# An Analysis of the Correlation between Lead Released from Galvanized Iron Piping and the Contents of Lead in Drinking Water

## Summary Report

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# 1.0 Executive Summary

The District of Columbia Water and Sewer Authority (DCWASA) retained the services of HDR Engineering Inc. (HDR) to evaluate residential profiles conducted at twenty-one homes following various stages of lead service line replacement (LSR). The evaluation focused on identifying and characterizing the co-occurrence between lead and iron release from old galvanized plumbing, which had first been recognized by DCWASA in recurring instances of elevated concentrations of both metals in Lead and Copper Rule (LCR) compliance monitoring. Laboratory testing and surface analyses of corrosion scales on galvanized pipes were included in this study. It need be recognized that while this study makes constant reference to galvanized pipe, in reality the pipe in question has long since lost its galvanization (zinc) layer, and is in reality a mild steel pipe with substantial accumulation of iron-rich corrosion scale.

HDR's evaluation of the 62 residential profiles conducted by DCWASA indicated that galvanized iron in-home plumbing can contribute substantial particulate lead to residential drinking water taps, and should be considered as potentially important a lead source as lead service lines themselves. Jar tests, and corrosion scales analysis established that lead rich iron corrosion scales can form as the result of decades of accumulation of lead-iron precipitates on interior pipe surfaces. Pipe loop experiments confirmed that lead release from galvanized iron scales can be exacerbated by an array of physical and hydraulic disturbances even after trends of diminishing release appear to indicate that available lead on an iron scale surface has been exhausted. The following key points were identified during this study:

- Lead adsorbed to galvanized iron corrosion scales can persist and be mobilized to household taps after full replacement of lead service lines, potentially for the remaining service life of the galvanized plumbing.
- In the group of homes with galvanized plumbing that participated in this study, more particulate lead was released per unit surface area of that plumbing than was released from lead service lines. Comparable lead release from other in-home plumbing materials was not observed
- A correspondence between elevated lead and elevated iron release from galvanized plumbing was consistently observed in household profiles. A moderately strong linear correlation was observed between lead and iron release overall.
- Very strong linear correlations between lead and iron were observed in particular locations. The slopes of regression lines varied from location to location indicating different lead contents in galvanized iron corrosion scales.
- The potential for lead release from galvanized plumbing at a given home must be assessed on an individual basis because lead content in corrosion scales will vary according the nuances of exposure and piping configuration at that location.
- Lead bears a strong adsorptive affinity for both ferric and ferrous iron compounds, and scale formation by co-precipitation and deposition of lead-iron precipitates can potentially result in accumulation over decades of a crystalline iron corrosion scale rich in lead.
- Lead content can vary substantially in galvanized iron pipe corrosion scales depending on the history of the premise piping. A range from a few hundredths of a percent to as high as eight percent lead by weight was observed in galvanized plumbing from different homes where lead service lines have been in place.
- Lead can reside in the deepest layers of galvanized iron corrosion scales indicating the potential for continued lead release for the remaining service life of the pipes.

- Non-iron lead minerals such as apatites, carbonates and oxides of lead can exist in isolated deposits embedded in iron corrosion scales, indicating the potential for multiple adsorptive mechanisms including physical capture that will influence scale characteristics and lead release.
- Lead release from galvanized plumbing will likely be exacerbated substantially by physical disturbance, especially immediately following a partial or full service replacement.
- Exacerbated lead release due to physical disturbance subsides over time following the disturbance, but the rate of diminishing lead release varies from location to location.
- Lead release from galvanized plumbing that appears to have subsided to some stable level over time following physical disturbance may increase again due to subsequent disturbances.
- Lead release from galvanized plumbing can be exacerbated by hydraulic disturbances such as water hammer or increased flow velocity.

Because lead accumulates over decades, and is likely distributed across the full thickness of a galvanized iron corrosion scale, the only way to ensure that lead is not mobilized from plumbing to tap in a given home is to fully replace the old galvanized plumbing.

After a full lead service replacement, lead release from in-home plumbing should be evaluated. If the evaluation indicates mild to low risk of lead exposure, and the homeowner cannot afford to fully replace galvanized plumbing, the following partial mitigation measures might be considered:

- Replace portions of in-home galvanized plumbing (pipe runs) associated directly with drinking water taps such as the kitchen sink or bathroom sinks.
- Identify lead release associated with existing faucets and replace with new non-lead bearing faucets.
- Install water hammer dampers where water hammer has been an issue historically.

If a post-LSR evaluation indicates high risk of lead exposure, especially in homes with small children, the homeowner should seriously consider replacing the in-home plumbing. Given the evidence that galvanized plumbing can contribute substantial lead to drinking water taps, DCWASA may wish to consider extending their existing financial assistance program to include specific provisions for replacement of galvanized plumbing where elevated lead release persists beyond full lead service replacement.

# 2.0 Background

In November 2000, the Washington Aqueduct (WA), which treats the water distributed by the District of Columbia Water and Sewer Authority (DCWASA), switched disinfectants from chlorine to chloramines in order to reduce disinfection byproducts (DBP). Following the disinfectant change, DCWASA observed elevated distribution system lead levels in their Lead and Copper Rule (LCR) compliance monitoring, exceeding the lead action level of 15 ppb between 2002 and 2004. Residential profiles, conducted in winter 2004 isolated the predominant source of distribution system lead to lead service (Schock & Giani – 2004). Additional residential profiles conducted during a chlorine burn in spring 2004 provided preliminary data indicating that high chlorine residuals added at the treatment plant in the 1990's created a lead (IV) oxide scale on DCWASA lead service lines. Subsequent investigations (Boyd et al. 2009) have confirmed that reduction in oxidation/reduction potential (ORP) associated with the switch to chloramination destabilized these lead oxide scales, mobilizing lead to the distribution system.

In 2004, WA began adding orthophosphate to average concentrations of 3.2 mg/L as $PO_4$ to inhibit corrosion and control lead release. Within 10 months, the 90th percentile of samples from residential taps decreased from 54 ppb to 15 ppb. In tandem with orthophosphate addition at WA, DCWASA implemented a program of lead service line replacements (LSR) which has encountered obstacles including annual expenditures of over $20 million as well as continual residential traffic disruption. The effectiveness of the LSR program has been questioned due to the fact that DCWASA can only legally replace the portion of an existing service line that lies within the public right of way (partial LSR), leaving lead service piping remaining on the private side. Evaluation of sample results taken from multiple utilities after partial LSR and full LSR (where lead service on the private side of the property line is replaced) has indicated that partial LSR is less effective than full LSR, and often results in only minimal improvement in first-draw lead levels, and only minor improvements in total mass of lead released after two months at a given site (Sandvig et al – 2008).

DCWASA continues to invest considerable resources in studying sources of lead in drinking water and evaluating the effectiveness of techniques to control it. In 2008 DCWASA conducted a detailed review of LCR data from 2006 and 2007 (Giani, 2008). The following findings were noted:

- 90th percentile lead levels were between 10 ppb and 15 ppb.
- Several homes consistently exceed 15 ppb from first draw samples.
- At least 75% of the homes that exceeded the lead action level for either first draw and/or second draw lead levels had elevated iron (> 79 ppb) in the same sample.

DCWASA conducted a preliminary investigation at one of the homes that consistently exceeded the 15-ppb LCR action level using industry accepted residential profile procedures (Giani, et al – 2004). At the time the profile was conducted, the private and public sides of the service line were lead, and the majority of in-home plumbing was galvanized iron installed in the mid twentieth century. Results indicated elevated particulate lead and iron concentrations in 1-liter samples associated with in-home plumbing. Particulate lead levels in in-home plumbing samples were higher than in those associated with the lead service, while dissolved lead concentrations were higher in samples associated with the lead service than in those associated with in-home plumbing. Additional preliminary investigations at other homes produced similar results, leading to the conclusion that lead may adsorb to, and be released from iron corrosion scales inside galvanized iron pipes.  Subsequently, in-home galvanized plumbing may serve as a potential source of lead released to drinking water.

To investigate the possible relationship between lead and iron release, and to further evaluate the effectiveness of LSR, DCWASA encouraged home owners at sites targeted for service replacement to participate in a study.  Homes with galvanized iron and/or copper in-home plumbing throughout the DCWASA service area participated.  This paper summarizes the results of these residential profiles.  In addition, this paper summarizes supplementary laboratory investigations including jar tests and pipe loop experiments conducted to further develop an understanding of the mechanisms and kinetics of lead sorption and release from galvanized iron plumbing.

## 2.1 Literature Review

The sorptive capacity of hydrous ferric hydroxides is well understood, and, indeed, iron based coagulation is widely employed in drinking water treatment processes. Alternatively, the adsorption of trace inorganic constituents to iron corrosion scales in distribution system piping is a phenomenon that has not gained serious attention until recently.  While the occurrence of lead and other trace metal deposits within iron corrosion scales has been observed in various studies, little is understood regarding the potential for iron corrosion scales in household plumbing to serve as a source of lead in drinking water.

In 2008, Sandvig et al reported the results of a collaborative investigation of contributions of service line and plumbing fixtures to lead levels at residential taps.  Several utilities contributed results of LCR compliance sampling, more general sampling, case studies, and other field data to the study.  Results indicated that lead service lines were the major source of lead measured at the tap (50 to 75 percent of the total mass of lead measured). The impact of full and partial lead service replacement on lead measured at the tap was a major component of this study.  While full replacement was generally effective at reducing to total mass of lead released, partial replacement was identified as having only limited effectiveness in reduction of lead release.  In both full and partial replacements, elevated particulate lead was observed in the days immediately following the replacement.  The elevated particulate release was identified with disturbance of the existing service lines and premise plumbing.  Premise piping was identified as contributing up to 35% of the total mass observed at household taps in the study.  It was hypothesized that this large contribution, second only to that from service lines, was the result of “seeding”, described as the migration lead from service lines through the system and being incorporated into scales on household plumbing.  Seeded lead could then be released over time depending on hydraulic, physical, or chemical conditions.  The hypothesis was not confirmed, nor was an attempt made to identify seeding with particular premise piping materials.

The mechanism of by which seeding of lead to iron corrosion scales may occur is not fully understood.  It is likely a combination of factors including physical sorptive processes involving weak van der Waals forces, chemical sorption, and co-precipitation resulting in a mixed lead-iron solid phase, and physical capture of mobilized particulate lead precipitates within the rough surface of iron scales.  The potential for co-precipitation is supported by a series of isotherms constructed by Dzombak (1990) fitting cation sorption data sets for dissolved lead ($Pb^{2+}$) and ferric oxide.  In all of the isotherms describing various molar ratios of iron to lead presented, effectively all of the lead present binds to iron at pH lower than typical drinking water levels (pH<6.5).

Despite the potential for co-precipitation and deposition of iron-lead solid phase to internal iron piping surfaces, relatively few observations of lead deposits in iron corrosion scales of a magnitude meaningful to potential lead release have been recorded. Reiber and Dostal (1997) obtained samples of cast iron and galvanized iron pipes from a distribution system in Fremont Nebraska following occurrences of unusually high levels of particulate iron, lead, copper, and arsenic at residential taps. Lead was detected at up to 6 parts per thousand within the corrosion scales in the pipe samples, though lead levels in the treated water were non-detect. Copper levels within the corrosion scales were of a similar magnitude. These results indicate clearly that under certain circumstances lead and copper deposits are very likely to form within iron corrosion scales.

# 3.0 Part 1 – Lead Profiling in DCWASA

## 3.1 Methodology

In 2004 DCWASA developed and refined an industry accepted method for conducting lead profile sampling of residential plumbing (Giani et al 2004), which was expanded and executed during the course of this study. The procedure, described in detail later in this section, involves identifying consecutive 1-L residential tap samples with specific segments of the in-house and service plumbing on a cumulative volume basis. A typical profile includes 10 to 15 consecutive samples collected after a six-hour stagnation period representative of all plumbing and service between the sample tap and main. Samples taken for profiles used in this study were analyzed for total and dissolved lead, iron, copper, zinc, cadmium, and, in some cases, manganese. Profiles were evaluated to identify trends in metal release from lead and copper service, as well as galvanized iron, copper, and other in home plumbing materials over time in relation to partial and full lead service replacement.

Twenty-one DCWASA service area homes were profiled in this study, of which five underwent full sequential sampling after partial and full LSR. Full sequential sampling involves multiple profiles conducted over a minimum of 60 days following partial LSR, and then again following full LSR. In all cases in this study, partial LSR is to be understood as replacement of lead with copper service pipe within the public right of way while full LSR is the replacement with copper of the remaining lead service on the private side of the property line. Full sequential sampling at DCWASA, from pre-partial LSR to post-full LSR, includes four phases.

- **Phase 1** - **Plumbing Survey**: DCWASA personnel conducted a walkthrough plumbing materials survey of the residence selected for study. During the walkthrough the length, diameter and material of in-homing plumbing and service lines were identified, as were meter location, riser lengths, and plumbing fixture materials. The volume of all sections of in-home and service piping were calculated and converted to liters so that consecutive 1-L samples taken at the tap could be identified with specific portions of the in-home and service plumbing on the basis of cumulative volume.
- **Phase 2 – Pre LSR Profile:** DCWASA personnel conducted a full water quality profile of the selected home as described in the sampling protocol potion of this section of the report. Sampling following induced water hammer (also discussed under sampling protocol) was also conducted at the time of the profile; in some cases a full sampling profile was collected following water hammer. Part way through the study, in late 2007, high flow velocity samples, conducted with the tap fully open, were also collected.
- **Phase 3 – Partial LSR Sequential Profiling**: Following DCWASA replacement of the public portion of the lead service line, the plumbing survey information from phase 1 was updated and full water quality profiles were conducted periodically for at least sixty (60) days after following the partial LSR. The first profile was conducted within 72 hours of the partial LSR. Additional profiles were conducted 14 to 18 days, 1 month (29 to 36 days), and 2 months (59 to 66 days) following the replacement.

- **Phase 4 - Full LSR Sequential Profiling:** Following DCWASA replacement of the private portion of the lead service line, the plumbing survey information from phase 1 was updated and full water quality profiles were conducted periodically for at least sixty (60) days after following the full LSR. The same periodicity of sampling described in Phase 3 was followed. If profiling indicated continuing lead release from in-home plumbing after full LSR, DCWASA recommended replacement of that plumbing to the home-owner.

### 3.1.1 Sampling protocol

Profile sampling was conducted following the completion of the plumbing survey. Plumbing surveys were updated after each stage of LSR, and include measurement and verification of all in-home and service plumbing materials. Meter location, riser lengths, and plumbing fixture materials were identified and recorded. Pictures were taken to identify the condition of the external surface of the residential plumbing material. The service line materials, lengths, and diameters were determined during the LSR and recorded by the on-site engineer. In cases where sampling was conducted prior to partial LSR and the service material could not be verified on-site, service material and dimensions were based on DCWASA historical records. Where historical records do not indicate service line diameters, lead service lines are assumed ¾-in unless 1-in pipe is observed at the point of entry. The volume of all sections of in-home and service piping were calculated and converted to liters so that consecutive 1-liter samples taken at the tap can be identified with specific portions of the in-home and service plumbing on the basis volume. Plumbing survey results for each home surveyed include a pictorial summary of plumbing arrangement by material and a summary table of piping material, dimensions, and calculated volume, and are provided in the appendices of this report.

Profile sampling includes a single pre-stagnation sample as well as a full profile consisting of approximately 15 consecutive 1-liter samples collected following a 6-hour minimum stagnation period. All samples were collected at the residence's kitchen tap, or, in one case, at a bathroom tap that was the primary use sink while the kitchen was being remodeled. Sample times were coordinated with customers in advance to ensure that no taps were opened in the customer home during the stagnation period. Immediately following the profile sampling, high velocity flush samples (from late 2007 on) and water hammer samples were also collected. Sampling follows the protocol outlined below:

### 3.1.2 1-liter pre-stagnation sample

- Aerator at the tap is removed in order to capture particulate metals.
- Tap is opened and pipes are flushed for at least 5 minutes.
- Following 5-minute flush, a 1-liter sample is collected in a vacuum flask rinsed with 1:1 nitric acid, followed by deionized water. The sample is representative of the water in the pipes at the start of stagnation. Any changes in chlorine, metals, HPC etc. after stagnation can be compared to this sample.
- Water Quality Analysis is conducted on 100 mL volume removed from the sample including total chlorine, pH and temperature:
- Approximately 300 mL of the sample is filtered through a 0.45 micron membrane pre-rinsed with deionized water.

Filtered and un-filtered samples are acidified with nitric acid.

### 3.1.3 Post 6-hour stagnation sample collection procedure

- Sample bottles are pre labeled and rinsed with 1:1 nitric acid, followed by deionized water at the WA lab, prior to the sample date. The number of liters sampled depends on the volume between the tap and main as calculated using plumbing survey information.
    - The first liter sampled consists of 4 individual 250-mL samples.
    - The remaining samples of volumes associated with in-home and service plumbing are collected in consecutive 1-liter samples.
    - 2 – 3 additional samples are collected of water associated with the service main after the full volume of in-home and service plumbing calculated in the survey are collected.
    - For each sample bottle prepared, a corresponding bottle is prepared to contain the filtered portion of the sample.
- After all sample bottles are prepared and arranged in the order to be collected, the tap is opened slowly. Samples are drawn at a rate of 250-mL/6-seconds.
- Sampling is conducted continuously without closing the tap or altering flow velocity until all prepared bottles are filled.
- Immediately following completion of profile sample collection, water quality including total chlorine, pH, and temperature is measured in a portion of a sample associated with in-home plumbing.
- Following completion of the profile sampling, the faucet is opened to full, and allowed to run for five minutes. After five minutes, five additional 1-liter samples are collected without shutting the faucet. These samples are representative of high flow velocity, and are intended to indicate the effects of scouring on metal release from in-home and service piping. High flow velocity sampling was commenced part-way through the study in late 2007.
- Following collection of the high velocity samples, the faucet is closed rapidly to create a water hammer effect within the plumbing. The faucet is opened full and rapidly closed several times to repeat this effect, and then a 1-liter sample is collected. These samples are representative of the effect that hydrodynamic disturbances caused by sudden changes in pressure might have on in-home plumbing corrosion scales. In 2008, toward the end of the profile sampling conducted for this study, full profiles were conducted following the induced water hammer to determine the effects of water hammer on different sections of the service and in-home piping, and to provide a non-stagnated profile to compare to the stagnated profile already collected.
- Portions of each sample are filtered through a 0.45 micron membrane pre-rinsed with deionized water.
- Filtered and un-filtered samples are acidified with nitric acid.

### 3.1.4 Sample Analysis

All samples collected were analyzed at the Washington Aqueduct Lab using ICP-MS for lead, iron, copper, zinc, and cadmium. These metals represent the suite of elements associated with lead and copper service lines as well as galvanized iron piping. In some cases manganese was also analyzed. Tables summarizing all samples collected at each home and the metals analyzed in each sample are provided in the appendices of this report.

### 3.1.5 Data Used in the Study

Five homes underwent full sequential sampling for this study, representing the core of the sample data collected. All of these homes had some galvanized iron in-home plumbing. At three of them, the in-home plumbing was almost entirely composed of galvanized iron. At the other two, in-home plumbing consisted primarily of copper or some other material with a few feet of galvanized iron pipe near the point of entry.

At the 16 other homes that participated in the study, plumbing surveys were performed and a single profile was conducted. These 16 homes represent a cross section of homes containing full or partial lead service covering the geographic extent of the DCWASA service area. Most homes included in the study were selected initially because of a history of elevated lead or iron release. Some homes were specifically targeted for study because of elevated iron release identified in conjunction with elevated lead release in LCR compliance monitoring samples; this group includes the five homes with galvanized iron plumbing where full sequential sampling was conducted. Other homes were specifically selected as a control group; these include homes where in-home plumbing is composed entirely of copper or some material other than galvanized iron. One home was selected with galvanized in-home plumbing where there was never a lead service line.

For this study, the twenty-one participating homes have been organized into three series of profiles by in-home plumbing material. Each home has been designated an alpha-numeric tag indicating which group the home is a part of. The “G” series comprises 9 homes whose in home plumbing is composed almost entirely of galvanized iron. Full sequential profiling was conducted at homes G-1, G-2, and G-3 from this group. The “M” series comprises 4 homes with mixed in-home plumbing, less than half of that plumbing being of galvanized iron. Of these homes, full sequential profiling was conducted at home M-1, while sequential profiling occurred following full LSR at home M-2. The “C” series, comprising 8 homes, represents the control group, where in-home plumbing is composed primarily of copper and no galvanized iron is present. Table 1 summarizes the full set of homes profiled, including alpha-numeric tags, descriptions of in-home plumbing materials, and the number of profiles conducted at various stages of LSR.

## Table 1: Summary of 21 DCWASA homes participating in study

| Home I.D. | In-House Plumbing Material(s) | Pre LSR Profile | Partial LSR Profiles | Full LSR Profiles |
|---|---|---|---|---|
| G-1 | Galvanized Iron | Yes | 2-day, 2-weeks, 4-weeks, 8-weeks | 1-day, 2-weeks, 4-weeks, 8-weeks, 6-month |
| G-2 | Galvanized Iron | Yes | 2-days, 2-weeks, 4-weeks, 8-weeks | 1-day, 1-week, 2-weeks, 4-weeks, 8-weeks |
| G-3 | Galvanized Iron | Yes | 1-day, 2-weeks, 4-weeks, 8-weeks | 1-day, 1-week, 2-weeks, 4-weeks |
| G-4 | Galvanized Iron | Yes - Existing Copper Service | No | No |
| G-5 | Galvanized Iron | Yes | No | No |
| G-6 | Galvanized Iron | Yes | No | No |
| G-7 | Galvanized Iron | No | Yes | No |
| M-1 | Copper (with 2.8-ft galvanized iron near service line connection prior to Full LSR) | Yes | 1-day, 2-weeks, 4-weeks, 8-weeks | 2-day, 1-week, 2-weeks, 4-weeks, 8-weeks, 9-month |
| M-2 | Copper (with 1.3-ft galvanized iron near service line connection prior to Full LSR) | No | One profile prior to full LSR; 4 years after partial LSR. | 2-days, 2-weeks, 4-weeks, 6-weeks, 10-weeks |
| M-3 | Copper (1-ft Galvanized iron) | Yes | No | No |
| M-4 | Copper (with 2.3-in. galvanized iron near tap) | Yes | No | No |
| M-5 | Copper (with 4.4-ft. galvanized iron near tap) | No | No | One profile conducted 18 months after full LSR. |
| M-6 | Copper (with 1.9-ft. galvanized iron near tap and entry point) | Yes | No | No |
| C-1 | Copper | No | One profile collected 13 months after partial LSR. | No |
| C-2 | Copper | No | One profile collected 2 months after partial LSR. | No |
| C-3 | Copper | Yes | No | No |
| C-4 | Copper | Yes | No | No |
| C-5 | Copper | Yes | No | No |
| C-6 | Copper | Yes | No | No |
| C-7 | Copper | Yes | No | No |
| C-8 | Copper | Yes | No | No |

Table 2 organizes the 62 profiles summarized in Table 1 by data series and the time the profile was conducted relative to stage of lead service replacement. The 2,624 filtered and unfiltered samples associated with the 62 study profiles are summarized by data series and the stage of lead service replacement in Table 3. Table 4 organizes the 2,624 samples by data series and associated piping or sampling method.

**Table 2: Study profiles organized by data series and time conducted relative to stage of lead service replacement**

| | **Number of Profiles Performed** | | |
|---|---|---|---|
| | **G-Series** | **M-Series** | **C-Series** |
| Pre-LSR | 6 | 4 | 6 |
| After Partial LSR | 13 | 5 | 2 |
| After Full LSR | 14 | 12 | 0 |
| Total | 33 | 21 | 8 |

**Table 3: Samples organized by data series and time conducted relative to stage of lead service replacement**

| | **Number of Individual Samples Collected (Filtered and Un-filtered)** | | |
|---|---|---|---|
| | **G-Series** | **M-Series** | **C-Series** |
| Pre-LSR | 188 | 164 | 252 |
| After Partial LSR | 544 | 184 | 86 |
| After Full LSR | 670 | 536 | 0 |
| Total | 1402 | 884 | 338 |

**Table 4: Samples organized by data series and associated piping or sampling method**

| | **Number of Individual Types of Samples (Filtered and Un-filtered)** | | |
|---|---|---|---|
| | **G-Series** | **M-Series** | **C-Series** |
| In-Home Plumbing (Including Pre-Stagnation Samples) | 434 | 274 | 96 |
| Service Piping (Lead or Copper) | 562 | 260 | 82 |
| Main | 100 | 198 | 64 |
| High Flow Velocity | 240 | 110 | 80 |
| Water Hammer | 66 | 42 | 16 |
| Total | 1402 | 884 | 338 |

### 3.1.6 Data Evaluation

To compare metal released from one site to another or one plumbing material to another, sample data has been normalized to mass release per unit interior surface area of pipe per profile (Eq. 1). In charts presented in this report, normalized mass release is always shown in units of µg/m$^2$.

$$Normalized\_Mass\_\mathrm{Re}lease = \frac{\sum m_i}{A_i n_i} \qquad \text{Eq. 1}$$

Where:
$m_i$ = sum of mass collected in n profiles from a plumbing material at location *i*
$A_i$ = total exposed internal surface area of a plumbing material
$n_i$ = number of profiles conducted at location *i*

Where an average mass release per plumbing material across a range of homes is presented (i.e. average particulate lead release from galvanized iron plumbing in G-series homes), it has been calculated as a weighted average (Eq. 2).

$$Average\_Normalized\_Mass\_\mathrm{Re}lease = \frac{\sum_i \frac{m_i}{A_i n_i}(n_i)}{\sum_i n_i} = \frac{\sum_i \frac{m_i}{A_i}}{\sum_i n_i} \qquad \text{Eq. 2}$$

To provide a point of reference in charts showing mass release in µg/m$^2$, a dashed line has been included at 71.5 µg/m$^2$, which represents the lead release per unit area at which a 1-liter sample collected from a ¾" pipe would exceed the LCR action level of 15 ppb.

Where coefficients of correlation between two sets of data are presented, they have been calculated using Equation 3.

$$Correlation(X,Y) = \frac{\sum (\mathrm{x} - \bar{\mathrm{x}})(y - \bar{y})}{\sqrt{\sum (x - \bar{x})^2 (y - \bar{y})^2}} \qquad \text{Eq.3}$$

Where:
$\bar{x}$ = mean value of data set x
$\bar{y}$ = mean value of data set y

## 3.2 Results

### 3.2.1 Lead Release Overview

To provide an overview of the results from the 62 profiles conducted for this study, average total and dissolved lead release per unit area were calculated for in-home plumbing, lead service lines, and copper service lines from each of the series of data sets. Samples from 61 of the 62 profiles conducted were used to calculate these fluxes. Profiles from home C-8, where the service material is unknown, were excluded from this analysis. Table 5 summarizes the number of profiles conducted per type of piping for each sample group.

**Table 5: Summary of Profiles Conducted by Piping Material**

| | Number of Profiles Conducted | | |
|---|---|---|---|
| Type of Piping | G-Series | M-Series | C-Series |
| In-home | 33 | 21 | 7 |
| Pb Service | 18 | 9 | 7 |
| Cu service | 28 | 17 | 2 |

Figure 1 shows the normalized mass release from each type of piping for the G-series, M-Series, and C-Series, respectively. Particulate lead release from both in-home and lead service lines exceeded the level equivalent LCR action level in G-series homes. More particulate lead mass was released per profile from galvanized in-home plumbing than lead service lines in the G-series results. Particulate lead release from galvanized in-home plumbing was an order of magnitude higher than that from predominantly copper and copper in-home plumbing in the M and C series, respectively. Normalized particulate lead release was also much higher from lead service lines in G-series homes than in the M or C series. Inspection of individual profiles for G-series homes (Appendices 1-9) indicates that much of the particulate lead release associated with lead service was collected in samples adjacent to the in home plumbing. Effects of advection or collection of samples including partially in-home and partially service line volumes may contribute to the discrepancy between lead service line particulate release between the G-series and the other series.

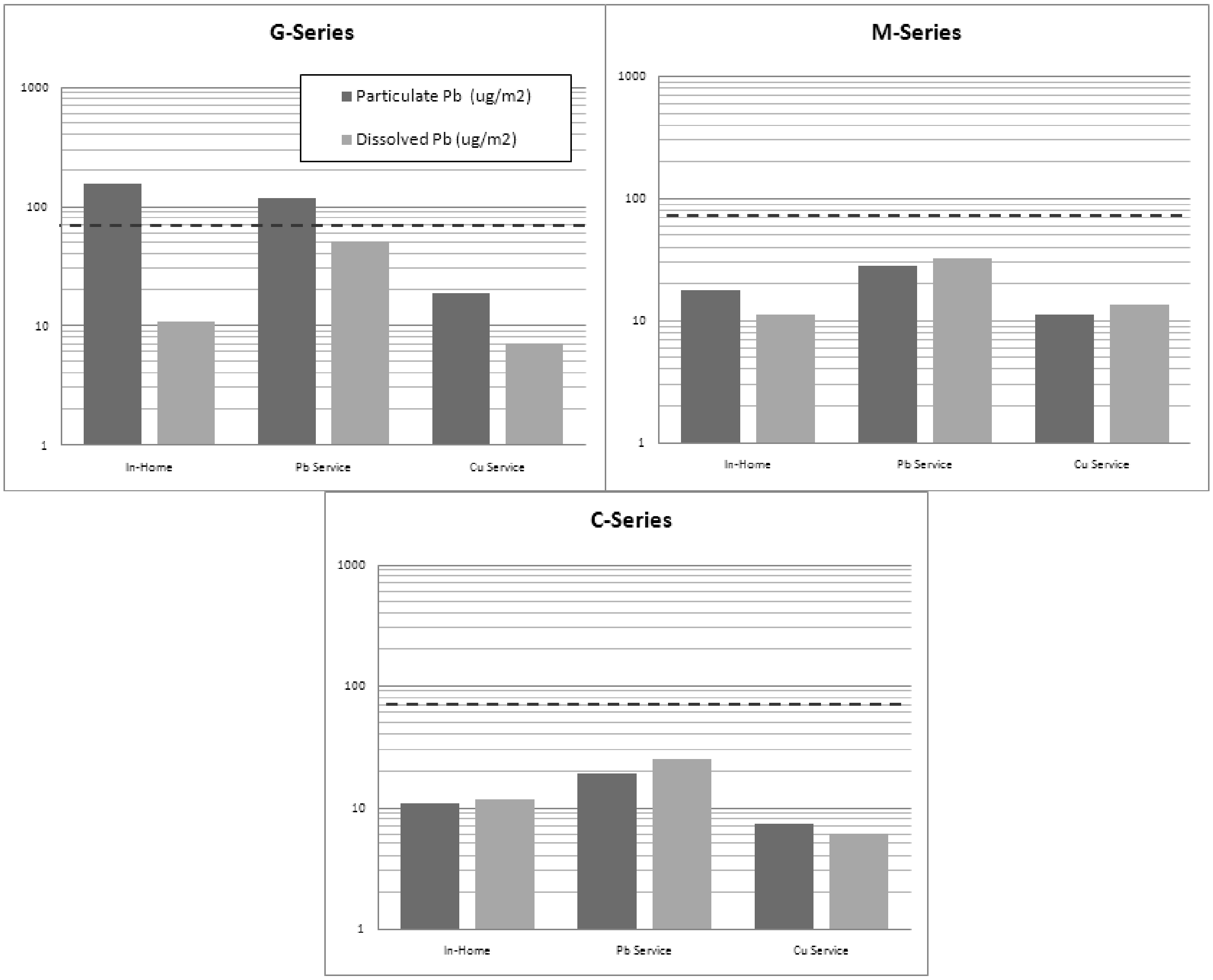

**Figure 1: Normalized lead mass released per area of in-home plumbing, lead service, and copper service in G-series, M-series and C-series profiles**

Dissolved lead associated with galvanized piping was not meaningfully different than that associated with copper or mixed in-home plumbing. In all three series, the greatest contribution of dissolved lead mass was from lead service lines. Total lead release was greater from lead service lines than galvanized iron plumbing in the G-series because the dissolved fraction from service was larger.

These results do not account for differences in lead mass release from location to location, nor do they account for differences before and after partial or full LSR. Figure 2 shows normalized particulate lead release per profile from a unit area of in-home plumbing in µg/m$^2$ on the basis of location and relative to LSR phases. The equivalent LCR action level mass release per volume has been added as a point of reference. An "X" near the site label on the vertical axis indicates that no samples were collected at the site during that stage of LSR.

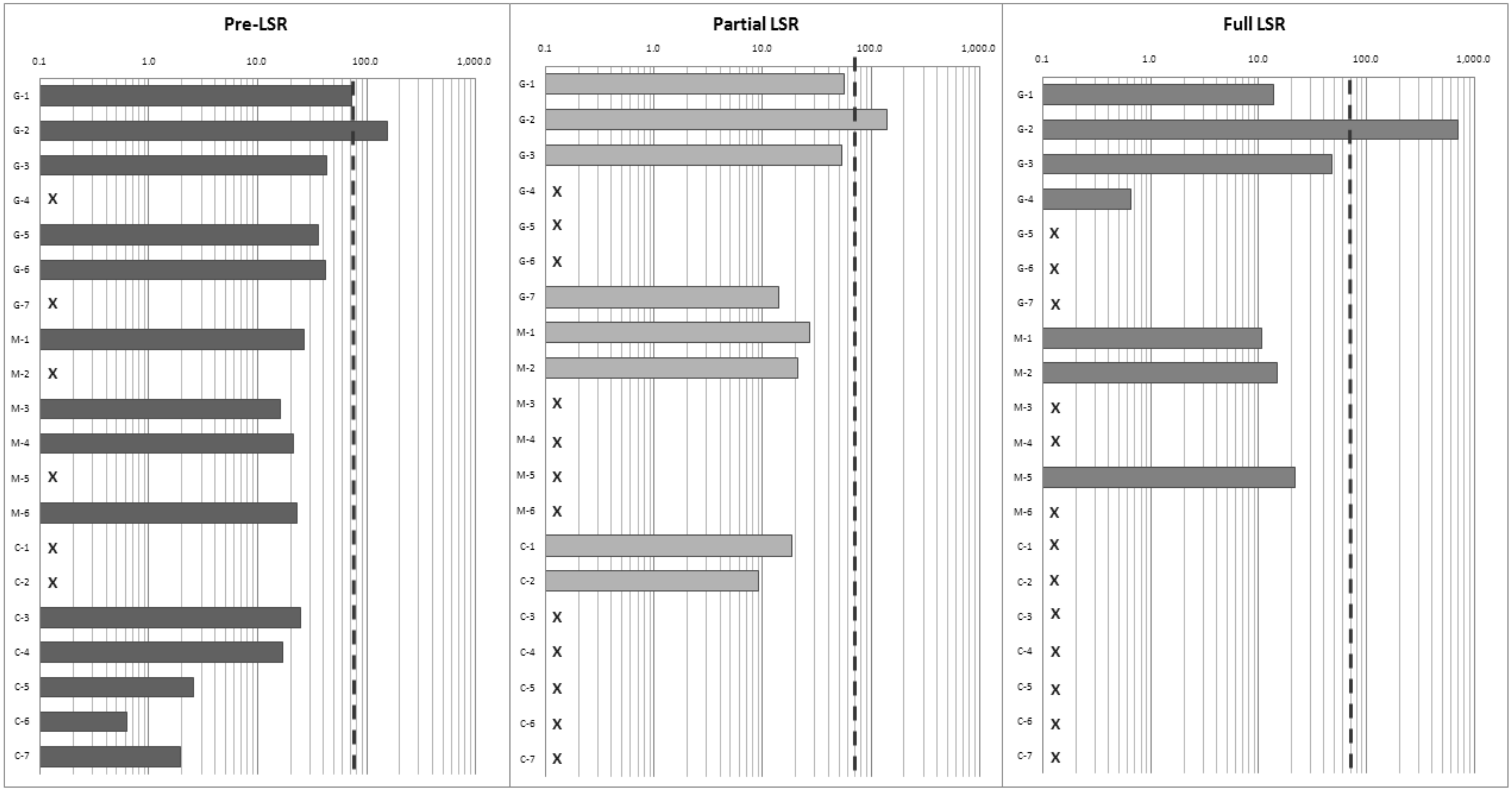

Figure 2: Average particulate lead release in µg/m$^2$ of in-home plumbing per profile for each home participating in the study at various stages of lead service replacement. An “X” near the site label on the vertical axis indicates that no samples were collected at the site during that stage of LSR.

These results show that particulate release from galvanized iron in-home plumbing varies in behavior from location to location. Lead release at location G-1 decreases substantially after full LSR. However, at site G-3 full LSR provided minimal improvement, while at site G-2 full LSR appears to have exacerbated lead release. In the other G-series sites, where fewer profiles were conducted, particulate lead release from in-home plumbing is generally only slightly higher than that from mixed or copper plumbing at the M and C locations. Homes G-1, G-2, and G-3 are the three homes in the G-series where full sequential profiling was conducted, and high lead release associated with multiple profiles conducted at these homes represents the major contribution to the normalized averages shown in Figure 1.

### 3.2.2 The Impact of Time on Lead Release

Inspection of particulate lead release from sequential profiles evaluated in this study indicates that particulate lead release is generally exacerbated immediately after LSR. The contribution of extreme release events immediately after LSR must be considered when evaluating averages at a specific location over an extended period such as those shown in Figure 2. Sandvig et al (2008) associated exacerbated post-LSR release with physical disturbance of the corrosion scales on premise piping during service replacement. Exacerbated release can be observed in almost all profiles in this study conducted immediately following partial or full LSR where galvanized iron plumbing was present. However, in almost all of these cases a decreasing trend in particulate lead release from galvanized pipes was observed as time between the LSR and the profile date increased. Figure 3, showing sequential sampling profiles following full LSR at home G-2, exemplifies this phenomenon. Lead release from in-home plumbing is highest immediately following full LSR, then decreases in the following weeks. Note that in this home particulate lead release continues at concentrations at or above the LCR action level throughout the sample volume associated with the in-home galvanized plumbing even eight weeks following full LSR.

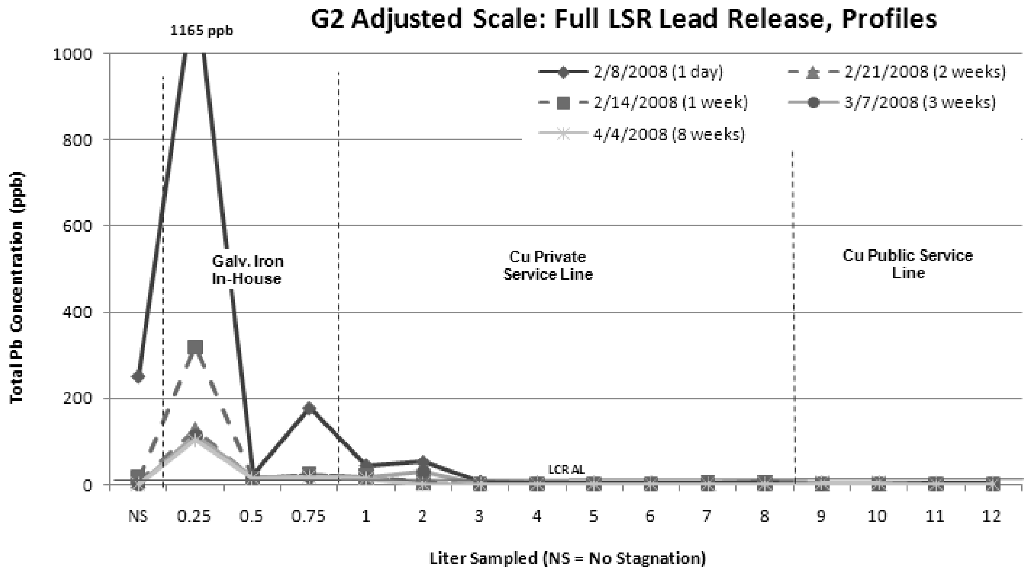

**Figure 3: Decrease in particulate lead release from galvanized plumbing with elapsed time at Site G-2.**

The decreasing trend in lead release from in home plumbing appears to be less consistent after a partial LSR. Normalized particulate lead release from in-home plumbing in four homes sampled multiple times following partial LSR is depicted in Figure 4. Figure 5 shows particulate lead release in the same four homes following full LSR. Following full LSR, particulate release appears to decrease at a consistent rate after 14 days in all four homes; however the magnitude of the rate of change in release varies substantially from home to home.

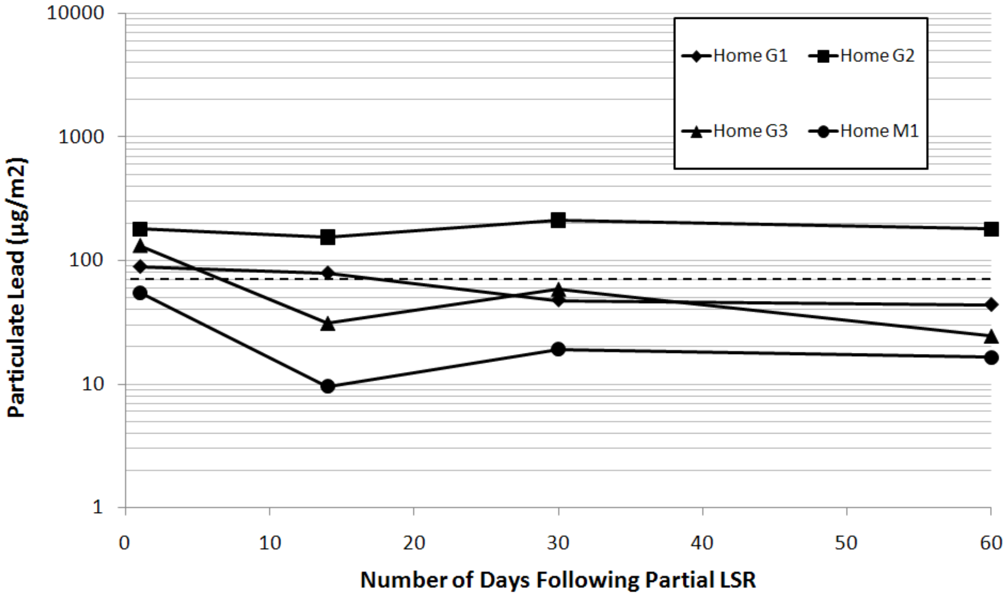

**Figure 4: Normalized Particulate Lead Release over Time from In-Home Plumbing following Partial LSR**

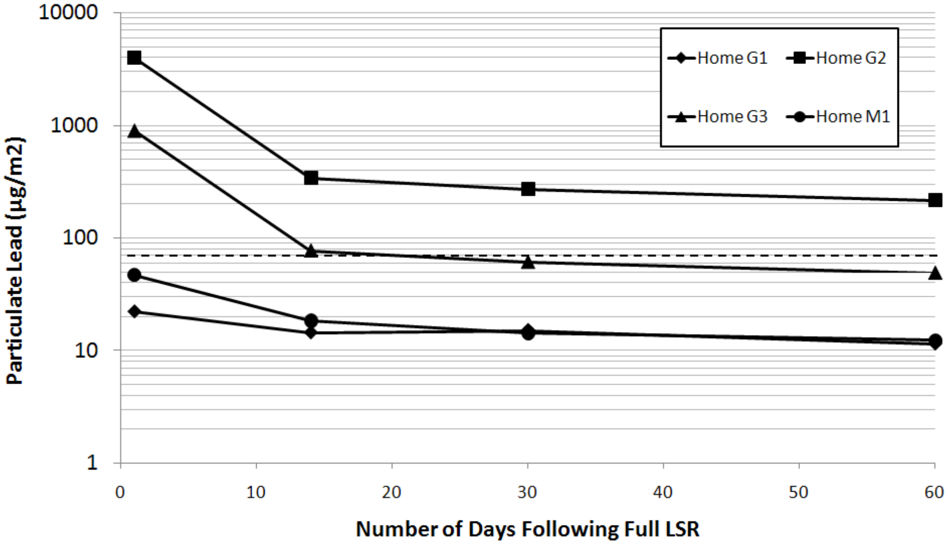

**Figure 5: Normalized Particulate Lead Release over Time from In-Home Plumbing following Full LSR**

### 3.2.3 The Impact of Hydrodynamic Disturbance on Lead Release

Hydrodynamic disturbances may also have an impact on particulate release from galvanized plumbing. In Figure 6, particulate lead and iron release have been normalized to area sampled for high flow velocity and water hammer samples collected at the time that profile sampling was conducted. Normalized particulate lead and total iron release in high velocity samples and water hammer samples are compared with normalized release from in-home plumbing calculated from profile samples (from Figure 1).

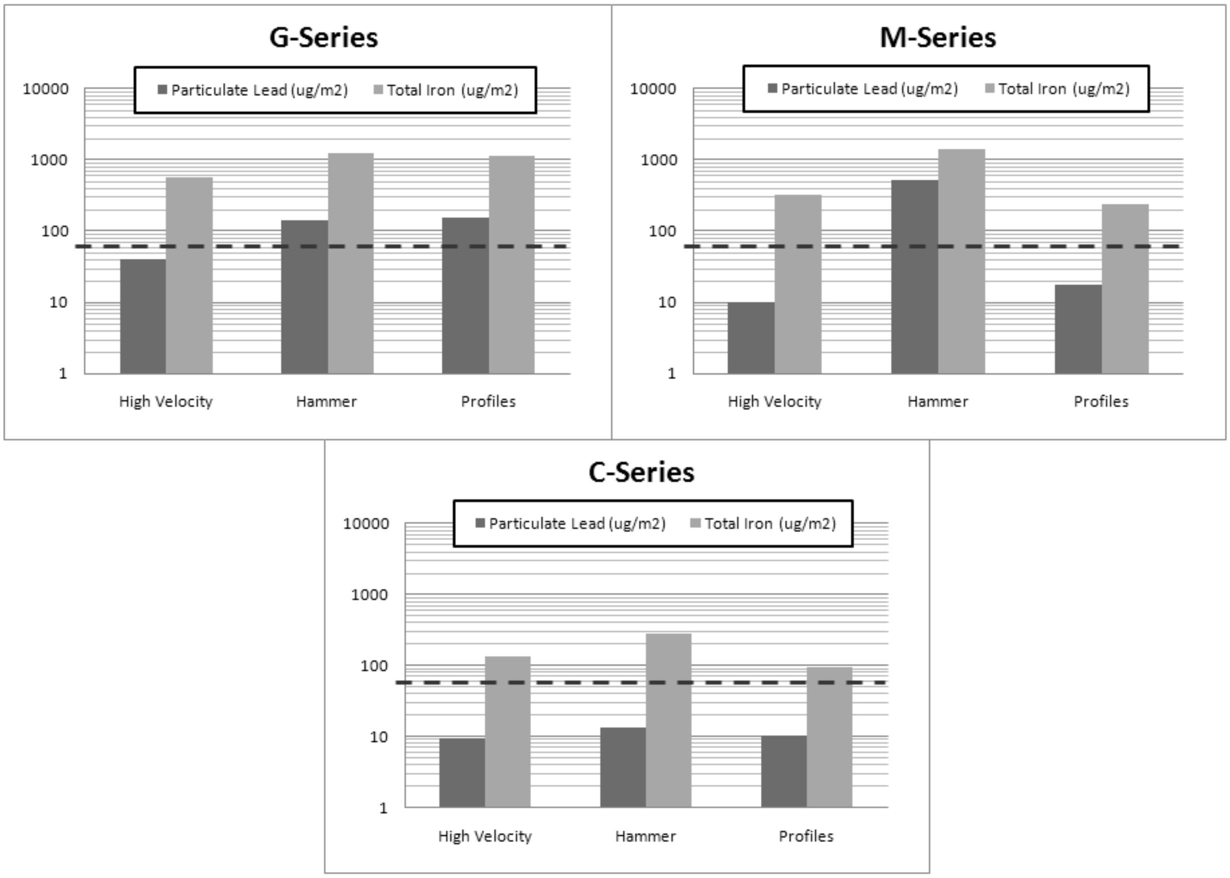

**Figure 6: Normalized particulate lead and iron release (μg/m$^2$) observed in high flow velocity samples, water hammer samples and in-home plumbing profiles conducted at G-Series, M-series, and C-series homes**

High flow velocity did not cause a meaningful increase in particulate lead or total iron release relative to that observed in the post-stagnation plumbing profiles in any of three series of sample sets. However, particulate lead release in high velocity sampling was 4 times greater in G-series homes than that observed in the M and C series. Water hammer produced lead release from galvanized plumbing similar to that observed in the profiles, at a level higher than the equivalent LCR action limit. In the M series, water hammer appears to have caused an order of magnitude increase in particulate lead release relative to that observed in profile sampling in the same series. However, inspection of the M series sampling data indicates that this increase is almost entirely attributable to a single sample from home M-3 on 11/26/2007 (Figure 7), where 2.2 mg/L of lead and 4.5 mg/L of iron were detected in the first 1-liter sample collected after induced water hammer. This first sample liter was part of a full profile conducted immediately following the induced water hammer on this sample date. Concentrations in 14-liters of additional samples collected were substantially lower, at averages of 22.1 and 61.7 µg/L of lead and iron, respectively. If the values from the M-3 samples are omitted from the normalized mass release calculations, average mass release from the M-series are 28 and 198 µg/m$^2$ for lead an iron, respectively. These values are comparable to those observed for the M-series profiles.

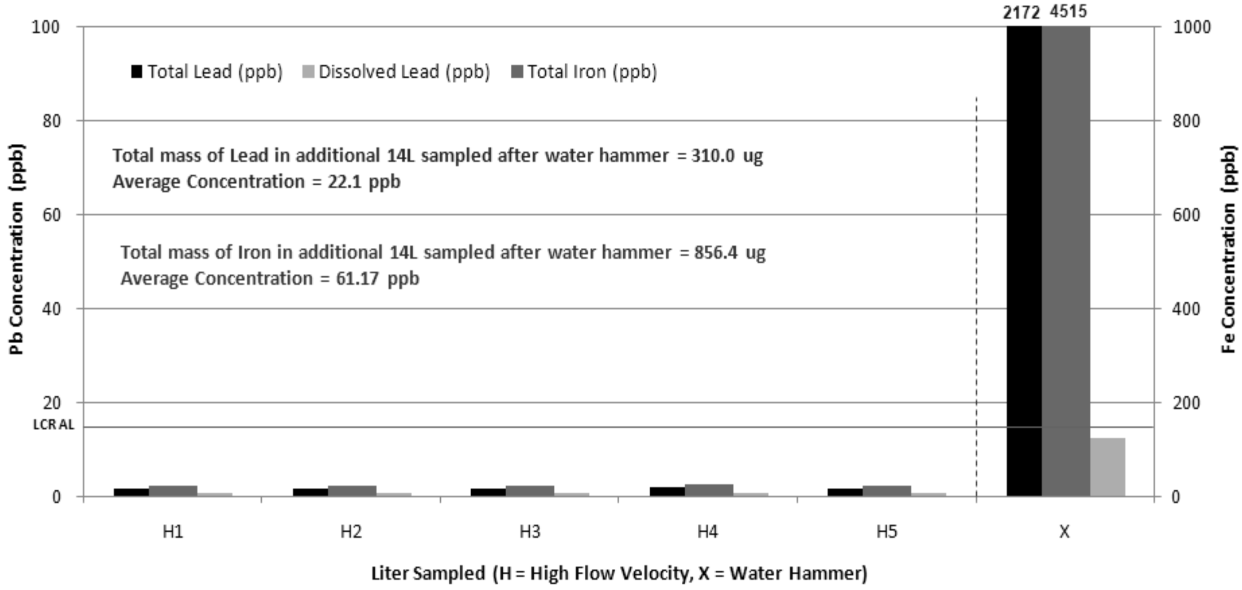

**Figure 7: High Velocity and Water Hammer Samples Collected at Home M-3 on 11/26/2007**

### 3.2.4 The Correlation between Particulate Lead and Iron Release

Corresponding spikes in both lead and iron such as that shown in Figure 7 are not isolated events. Profiles assembled in the appendices of this report include numerous instances of simultaneous elevations in both lead and iron release. Figure 8 shows the distributions of particulate and dissolved lead release (x-axis) versus total iron release (y-axis) in all samples collected at G-series homes. The coefficient of linear correlation between iron and particulate lead (R$^2$ value) is only 0.5 indicating that the linear correlation in this data is not particularly strong. It should be noted, however, that evaluation of a linear coefficient implies uniform adsorption and release, which is not consistent with the variation from site to site observed in Figure 2. The distribution between particulate lead and iron under 100 and 1000 µg/L, respectively, has a high degree of scatter. Nonetheless, a relationship is evident. Every point where iron is over about 1 mg/L corresponds to an elevated concentration of particulate lead. In addition, distinct portions of the distribution above and below the regression line appear linear. When distributions from locations G-2 and G-3 are viewed separately, as shown in Figure 9, linear correlations become stronger, and two apparently linear portions of the distribution shown above and below the regression line in Figure 8 appear to be associated with individual sites.

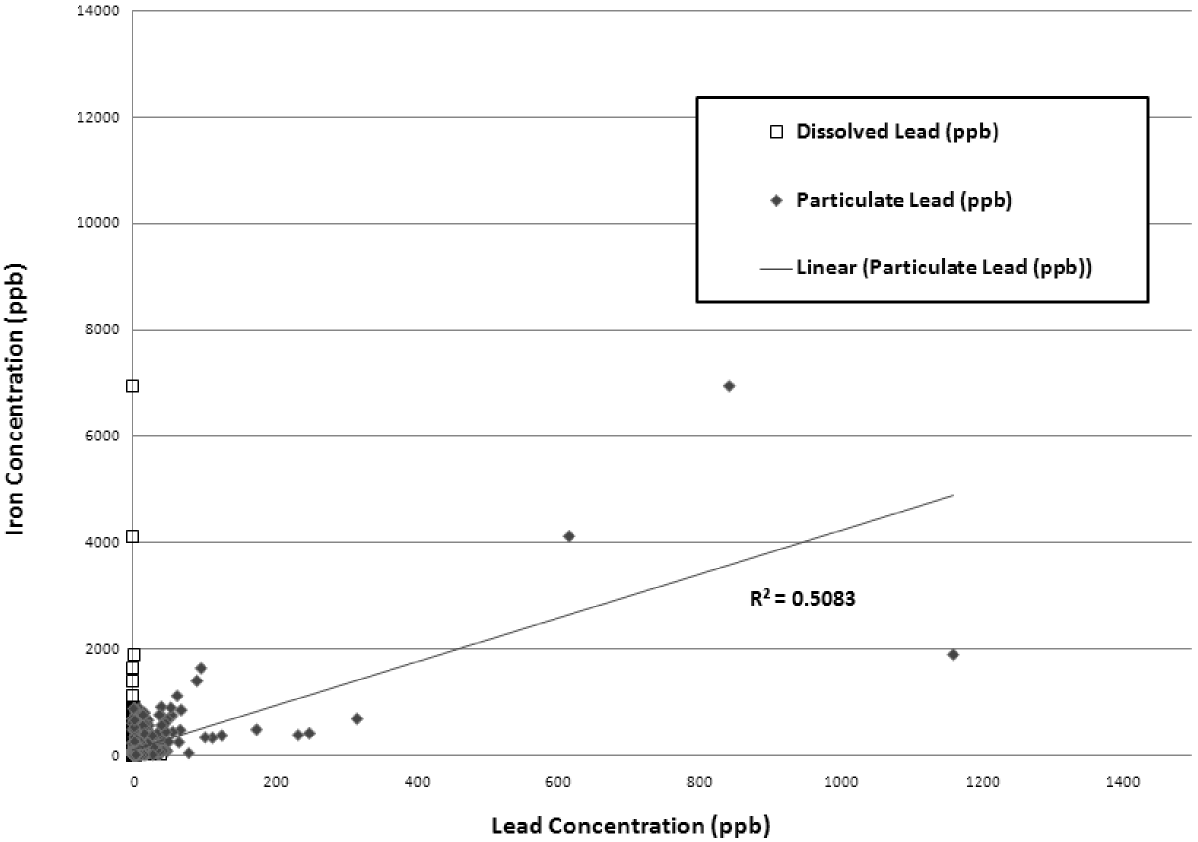

Figure 8: Distributions of particulate and dissolved lead versus iron concentrations in samples collected from G-series locations

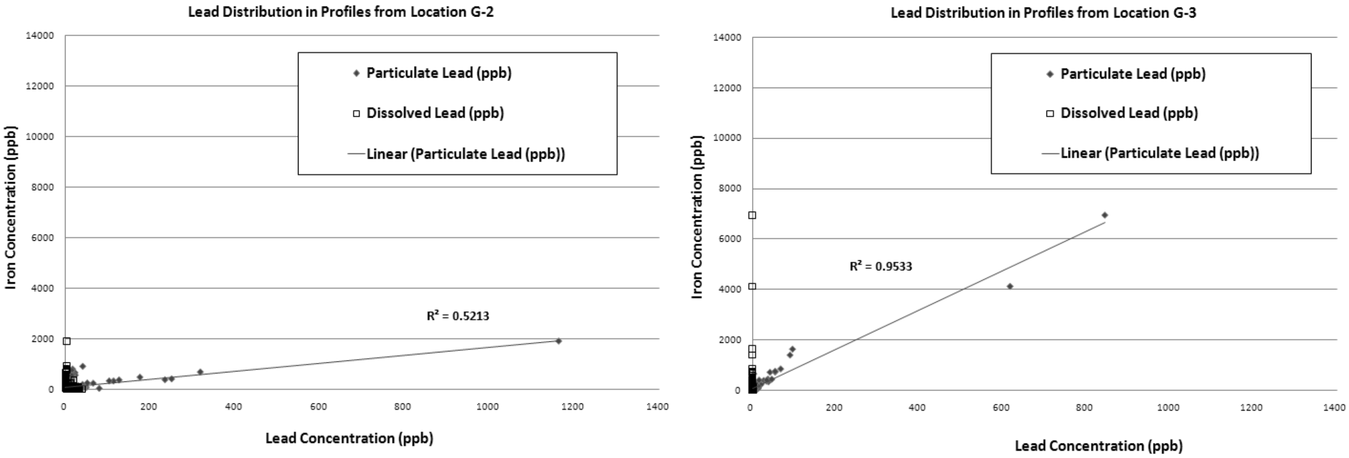

Figure 9: Distributions of particulate and dissolved lead versus iron concentrations in samples collected from locations G-2 and G-3

To investigate the variation in correlation that might occur from site to site, calculated correlation coefficients between particulate lead and iron as well as dissolved lead and iron in the G-series, M-series, and C-series of sample sets are shown in Figure 10. Coefficients were calculated using all samples from each profile including samples associated with the water main, as well as first draw samples, water hammer samples, and high flow velocity samples. The figure includes coefficients calculated at individual locations within a data series as well as the overall coefficient calculated for the series.

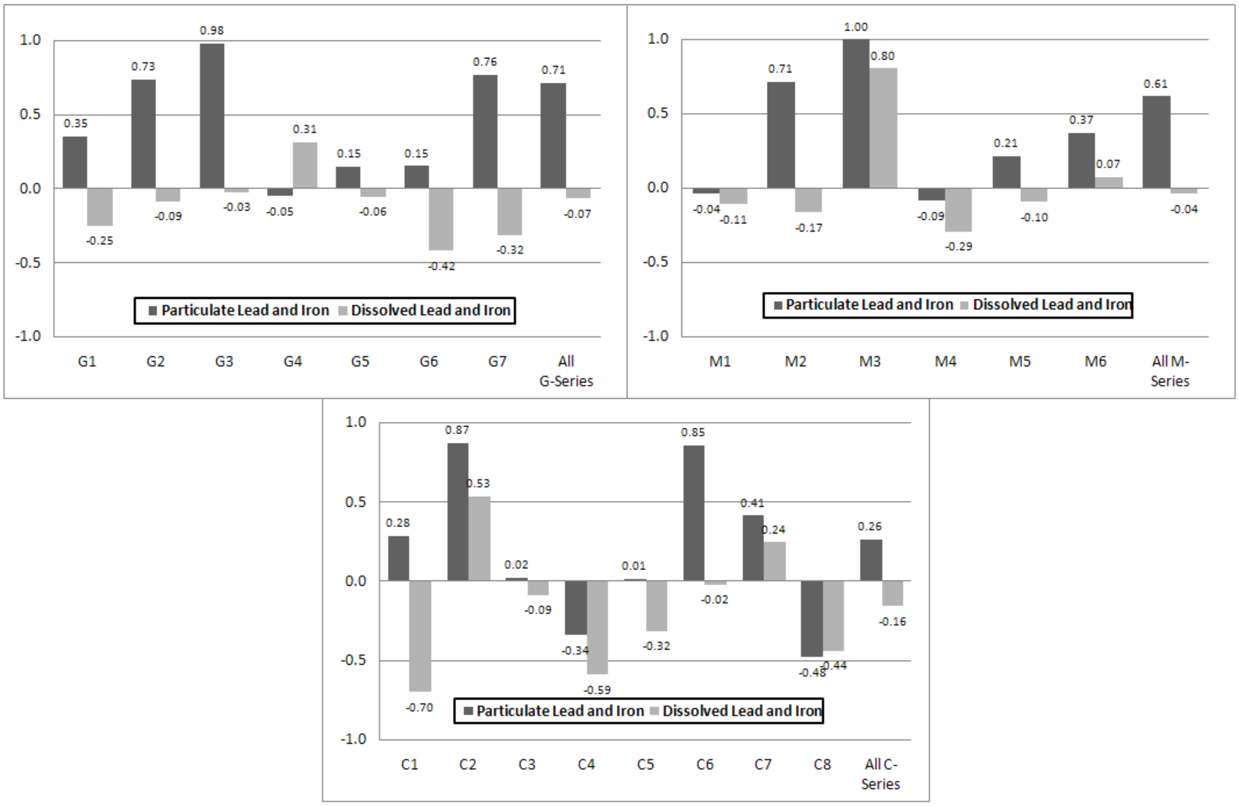

**Figure 10: Calculated correlation coefficients for individual homes and for the G, M, and C-series overall**

Variation in coefficients of correlation from home to home is evident upon inspection of Figure 10. Even in the C-series, where there is no source of iron release from in-home plumbing, the correlation is stronger overall than that for dissolved lead. The coefficient of correlation between particulate lead and iron for 701 samples in the G-series sample set is 0.7, which is reasonably strong over a large sample size. It is 0.6 and 0.3 for the M-series of 400 samples and C-series of 169 samples, respectively.

### 3.2.5 Other Metals

Figure 11 summarizes calculated coefficients of correlation between lead and iron, dissolved lead and iron, lead and zinc, lead and copper, iron and zinc, and iron and copper. These coefficients are calculated from the entire set of 1313 samples collected; including water hammer, high velocity, first draw, and distribution main.

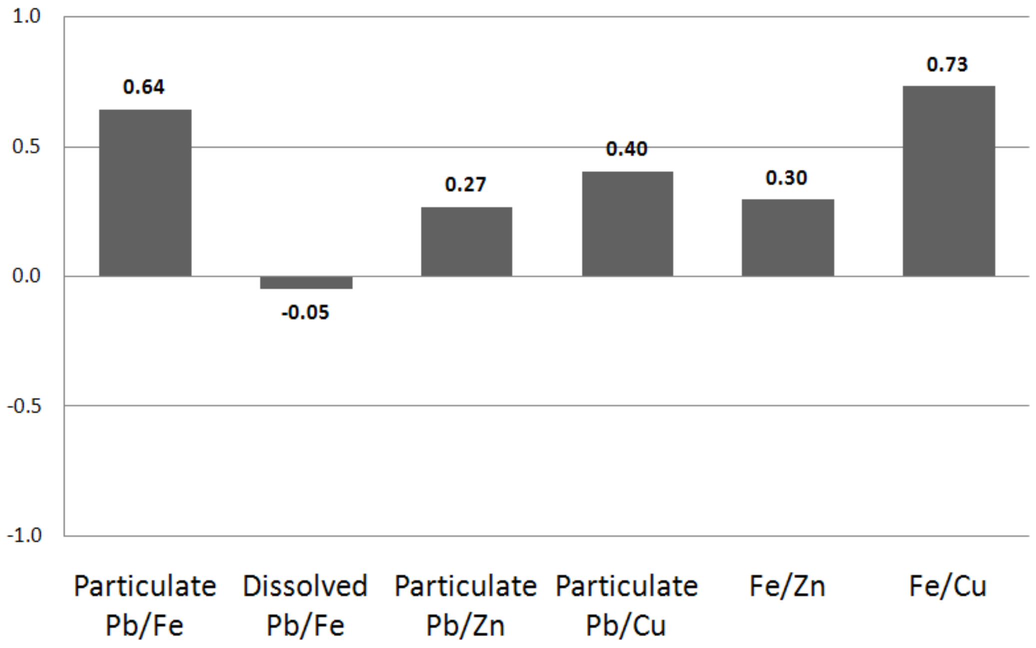

**Figure 11: Calculated Correlation Coefficients between Iron, Lead, Copper, and Zinc**

The correlations of zinc with lead and iron are both relatively low. This is likely the result of the fact that the galvanized plumbing is decades old and almost all of the zinc is probably gone. Areas with high zinc seem to correspond to samples associated with sink fixtures in the samples collected during the study. The correlation between iron and copper is 0.73, which is higher than the correlation between iron and lead at 0.64.

## 3.3 Discussion – Part I

The average lead mass per unit area calculations based on 34 profiles from all stages of service replacement revealed that particulate lead release from galvanized in-home plumbing in the G-series was greater than particulate release from lead service lines. Particulate release from galvanized in-home plumbing, alone, was great enough to exceed the LCR action level, though the dissolved fraction of lead was low relative to that associated with lead service. These results are a meaningful indication that galvanized iron corrosion scales can serve as source of lead in drinking water, and, indeed, the number of profiles upon which the calculations were based conveys a degree of statistical significance.

Lead release has been represented in dissolved and particulate fractions throughout this portion of the study. Dissolved lead release from galvanized plumbing was on average a full order of magnitude less than the particulate form, and thus the emphasis in evaluating results here has been on the particulate fraction. These fractions are based on a simple 0.45 micron filtration step performed in the field, and their meaning is subject to some interpretation. This is especially true regarding particulate lead, which might include a range of lead complexes and minerals. Mobilized mineral forms might be lead carbonates, oxides, or appetites associated with lead corrosion scales. Alternatively they might include ferric or ferrous lead complexes either mobilized directly from a lead-rich iron corrosion scale matrix, or resulting from adsorption of dissolved lead to mobilized iron in solution. Categorization into particulate and dissolved does not allow the characterization of the particles.

Importantly, results of the study indicate that the release of particulate lead from galvanized plumbing can differ substantially in magnitude and behavior from one location to another. In some cases lead release persists at levels exceeding the LCR action level months after full lead service replacement. In other cases where full lead service lines remain in place, lead release from galvanized plumbing appears minimal. Differences in sensitivity to physical and hydrodynamic disturbances were also observed, making it difficult to develop any but the broadest characterizations of the phenomenon of lead release from galvanized plumbing.

Though the magnitude varied substantially with location, a trend of exacerbated lead release from galvanized plumbing immediately following any stage of LSR was observed consistently. In all cases where exacerbation was observed immediately following full replacement, a consistent decreasing trend over subsequent weeks followed. Yet the differences in magnitude of release from home to home make it impossible to develop a general timeframe after which any home with galvanized plumbing would see lead concentrations diminish to below 15 µg/L at the tap following stagnation.

The locational variation in lead release from galvanized plumbing also makes it difficult to establish a consistent correlation between lead and iron release. A relatively strong linear relationship was observed at specific locations, yet the slope of regression lines differed meaningfully. The locational variation is likely indicative of varying lead content of the iron corrosion scale matrix from location to location, where lead content at any given location is a function of the unique history of exposure associated with the premise plumbing. Various factors such as piping configuration or hydrodynamics might contribute to variations in the lead content of galvanized corrosion scales from home to home.

In addition to the evaluation included in this study of the contribution of physical and hydrodynamic forces to these results, other potential factors posited elsewhere, such as lead release from lead bearing faucet components and the potential impact of galvanic coupling of lead and copper service lines, deserve some consideration within this context.

The numerous profiles conducted for this study include multiple instances of elevated lead release within the first 250-mL sampled following stagnation. It is likely that this represents a contribution from lead bearing materials within faucet valves. No effort was made in this study to separate the impacts of lead bearing faucets from general lead release from in home plumbing. In depth investigation of faucet valve materials was not conducted, nor could the variations in faucets from home to home be adequately assessed. As such, accurately quantifying the contribution to lead release of lead bearing materials within faucet valves was not possible. It stands to reason, however, that faucet contributions to lead release associated with in-home plumbing were not meaningfully greater in C-series or M-series homes than those with predominantly galvanized plumbing, and that differences in normalized lead release reported here accurately reflect the contribution to lead release directly from galvanized corrosion scales.

As with lead bearing faucet contributions, no effort was made to quantify the impacts, if any, of galvanic coupling between lead and copper service lines to lead release from lead service lines. While not a focus of this study, it should be noted that in visual inspections of the 21 profiles conducted following partial LSR, in no case was localized elevated lead release observed at the lead/copper junction.

Other factors that could potentially obscure interpretation of residential profile results include the effects of advection and diffusion, and the potential for particulate release into the flow stream as volumes of water move through discrete sections of plumbing toward the sample point. No attempt to quantify the effects of advective and diffusive forces was made in evaluation of these data sets. Visual inspection of individual profiles reveals multiple instances where a consistent level of dissolved or particulate metal strongly associated with a portion of premise piping includes a tail that carries over into an adjacent portion of piping. Thus, dissolved lead associated with a service line, might have an impact on in-home plumbing near the point of entry due to advection or diffusion. Likewise, particulate metal associated with in-home plumbing may manifest in sample volumes associated with service piping near the point of entry.

Though the impact of scouring, evaluated via high velocity sampling, appeared relatively minimal in the field study, it should be noted that mobilization of exposed, friable iron corrosion scale material might occur as sample volumes associated with other portions of premise plumbing move through in-home plumbing to the sample point. During sampling, flow rates are minimized to reduce this occurrence, but the phenomenon must be noted.

Interestingly, though not a primary focus of this study, copper appears to bear a more consistent linear correlation with iron release from galvanized plumbing than does lead. This indicates the likely existence of a strong affinity for copper with iron corrosion scales. Due to the much higher action level for copper in the LCR, release of copper from galvanized plumbing may not present as great a concern for compliance issues as the lead phenomenon, but the perceived affinity should be considered when evaluating trace metals release from galvanized plumbing at any particular home.

Given the variation in behavior and magnitude of lead release results observed in the profiles conducted for this study, a greater understanding of sorptive properties and surface morphologies is desired to adequately characterize the relationships that determine lead release from galvanized iron corrosion scales. The second portion of this study was conducted in order to develop this understanding.

# 4.0 Part II – Investigation of Lead Sorption to Ferric and Ferrous Iron and Characterization of Galvanized Iron Corrosion Scales

## 4.1 Jar Testing

### 4.1.1 Methodology

Simple jar tests were conducted to investigate the adsorption of lead to ferric and ferrous iron. The potential affinity of lead and other trace drinking water constituents to adsorb to ferric iron corrosion scales has been previously noted. Whether the affinity exists to the same degree for adsorption to ferrous corrosion scales is less well understood.

Three jars were prepared using deionized water at pH 7.5 and alkalinity 25 mg/L-$CaCO_3$. Jars were 2-L molded, acrylic jars manufactured by Phipps and Bird. Lead nitrate ($Pb(NO_3)_2$) was added to the jars to produce lead concentrations of 100 mg/L. In a preliminary attempt at the experiment, it was determined that up to 35% of dissolved lead added to the polymer jars could adsorb to the sides of the containers. The jars were allowed to sit over night until observed dissolved lead concentrations stabilized. Solid ferrous sulfate ($FeSO_4$) was added to Jar 1 to produce an iron to lead molar ratio of 100:1. Ferric oxide ($Fe_2O_3$) was added to Jar 2 to produce the same iron to lead molar ratio. Ferrous sulfate was selected to represent freshly corroded iron, and ferric oxide was selected to represent a developed iron scale. No iron was added to Jar 3. All three jars were stirred at 100-rpm for 15 minutes in a standard jar tester with 1" x 3" paddles. Dissolved lead samples were then taken from each jar. Nitric acid was added to each jar to $pH < 2.0$, and the jars were allowed to sit overnight to mobilize any lead that had sorbed to the sides of the containers. Total lead samples were analyzed after 24 hours.

Total lead was measured in unfiltered samples from the jars using a Hach HSA-1000 bench-top lead test. Dissolved iron concentration was measured by filtering an aliquot of solution from the jar through a 0.45 micron membrane pre-rinsed with deionized water, followed by analysis with the Hach unit. Hach unit measurements were verified by Amtest laboratories of Kirkland, WA using ICP-MS.

### 4.1.2 Results

Figure 12 summarizes dissolved lead concentrations observed prior to addition of iron compounds, after iron addition and mixing. Total lead concentrations measured following the 24-hour nitric acid bath are also provided. Both ferrous and ferric iron adsorbed effectively all of the observed dissolved lead concentration in Jars 1 and 2, respectively. 96% of the observed dissolved lead was adsorbed to ferrous iron in Jar 1 after 15 minutes of mixing. 98% of the observed dissolved lead was adsorbed to ferric iron in Jar 2 after 15 minutes of mixing. Between 18% and 30% of dissolved lead added to the solutions likely adsorbed to the plastic containers prior to iron additions.

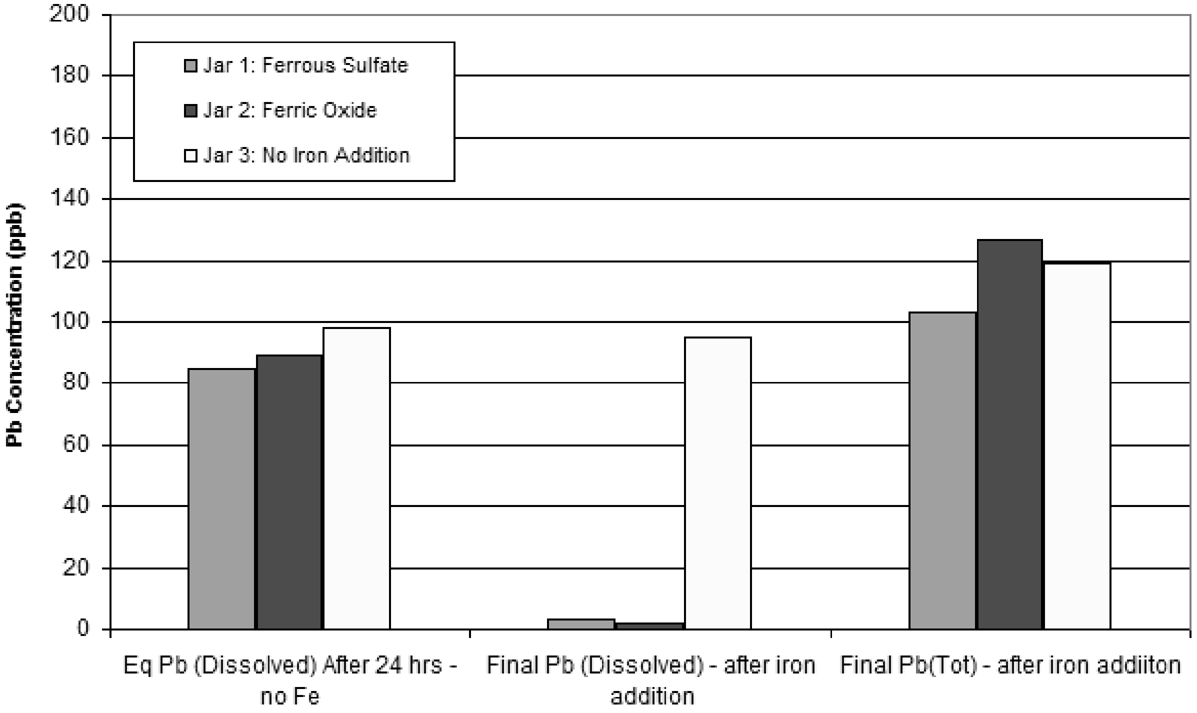

**Figure 12: Results of Jar Tests showing Adsorption of Dissolved Lead to both Ferric and Ferrous Iron**

## 4.2 Galvanized Iron Surface Analyses

Corrosion scales from five segments of galvanized iron in-home plumbing removed from homes throughout the DCWASA service area were analyzed using Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDAX) at the University of Cincinnati Department Of Geology. Results revealed widely varying amounts and types of lead incorporated into the corrosion scales, not only between pipes collected at different locations, but between discreet areas of the corrosion scale surface within the same pipe.

Iron scales observed were typically rough textured, ranging from orange-brown to black in color. Typically the scales were composed of two distinct layers; a soft outer layer contacting the water and a harder under-layer closer to the pipe metal. The hard under-layer was typically composed of well-crystallized goethite, a ferric oxy-hydroxide, and magnetite, an oxide with ferric and ferrous components. The softer, outer layer was typically composed of less well-crystallized goethite and manganese oxide as shown in Figure 13.

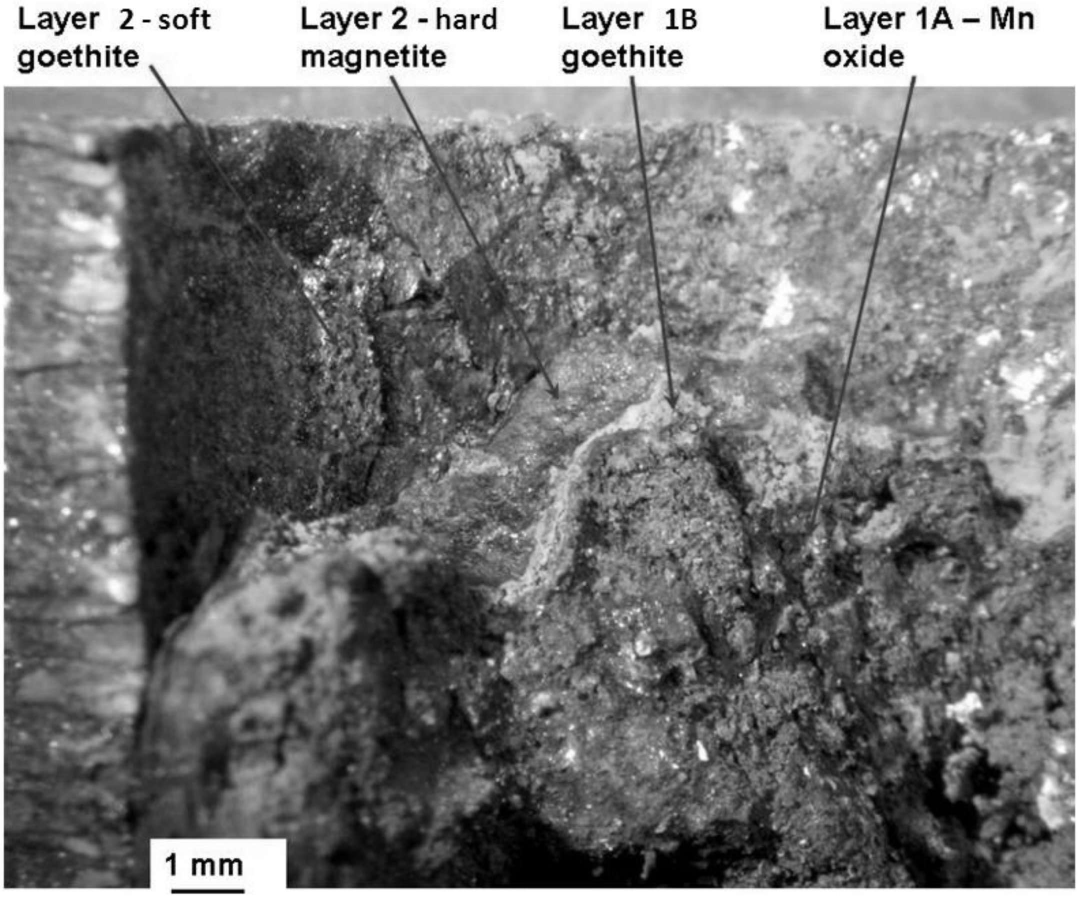

**Figure 13: Soft outer layer (Layer 1) and hard inner layer (Layer 2) observed in corrosion scales on galvanized iron pipes removed from DCWASA service area.**

Portions of both layers were scraped from the surface of the pipes for bulk chemical analysis. Hard layers were separated into magnetite and non-magnetite fractions. Individual tubercles were also separated from the corrosion scales and divided into outer shells and inner shells.

The composition of both the hard and soft scale scrapings included lead ranging from a few hundredths of a percent to as high as 8.3 percent by weight. Lead-rich compositions were typically accompanied by elevated manganese and phosphorus within the scale matrix, especially in the soft outer layers. These scales were also often rich in copper. Typically lead, manganese, and phosphorus composition of tubercles was somewhat lower than the hard and soft scale layers.

The surface analysis indicated a non-homogeneous distribution of lead and other trace metals. A number of discrete, lead rich deposits were observed in which lead minerals such as pyromorphite and apatite were identified. In such cases these minerals appear to be embedded within the scale matrix near the surface (see Figure 14). On other pipes, where phosphorus and vanadium composition is less, lead oxide and carbonate minerals appear embedded in discrete deposits. The weight percent of lead at many such deposits observed was as high as 35%.

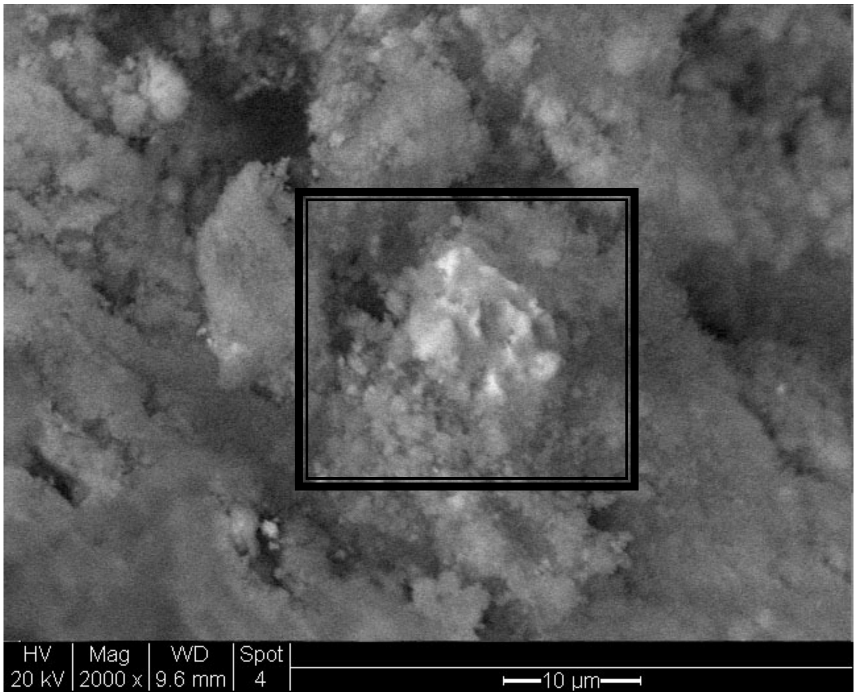

**Figure 14: Discrete micron-sized Pb-rich spot on corrosion scale surface likely consisting of the mineral Pyromorphite.**

## 4.3 Discussion – Part II

Results of jar testing establish a strong affinity of dissolved lead with both ferric and ferrous iron. Whether adsorption of dissolved lead to iron is through true chemical bonding and formation of oxides or hydroxides of lead-iron complexes, or through some other mechanism akin to physical capture is unclear. Regardless of the mechanism, dissolved lead will sorb to mobilized iron, ferric or ferrous, and in turn settle out of solution. This co-precipitative process would almost certainly occur inside galvanized plumbing, leading to deposition of lead-rich iron complexes on pipe surfaces forming corrosion scales. It stands to reason that, over time, a thick, lead-rich corrosion scale could develop on galvanized in-home plumbing, presenting the potential for lead release from these scales for prolonged periods, even after other sources of lead (i.e. lead service lines) have been removed. Factors such as pipe orientation, length, or degree of passivation of a lead service line, or in-home hydraulics might impact the degree to which lead is incorporated into galvanized plumbing scales at a given location. These deductions are consistent with results from Part I, indicating that the slopes of regression lines establishing the relationship of iron and lead release vary from one location to another.

These deductions are also supported by the results of the surface analysis performed. While some galvanized pipes had corrosion scales with lead content only to a few hundredths of a percent by weight, others contained lead to 7 or 8 percent throughout all layers of the corrosion scale. High lead was observed in the crystalline matrix of even the deep, hard, under layer of analyzed scales, indicating the likelihood of a long history of lead/iron co-precipitation processes contributing to the formation of the scales.

The presence of relatively large (i.e. 1-micron) particulate lead minerals such as lead apatites, oxides, and carbonates, embedded within the iron scale matrix establishes the potential for adsorption by physical capture within the rough corrosion scale surface. The presence of lead within iron corrosion scales, then, cannot be interpreted as solely the result of a particular sorptive mechanism. Rather, the spectrum of sorptive mechanisms, physical and mechanical, must be considered possible contributors to the formation of lead rich iron corrosion scales.

As formation of lead-rich iron corrosion scales may be influenced by multiple factors, lead release may occur through a variety of mechanisms. The most likely would appear to be through mobilization to solution of lead-iron compounds from the soft, exposed, outer layer of the corrosion scale during periods of stagnation. This would likely lead to a relatively constant ratio of lead to iron observed at the tap for a given location, based on the lead content of corrosion scales developed over time. Alternatively, physical or hydrodynamic disturbance of the corrosion scale could dislodge embedded particulate lead minerals, causing potential spikes in lead observed at the tap. Such forces would also contribute to scouring of the soft outer scale layer, leading to predictable lead/iron ratios observed at the tap.

Lead iron ratios implied by the slope of regression lines applied to sample results from various sites profiled in Part 1 are much higher than the 8% content observed at the high end of the range of pipes subjected to surface analyses. This raises questions about the specific mechanisms of release. Lead may be released from the scale matrix preferentially, without corresponding iron release proportional to the lead content of the scale. Various explanations for how this could occur might be presented. The presence of phosphates in the water, for example, could induce mobilization of lead from iron scales in the formation of highly insoluble lead-apatite minerals. Such hypotheses, based on the surface analyses and behavior observed in Part 1 of the study remain highly conjectural. Additional investigation is required into mobilization of lead from iron scales before a comprehensive theory can be developed. Such investigations are the focus of Part 3 of this study.

# 5.0 Part III – Pipe Loop Investigations of Lead Release from Iron Corrosion Scales

To further investigate the behavior and mechanisms of mobilization of lead from galvanized household plumbing two pipe-loop experiments were performed. The first, conducted at DCWASA, utilized 1-foot lengths of ¾” galvanized iron plumbing removed from homes in the DCWASA service area with existing lead service lines. The second, performed at the HDR Applied Research and Technology Center (ARTC) in Redmond, WA, utilized new, artificially corroded, black iron piping to which lead was adsorbed within the laboratory setting.

## 5.1 DCWASA Pipe Loops

### 5.1.1 Methodology

Four pipe loops were constructed using 1-foot segments of ¾" galvanized iron plumbing removed from separate homes from throughout the DCWASA service area. Each home had existing lead service lines at the time the plumbing segments were removed. Each pipe loop consisted of a 10 L Nalgene reservoir, a ball valve, and flow meter for flow control, and a dedicated circulation pump. These elements were connected with 1/4" I.D. chemical resistant flexible tubing in the configuration shown in Figure 15.

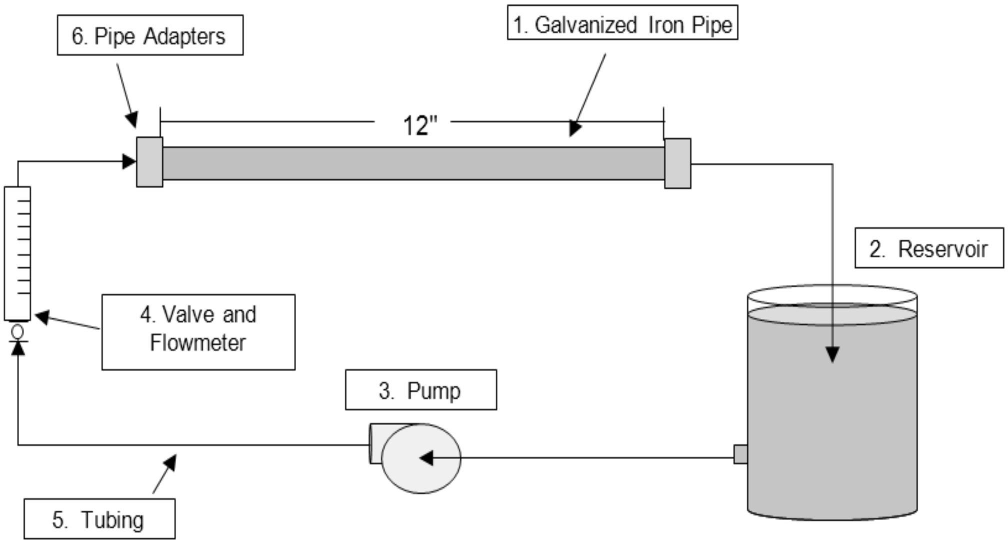

**Figure 15: Schematic of DCWASA Pipe Loop Configuration**

Treated DCWASA drinking water from the WA was used in each of the four loops. Water was changed once per week, at which time pH, total chlorine, and phosphate were measured. The same parameters were also measured at the end of each week. Typical water quality observed during testing was consistent with the following:

- pH = 7.4 – 7.8
- NH2Cl = 3.0 - 3.5 mg/L-Cl2
- Orthophosphate = 2.5 - 3.0 mg/L-PO4

Water changes and sampling involved the following steps:

1. **First Flush Sample**: New water was added to the reservoir and circulated through the loop. The first flush of water through the pipe segment invariably caused increased lead release, visibly distinguished by reddish discoloration. Several hundred milliliters of first flush water was diverted from discharging to the reservoir until the reddish discoloration in the effluent subsided. Filtered and un-filtered samples of first flush effluent were collected each week throughout the course of testing.

2. **End of Week Filtered Sample**: Prior to changing the water after running the pipe loop for one week, the reservoir was mixed and a filtered sample was collected.
3. **Shut Down and Acid Rinse**: After collecting a filtered sample from the reservoir, the pump was turned off and the pipe and tubing water was siphoned to the reservoir. The reservoir was acidified to pH < 2.0 to mobilize any lead that may have adsorbed to the sides of the reservoir. The acidified reservoir was allowed to sit for 24 hours and a sample from the reservoir was collected. Following collection of the acidified reservoir sample, the reservoir was emptied and rinsed, and new water was added.

All filtered samples were collected by filtering an aliquot of solution through a 0.45 micron membrane pre-rinsed with deionized water. Filtered and unfiltered samples were acidified and sent to WA for analysis. Each sample was analyzed with ICP-MS for lead, iron, zinc, manganese, and copper.

### 5.1.2 Phases of Testing

Each of the four pipe loops was subjected to an identical sequence of test phases developed to identify various hydrodynamic impacts on lead release from galvanized iron plumbing. Prior to commencement of the first test phase, each pipe loop was run continuously for a 2 week conditioning period. During this period, iron release from the newly excavated pipe segments was elevated to an extent that bore little resemblance to actual in-home plumbing. Water was changed during the conditioning period based on visual inspection of the deepening red color in the reservoir water. When the corrosion scales had attained greater stability, and iron release had subsided, regular testing and sampling, as described above, commenced. The sequence of three test phases following the conditioning period proceeded as follows:

1. **Phase 1, Continuous flow**: Each week, the four pipe loops were subjected to continuous flow at a rate of 0.5 gallons per minute. This phase continued for 8 full weeks.
2. **Phase 2, Stagnation/Flow**: To approximate flows observed in actual homes, a daily regimen of alternating stagnation and flow was conducted. The regimen consisted of 8 hours of stagnation followed by 16 hours of flow at 0.5 gallons per minute. Phase 2 continued for 5 full weeks.
3. **Phase 3, High Flow Velocity:** During phase 3 constant, flow was increased to greater than 1.5 gpm, and maintained continuously for the duration of each week. The elevated flow rate was implemented to examine the potential effects of scouring action on corrosion scales. Phase 3 continued for 8 full weeks.

## 5.2 Results

As with results of profiling at homes in the distribution system, behavior from pipe loop to pipe loop was varied. The results are discussed individually for each pipe.

### 5.2.1 Loop 1

Figure 16 summarizes total lead and iron concentrations measured at the end of each week in the acidified reservoir of pipe loop 1.

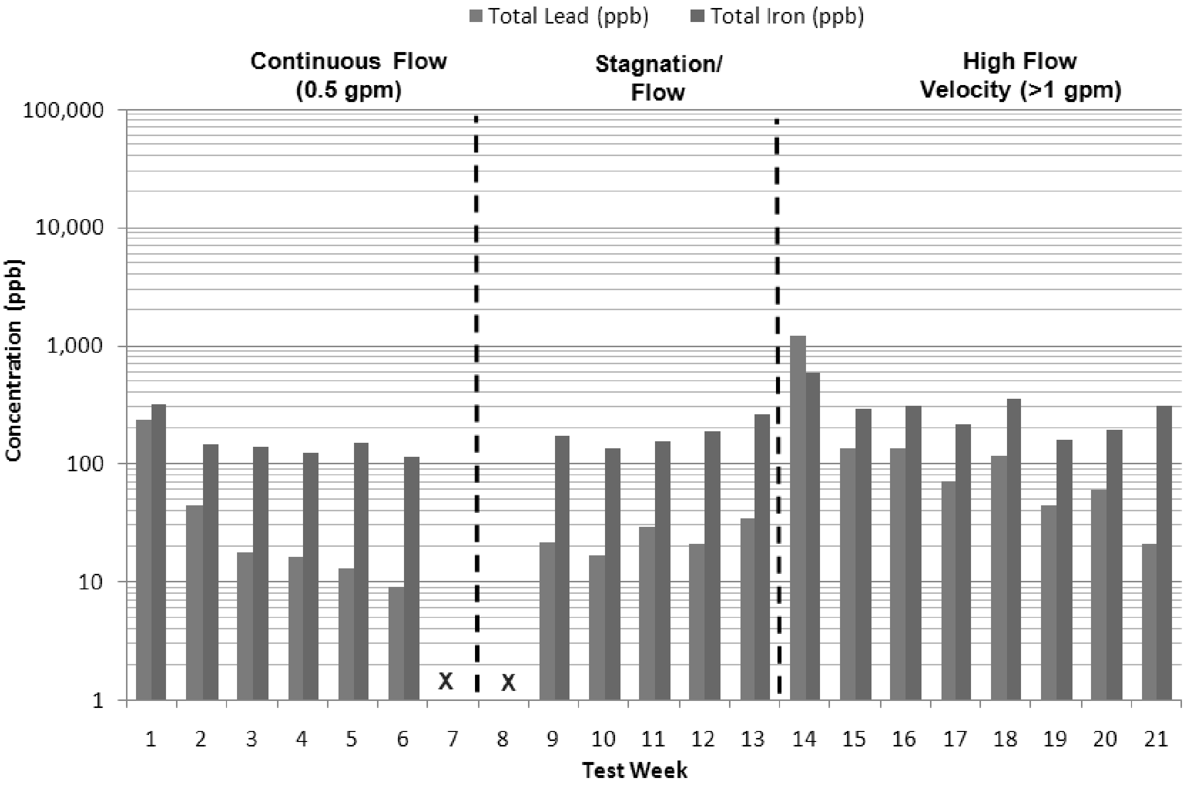

**Figure 16: End of Week Total Lead and Iron Concentrations from the Pipe loop 1 Reservoir following Acidification**

During the first phase of testing, a gradual decrease in lead release from loop 1 is evident. In the first week of testing, lead release was very high relative to the following weeks of this phase. A decreasing trend in release is evident in weeks 2 through 6. Acidified samples were not collected during weeks 7 and 8.

During the second phase of testing, lead release appears somewhat elevated in relation to phase 1 results. The apparent trend of decrease in lead release observed through phase 1 is no longer apparent in phase 2, where concentrations observed in weeks 11 through 13 are generally higher than those observed during weeks 9 and 10.

A meaningful increase in lead release is apparent during phase 3 where flow rate was substantially increased. A general trend of decreasing lead release is observed during this period, but in weeks 19 and 20, measured concentrations remain much higher than typically observed in previous phases.

In general, first flush samples (results provided in appendix) followed similar release trends throughout testing, though magnitudes of iron and lead concentrations observed were typically twice as high as those observed in acidified reservoir samples. Zinc release trended similarly, but manganese release varied little from week to week.

### 5.2.2 Loop 2

Figure 17 summarizes total lead and iron concentrations measured at the end of each week in the acidified reservoir of pipe loop 2.

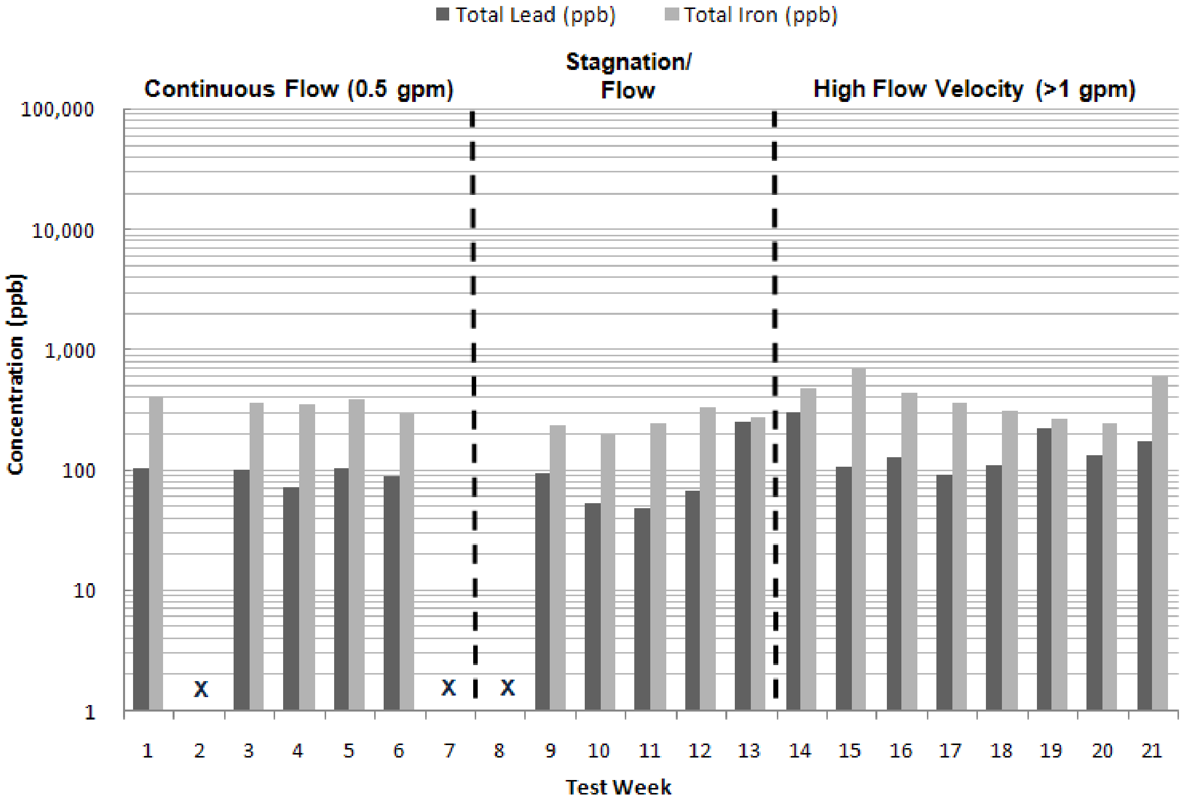

**Figure 17: End of Week Total Lead and Iron Concentrations from the Pipe loop 2 Reservoir following Acidification**

Overall, lead release from loop 2 was greater than that observed in loop 1. The increase in observed iron release relative to loop 1 is less pronounced. Unlike loop 1, a decreasing trend in lead release was not observed during phase 1. An overall decrease in observed lead release is apparent in weeks 10 through 12, but a great increase in lead release was observed during weeks 13. Similar to loop 1, a meaningful increase in lead release occurred when the flow rate was increased during phase 3.

In general, first flush samples (results provided in appendix) were much higher than acidified reservoir samples, and release trends differed. An overall decrease in release is apparent in first flush samples collected during phase 3.

### 5.2.3 Loop 3

Figure 18 summarizes total lead and iron concentrations measured at the end of each week in the acidified reservoir of pipe loop 3.

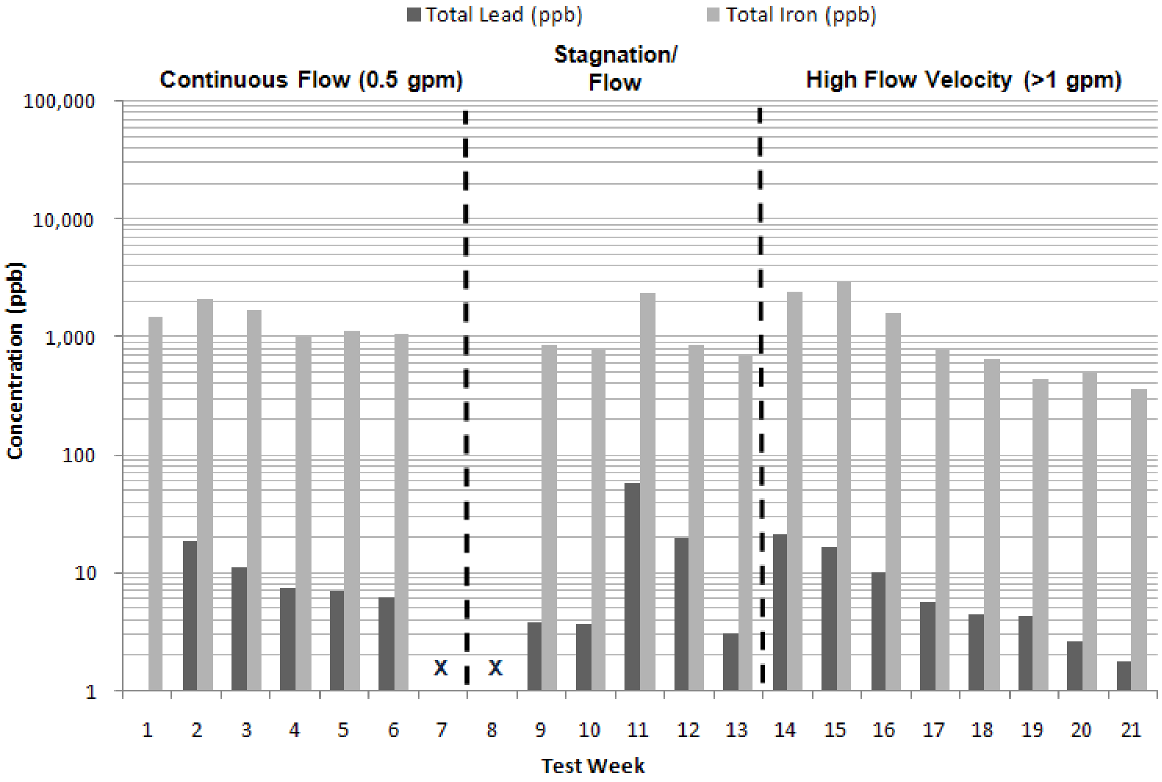

**Figure 18: End of Week Total Lead and Iron Concentrations from the Pipe loop 3 Reservoir following Acidification**

Overall, loop 3 released less lead and more iron across all three test phases than did loops 1 or 2. Lead release from loop 3 exhibited decreasing trends in phases 1 and 3. Lead release during phase 2 varied substantially from week to week.

First flush lead release from loop 3 was also much lower than that from loops 1 and 2 (Results provided in the appendix). A decreasing trend appears over the first few weeks

### 5.2.4 Loop 4

Figure 19 summarizes total lead and iron concentrations measured at the end of each week in the acidified reservoir of pipe loop 4.

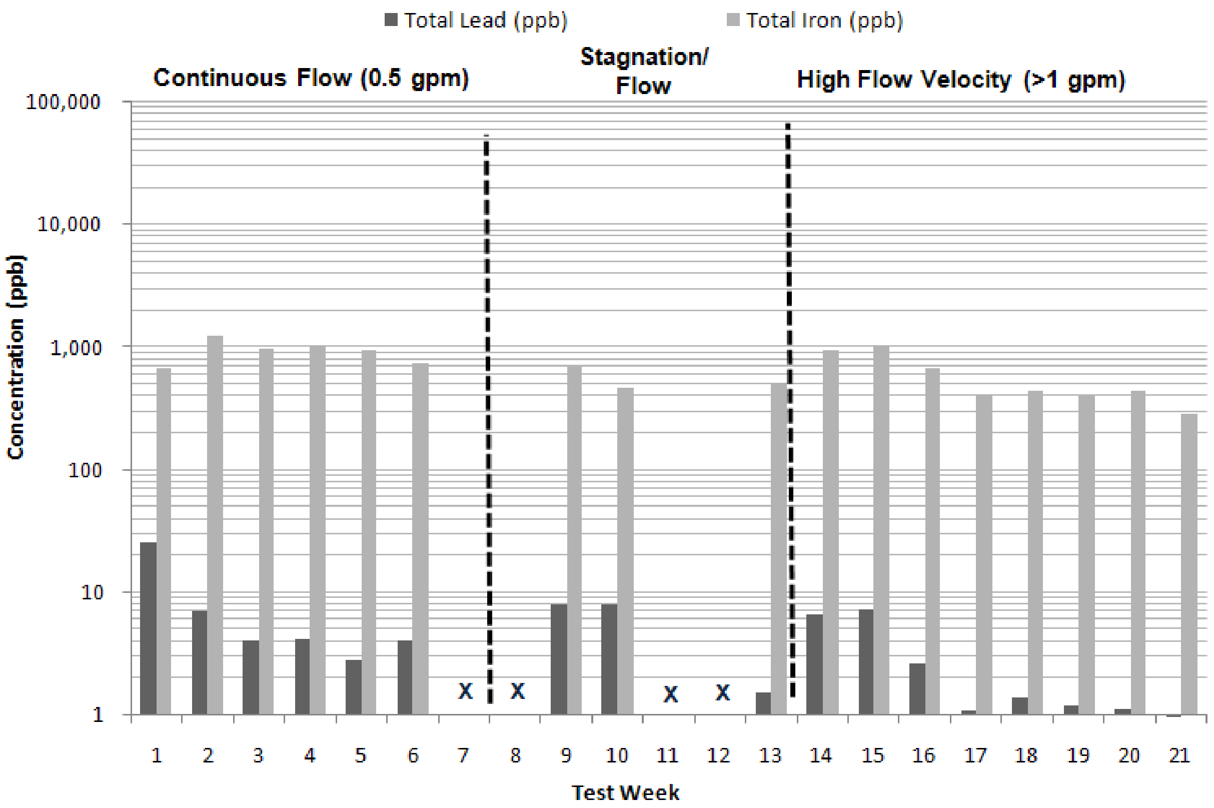

**Figure 19: End of Week Total Lead and Iron Concentrations from the Pipe loop 3 Reservoir following Acidification**

Lead release from loop 4 was minimal. With the exception of a decreasing trend during phase 1, no meaningful changes in lead release occurred throughout testing. First flush samples were likewise minimal (results provided in appendix).

## 5.3 ARTC Pipe Loops

### 5.3.1 Methodology

The four pipe loops constructed at ARTC in Redmond, WA were developed to examine adsorptive processes as well as release. The principal difference between the ARTC loops and the DCWASA loops was the pipes themselves. The ARTC loops used new 1-foot segments of ¾" black iron, typically used for gas piping. These loops were subjected to an accelerated corrosion process, in which they were filled with 1.0 normal hydrochloric acid and allowed to stand overnight. The pipes were emptied and exposed to the atmosphere for 8 hours before being refilled with HCl for another overnight stagnation. The process was repeated several times until a thick corrosion scale formed on the inside of the pipes, as shown in Figure 20. Each pipe was subjected to identical treatment.

Figure 20: Iron Corrosion Scale Developed on Black Iron Pipes for Pipeloops at ARTC

The pipe loop apparatus differed from those constructed at DCWASA only in reservoir size and the addition of a bypass. ARTC reservoirs were 20 liters. A valved bypass was installed around the pipe segment in each loop, so that during acidification of the reservoir the pipe could be closed off, while recirculation of the acidified solution was continued. By this mechanism, lead and iron that may have adsorbed to the inside of the pipe loop tubing could be scoured away and accounted for in sampling. A sample tap was also added to each loop to drain the water that stagnated in the pipe segment during acidification and bypass. Materials used in DCWASA and ARTC loops were otherwise identical.

Sampling at ARTC followed a similar protocol to that at DCWASA as well. A first flush sample, consisting of an aliquot taken from the first 500 mL to flow through the pipe after a water change, was taken at the beginning of each week. An acidified reservoir sample was collected at the end of each week of testing, as well. Because of the addition of the bypass to ARTC pipe loops, water stagnated within the pipe segment during the acidification process. This stagnated water was sampled at the end of the acidification process, prior to the water change each week.

### 5.3.2 Loop characterization and Phases of Testing

Prior to commencing test periods, each loop was subjected to an identical four week “dosing” phase, in which high concentrations of dissolved lead were produced in each loop by sequential additions of a $Pb(NO_3)_2$ solution (40 mg/L-Pb) with the intention of promoting the adsorption of lead to the fresh iron corrosion scales. During the dosing phase, water was changed on a weekly basis, at which time the reservoir was acidified overnight while the pipe bypass was used to avoid exposing the pipe itself to the acidified water. The acidified reservoir water and stagnated water in the pipe were sampled prior to the water change. A mass balance was conducted by subtracting the lead captured in the reservoir and stagnated pipe samples from the known mass of lead added to each pipe. Over the course of the four week dosing period, the four pipes exhibited near identical behavior. Table 6 summarizes the total lead mass added to each loop over four weeks, the total lead mass captured in acidified reservoir and stagnated pipe samples, and the calculated total lead mass remaining on each pipe at the end of the dosing phase. Each pipe retained approximately 25% of the lead it was exposed to.

**Table 6: Summary of lead mass added, lost, and adsorbed to iron pipes during the four-week dosing phase**

| | Loop 1 | Loop 2 | Loop 3 | Loop 4 |
|---|---|---|---|---|
| Total Pb mass added to loop in solution (mg) | 88.3 | 88.0 | 88.3 | 88.7 |
| Total Pb mass observed in samples (mg) | 64.6 | 66.5 | 67.1 | 63.6 |
| Total Pb mass remaining on loop (mg) | 23.6 | 21.5 | 21.2 | 25.1 |
| % of Pb mass remaining on Pipe | 27% | 24% | 24% | 28% |

Following the dosing phase, a six-week test period was commenced during which lead release was monitored under a variety of water quality and hydrodynamic conditions. The water quality and test phases associated with each loop were developed to examine the effects of disinfection, corrosion control, and hydrodynamics on lead release from iron corrosion scales as follows:

- Loop 1: Control loop with baseline water quality consisting of dechlorinated Seattle tap water with alkalinity ~20 mg/L-$CaCO_3$ and pH adjusted to 7.0. Water quality was never changed in this loop.
- Loop 2: ORP loop designed to study the impact of changing disinfectant levels on lead release from iron corrosion scales. Testing was broken into two phases: 1) elevated chloramines at 5-mg/L-$Cl_2$, and 2) elevated free chlorine at 5-mg/L-$Cl_2$
- Loop 3: Corrosion control loop designed to study the impact of elevated pH and orthophosphates on lead release from iron corrosion scales. Testing was broken into two phases: 1) pH elevated to8.3, and 2) orthophosphate at 3 mg/L-$PO_4$ and pH 7.5
- Loop 4: Hydrodynamics loop designed to study the impact of flow changes and physical disturbance on lead release from iron corrosion scales. Testing was broken into three phases: 1) Alternating 8 hours of flow with 16 hours of stagnation, 2) alternating flow rate changes between 24 hours at 1.0 gpm and 24 hours at 0.25 gpm, and 3) physical disturbance caused by rapidly closing a solenoid valve installed just down-stream of the pipe in the loop. Water hammer samples were collected by discharging 500-mL from the loop to a graduated cylinder following 4 consecutive closures of the solenoid valve when pipe flow was at over 1.0 gpm.

The six week schedule of water and hydrodynamic changes in each loop is summarized in Table 7.

**Table 7: Summary of water quality and hydrodynamic changes implemented in each loop during the six-week test period**

| | Loop 1 | Loop 2 | Loop 3 | Loop 4 |
|---|---|---|---|---|
| Purpose | Control | Elevated ORP | Corrosion Control | Hydrodynamics |
| Flow Rate | 0.25 gpm | 0.25 gpm | 0.25 gpm | Varies |
| pH | 7.0 | 7.0 | Varies | 7.0 |
| Week 1 | No Change | $NH_2Cl$ @ 5.0 mg/L-$Cl_2$ | pH 8.3 | Stagnation/Flow |
| Week 2 | | | | |
| Week 3 | No Change | | | Flow Velocity Changes |
| Week 4 | | Free Chlorine @ 5 mg/L-$Cl_2$ | Orthophosphate at 3 mg/L-$PO_4$ and pH 7.5 | |
| Week 5 | No Change | | | Physical Disturbance |
| Week 6 | | | | |

Following the completion of week six, the loops were acidified to pH < 1.0, and run for five days with the by-pass closed to expose the interior of the pipe to the acidified water. This was done to mobilize as much of the lead remaining on each pipe as possible to determine the mass that remained on each loop following the six week test period.

## 5.4 Results

Overall, results were very similar from loop to loop. More than half of the lead mass remaining on each pipe at the end of the dosing phase was released into the reservoir or appeared in the stagnated loop and first flush samples collected at the end of the first week of testing. In the different types of samples collected from each loop during the second week of testing, observed lead concentrations were consistently about an order of magnitude lower than in the first week without a meaningful change in observed iron concentrations. A second meaningful decrease in lead release was observed during the second week of testing. During the following weeks, observed sample concentrations leveled out, decreasing only slightly over time, though some exceptions to the trend were observed in Loops 3 and 4. Iron release changed little from week to week. Impacts of changing water quality or hydrodynamic conditions were less obvious than general trends in lead release observed in every loop.

### 5.4.1 Loop 1 – Control

The trends in lead release from Loop 1 are typical of the overall trends observed. Figure 21 summarizes lead and iron release observed in acidified reservoir samples over the six week test period. Lead release decreased by nearly two orders of magnitude during the first two test weeks followed by little to no change in concentrations observed in remaining weeks. By the end of the third week lead release was at about 10-ppb after a full week, where it would remain until the end of testing. Iron release between 3 and 5 mg/L throughout testing.

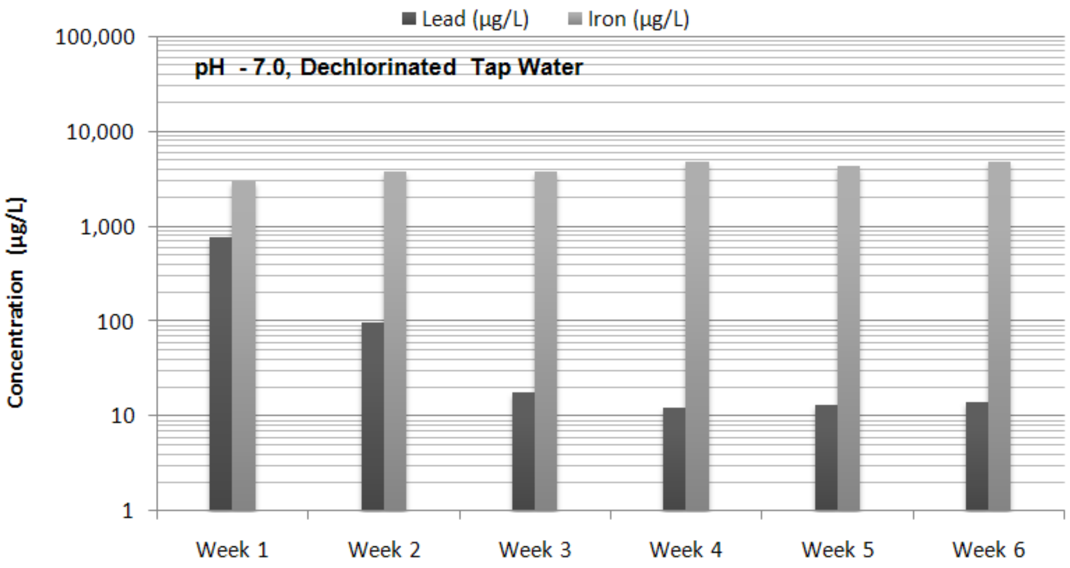

**Figure 21: Lead and iron release observed in acidified reservoir of Loop 1 at the end of each of six test weeks**

A similar trend was observed in stagnated pipe samples, though the magnitude of the observed concentration from week to week was about an order of magnitude higher than that observed in the acidified reservoir. Iron release in these samples is also notably higher, and displays little meaningful variation from week to week. The results of stagnated pipe samples are provided in Figure 22, the vertical scale of which has been increased by an order of magnitude to compensate for the higher concentrations observed. Note that a stagnated pipe sample was not collected following week 6.

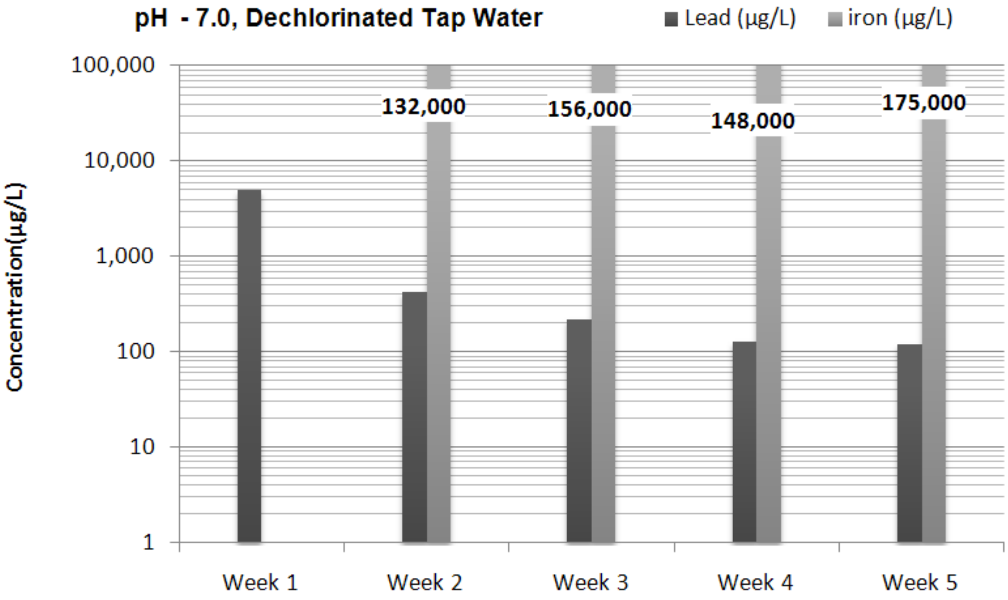

**Figure 22: Lead and iron release observed in stagnated pipe samples from Loop 1 at the end of each of six test weeks**

A decreasing trend in lead release over time was also observed in first flush samples collected from Loop 1. Overall, the magnitudes of concentrations observed in these samples were slightly higher than those observed in the acidified reservoir. A chart summarizing first flush sample results for Loop 1 is provided in Appendix 24 of this report.

Figure 23 shows the calculated lead mass remaining on pipe 1 at the start of each of the six test weeks, and includes the calculated remainder after the pipe was exposed to acidified water following week 6. These results typify the magnitudes of remaining lead observed on each of the four pipe loops from week to week during testing.

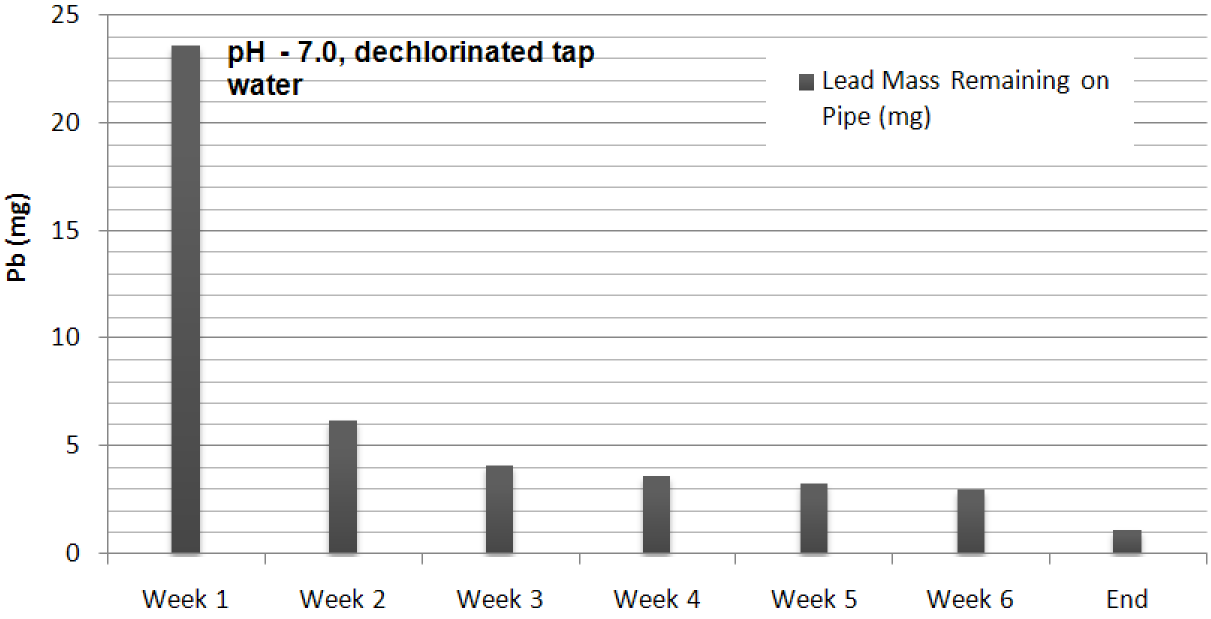

**Figure 23: Weekly calculated value of lead mass remaining on the Pipe 1 at the start of each of the six test weeks and after exposure to acidified water at the end of testing.**

### 5.4.2 Loop 2 – Elevated ORP

Magnitudes of lead and iron release observed in all samples collected from Loop 2 during the six week test period were similar to those observed in Loop 1. A slight increase in observed lead concentrations in acidified reservoir samples was observed in samples from weeks 5 and 6 after switching from chloramines to free chlorine. Given that this increase was no more than 6 µg/L, the results may not be significant. However, similar increasing trends were observed in week 5 stagnated pipe and first flush samples. Iron release underwent no meaningful changes in reservoir samples during weeks 5 and 6, but increases in lead release correspond to increases in iron release in stagnated pipe samples from these weeks. Figure 24 summarizes lead and iron release observed in Loop 2 acidified reservoir samples over the six week test period. Figure 25 summarizes lead and iron release observed in Loop 2 stagnated pipe samples over the six week test period. Results for first flush and weekly mass balance calculations are provided in the appendices.

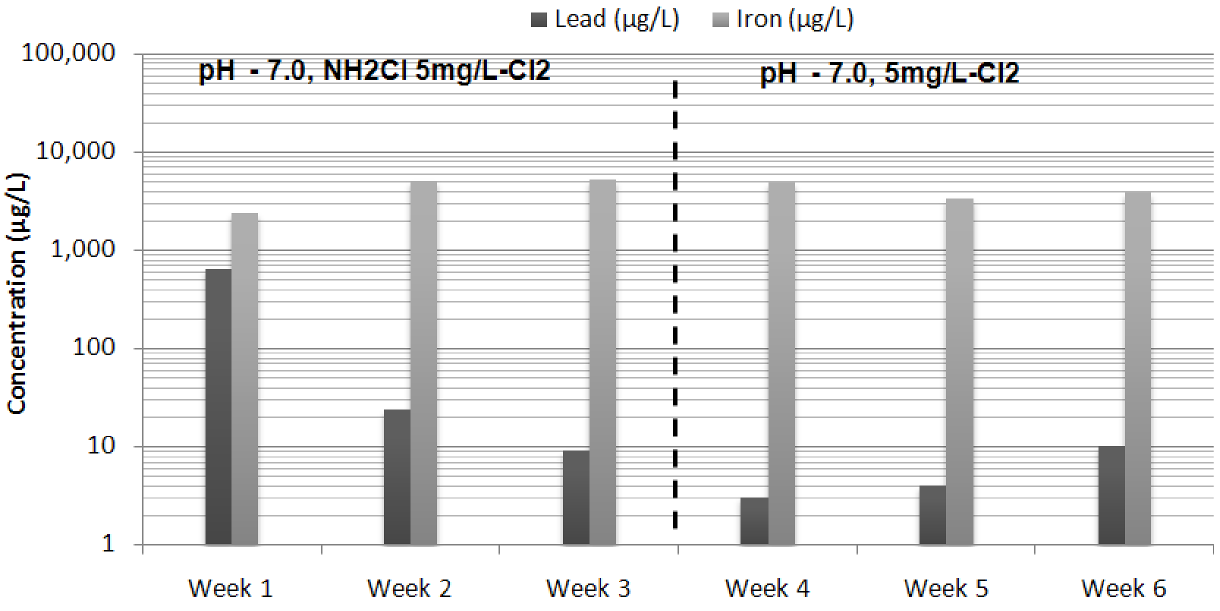

Figure 24: Lead and iron release observed in acidified reservoir of Loop 2 at the end of each of six test weeks

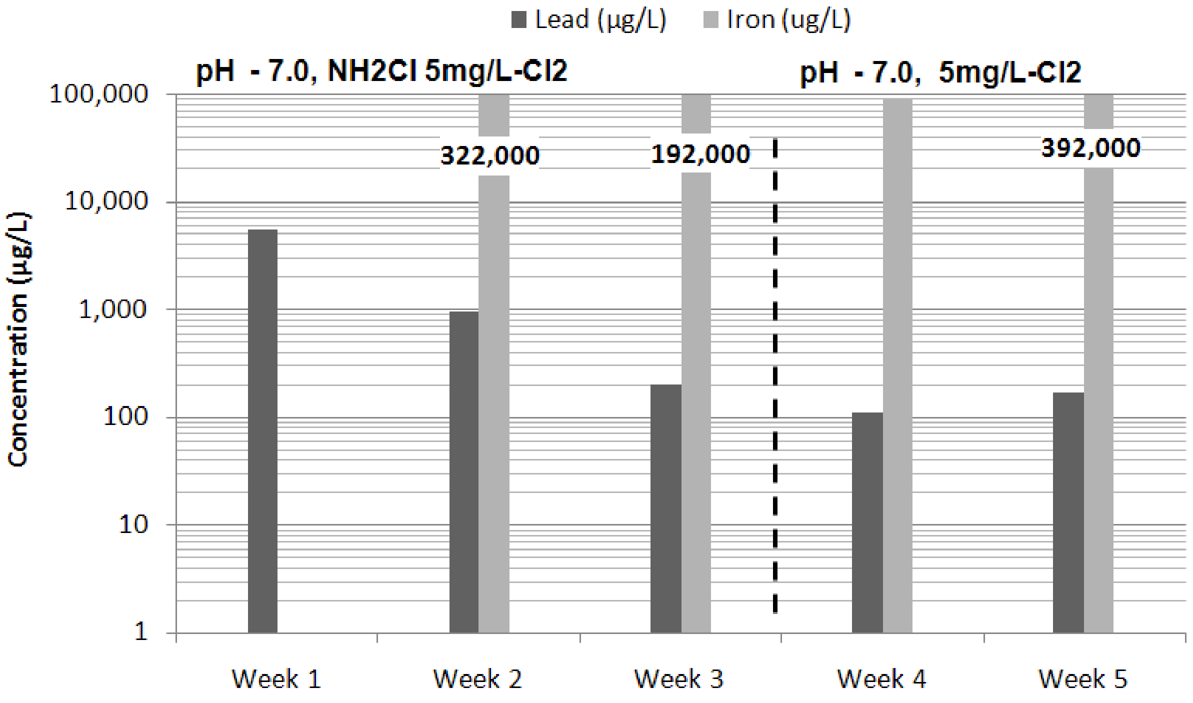

Figure 25: Lead and iron release observed in stagnated pipe samples from Loop 2 at the end of each of six test weeks

### 5.4.3 Loop 3 – Corrosion Control

Lead release observed in Loop 3 acidified reservoir samples were similar in magnitude and overall trend to those observed in Loops 1 and 2. In acidified reservoir, stagnated pipe, and first flush samples lead release increased after the switch to orthophosphate in Week 4. During the following weeks, lead release decreased incrementally. Though the changes in magnitudes were slight, increases and decreases in iron release consistently corresponded to changes in lead release. These trends are most evident in the set of stagnated pipe samples shown in Figure 26. Figures showing acidified reservoir samples, first flush samples, and mass remaining calculations are provided in the Appendices.

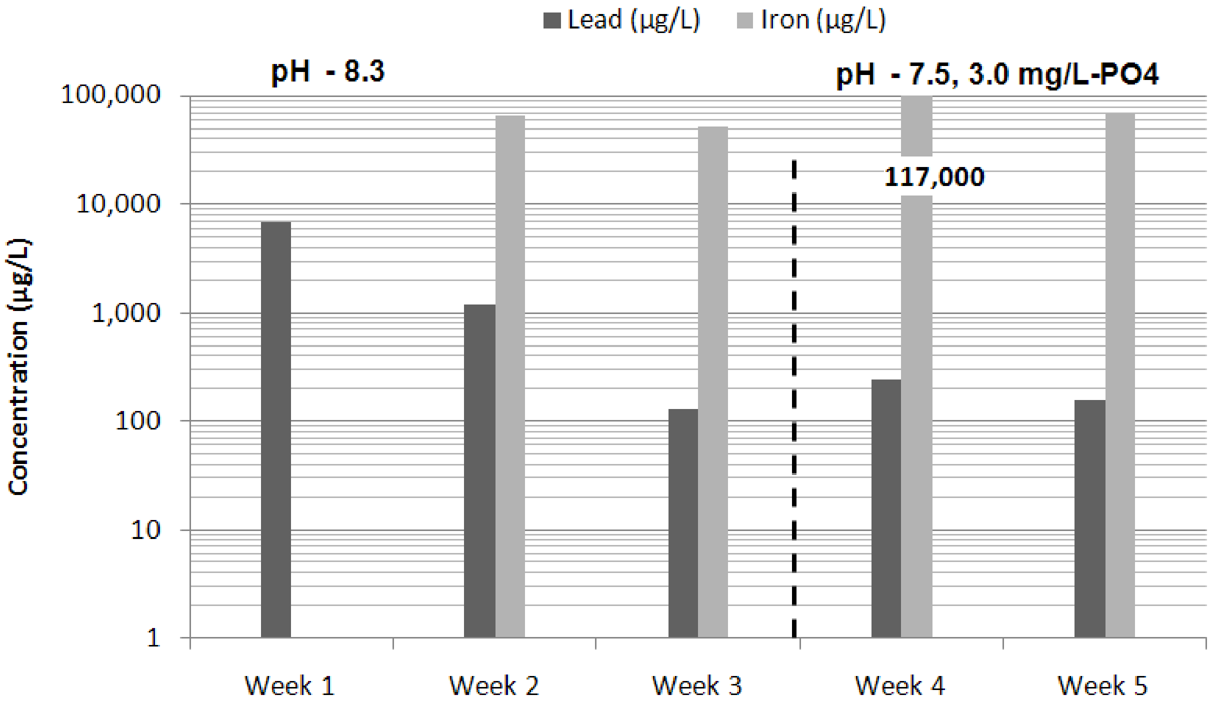

**Figure 26: Lead and iron release observed in stagnated pipe samples from Loop 3 at the end of each of six test weeks**

### 5.4.4 Loop 4 – Hydrodynamics

During the stagnation/flow regimen implemented during weeks 1 and 2 of testing, trends in lead and iron release differed negligibly from those observed in the other three loops. In reservoir, stagnated pipe, and first flush samples, release was minimal after switching to the regimen of alternating flow velocity in Week 3, but spiked during after the second week of alternating flows (Week 4). Iron release followed the same trend during Weeks 3 and 4. Figure 27 shows lead and iron release observed in Loop 4 acidified reservoir samples over the six week test period. Stagnated pipe samples, first flush samples, and mass remaining calculations are provided in the Appendices.

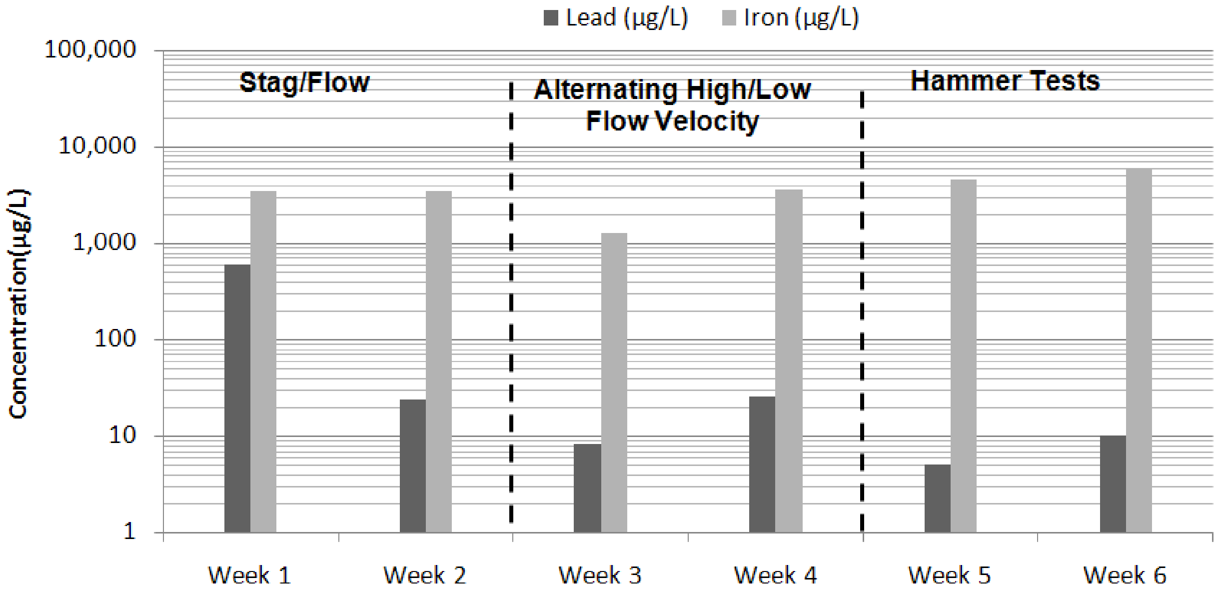

**Figure 27: Lead and iron release observed in acidified reservoir samples from Loop 4 at the end of each of six test weeks**

Water hammer tests were performed once per week during Weeks 5 and 6. During the remainder of Weeks 5 and 6, flow was maintained at 0.25 gpm, and results of the hammer tests may not be evident in the acidified reservoir data. Results of the hammer tests are shown in Figure 28. During each water hammer test one sample was collected prior to the test, and one sample was collected from the first 500-mL to discharge to a graduated cylinder following the sequential opening and closing of the in-line valve. During the Week 5 test, an increase from 8 to 9 µg/L of lead was observed in the pre- and post-hammer test samples; constituting a negligible difference. Iron concentrations before and after the hammer test during Week 5 were also minimal. During week 6 an increase in lead release from 11 to 19-µg/L was observed accompanied by an increase in iron release by a factor of 6.

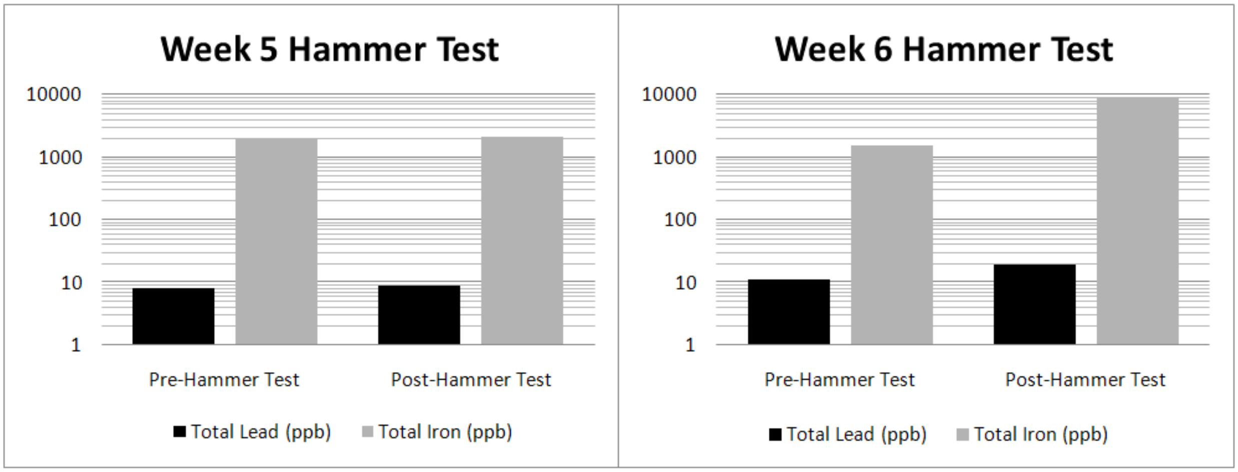

**Figure 28: Results of hammer tests performed on Loop 4 during Weeks 5 and 6 of testing.**

## 5.5 Discussion – Part III

The results of the four pipe loops tested at DCWASA reflect the diversity of results observed in household profiles discussed during Part 1. Pipes 1 and 2 both acted as consistent sources of lead release throughout testing. While Pipe-loop 1 showed signs of gradually decreasing lead release through the constant flow phase of testing, as soon as the flow regimen was changed to alternating stagnation flow, the release increased. Release increased again, to as high as 100 ppb, following the change to increased flow velocity, after which a trend of decreasing lead release was identified. Loop 2 released consistently elevated lead (i.e. > 100 ppb) throughout the constant flow and stagnation/flow phases, without visible trends of decrease. Increased flow velocity increased lead release notably.

Both Loop 1 and 2 showed evident sensitivity to high flow velocity, presumably due to the effects of scouring. This is somewhat contrary to the results of high flow sampling conducted during profiling in Part 1, where the impact of scouring appeared minimal. This reinforces the capacity for hydrodynamic forces to act as a driver of lead release from galvanized plumbing. Both loops also continued to release elevated lead, even after a decreasing trend was observed. This behavior implies that decreasing trends observed in Part 1 following full LSR may not be indicators that the source of lead (i.e. galvanized plumbing) is actually being exhausted. Rather, this behavior indicates that galvanized plumbing may serve as a source of lead indefinitely; a source that could be activated by a number of physical or hydraulic factors to which household plumbing is commonly subjected. This notion is reinforced by the surface analysis in Part 2, where substantial lead content was observed even in the deepest layers of the iron corrosion scales.

Alternatively, Loops 3 and 4 released comparatively little lead for most of the test duration. Both displayed some sensitivity to changes in flow regimen, but the overall magnitude of release in both loops remained low, relative to the behavior of Loops 1 and 2. All four loops contributed elevated lead during first flush sampling, further establishing the potential for hydraulic forces to drive lead and iron mobilization. Iron release from all four DCWASA loops trended similarly, in most cases, to lead release, implying a relatively stable relationship between the two metals. Iron release magnitudes were similar from loop to loop, overall, while lead release magnitudes varied meaningfully, overall. These results substantiate the observations in Parts 1 and 2, that lead release from galvanized iron scales will vary from location to location based on the specific history of the premise plumbing.

The pipes used in the ARTC loops had identical histories of corrosion and lead exposure. Lead release during testing was, likewise, nearly identical in magnitude and behavior. Each loop adsorbed about 25% of the lead it was exposed to during the dosing phase. Most of the lead recovered during this phase was recovered in samples taken from water that stagnated in the pipe during water changes. The majority of the lead that remained on each pipe was mobilized to solution during the first, and, to a lesser extent, the second weeks of testing. After the second week of testing, lead release from each pipe remained low.

Stagnation of water in the pipe during water changes continued to induce the greatest amount of lead release as testing continued, though the trends in release recapitulated the trends observed in release to the reservoir. Overall the trend of substantial lead release during weeks 1 and 2 followed by stable low level release in reservoir, stagnation, and first flush samples overshadowed any impacts of changing water quality or flow regime. Importantly, no substantial decrease in iron release was observed corresponding to the decrease in lead release over the first weeks of testing. No apparent correlation between releases of these metals was observed until lead release had stabilized after week 3.

These results, and the known history of short term but intense lead exposure of each pipe, indicate that the lead that sorbed to the pipes remained, for the most part, at the surface of the iron corrosion scales. Because of the absence of a long history of scale formation as would occur in household plumbing under conditions conducive to co-precipitation and other sorptive mechanisms, the potential for these pipes to act as a source of lead over a prolonged period did not exist. By the time that changes in water qualities and flow regimes were implemented in these tests, enough of the lead had sloughed off of the scale surface, that results meaningful to understanding desorptive mechanisms could not be clearly ascertained.

# 6.0 Recommendations for Mitigation

In-home plumbing can potentially be as much or more of a source for lead in residential drinking water as lead service lines. Lead release from galvanized plumbing can persist even after full lead service replacement. Evaluation of residential profiles and observations of trace metal content and surface morphology of corrosion scales have indicated that lead release from galvanized iron premise plumbing will vary substantially in magnitude and behavior from one location to another. This variation is highly dependent on a number of factors, including the configuration and relationship of source and in-home plumbing, contributing to the individual history of the premise plumbing. The degree to which remediation is required, thus, will also vary by location.

Substantial lead content may persist in the deepest strata of iron corrosion scales. Up to 8% lead content by weight has been observed in the crystalline matrix constituting the deep layers of such scales. This represents decades of accumulated lead that could likely persist for the remaining service life of existing galvanized pipes. Alternatively, relatively small lead contents have been observed in other galvanized pipes from DCWASA homes, presumably indicating less risk for mobilization of lead from these pipes. The amount of lead release from a given home's galvanized plumbing is likely to depend on the lead content in the upper, more exposed layers of the corrosion scales. Even in homes where relatively little lead release from in-home plumbing is observed, the potential exists for deposits of lead in the deeper scale layers, which could potentially result in elevated release should the scales undergo a transformation due to external disturbances of a physical or even hydraulic nature. The only way to fully ensure that lead is not mobilized from galvanized plumbing in a given home is to fully replace the galvanized plumbing.

To the extent possible, the potential for lead release from a home's plumbing should be evaluated before plumbing replacement and its associated expense is undertaken. If evaluations indicate that lead release is consistently low (i.e. less than the 15 ppb LCR action level) shortly after full lead service replacement, immediate replacement of in-home plumbing may not be urgently required. Nonetheless, the homeowner should be educated about the potential for lead release from the galvanized plumbing, and, at a minimum, take into account the following considerations regarding whether or not to proceed with plumbing replacement:

- Lead release from galvanized plumbing can be exacerbated due to physical disturbances. Any modifications or improvements to the plumbing, including water heater installations or even fixture replacements, could potentially lead to short term spikes in lead release.
- Lead release from galvanized plumbing can be exacerbated due to hydraulic disturbances. If the configuration of a home's piping is conducive to frequent water hammer, the homeowner should recognize this as a potential cause of periodic lead release events.
- Young children are more vulnerable to the detrimental effects of lead. If there are young children in the home, especially infants, the risk of retaining existing plumbing should be considered greater, and any steps that can be taken to mitigate lead release from in-home plumbing should be taken.

To some homeowners the financial burden associated with a plumbing replacement will be excessive. If evaluations of lead release following full lead service replacement indicate mild but consistent risk (i.e. persistent lead levels at or near 15 ppb), the following partial mitigation measures should be considered where full plumbing replacement is not financially feasible:

- A variety of at-the-tap treatment technologies are available commercially. Use of an NSF certified filter on tap water prior to drinking should substantially reduce particulate lead concentrations.
- Replace portions of in-home galvanized plumbing associated directly with drinking water taps such as the kitchen sink or bathroom sinks. This measure may help to reduce lead mobilized to these taps, thereby reducing the amount of lead ingested by residents.
- If water hammer has historically been an issue at a home where lead release from galvanized plumbing has been identified, installation of water hammer dampers is recommended.

In homes where evaluations indicate consistent high risk lead levels weeks to months following lead service replacement (i.e. persistent lead levels above 15 ppb), full plumbing replacement may be the only feasible means of remediation. Where the homeowner cannot afford a plumbing replacement, especially if small children are living there, financial assistance may be available through various organizations. DCWASA may wish to consider extending their existing financial assistance program to include specific provisions for low interest loans for replacement of galvanized plumbing where elevated lead release persists beyond full lead service replacement.

# 7.0 Summary and Conclusions

## 7.1 Part 1

In Part 1, 62 residential profiles conducted at homes in the DCWASA service area were evaluated. Particulate lead release in samples from these homes was greater than that from lead service lines. The magnitude of the contribution of lead from galvanized plumbing varied widely from home to home. Linear correlation between lead and iron release was reasonably strong in individual homes where lead release was elevated, but in other homes the calculated correlation was relatively weak suggesting non-uniform adsorption and release. The ratio of iron to lead release varied from home to home as well; weakening, to some extent, the overall correlation. It was determined that the degree to which galvanized plumbing will act as a source of lead, even after full lead service replacement, is subject to the particular history of a site's premise plumbing, and should be evaluated on a home to home basis.

Evidence that a variety of physical and hydrodynamic factors, including water hammer, high flow velocity, or scouring, and physical disturbance during LSR, may exacerbate lead release from galvanized plumbing. Physical disturbance caused the most acute increases in elevated lead and iron, particularly immediately following LSR; but decreasing trends were observed consistently in lead release following full replacement. The magnitude of lead release and the rate of diminishing release varied widely from home to home, making it impossible to generalize the phenomenon to a quantitative relationship.

The impacts of some factors, including lead release from lead-bearing faucet valves and the effects of diffusion and advection may have affected some results. Nonetheless, results were statistically significant regarding galvanized plumbing as a source of lead.

The impacts of other trace metals, though not a focus of the study, included the finding that copper, potentially, has a very strong affinity for adsorption to iron corrosion scales. Correlations between iron and lead with other metals such as zinc were weak. In the case of zinc, this is likely because the zinc in the old galvanized plumbing is already gone.

The following conclusions were drawn from the evaluation of Part 1:

- Galvanized iron plumbing can serve as a sink/source for lead due to historical capture of lead released from upstream service lines.
- The magnitude and behavior of lead release from galvanized plumbing varies substantially form location to location.
- Correlations of lead and iron release vary from location to location. Good linear correlations may occur at a specific site, but the slope (i.e. ratio of iron to lead) will vary from site to site.
- Lead can be mobilized from iron corrosion scales to a meaningful extent. When it does, it can exceed the LCR Action Level even when no other source of lead is present.
- Stagnation can exacerbate lead release from galvanized plumbing.
- Water hammer can potentially exacerbate lead release from iron corrosion scales.
- Physical disturbance, especially during LSR, can lead to acute elevation of lead release from galvanized plumbing.

- After a physical disturbance, lead and iron release from galvanized plumbing exhibit consistent trends of decrease, but with variability from site to site.
- Lead may persist in galvanized corrosion scales for extended periods.

## 7.2 Part 2

Jar tests and surface analyses were performed to investigate sorptive mechanisms and identify the morphology of iron corrosion scales extracted from the DCWASA service area. Results of the jar test investigation established the potential for formation of lead-rich corrosion scales through a co-precipitative process, whereby dissolved lead is adsorbed to mobilized ferric or ferrous iron and deposited on the inner pipe surface. The affinity between dissolved lead and iron was determined to be very strong, whatever the specific adsorptive process.

SEM and EDAX surface analysis performed on five galvanized pipes from the DCWASA service area confirmed the diversity of corrosion scale content and surface morphology from one location to the next. Lead-rich and relatively lead-poor scales were observed. In lead-rich scales, lead content was as high as 7 or 8% by weight, even in the deepest, most crystalline layers of existing scales. These results confirm the possibility that a galvanized pipe might serve as a lead source for many years or even indefinitely.

In addition, an array of particulate non-iron-based lead minerals such as lead-apatites, carbonates and oxides were observed to be embedded in discrete areas of scale surfaces. The observation indicates the potential for spikes in particulate release should these mineral deposits become dislodged from the iron corrosion scale. It also provides evidence that an array of sorptive mechanisms, from physical capture to long term co-precipitation, can participate in developing a lead rich iron corrosion scale over time.

The following conclusions were drawn from the evaluation of Part 2:

- A variety of sorptive mechanisms can participate in developing a lead rich iron corrosion scale over time
- The potential exists for deposition of lead-iron complexes resulting from co-precipitation to form lead-rich, crystalline corrosion scale layers
- Lead rich crystalline layers may be found throughout the entire depth of a given iron corrosion scale
- Great variation in lead content may occur from location to location within a given distribution system, which emphasizes the importance of evaluation of galvanized iron as a lead source on a site-by-site basis.
- Great variation in lead content may occur from location to location within a given segment of corroded galvanized pipe. Layers of iron deposition may vary in lead content, and very high lead contents may be observed in isolated locations where defined non-iron mineral deposits are embedded in the scale matrix.
- The presence of other elements such as phosphorus and manganese on scale surfaces may alter the relationship of lead and iron within an existing scale or at the surface of the scale.

## 7.3 Part 3

In Part 3 of the investigation, pipe loop experiments, each consisting of four separate pipe loops, were conducted at DCWASA and at the HDR ARTC facility in Redmond, WA. The DCWASA loops used galvanized pipe taken from four different homes within the DCWASA service area. The HDR loops used new, black iron, artificially corroded in the laboratory, and subjected to very high concentrations of dissolved lead for a 1 month dosing period.

Results of the DCWASA loops reinforced the observation from Part 1 that variation in the capacity for galvanized plumbing to act as a lead source is typical. Two pipes acted as persistent sources of elevated lead release. The other two pipes' lead release remained low, even after a week of release and recirculation to a small reservoir. All four pipes exhibited sensitivity to hydrodynamic changes including flow rate changes and stagnation flow regimens. Multiple instances of decreasing trends in lead release were observed, which ended abruptly when the pipe surface was agitated due to a change in flow regimen. The changes induced spikes in lead release, followed by gradual decreasing trends in release similar to the initial trend, or to the trends observed following full LSR at several homes where post-LSR profiles were conducted in Part 1. These pipes exhibited a meaningful sensitivity to stagnation with respect to lead mobilization.

The pipes in the ARTC loops, unlike the DCWASA loops, did not have a long history of exposure and scale formation as would occur in household plumbing under conditions conducive to co-precipitation and other sorptive mechanisms. These were corroded in an accelerated laboratory procedure, and then exposed to very high levels of dissolved lead over a relatively short period. While a substantial amount of lead sorbed to the corrosion scales on these pipes, the majority of the sorbed lead sloughed off with the first two weeks of testing. All four pipes exhibited near identical lead release behavior. These pipes exhibited a substantial sensitivity to stagnation with respect to lead mobilization. Other impacts, especially the addition of phosphates and subjection to water hammer created notable increases in lead release, but by the time these test were performed much of the lead had already come off of the pipe surface.

The following conclusions were drawn from the evaluation of Part 3:

- The capacity of galvanized plumbing to act as a source of drinking water lead is a function of the history of exposure and interplay of adsorptive processes during long-term formation of the iron corrosion scale matrix.
- Decreasing trends of lead release observed over time under relatively constant conditions, can be readily agitated, renewing the process of elevated lead release and gradual suppression, by a number of physical and hydraulic influences.
- A lead-rich corrosion scale may continue to serve as a lead source in drinking water long after all other sources of lead have been removed.

# 8.0 References

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