current study was to determine the effect of connection methods on lead release resulting from galvanic corrosion. A secondary objective was to evaluate the effect of CSMR on lead release. To best represent actual field PLSLRs, aged lead pipes were connected to new copper tubing with commercially available couplings.

MATERIALS AND METHODS

Materials. Laboratory experiments were conducted with aged lead pipes that were harvested from the Washington, D.C., distribution system after being used for more than 100 years. Pipes had inner diameters of 0.75 in. and outer diameters of 1.18–1.2 in. Pipes were cut into 24-in. sections for use in experiments; the remaining portions were used for scale characterization.

Type K copper tubing (1-in. inner diameter, 1.125-in. outer diameter) was cut into 24-in. sections. Polyvinyl chloride (PVC) pipe sections with inner diameters of 1 in. were cut into 24-in. sections and used in lead-free control experiments. Couplings used in the experiments were brass compression fittings,¹ brass dielectric fittings,¹ and all-plastic polypropylene fittings² (Figure 1). All solu-

FIGURE 1 Three types of couplings used to connect the lead pipes and copper tubing



tions were prepared using ultrapure water (resistivity > 18.2 M Ω -cm). Reagent-grade calcium chloride, magnesium chloride, magnesium sulfate, disodium hydrogen phosphate, sodium fluoride, sodium chloride, sodium bicarbonate, sodium hypochlorite, sodium hydroxide, and nitric acid (HNO₃) were purchased and used for preparing synthetic tap water.

Pipe conditioning. Six harvested lead pipes were conditioned with water prepared to simulate the chemistry of the Washington, D.C., distribution system (Table 1) before being connected to copper tubing. The pipes were first conditioned in a static “dump-and-fill” mode for at least eight weeks. This mode was operated following the procedures described elsewhere (Wang et al, 2012). Samples were collected periodically and preserved for dissolved (filtered with 0.45- μ m polyethersulfone syringe filters) and total lead analysis. After being conditioned in the static mode, the pipes were conditioned in a recirculating flow mode for four weeks. A similar protocol was then used when the lead pipes were connected to the copper tubing. Briefly, the system was operated on a daily cycle during weekdays. Each Monday morning, freshly made synthetic Washington, D.C., water was fed into the pipes from a 10-L polyethylene reservoir. The weekday cycle for Monday–Thursday started with a 6-h stagnation period, then 15 min of flow, a 17.25-h stagnation period, and finally 30 min of flow. On Fridays, the 17.25-h stagnation period was replaced with a 65.25-h stagnation period (i.e., over the weekend)—hereafter referred to as the 65-h stagnation period—as the last phase of the weekly cycle. The reservoirs were then replaced with clean ones with freshly made solutions to begin the next week’s cycle. A peristaltic pump³ provided a flow rate of 5.2 L/min, which corresponded to a water velocity of 1 fps in the lead pipes. A 50-mL water sample was collected each Monday, Wednesday, and Friday before the 6-h stagnation period and on Friday before the 65-h stagnation period. Following the corresponding stagnation periods, 50-mL poststagnation samples were collected and mixed evenly. Ten millilitres were used for total lead analysis, 10 mL for



During this research four couplings were used to connect lead pipe and copper tubing.

dissolved lead analysis, and the remaining 30 mL for pH and combined chlorine measurement. The orthophosphate concentration was determined for selected samples. For pipe conditioning and experiments with couplings (discussed in the subsequent section), the pH and orthophosphate concentrations in the reservoir were stable over the one-week cycle. The concentrations of combined chlorine decreased over time, so the chlorine concentrations were regularly adjusted back to the target value.

Experiments with couplings. After being conditioned in the recirculating flow mode for four weeks, the lead pipes were connected to new copper tubing using the different couplings and placed in the recirculation system (Figure 2) and operated for at least six weeks. Flexible plastic tubing was used to connect the sampling valves, flow meters, and the recirculation reservoirs. The overall timeline of all experiments conducted is shown in Table 2. Two pipes were connected to copper tubing using the plastic couplings (plastic 1 and plastic 2), two using the brass couplings (brass 1 and brass 2), and two using the brass-dielectric couplings (brass-die 1 and brass-die 2). The numbers 1 and 2 refer to duplicates. The electrical resistance between the lead and copper was measured by a handheld multimeter. For the plastic-coupled and brass dielectric-coupled systems, the resistance was high (> 1 M Ω) between the lead pipes and copper tubing, indicating no electrical connection; little resistance (< 4 Ω) was observed for the brass-coupled pipe assemblies. The brass dielectric couplings still have the potential for dissimilar metal couples of copper/brass and brass/lead. The internal volumes of the lead pipe and copper tubing sections were 180 and 310 mL, respectively.

After six weeks of the experiment, for pipes with plastic and brass couplings, the CSMR was increased from its original value of 0.7 to an elevated value of 7.0 for four weeks (Table 1). The value of 0.7 is near the boundary where CSMR was found in previous research to result in enhanced galvanic corrosion (Nguyen et al, 2011c).

The opportunity for galvanic corrosion to occur was also introduced for the lead–copper assemblies with the plastic couplings by electrically connecting them with an external wire as was done in previous work (Triantafyllidou & Edwards, 2011). Unlike connections made using brass or brass dielectric couplings, this connection allows for the measurement of galvanic current

TABLE 1 Water composition of synthetic Washington, D.C., water with original and elevated CSMR

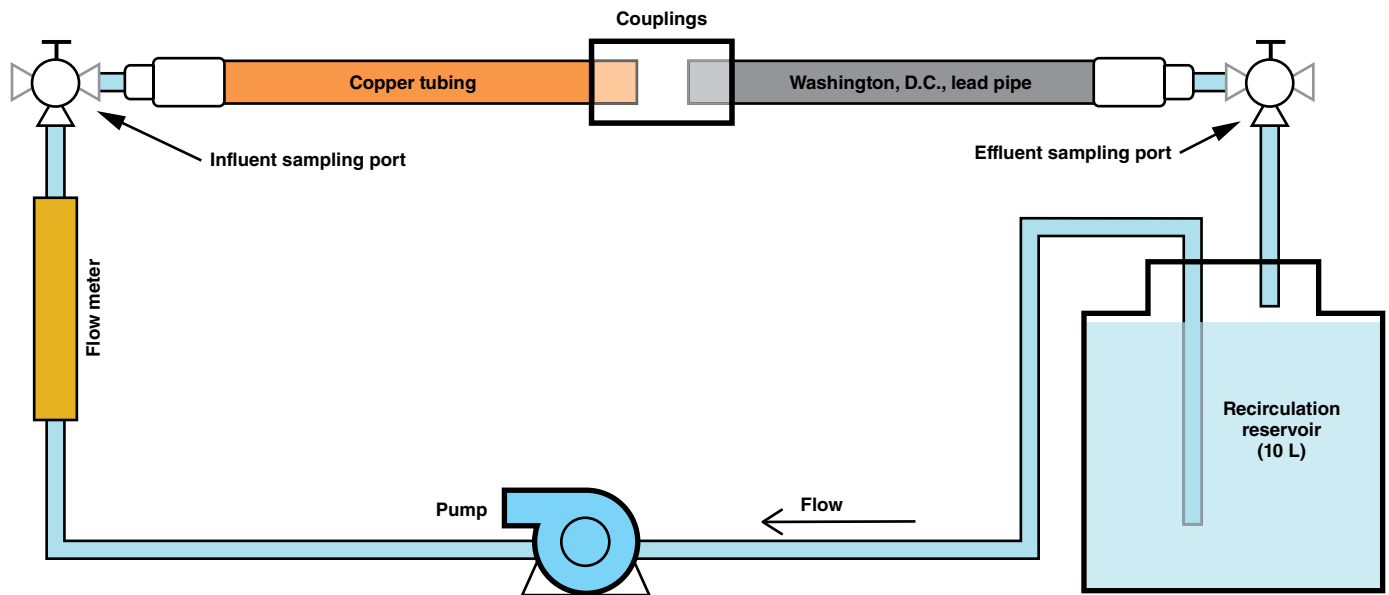
Water Composition Parameter*	Range for Washington, D.C., Water†	Synthetic Water (Original CSMR)	Synthetic Water (Elevated CSMR)
pH	7.7–7.8	7.7	7.7
Combined chlorine—mg/L as Cl ₂	3.6–3.8	3.7	3.7
Total alkalinity—mg/L as CaCO ₃	49–99	76	76
TDS—mg/L	126–261	310	310
Total hardness—mg/L as CaCO ₃	114–155	128	128
Chloride—mg/L	30–56	34	77
Sulfate—mg/L	40–71	50	11
Orthophosphate—mg/L as PO ₄	2.4–2.8	2.5	2.5

CaCO₃—calcium carbonate, Cl₂—chlorine, CSMR—chloride-to-sulfate mass ratio, PO₄—phosphate, TDS—total dissolved solids

*No natural organic matter was added in the synthetic Washington, D.C., water.

†Ranges are for the Dalecarlia Water Treatment Plant, Washington, D.C.

FIGURE 2 Schematic view of the recirculating flow system with lead pipes and copper tubing connected with brass, brass dielectric, and plastic couplings



Adapted from Wang et al, 2012

The pipe assembly was inclined at an approximate 15° angle, with the lead end above the copper end.

by placing a multimeter in the externally wired connection. These systems are referred to as plastic-ex 1 and plastic-ex 2. This connection was made for the same pipes that had been studied with the plastic couplings (without the externally wired connection) at first with the original CSMR and then at the elevated value. Before the connection with the external wires was made, the lead-copper assemblies were operated using the water with the original CSMR for four weeks. Then, after the pipe assemblies were operated with the external connection for six weeks, the wires were disconnected, and lead release was monitored for one more week.

To test for lead release from the couplings themselves, control experiments were performed using PVC pipes connected to copper tubing. Experiments were operated for four weeks using the same weekly cycle used for the experiments with lead pipes and copper tubing.

In both the experimental lead-copper and control PVC-copper experiments, influent samples of 50 mL were collected at a flow rate of 1 L/min on each Monday and Thursday before the 6-h stagnation period and on Friday before the 65-h stagnation period. Poststagnation samples of approximately 500 mL were collected directly from the pipe assemblies at a flow rate of 1 L/min following the Monday and Thursday 6-h stagnation periods and the weekend 65-h stagnation period. A profile sampling approach was performed once for each lead-copper pipe assembly using the synthetic Washington, D.C., water with the original CSMR. Seven consecutive 70-mL samples were collected for each lead-copper pipe assembly at the effluent sampling port

(Figure 1) near the lead end of the assembly after 6 h of stagnation in an effort to identify possible localization of lead release along the pipe sections.

After the old reservoir was replaced with a new one each Monday, the solution in the old reservoir was acidified to pH < 2 by the addition of 10 mL concentrated HNO₃ and was allowed to sit at this low pH for at least 24 h before a 10-mL sample was collected for total lead and copper analysis. This acidification was intended to mobilize and dissolve particulate or adsorbed lead that may have accumulated in the reservoirs during the previous week.

Characterization of lead pipe scales. X-ray diffraction (XRD) was performed to identify the crystalline phases of the corrosion products from both the as-received pipes and pipes after the experiments with couplings. For XRD, 6-in. portions of the pipes were cut longitudinally, and scales were scraped from the inner surfaces of the pipes using a metal spatula. For imaging, a 1-in. section of the as-received pipe was filled with epoxy. Then the cross-section was cut, and the thickness of the pipe scales was observed using scanning electron microscopy (Figure 3). The elemental composition of the scales from the as-received pipes was quantified by digesting 0.01 g of the scale in a mixture of 2.5 mL concentrated hydrochloric acid and 10 mL concentrated HNO₃ that was heated to 100°C for 4 h and then processing the digestate for analysis.

Analysis methods. Concentrations of lead and copper were measured by inductively coupled plasma mass spectroscopy.⁴ The samples were acidified to 2% HNO₃ and allowed to sit for at least

TABLE 2 Timeline of the experiments conducted in the recirculating flow mode

Connection Methods*	Experimental Dates					
	Aug. 29–Sept. 26, 2011 (4 weeks)	Sept. 26–Nov. 7, 2011 (6 weeks)	Nov. 7–Dec. 5, 2011 (4 weeks)	Dec. 5, 2011–Jan. 2, 2012 (4 weeks)	Jan. 2–Feb. 13, 2012 (6 weeks)	Feb. 13–20, 2012 (1 week)
Plastic with and without external wires	Conditioning of lead pipe	No wire CSMR = 0.7 (plastic 1 and 2)	No wire CSMR = 7 (plastic 1 and 2)	No wire CSMR = 0.7 (plastic 1 and 2)	With wire CSMR = 0.7 (plastic-ex 1 and 2)	No wire CSMR = 0.7 (plastic 1 and 2)
Brass	Conditioning of lead pipe	CSMR = 0.7 (brass 1 and 2)	CSMR = 7 (brass 1 and 2)			
Brass dielectric				Conditioning of lead pipe	CSMR = 0.7 (brass-die 1 and 2)	

Brass—two pipes connected to copper tubing with brass couplings, brass-die—two pipes connected to copper tubing with brass-dielectric couplings, CSMR—chloride-to-sulfate mass ratio, plastic—two pipes connected to copper tubing with plastic couplings, plastic-ex—two pipes connected to copper tubing with plastic couplings and electrically connected with external wire

*Experiments were conducted in duplicate.

24 h before analysis. XRD was conducted on a diffractometer⁵ using X-rays with a wavelength of 1.54 Å. Electron microscopy images were acquired on a field emission scanning electron microscope.⁶ Total chlorine and orthophosphate concentrations were determined by the standard *N,N*-diethyl-*p*-phenylenediamine and ascorbic acid colorimetric methods (4500-Cl G and 4500-P E; *Standard Methods*, 1998, respectively) with a spectrophotometer.⁷ Solution pH was measured with a pH electrode and pH meter.

RESULTS AND DISCUSSION

Pipe-scale characterization. Using XRD, the Pb(II) phosphate hydroxylpyromorphite [i.e., $\text{Pb}_5(\text{PO}_4)_3\text{OH}$] was observed as the dominant solid phase in the scales of both the as-received pipes and pipes after the experiments. $\text{Pb}_5(\text{PO}_4)_3\text{OH}$ has been observed in distribution systems using phosphate as corrosion inhibitors (Schock et al, 2005a). Orthophosphate has been added since 2005 in Washington, D.C., water treatment plants to mitigate lead release to tap water. In addition, the Pb(IV) oxides scrutinyite and plattnerite were both observed. Lead oxide has been found in distribution systems in which free chlorine was consistently used as the disinfectant (Kim & Herrera, 2010; Schock et al, 2005b). Washington, D.C., used free chlorine as the disinfectant persistently before 2001 and now still operates with free chlorine for about one month per year to limit the growth of biofilms.

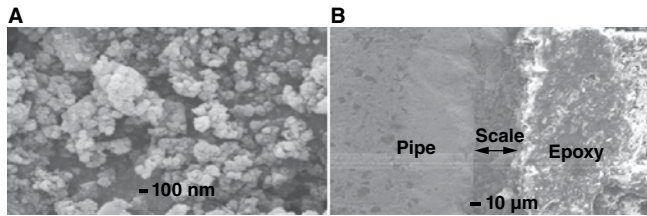
The dominant solids present in the pipe scales were aggregates of small roughly spherical particles with primary sizes < 100 nm (Figure 3, part A). The shape observed in the current work was consistent with that of Pb(IV) oxide or $\text{Pb}_5(\text{PO}_4)_3\text{OH}$, both of which have been previously observed as small, roughly spherical particles or aggregates (Wang et al, 2010; Giammar et al, 2008). The thickness of the scale was approximately 40 μm . Xie and Giammar (2011) conditioned new lead pipes for eight months in the laboratory and observed development of a pipe scale with a similar thickness of 24 μm , whereas a 300- μm -thick pipe scale was observed in a pipe harvested from a water utility in Illinois (Nadagouda et al, 2011).

Completely digesting the scales determined that trace amounts of aluminum (15 mg/g), manganese (Mn; 5 mg/g), vanadium (V; 4 mg/g), and copper (6 mg/g) were present. Mn and V have been observed in the scales of the lead pipes in the distribution system of Providence, R.I. (Wang et al, 2012). Gerke et al (2009) analyzed the scales of 15 lead pipes from eight distribution systems and found that most scales contained V with a concentration of 2–8 mg/g. They suggested that V can be present in the form of vanadinite. Because of the similarity of vanadate (VO_4^{3-}) to phosphate (PO_4^{3-}), VO_4^{3-} may even partially substitute for PO_4^{3-} in the lead phosphate solids in the pipe scales.

Lead release during pipe conditioning. In the recirculating flow mode, the average dissolved lead concentrations following 6-h stagnation periods were quite similar for the six pipes, with the average value around 15 $\mu\text{g/L}$ (Figure 4, part A). Increasing the stagnation time to 65 h did not affect the dissolved lead concentrations except for one pipe, suggesting that the water in the pipes may have reached equilibrium with respect to the corrosion products within 6 h. On the basis of available thermodynamic data (Benjamin, 2002; Schecher & McAvoy, 1998), the dissolved lead concentration in equilibrium with $\text{Pb}_5(\text{PO}_4)_3\text{OH}$ in the Washington, D.C., water chemistry is estimated to be 9 $\mu\text{g/L}$, which was close to the measured value. The total lead concentrations were only slightly higher than the dissolved lead concentrations (Figure 4, part B), indicating that most of the lead released during the conditioning period was dissolved and not particulate at the sampling flow rate of 1 L/min. Flow rate can strongly affect particulate lead release, and it is anticipated that greater amounts of particulate lead would be released at a higher flow rate (Cartier et al, 2012).

Lead and copper release from the coupling experiments. After connecting lead pipes to copper tubing using the different types of test couplings, dissolved and total lead concentrations were monitored regularly each week. The lead concentrations after stagnation were lowest for the plastic-coupled systems, which had no galvanic connection of the copper and lead, and highest for

FIGURE 3 Electron micrographs of the morphology of the scales from pipe 4A (A) and a cross-section of pipe 4C (B) after filling with epoxy and polishing



Pipe labels 4A and 4C are identification codes of the pipes based on their harvesting address. Pipes that have the same first digit in their codes are from the same location.

the brass-coupled pipe assemblies (Figure 5). Concentrations for the brass dielectric and externally wired plastic couplings were between these two bounding cases and also had more variation between duplicates.

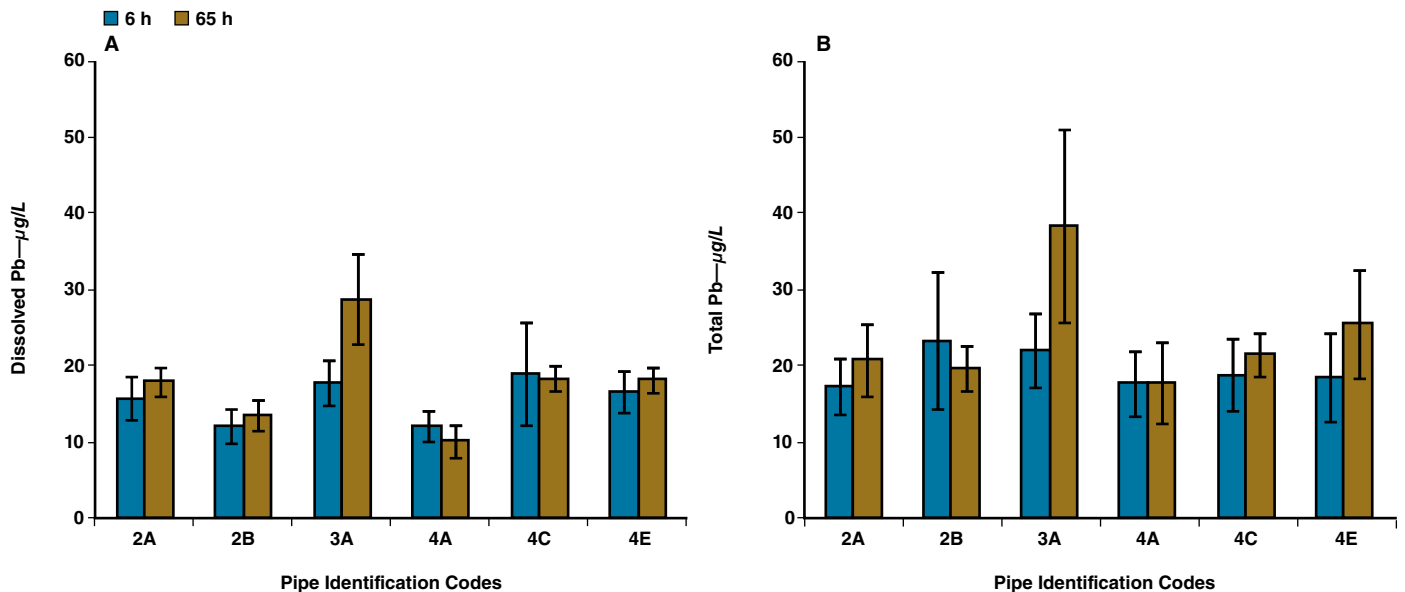
Effect of coupling types. Total lead and dissolved lead concentrations were quite similar for the plastic-coupled systems, suggesting that lead was present primarily in the form of dissolved lead. The total lead concentrations in the plastic-coupled systems following 6-h stagnation periods were stable over the six weeks

of experiments, with an average concentration of approximately 10 μg/L (Figure 5). Increasing the stagnation time to 65 h only slightly increased the total lead concentrations. These lead concentrations were close to the predicted equilibrium solubility of $Pb_5(PO_4)_3OH$ and similar to values measured during pipe conditioning. Because lead pipes and copper tubing were not galvanically connected in the plastic-coupled systems, the lead concentrations provided a baseline for comparison with systems in which galvanic corrosion was possible.

Connecting the lead and copper with external wires in the plastic-coupled systems allowed galvanic corrosion. Enhanced lead release was only observed for one of the duplicates (plastic-ex 2; Figure 5). On average, the total lead concentrations for plastic-ex 2 were eight times higher than without the wired connection (Figure 5). The enhancement of lead release for one of the externally wired connections can be attributed to galvanic reactions between lead and copper during the period of the wire connection (days 98–140, as shown in Figure 6). Removing the wires immediately decreased the lead concentrations to the levels without the wired connection (Figure 6). The duplicate pipe assemblies behaved almost identically before the external connection was made, but one of the pipes was much more susceptible to galvanic corrosion upon connection. Differences in the structure and spatial distribution of scales of corrosion products on the pipes could be responsible for the observed differences.

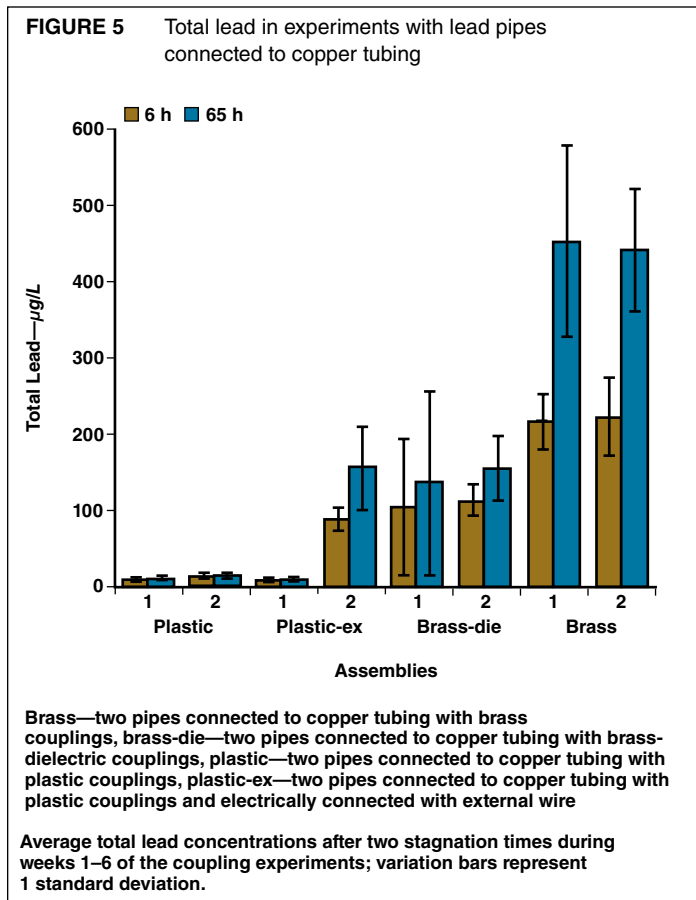
Galvanic current was detected in both of the externally wired plastic-coupled systems, and the current did not decrease over

FIGURE 4 Dissolved (A) and total (B) lead concentrations in the recirculating-flow conditioning mode



Pb—lead

Average values in the effluent after 6- and 65-h stagnation periods during weeks 1–4 of the recirculating-flow conditioning mode. Labels 2A, 2B, 3A, 4A, 4C, and 4E are identification codes of the pipes based on their harvesting address. Pipes that have the same first digit in their codes are from the same location. Variation bars represent 1 standard deviation.



the six weeks of the experiments. For plastic-ex 1, the average galvanic current during the stagnation periods was 18 μA ; plastic-ex 2 had a higher average galvanic current of 40 μA . Galvanic current has also been measured in previous studies with new lead pipes. By connecting lead pipes and copper tubing using external wires, Cartier et al (2012) found that the galvanic current persisted for seven months with an average value around 25 μA during stagnation. Using a similar connection method but different water chemistry, Triantafyllidou and Edwards (2011) reported galvanic currents during stagnation as high as 87 μA . Both in current and previous works (Cartier et al, 2012; Triantafyllidou & Edwards, 2011), the observed lead release was much lower than the total amount of $\text{Pb}(0)$ oxidation to $\text{Pb}(II)$ that is predicted from the galvanic current using Faraday's law. The total electron transfer from the lead to the copper is measured in the galvanic current, and this can then be used to determine the amount of lead oxidation that occurred. Even for plastic-ex 2, which had elevated lead release, only 1.5 and 3.0% of the $\text{Pb}(II)$ generated by galvanic corrosion was released as dissolved and particulate lead to water after 6 h of stagnation, respectively. In the current work, the galvanic currents during recirculation were typically 50% higher than those during stagnation periods, which was consistent with a previous observation that the galvanic current was 25–50% higher during flow than during stagnation (Cartier et al, 2012). For galvanic corrosion to occur, transport of solutes from the anode to the cathode must occur; for stagnant water, this trans-

port is controlled by diffusion, but for conditions with flow, the transport can be provided much more rapidly by convection.

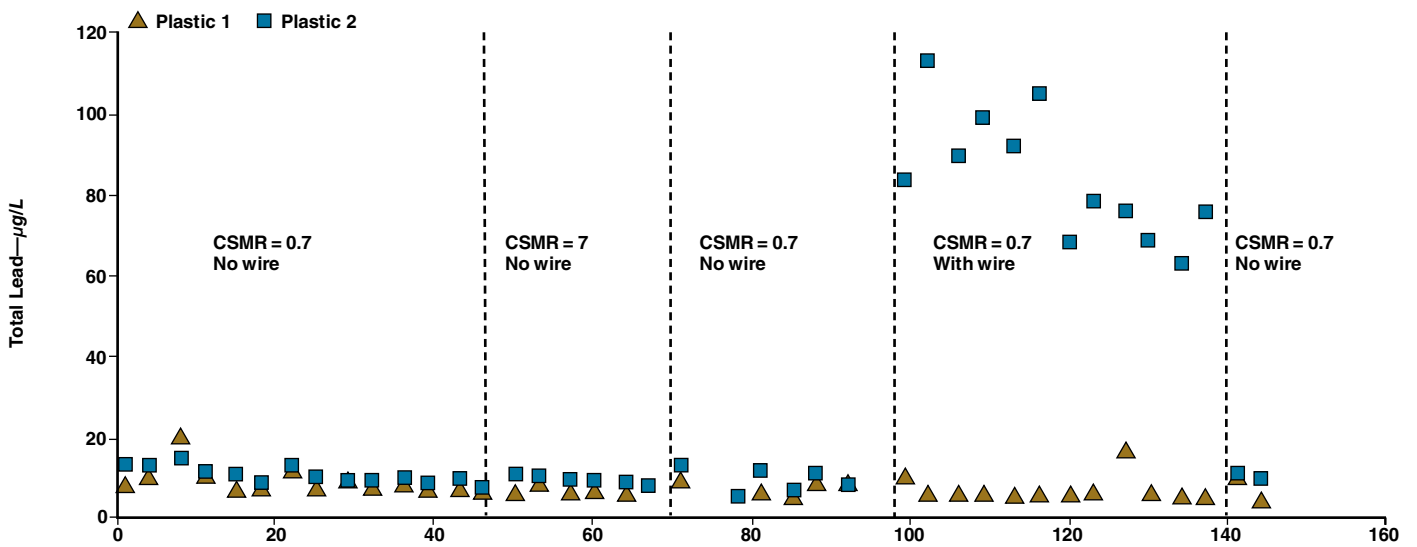
Greater amounts of total lead were released from the brass dielectric-coupled systems than from the plastic-coupled ones (Figure 5). The total lead concentrations continuously decreased for one of the duplicate assemblies (brass-die 1), but were stable for the other (brass-die 2). As with the externally wired plastic connections, the difference in the behavior of the duplicate assemblies with brass dielectric couplings may be attributed to microscale differences in the composition and distribution of the pipe scales. On average, the total lead concentrations following 6-h stagnation periods were an order of magnitude higher for the brass dielectric-coupled systems than for the plastic-coupled ones. Increasing the stagnation time to 65 h further enhanced the total lead release.

Although the brass dielectric couplings themselves may be a source of lead, lead pipe-free control experiments suggested that $< 20 \mu\text{g/L}$ of lead was released from the couplings following 6-h stagnation periods and that the lead was primarily in the form of dissolved lead (Figure 7). Therefore the majority of the lead release in the brass dielectric-coupled systems may be attributed to galvanic corrosion. With the brass dielectric couplings, the lead and copper are not galvanically connected, but brass and lead can act as a galvanic couple with lead serving as the anode.

The highest lead release was observed in the brass-coupled pipe assemblies (Figure 5). Compared with those in plastic-coupled systems, the total lead concentrations in the brass-coupled systems were about 20 times higher following 6-h stagnation periods. Increasing the stagnation time from 6 to 65 h doubled the total lead concentrations in the brass-coupled systems. The brass couplings themselves would release $< 20 \mu\text{g/L}$ total lead (Figure 7); therefore, the much greater lead release was primarily the result of galvanic corrosion. For both stagnation times, the duplicate brass-coupled assemblies were in agreement over the entire experiment duration. Clark et al (2011) also observed that more lead was released from brass-coupled systems than from systems in which the lead and copper were directly connected.

Copper release was monitored for the systems with different couplings and can provide complementary data to lead release regarding galvanic corrosion. Dissolved and total copper concentrations were similar for all of the systems, so most copper was dissolved. Although copper concentrations were similar for all the pipe assemblies after 6 h of stagnation, increasing the stagnation time to 65 h resulted in a different trend of copper release (Figure 8). The highest copper release was observed for the plastic-coupled systems in which no galvanic corrosion could occur. Relative to the plastic-coupled systems, the copper concentrations decreased in the order of systems connected with brass dielectric, externally wired plastic, and brass couplings. These results are consistent with copper acting as the cathode that is protected from corrosion by its galvanic interaction with lead or brass. For plastic-ex 1, although lead release was not enhanced because of galvanic corrosion, copper concentrations after 65 h of stagnation were decreased relative to those before making the external galvanic connections (plastic) and were comparable with those for plastic-ex 2 (Figure 8). Combined

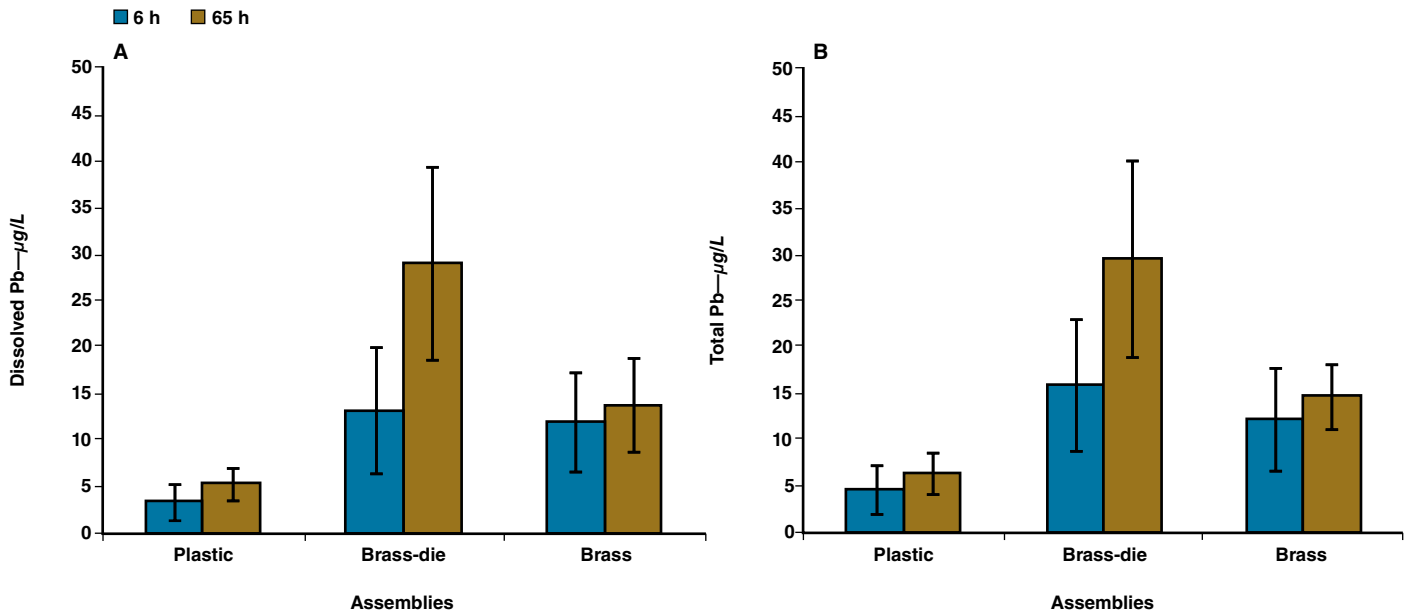
FIGURE 6 Total lead concentrations after 6-h stagnation periods over five months with plastic-coupled pipe assemblies



CSMR—chloride-to-sulfate mass ratio

The plastic-coupled pipe assemblies were used to investigate the effects of CSMR and the external electrical connection of the lead pipe and copper tubing.

FIGURE 7 Lead in control experiments (no lead pipe)—dissolved lead (A) and total lead (B)



Brass—two pipes connected to copper tubing with brass couplings, brass-die—two pipes connected to copper tubing with brass-dielectric couplings, plastic—two pipes connected to copper tubing with plastic couplings

Average dissolved and total lead concentrations after 6- and 65-h stagnation periods during weeks 1–4 of the experiments with polyvinyl chloride pipes connected to copper tubing. The lead released in these control experiments was almost entirely in a dissolved form. Variation bars represent 1 standard deviation.

with the measurements of galvanic current, these results regarding copper concentrations confirm that a galvanic interaction did still occur in the plastic-ex 1 system even though lead release was not increased.

Distribution of released lead between dissolved and particulate.

The distribution of total lead between particulate and dissolved was examined for all of the pipe assemblies (Figure 9). Dissolved lead was more abundant than particulate lead for the plastic-coupled systems. When the externally wired connection was made for plastic-ex 2, the increased lead concentration was primarily in the form of particulate lead, and more than 80% of the total lead was particulate (Figure 9). Triantafyllidou and Edwards (2011) determined lead release by connecting lead pipes and copper tubing with external galvanic wires. They observed that in stagnant water 70–99% of total lead was in particulate forms.

Compared with the plastic-coupled systems, both dissolved and particulate lead concentrations increased significantly for the brass-coupled and brass dielectric-coupled systems. For the brass-coupled systems, particulate lead became more dominant, and typically 52–72% of the total lead was particulate in the water after 6 h of stagnation. By examining the lead release from a similar pipe loop with a different water chemistry, Wang et al (2012) reported that 55–93% of the total lead was in particulate form in brass-coupled pipe assemblies. For the systems with brass dielectric couplings, the concentrations of dissolved and particulate lead were comparable.

The enhancement of particulate lead release from galvanic corrosion may be due to the formation of new particles or destabilization of the existing pipe scales (Wang et al, 2012). Destabilization of pipe scales could be strongly affected by the flow rate. Cartier et al (2012) found that increasing the sampling flow rate from 1.3 (laminar flow) to 8 L/min (turbulent flow) increased the particulate lead release by a factor of 3–40. They concluded that portions of the pipe scales may be more easily released at turbulent flow conditions. Given the sampling flow rate of 1 L/min and the pipe diameter, the flow pattern was laminar flow (Reynolds number = 1,140) in the current work. The total lead concentrations were quite stable over time and not noisy as would be expected for episodic detachment of lead-rich particles; therefore, the particulate lead release in the galvanically coupled systems of the current work is probably the result of the formation of new particles. In a related pilot-scale study to determine the effect of galvanic corrosion using Washington, D.C., lead pipes, a higher sampling flow rate of 10 L/min was used. The total lead concentrations were much noisier, which may be due to the destabilization of the existing scales (Welter et al, 2013).

In the recirculating flow experiments, the particulate lead released during recirculation can accumulate in the reservoir (Arnold et al, 2011; Nguyen et al, 2010b). To evaluate the amount of particulate lead that had accumulated in the reservoir, the water in the reservoir after a one-week cycle of stagnation and recirculation was acidified. In all cases, a significant increase (2.5–4.5 times) in total lead concentrations was observed following acidification of reservoirs that served pipe assemblies in which galvanic corrosion occurred. Typically, more than 80% of the total lead was in particulate form. The total lead that had accu-

mulated in the reservoirs followed the same trend as the stagnation samples, with concentrations increasing in the order of plastic, externally wired plastic, brass dielectric, and brass (compare Figures 5 and 10). By quantifying lead release in a similar experimental setup but with a different water chemistry, Wang et al (2012) found that for the brass-coupled pipe assemblies, the total lead concentrations in the reservoir increased by 3.5 times after acidification. In previous work with systems that had lead and copper connected by external wires, the total lead concentrations in the acidified reservoir were three–eight times those prior to acidification (Arnold et al, 2011; Nguyen et al, 2010b).

The total mass of lead released during one week is the sum of the lead released during stagnation and during recirculation. This total can be determined from the lead measurements in the three poststagnation samples collected each week and in the acidified reservoir. The reservoir includes lead released during stagnation periods for which samples were not collected and during the 45 min of recirculating flow that occurred each weekday. For one week, the total lead released from stagnation can be estimated as the sum of lead release from five 6-h, four 17-h, and one 65-h stagnation periods. Poststagnation samples were collected after 6- and 65-h stagnation periods, but not after 17-h stagnation periods. Assuming that all five 6-h stagnation periods had the same lead release and that similar amounts of lead were released during the 17- and 65-h stagnation periods, the total stagnation periods for a week contributed approximately 33, 51, 29, and

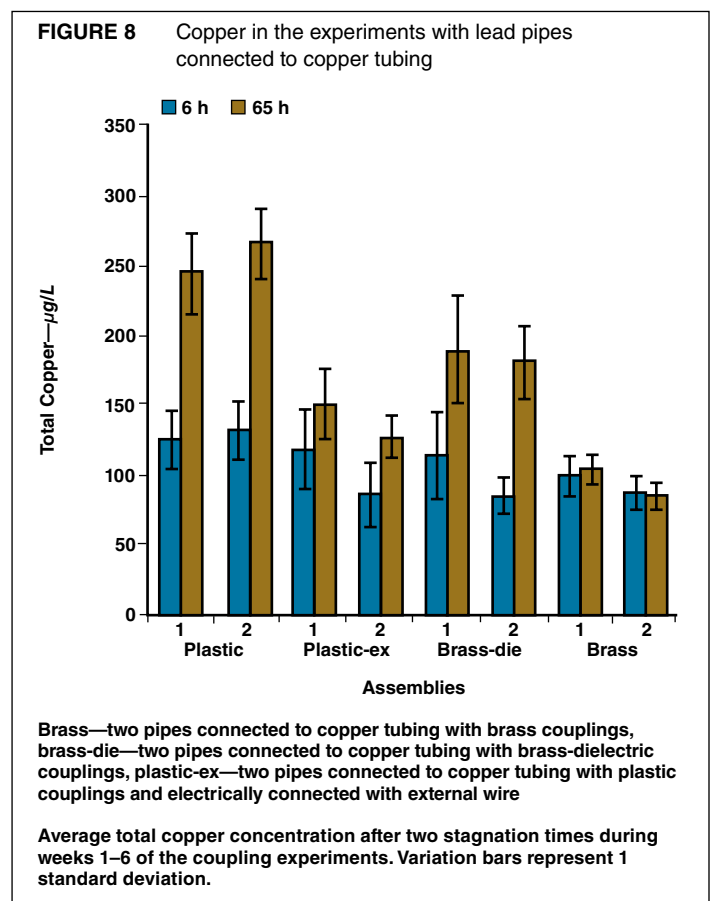
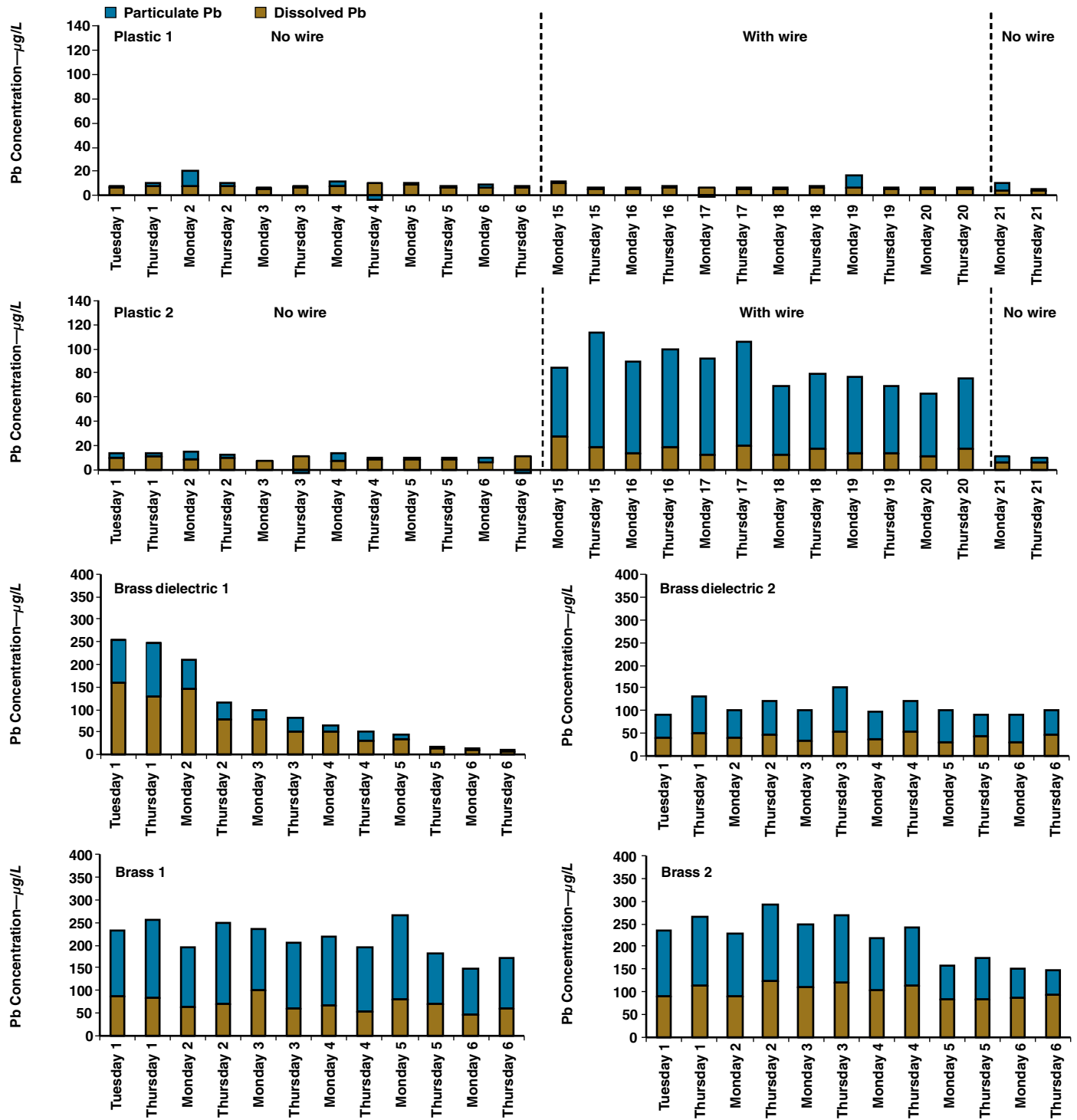
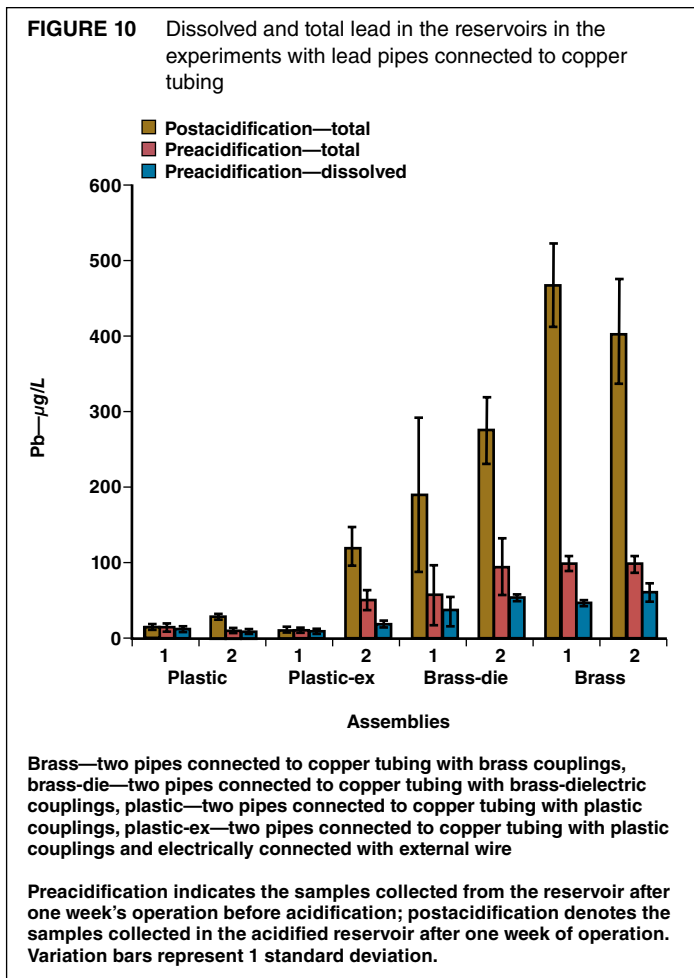


FIGURE 9 Distribution of dissolved and particulate lead in the experiments with lead pipes connected to copper tubing



Pb—lead
 Dissolved and particulate forms of lead during weeks 1–6 of experiments with lead pipes connected to copper tubing following 6-h stagnation periods using plastic (with and without externally wired connections), brass-dielectric, and brass-compression couplings. The x-axis indicates the day of the week and the week of the experiment.



40% of the total lead released (stagnation + flow) for plastic-coupled, plastic-ex 2, brass dielectric-coupled, and brass-coupled pipe assemblies, respectively. Therefore, it can be concluded that significant amounts of lead were released during stagnation as well as during flow.

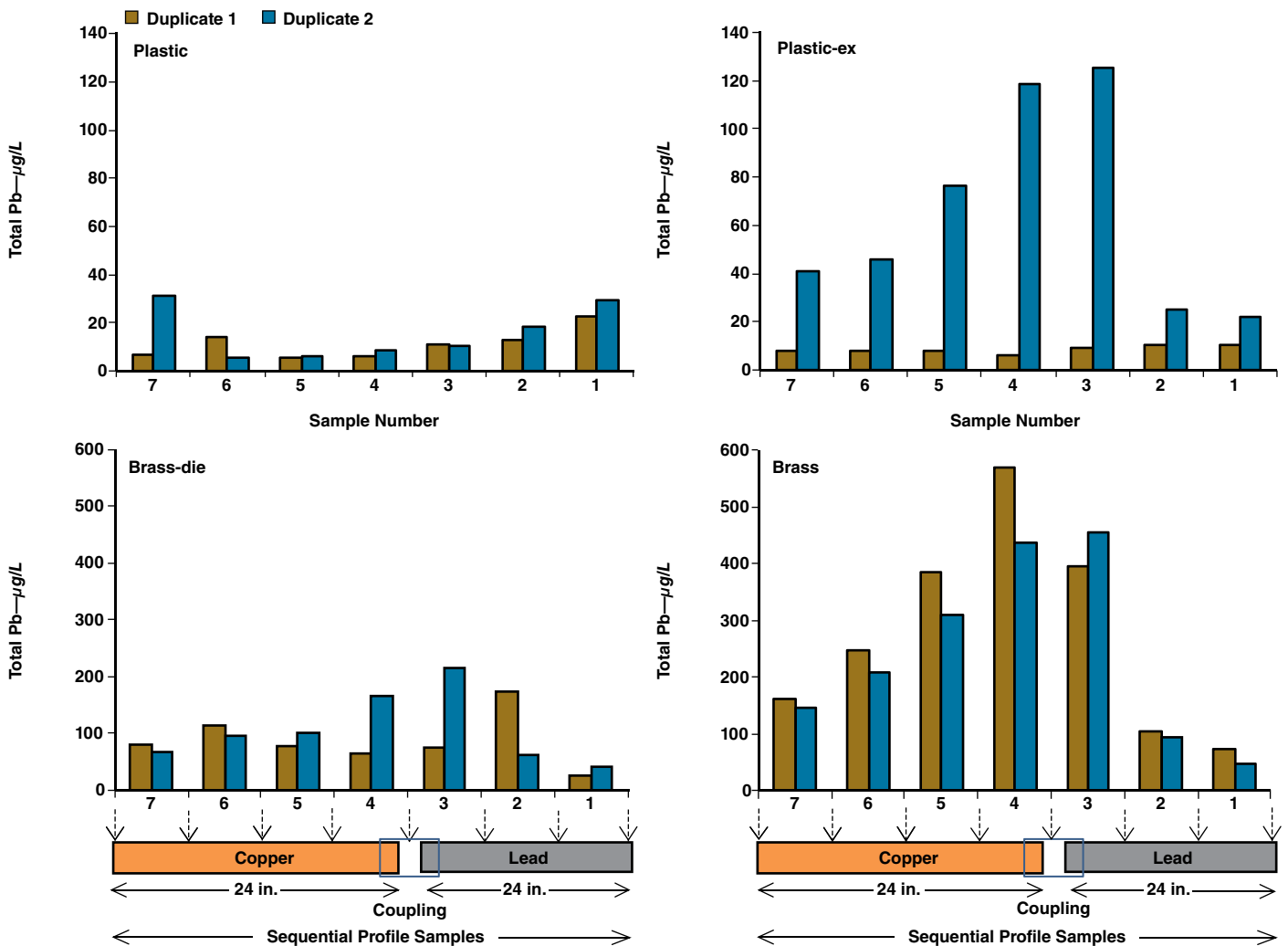
Results of lead release from both stagnation and flow suggested that galvanic corrosion enhanced total lead release for at least six weeks. Triantafyllidou and Edwards (2011) observed that lead concentrations were increased for several months in stagnant water in a system where lead and copper were galvanically connected by external wires and suggested that the enhancement was due to galvanic corrosion. Cartier (2012) monitored lead release in a once-through flow system following connection of lead pipes to copper tubing with brass couplings and found that galvanic corrosion enhanced lead release for at least eight weeks. Muylwyk et al (2011) measured lead concentrations in tap water in Guelph following PLSLRs (94 replacements tracked) and found that for some locations high lead concentrations persisted for at least 18 months. Further studies focused on lead release over months to years are needed to better evaluate the duration of galvanic corrosion.

Localized nature of galvanic corrosion. Localized lead release caused by galvanic corrosion was evaluated by a sampling approach that collected water from different zones of the pipe

assemblies after a 6-h stagnation period (Figure 11). For the plastic-coupled systems, lead concentrations were highest near the end of the lead pipe and decreased along the portion to the copper tubing. For the brass-coupled, brass dielectric-coupled, and one of the externally wired plastic-coupled systems, the highest lead concentrations were observed in the zones near the coupling connections, indicating that galvanic reactions were most dramatic in the regions near the couplings. For the three systems in which galvanic corrosion occurred, moving away from the junction higher lead concentrations were observed in the copper tubing sections than in the lead pipe sections. The aged lead pipes may provide a sink for lead in scavenging lead from solution. Previous observations also concluded by using various techniques that galvanic corrosion was a localized phenomenon. Boyd et al (2012) measured the open-circuit potential of directly connected lead pipe and copper tubing and observed that the galvanic effect was most pronounced within the first few centimetres next to the lead-copper junction. Several studies measured the galvanic currents between lead pipes and copper and found that the galvanic currents were highest within the first few inches next to the junctions and decreased with increasing distance (St. Clair et al, 2012; Clark et al, 2011; Triantafyllidou & Edwards, 2011). By measuring lead release from brass-coupled pipe assemblies, Wang and colleagues observed higher lead release near the brass couplings because of galvanic corrosion (Wang et al, 2012). In the current work, the lead concentrations near the couplings were five to eight times higher than at the lead ends of the systems in which galvanic corrosion occurred. Although these high local lead concentrations may not increase the overall lead exposure at the tap for locations at which a large portion of the lead service line is replaced, the maximum lead concentration that a consumer might be exposed to could be increased significantly. Further, depending on the relative amounts of lead pipe remaining after a PLSLR, the increased local release of lead resulting from galvanic corrosion may outweigh the decrease in lead associated with the removal of a portion of the lead pipe.

Effect of CSMR. For both the plastic- and brass-coupled systems, increasing the CSMR from 0.7 to 7.0 increased neither the total lead release following 6- and 65-h stagnation periods nor the total lead concentrations in the acidified reservoir after a one-week cycle of flow and stagnation (Figure 12). This observation is expected for the plastic-coupled pipe assemblies because no galvanic corrosion would occur in that system. However, for the systems having galvanic connections, CSMR was previously found to be an important parameter that affected the extent of galvanic corrosion (Triantafyllidou & Edwards, 2011; Nguyen et al, 2010a; Edwards & Triantafyllidou, 2007; Gregory, 1990). Previous studies have suggested that the threshold value of the CSMR above which lead release from galvanic corrosion could be accelerated was in the range of 0.6–0.77 (Nguyen et al, 2011b; Edwards et al, 1999). Based on chloride and sulfate concentrations that are representative of those in the water in the DC Water and Sewer Authority distribution system, the original CSMR was selected as 0.7 for the current study. It is possible that lower lead release would have occurred for the pipe assemblies if the CSMR was decreased to a lower value. Cartier (2012) determined lead

FIGURE 11 Profile sampling in the experiments with lead pipes connected to copper tubing



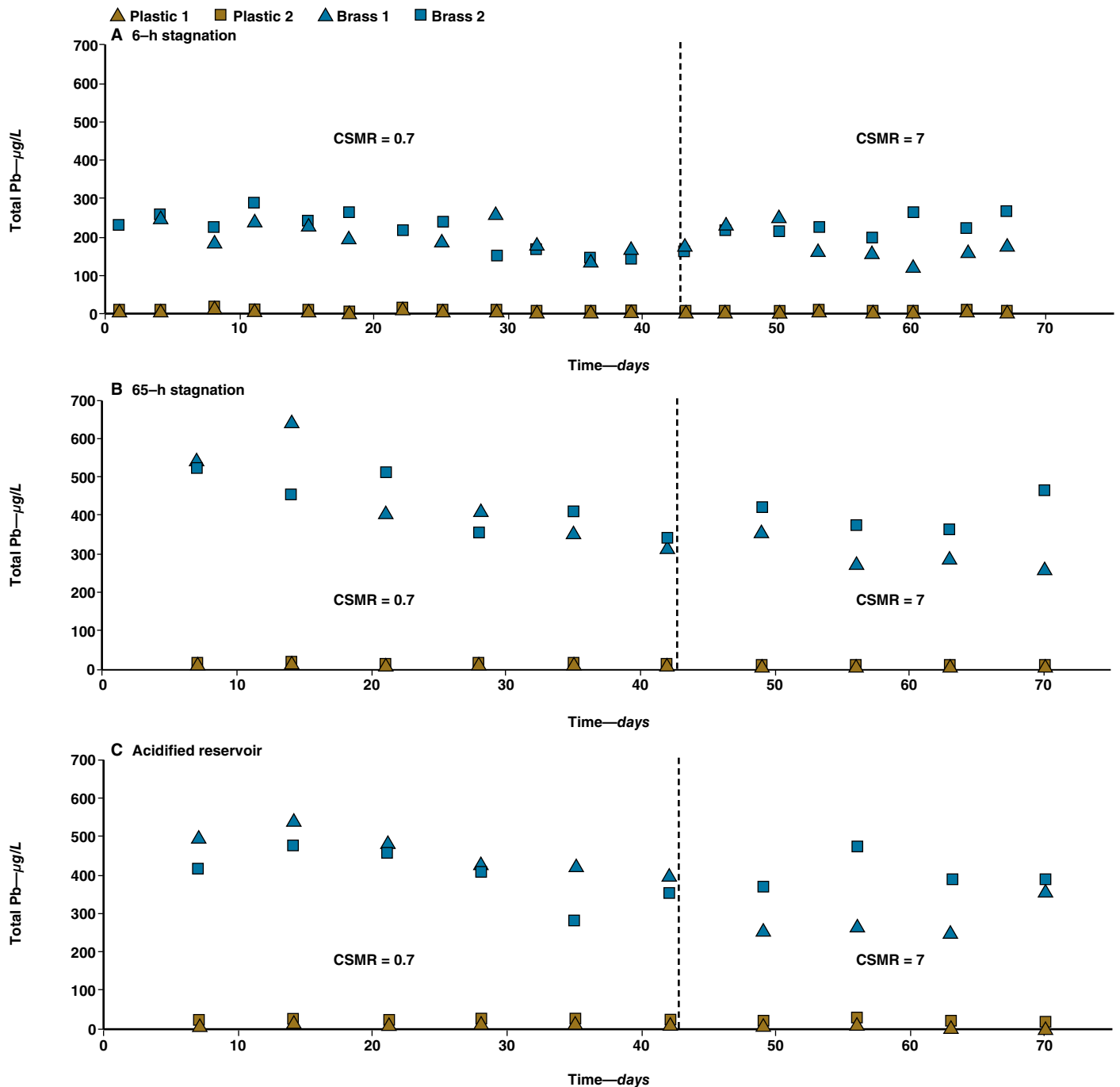
Pb—lead

Total lead concentration after 6 h of stagnation in different sampling regimes from experiments with lead pipes connected to copper tubing using plastic, plastic with external galvanic wires, brass-dielectric, and brass-compression couplings. Axis labels 7–1 indicate seven consecutive 70-mL samples as illustrated by the copper coupling–lead schematic below the charts. The inner diameters of the copper tubing and lead pipe were 1 and 0.75 in., respectively, so a given length of copper tubing holds more water than the same length of lead pipe. The copper tubing is shown as longer than the lead pipes to represent its larger volume.

release from galvanic corrosion in a once-through flow system by connecting lead pipes and copper tubing using brass couplings and found that decreasing the CSMR from 0.9 to 0.3 decreased lead release. In a system in which lead pipes and copper tubing were connected using external wires, the lead release increased by three to 12 times when the CSMR was increased from 0.2 to 16. However, even when the CSMR was low (0.2), lead release from galvanic corrosion was still significant (Triantafyllidou & Edwards, 2011). In the current study, the relatively high bicarbonate concentration combined with the presence of orthophosphate may have helped mitigate any enhanced lead release that could have been caused by increased galvanic corrosion at the elevated CSMR.

Environmental implications. By connecting aged lead pipes to new copper tubing with commercially available couplings, the current work demonstrated that galvanic corrosion occurred and enhanced lead release in the systems in which lead and copper were electrically connected. Even lead and brass may serve as a galvanic couple and result in an elevated lead release for at least six weeks. Therefore, to prevent galvanic corrosion following PLSLRs in the field, lead and copper would need to be connected with nonconductive plastic couplings. In addition, unlike new lead pipes, aged lead pipes may have quite different susceptibility to galvanic corrosion because of the spatial heterogeneity of the pipe scales. Given the pipe-to-pipe variability for the aged lead pipes,

FIGURE 12 Effect of CSMR in the experiments with lead pipes connected to copper tubing after 6-h (A) and 65-h (B) stagnation periods and in the acidified reservoir (C) after one week of operation



Brass—two pipes connected to copper tubing with brass couplings, CSMR—chloride-to-sulfate mass ratio, plastic—two pipes connected to copper tubing with plastic couplings, Pb—lead

decisions should be made conservatively with knowledge of the worst-case outcomes.

The experiments were performed for only six weeks for each condition, and sampling was performed at a relatively low flow rate (1 L/min), so caution should be taken in applying the conclusions of this work directly to the field. Long-term experiments

with higher sampling flow rates would provide more direct information on the duration of galvanic corrosion in the field. The current work found that increasing the CSMR from 0.7 to 7 did not increase lead release from galvanic corrosion, and future research could examine whether there would be benefits to lowering the CSMR below 0.7. Future research could also more

comprehensively evaluate the effects of other water chemistry parameters on galvanic corrosion.

CONCLUSIONS

The effect of coupling types and CSMR on lead release following simulated PLSLRs with copper was investigated using aged lead pipes harvested from Washington, D.C. Before conducting the experiments, the pipes were conditioned for a total of at least 12 weeks. Experiments with lead-copper pipe assemblies conducted for six weeks in a recirculating flow mode with intermittent stagnation periods yielded the following conclusions.

- The lowest lead release was observed for the plastic-coupled pipe assemblies, which served as a galvanic-free control. Relative to the plastic-coupled systems, the total lead release following stagnation increased in the order of systems coupled with plastic fittings with external wires, brass dielectric fittings, and brass fittings.
- The copper concentrations following 65 h of stagnation decreased in the order of systems coupled with plastic, brass dielectric, externally wired plastic, and brass, which was consistent with copper acting as the cathode in a galvanic interaction with lead.
- For the plastic-coupled systems with external wires, lead and copper can serve as a galvanic couple with lead serving as the anode, resulting in an increase of lead release and decrease of copper release, especially after 65 h of stagnation. Galvanic currents between lead and copper were detected, which confirmed the occurrence of galvanic corrosion.
- Duplicate pipe assemblies with plastic couplings with external connections and with brass dielectric couplings had different susceptibility to galvanic corrosion. It is possible that similar pipe-to-pipe variation could occur with other couplings as well.
- For the brass dielectric-coupled system, because brass itself only released limited lead, the enhanced lead release was probably the result of the galvanic reaction between lead and brass.
- For the systems with enhanced lead release, the increased lead amounts were primarily in particulate forms. Increasing the stagnation time from 6 to 65 h further increased the release of particulate lead, especially for the brass-coupled pipe assemblies.
- For the systems with enhanced lead release caused by a connection to copper tubing, lead concentrations were locally much higher in the regions closest to the couplings.
- For both the brass-coupled and plastic-coupled pipe assemblies, increasing the CSMR from 0.7 to 7 did not increase the amount of lead release.

ACKNOWLEDGMENT

The Water Research Foundation and the DC Water and Sewer Authority are the joint owners of the technical information on which this article is based. The authors thank the foundation and DC Water for their financial, technical, and administrative assistance (WaterRF Project Number 4349). The comments of three anonymous reviewers were valuable in revising an earlier version of this article. The authors thank Abigail Cantor for her helpful discussions of the data. Rob Richards and Gary Allen of O'Brien & Gere provided valuable assistance in acquiring the harvested lead pipes. A.Y. McDonald and Harco Fittings donated the tested

transition couplings. Scanning electron microscopy was done in the Center for Materials Innovation. Steve Picker, Zimeng Wang, and Ziming Yang provided valuable laboratory assistance.

ABOUT THE AUTHORS



Yin Wang is a postdoctoral research associate at the University of Illinois at Urbana-Champaign. At the time this article was written, Wang was a PhD student at Washington University in St. Louis. He has five years of research experience in drinking water quality and corrosion control, including three years of experience in galvanic corrosion.

Vrajesh Mehta is a graduate research assistant at Washington University in St. Louis. Gregory J. Welter is a technical director at O'Brien & Gere, in Bowie, Md. Daniel E. Giammar (to whom correspondence should be addressed) is an associate professor, Department of Energy, Environmental, and Chemical Engineering, Washington University in St. Louis, Campus Box 1180, 1 Brookings Dr., St. Louis, MO 63130; giammar@wustl.edu.

FOOTNOTES

- ¹Mac-Pak fittings, A.Y. McDonald, Dubuque, Iowa.
- ²UTC fittings, Harco-Philmac, Lynchburg, Va.
- ³MasterFlex L/S pump, Cole Parmer, Vernon Hills, Ill.
- ⁴Agilent 7500ce, Agilent, Santa Clara, Calif.
- ⁵Geigerflex D-MAXA diffractometer, Rigaku, The Woodlands, Texas
- ⁶7001LVF scanning electron microscope, JEOL, Tokyo, Japan.
- ⁷Lambda XLS+ spectrophotometer, PerkinElmer, Waltham, Mass.

PEER REVIEW

Date of submission: 10/22/12

Date of acceptance: 04/19/13

REFERENCES

- Arnold, R.B. Jr.; Raetz, M.; & Edwards, M., 2011. Effects of Alkalinity, NOM, and Orthophosphate on Galvanic Corrosion. Proc. 2011 AWWA Ann. Conf., Washington, D.C.
- Benjamin, M.M., 2002. *Water Chemistry*. McGraw-Hill, New York.
- Boyd, G.R.; Reiber, S.H.; McFadden, M.S.; & Korshin, G.V., 2012. Effect of Changing Water Quality on Galvanic Coupling. *Journal AWWA*, 104:3:45.
- Cartier, C., 2012. Identification and Treatment of Major Sources of Lead in Drinking Water. Doctoral dissertation, Department of Civil, Geological, and Mining Engineering, École Polytechnique de Montréal, Montréal, Québec, Canada.
- Cartier, C.; Arnold, R.B. Jr.; Triantafyllidou, S.; Prévost, M.; & Edwards, M., 2012. Effect of Flow Rate and Lead/Copper Pipe Sequence on Lead Release from Service Lines. *Water Research*, 46:13:4142.
- Clark, B.; Cartier, C.; Clair, J.S.; Triantafyllidou, S.; Prévost, M.; & Edwards, M., 2011. Lead Contamination of Drinking Water After Partial Lead Service Line Replacement With Copper Pipe: Bench Testing of Galvanic Impacts. Proc. AWWA 2011 Ann. Conf., Washington.
- DeSantis, M.K.; Welch, M.; & Schock, M., 2009. Mineralogical Evidence of Galvanic Corrosion in Domestic Drinking Water Pipes. Proc. AWWA 2009 Water Quality Technology Conf., Seattle, Wash.
- Edwards, M.; Jacobs, S.; & Dodrill, D., 1999. Desktop Guidance for Mitigating Pb and Cu Corrosion By-products. *Journal AWWA*, 91:5:66.
- Edwards, M. & Triantafyllidou, S., 2007. Chloride-to-Sulfate Mass Ratio and Lead Leaching to Water. *Journal AWWA*, 99:7:96.
- Gerke, T.L.; Scheckel, K.G.; & Schock, M.R., 2009. Identification and Distribution of Vanadinite (Pb₅(VO₄)₃Cl) in Lead Pipe Corrosion By-Products. *Environmental Science & Technology*, 43:12:4412.

- Giammar, D.; Nelson-Nguyen, K.; Xie, Y.; & Noel, J., 2008. Role of Phosphate in Mitigating Lead Release From Corrosion Products. Proc. AWWA 2008 Water Quality Technology Conf., Cincinnati.
- Gregory, R., 1990. Galvanic Corrosion of Lead Solder in Copper Pipework. *Water and Environment Journal*, 4:2:112.
- Kim, E.J. & Herrera, J.E., 2010. Characteristics of Lead Corrosion Scales Formed During Drinking Water Distribution and Their Potential Influence on the Release of Lead and Other Contaminants. *Environmental Science & Technology*, 44:16:6054.
- McNeill, L.S. & Edwards, M., 2002. Phosphate Inhibitor Use at US Utilities. *Journal AWWA*, 94:7:57.
- Muylywyk, Q.; Waller, M.; Spielmacher, A.; Olesiuk, J.; & Suffoletta, V., 2011. Full Versus Partial Lead Service Line Replacement and Lead Release in a Well-Buffered Groundwater. Proc. AWWA 2011 Water Quality Technology Conf., Phoenix.
- Nadagouda, M.N.; White, C.; & Lytle, D., 2011. Lead Pipe Scale Analysis Using Broad-Beam Argon Ion Milling to Elucidate Drinking Water Corrosion. *Microscopy and Microanalysis*, 17:2:284.
- Nguyen, C.K.; Stone, K.R.; & Edwards, M.A., 2011a. Nitrate Accelerated Corrosion of Lead Solder in Potable Water Systems. *Corrosion Science*, 53:3:1044.
- Nguyen, C.K.; Clark, B.N.; Stone, K.R.; & Edwards, M.A., 2011b. Role of Chloride, Sulfate, and Alkalinity on Galvanic Lead Corrosion. *Corrosion*, 67:065005:1.
- Nguyen, C.K.; Stone, K.R.; & Edwards, M.A., 2011c. Chloride-to-Sulfate Mass Ratio: Practical Studies in Galvanic Corrosion of Lead Solder. *Journal AWWA*, 103:1:81.
- Nguyen, C.K.; Stone, K.R.; Dudi, A.; & Edwards, M.A., 2010a. Corrosive Microenvironments at Lead Solder Surfaces Arising From Galvanic Corrosion with Copper Pipe. *Environmental Science & Technology*, 44:18:7076.
- Nguyen, C.; Stone, K.; Clark, B.; Edwards, M.; Gagnon, G.; & Knowles, A., 2010b. Impact of Chloride: Sulfate Mass Ratio (CSMR) Changes on Lead Leaching in Potable Water. Water Research Foundation Report. www.waterrf.org/PublicReportLibrary/4088.pdf (accessed May 6, 2013).
- Reiber, S.H. & Dufresne, L., 2006. Effects of External Currents and Dissimilar Metal Contact on Corrosion of Lead From Lead Service Lines. Report to US Environmental Protection Agency Region III. www.epa.gov/dclead/Grounding_Effects_Study_Final_November_2006.pdf (accessed May 6, 2013).
- Sandvig, A.; Kwan, P.; Kirmeryer, G.; Maynard, B.; Mast, D.; Trussell, R.R.; Trussell, S.; Cantor, A.; & Prescott, A., 2009. Contribution of Service Line and Plumbing Fixtures to Lead and Copper Rule Compliance Issues. Report 91229. Water Research Foundation, Denver.
- Schecher, W.D. & McAvoy, D.C., 1998. MINEQL+: A Chemical Equilibrium Modeling System, Version 4.0. Environmental Research Software, Hallowell, Maine.
- Schock, M.R.; Lytle, D.A.; Sandvig, A.M.; Clement, J.; & Harmon, S.M., 2005a. Replacing Polyphosphate With Silicate to Solve Lead, Copper, and Source Water Iron Problems. *Journal AWWA*, 97:11:84.
- Schock, M.R.; Scheckel, K.; DeSantis, M.; & Gerke, 2005b. Mode of Occurrence, Treatment, and Monitoring Significance of Tetravalent Lead. Proc. AWWA 2005 Water Quality Technology Conf., Québec City, Canada.
- St. Clair, J.; Stamopoulos, C.; & Edwards, M., 2012. Technical Note: Increased Distance Between Galvanic Lead: Copper Pipe Connections Decreases Lead Release. *Corrosion*, 68:9:779.
- Standard Methods for the Examination of Water and Wastewater*, 1998 (20th ed.). APHA, AWWA, and WEF, Washington.
- Swertfeger, J.; Hartman, D.J.; Shrive, C.; Metz, D.H.; & DeMarco, J., 2006. Water Quality Effect of Partial Lead Line Replacement. Proc. AWWA 2006 Ann. Conf., San Antonio, Texas.
- Triantafyllidou, S. & Edwards, M., 2011. Galvanic Corrosion After Simulated Small-scale Partial Lead Service Line Replacements. *Journal AWWA*, 103:9:85.
- US Environmental Protection Agency (USEPA), 2011. Science Advisory Board Evaluation of the Effectiveness of Partial Lead Service Line Replacement. [http://yosemite.epa.gov/sab/sabproduct.nsf/964CCDB94F4E6216852579190072606F/\\$File/EPA-SAB-11-015-unsigned.pdf](http://yosemite.epa.gov/sab/sabproduct.nsf/964CCDB94F4E6216852579190072606F/$File/EPA-SAB-11-015-unsigned.pdf) (accessed May 6, 2013).
- USEPA, 1991. Maximum Contaminant Level Goals and National Primary Drinking Water Regulations for Lead and Copper. Final Rule. *Federal Register*, 56:26460.
- Wang, Y.; Jing, H.; Metha, V.; Welter, G.J.; & Giammar, D.E., 2012. Impact of Galvanic Corrosion on Lead Release From Aged Lead Service Lines. *Water Research*, 46:16:5049.
- Wang, Y.; Xie, Y.; Li, W.; Wang, Z.; & Giammar, D.E., 2010. Formation of Lead(IV) Oxides from Lead(II) Compounds. *Environmental Science & Technology*, 44:23:8950.
- Welter, G.; Giammar, D.; Wang, Y.; & Cantor, A., 2013. Galvanic Corrosion Following Partial Lead Service Line Replacement, Draft Final Report. Project Number 4349. Water Research Foundation, Denver.
- Xie, Y. & Giammar, D.E., 2011. Effects of Flow and Water Chemistry on Lead Release Rates From Pipe Scales. *Water Research*, 45:19:6525.