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The impact of sampling approach and daily water usage on lead levels measured at the tap

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ABSTRACT

There are many sampling approaches available for lead (Pb) in drinking water. Selecting the best approach for its intended use is critical. The objective of this work was to compare water Pb levels collected by multiple sampling approaches from a model home plumbing system (HPS) that included an old Pb service line (LSL), as a function of daily water usage. Specifically, flushed, direct LSL, sequential profile, random daytime (RDT), first draw, and manual composite samples were compared, and daily water usage rates ranged from 1.4 to 120 gallons/day (5.3–454.2 L/day). Pb levels in water collected directly from the LSL reached equilibrium after 7–15.5 h of stagnation, and approximately 55% of the equilibrium Pb concentration was reached after 2 h. Sequential sampling accurately identified the LSL; however, the peak profile Pb concentrations were a fraction of the Pb measured directly from the LSL. Daily water usage patterns greatly impacted total Pb levels in all water sampling approaches, although manual composite and RDT samples were more sensitive to changes. Manual composite and RDT samples were equal to or greater than first draw samples throughout the study, and differences grew larger as water usage decreased.

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1. Introduction

Lead (Pb) is a neurotoxin that can cause permanent cognitive and behavioral impairments in children, and cardiovascular and kidney problems in adults (Fowler and DuVal, 1991; ATSDR, 2007; Lanphear et al., 2018). Lead in tap water originates from the corrosion or dissolution of leaded plumbing materials. Lead service lines (LSLs) are the largest contributor to total Pb in residences, responsible for up to 75–86% of the Pb seen at the tap (Sandvig et al., 2008; Lytle et al., 2019). Other Pb plumbing materials include brass fixtures and fittings (Kimbrough, 2001; Schock and Neff, 1988); leaded solder (Subramanian et al., 1995); and Pb goosenecks. Galvanized steel pipes can also leach Pb into drinking water (Sandvig et al., 2008).

Lead levels across communities and even within the same community are impossible to predict (Del Toral et al., 2013; Wang et al., 2014) because of the uncertain mix of leaded and non-leaded premise plumbing, unreliable LSL records (Karalekas et al., 1976), water quality variability, the variability of the leaded scale and its solubility, the degree to which the water has reached equilibrium with the leaded plumbing materials (Schock, 1990; Schock et al., 1996; Cardew, 2006), presence of Pb particles (Deshommes et al., 2010; Triantafyllidou et al., 2007), and variable water usage (Masters et al., 2015; Riblet et al., 2019; Salehi et al., 2020). Therefore, drinking water sampling is necessary to ascertain the amount of Pb leached into drinking water. Reasons to sample for Pb include regulatory compliance, evaluation of corrosion control effectiveness, Pb plumbing source identification, and human exposure assessment (Schock et al., 2019). Deciding which sampling approach to use to address specific questions is critical to obtaining a representative answer.

The most commonly used and most studied Pb sampling approaches include: first draw sampling, the method required for compliance sampling under U.S. Environmental Protection Agency's (EPA's) Lead and Copper Rule (LCR) (EPA, 1991a, 1991b, 1991c); flush sampling, which has been used to identify the presence of an LSL (Cartier et al., 2012; Deshommes et al., 2016; James et al., 2019; Riblet et al., 2019); profile or sequential sampling, which in-

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volves a series of 10–20 samples and can be used to develop a Pb profile (Giani et al., 2004; Clark et al., 2014; Lytle et al., 2019; Sandvig et al., 2008; Van den Hoven and Slaats, 2006); stagnation profile sampling, which determines how much lead is released after varying amounts of stagnation (Lytle and Schock, 2000; Schock et al., 1996); random daytime sampling (RDT), which is rooted in statistical risk evaluation and offers a broad view of a community's water quality (Cardew, 2003; Hayes, 2009; Hayes and Croft, 2012); and composite sampling, which is useful for assessing Pb exposure at individual houses and for data collection in Pb exposure studies (Clement et al., 2000). A detailed explanation of sampling approaches can be found in the SI Section S1.

Other studies have considered direct comparisons of Pb sampling methods. For example, RDT sampling has been shown to correlate closely with composite sampling (Hayes, 2009). Riblet et al. (2019) compared composite proportional sampling, flushed, first draw (after 30 min and 6 h) and RDT at twenty-nine homes with and without LSLs. They reported that RDT samples were most similar to proportional (exposure-based) samples whereas 6-hour first draw overestimated and flushed samples severely underestimated exposure sampling at sites with LSLs.

Consumer household drinking water usage patterns are recognized as important factors impacting Pb levels at the tap, but associated complexities (e.g., variability in flow rate, daily use rate, water stagnation) make it challenging to predict the extent of the impact and understand related mechanisms. Lower water usage and long stagnation times have been linked to elevated Pb release from Pb sources (Masters et al., 2015; Salehi et al., 2020). Water quality in plumbing may change over long stagnation periods as a result of chemical and biological reactions which can increase Pb solubility or enhance corrosion (Lytle and Schock, 2000; Korshin et al., 2000). Regularly moving water through LSLs and home plumbing prevents the loss of important water quality parameters such as disinfectant residuals and corrosion inhibitors (e.g., orthophosphate) that can be important in the geochemical stability of corrosion by-products or other solids enmeshed in pipe scale layers and can help remove loose Pb particles out of the pipe (Brown and Cornwell, 2015; ANSI/AWWA, 2017). The release of particulate Pb was attributed in part to low water usage in Flint homes, which led the state of Michigan and EPA to promote the "Flush for Flint" program (EPA, 2016) that encouraged residents to use repeated flushing as a means to remove Pb particles from household plumbing and improve the effectiveness of corrosion control treatment by preventing the loss of orthophosphate and disinfectant residuals in homes. Water usage patterns also impact mass-transfer factors including the rate Pb ions diffuse into the water from Pb sources and then transport Pb ions through plumbing (Burkhardt et al., 2020).

A critical need still exists to identify differences in the results and interpretation of Pb sampling approaches in drinking water and to better define appropriate sampling purposes. The objective of this work was to compare drinking water tap sampling approaches and associated Pb concentrations in water samples collected from a model home plumbing system (HPS) that contained an LSL. Direct LSL, flushed LSL, first draw, RDT, sequential profile, stagnation profile and manual composite sampling approaches were compared. The impact of water usage on Pb (particulate and soluble) concentration associated with sampling approaches was also examined.

2. Materials and methods

2.1. Home plumbing system

A home plumbing system (HPS) simulator (Fig. 1) was constructed in EPA's Andrew W. Breidenbach Environmental Research Center (AWBERC) (Cincinnati, OH) and has been in continuous operation since January 23, 2012. The number of fixtures, pipe length, and the daily water usage were modeled after a household of four (Cahalan and Lytle, 2017) based, in part, on earlier water use considerations (Mayer et al., 1999). The HPS was constructed with copper plumbing (using occasional leaded solder joints) and contained an electric hot water tank, four faucets (Pb content <8%), a toilet, and a shower (Fig. 1). The HPS was plumbed directly from AWBERC's cold water plumbing (Greater Cincinnati Water Works tap water) and was adjusted to a target pH of 7.5 using 0.6 M HCl to enhance corrosivity. A 6.7 ft (2.03 m) long LSL was added inline to the HPS (Supporting Information [SI] Figure S1) in February 2017, after removal from a home in Cincinnati and a conditioning period (i.e., HPS was by-passed) beginning in September 2016. Specific details regarding the design and construction of the HPS can be found in SI Section S2, whereas the procedure used to condition the LSL is described in Section S3. The results presented herein span September 2016-February 2019. This includes LSL sample results during a 6-month conditioning period where it was separate from the HPS, and HPS tap results after the LSL was installed inline with the rest of the HPS.

2.2. HPS water usage patterns and sampling procedures

The faucets were given specific usage-type designations: a washing machine/dishwasher (faucet 1), two-bathroom taps (faucet 2 and 4) and a kitchen tap (faucet 3) (faucet details in SI S2). Household activities such as filling a glass of water, flushing the toilet, showering or washing dishes were mimicked. Water usage volume and time of use during each action were recorded. As shown in Table 1, the HPS was operated under five different water usage patterns during the study: (1) whole house "*normal use*" (120 gallons/day [gpd] [454 L/day]), (2) *"reduced normal use"* (60 gpd [227 L/day], M-F), (3) single faucet "*minimum daily use*" (1.4 gpd [5.3 L/day], M-F), (4) single faucet "*minimum use"* (1.4 gpd [5.3 L/day], T and Th only, reduced normal use M, W, F), (5) "*minimum use 2x*" (2.8 gpd [10.6 L/day], T and Th only, reduced normal use M, W, F), and (5) "*minimum use 2x and toilet*" (6.6 gpd [25 L/day], T and Th only, reduced normal use M, W, F) (see Table 1). Specific details on how the water usage patterns were achieved can be found in Section S4.

Direct LSL, flushed LSL, first draw, RDT, sequential profile, stagnation profile and manual composite samples were collected from the HPS over the course of the study. Specific details of sampling including water filtration protocol are described in Section S5.

2.3. Analytical methods

A combination pH electrode (Hach 48,600, Loveland, CO) was used to measure temperature, pH, and dissolved oxygen (DO) upon each sample collection event. Free chlorine residual was measured immediately after water collection using a Hach spectrophotometer DPD method 80 (Hach DR/2000, Loveland, CO). Lead, copper, total phosphorus (total phosphate), sulfur (sulfate) iron, calcium and other inorganic elements were measured by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) per EPA method 200.7 (U.S. EPA, 1994). The detection limits for Pb, copper, and zinc were 2.0, 1.0, and 0.5 µg/L, respectively. Calculations and graphs used the detection limit as the sample concentration for samples below the detection limit. Anion samples were stored below 4 °C (39°F) and analyzed for orthophosphate and nitrate by ion chromatography (EPA method 300) (EPA, 1993), and total alkalinity was analyzed using potentiometric titration (method 2320 b.4.6)(APHA; AWWA; WEF, 2018). Baseline water quality analyses results are summarized in Table S1. With the exception of sequential samples, which only included unfiltered water, samples for Pb were

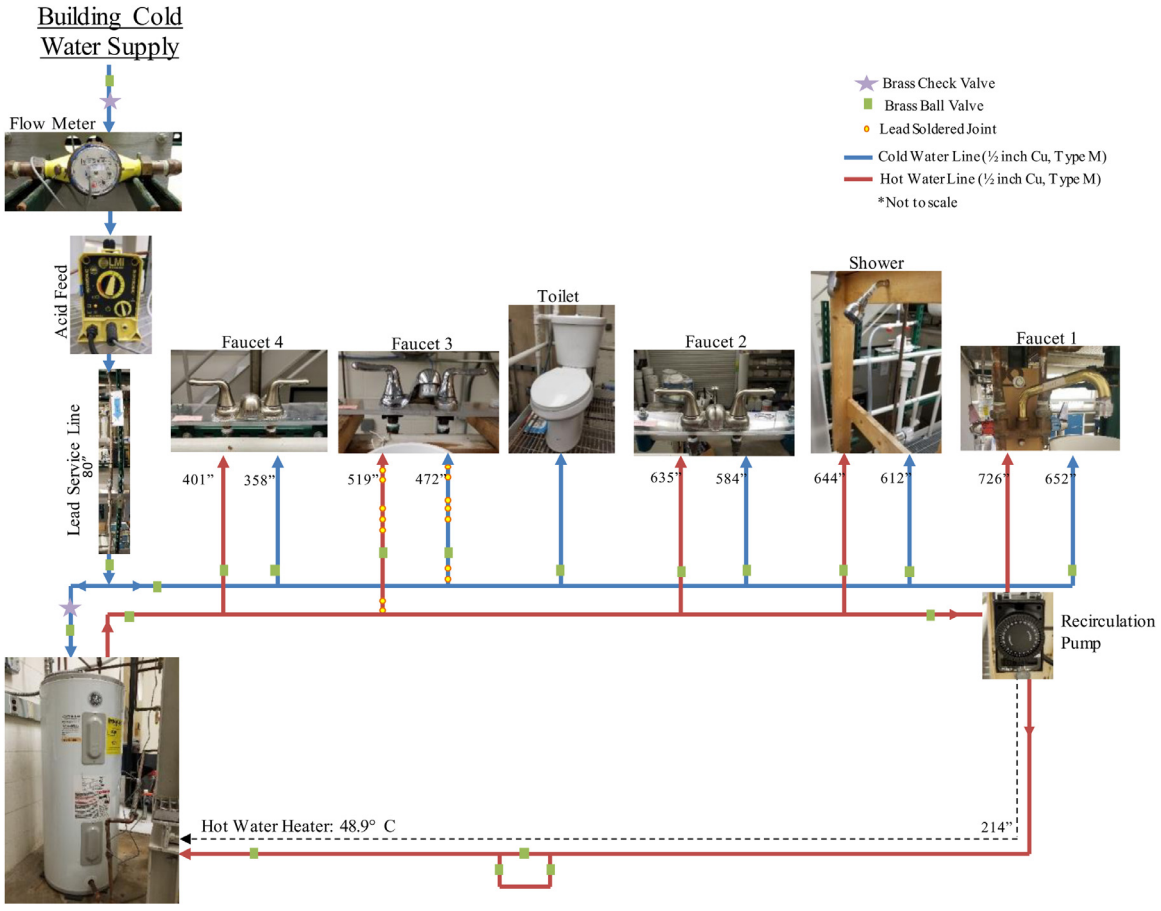
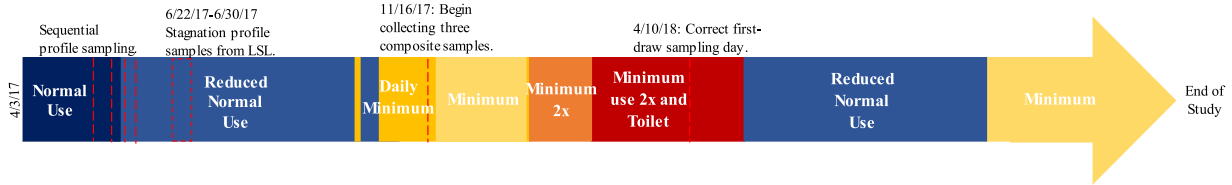


Fig. 1. Home plumbing system (HPS) schematic diagram. HPS components are mapped and distances of fixtures from the lead service line are noted (in inches). Cold and hot water lines are composed of ½ inch (12.7 mm) diameter type M copper pipes. The dashed line between the recirculation pump and the hot water heater represents the distance between the two components.

Table 1
Timeline and explanation of targeted water usage patterns at the home plumbing system after the conditioned lead service line was added. The timeline explains when each water use pattern occurred in relation to other events in the home plumbing system. Reduced normal use continued Mondays (M), Wednesdays (W) and Fridays (F) while minimum use, minimum use 2x, minimum use and toilet, and minimum use and pre-flush occurred on Tuesdays (T) and Thursdays (Th).

	Normal Use	Reduced Normal Use	Daily Minimum Use	Minimum Use		Minimum Use 2x		Minimum Use 2x and Toilet	
Date(s)	4/4/17-5/23/17	5/23/17-10/2/17; 10/6/17-10/16/17; 5/14/18-9/25/18	10/2/17-10/6/17; 10/16/17-11/22/17	11/22/17-1/9/18; 9/25/18-12/13/18		1/9/18-2/13/18		2/13/18-5/14/18	
Daily Use, Gallons	M-F 120	M-F 60	M-F 1.4	MWF	60	MWF	60	MWF	60
				T Th	1.4	T Th	2.8	T Th	6.6
Weekly Use, Gallons	600	300	7	183		186		193	
Pattern	A water usage goal was in place for each day, but fixtures were flushed on a random schedule	Same operation as Normal Use but the water usage goal was reduced by one half	Eight glasses of water from faucet 3, 200-400 mL	T/Th Eight glasses of water from faucet 3, 200-400 mL		T/Th-Eight glasses of water from faucet 3, 400-800 mL		T/Th -Eight glasses of water from faucet 3, 400-800 mL and three toilet flushes	



filtered by passing approximately 60 mL of water through a 0.2 µm polypropylene 25 mm syringe filter (GE Life Sciences, Marlborough, MA) to distinguish between soluble and particulate Pb.

2.6. Data analysis

Statistical analyses of data were performed using Sigmaplot® 13.0. The Mann-Whitney test was used to compare two groups, and the Kruskal-Wallis test was used for comparing more than two groups. A p-value less than 0.1 indicated a statistically significant difference between the medians of datasets. The Kruskal-Wallis test was followed by Dunn's test for performing multiple pairwise comparisons. Section S6 contains details regarding theoretical Pb solubility modeling.

3. Results

3.1. Flushed and stagnation sampling directly from LSL during conditioning and after installation to HPS

Excluding three elevated Pb samples collected within the first two weeks of conditioning, flushed total Pb levels averaged 5.4 ± 2.3 µg/L between September 28, 2016 and November 8, 2016 (Fig. 2a). During this time, soluble Pb averaged 3.7 ± 2.0 µg/L and was statistically different from total Pb ($U = 94$, $P < 0.001$) albeit a very small concentration. This indicated particulate Pb uptake as water passed through the LSL, likely through a "scouring" mechanism. Three elevated total Pb flushed samples (as high as 85 µg/L) near the beginning of the conditioning period were observed, while the corresponding soluble Pb levels were less than 7 µg/L (Fig. 2a). This observation suggested particulate Pb was likely a remnant of disturbances associated with pipe extraction, transport, and installation.

Water was also directly sampled from the LSL after 16–18 h (overnight) and 65 h (over the weekend) of stagnation (Fig. 3b) during the conditioning period. Total Pb levels in samples collected after 16–18 h of stagnation averaged 142 ± 45 µg/L. Soluble Pb levels averaged 126 ± 32 µg/L and were statistically different than total Pb levels ($U = 1101$, $p = 0.096$), indicating a significant but relatively small percentage (11% on average) of particulate Pb. Total Pb levels in stagnant samples collected after 65 h of stagnation averaged 153 ± 30 µg/L during this same time. Soluble Pb levels averaged 146 ± 24 µg/L and were not statistically different from total Pb ($U = 178$, $p = 0.21$). Statistically, there was no difference between mean Pb levels after 16–18 h (142 ± 45 µg/L) and 65 h (153 ± 30 µg/L), indicating Pb levels had already reached equilibrium at 16–18 h ($U = 632$, $p = 0.125$). Soluble Pb levels in stagnant samples from the LSL represent the highest soluble Pb level the HPS would measure in a drinking water sample. This observation would apply to any home with an LSL.

Pb was measured in stagnant water directly from the LSL after the LSL was connected to the HPS (Fig. 2b). Overall average Pb concentration during this time was 181 ± 115 for total Pb and 151 ± 75 for soluble Pb. Lead concentrations in the LSL during the entirety of this study were highly variable. Lead levels of water in direct contact with the LSL after it was added to the HPS, between April and July 2017, (normal use and reduced normal use protocol) were relatively consistent. During this period, total and soluble Pb concentrations averaged 155 ± 66 µg/L and 132 ± 37 µg/L, respectively (including a spike of 395 µg/L on June 15, 2017 and combining overnight and over-weekend samples). Pb was dominantly in the soluble form (89% of total Pb on average), and the overnight and over-weekend samples were indistinguishable (Fig. 2b). Unfortunately, direct LSL data was sparsely collected between July 11, 2017 and February 2018. During this period, only five days of LSL samples were collected (Fig. 2b). Routine direct LSL stagnant water sampling restarted in February 2018. Surprisingly, Pb levels were initially elevated in February to April 2018 to an average total and soluble Pb concentration of 345 ± 90 µg/L and 263 ± 39 µg/L, respectively, for the first 80 sampling days. During this period, soluble Pb accounted for 82% of the total Pb on average. In a short timeframe (approximately two weeks), Pb levels rapidly dropped and remained in a narrow range (May 2018 to the study end) to an average total and soluble Pb concentration of 111 ± 58 µg/L and 98 ± 52 µg/L, respectively. During this period, soluble Pb accounted for 92% of the total Pb on average. The reason(s) for the erratic LSL Pb release behavior could not be precisely identified initially, and the data gap further complicated interpretation. However, after close examination, a relationship between pH and soluble Pb became apparent (soluble Pb increased with decreasing pH), which was consistent with the predicted solubility trend of Pb(II) carbonate geochemistry (Figure S2). The reported pH values were measured directly from the LSL after-stagnation Pb sample, therefore, do not necessarily reflect the consistency of the pH control system but rather more accurately reflect equilibrium pH conditions at the time of soluble Pb sampling. The pH over the entire study period averaged 7.7 ± 0.3 (Table S1).

3.2. Stagnation profile sampling directly from LSL

A series of samples collected from the LSL over time resulted in a Pb concentration profile that increased logarithmically as stagnation time increased (Fig. 3) with Pb levels increasing most rapidly over the first two hours of stagnation. The Pb increase turned more gradual between 2 and 7 h of stagnation. Pb concentrations leveled off between 7 and 18.5 h of stagnation, thereby reaching an apparent equilibrium maximum concentration of approximately 164 µg/L.

A radial molecular diffusion model was fitted to the experimental Pb profile data to predict the concentration of Pb leached from a pipe over time (Fig. 3). The model assumed that the Pb concentration was constant along the pipe axis, sufficient oxidant was present to drive Pb oxidation, and molecular diffusion was the rate-limiting step. Model details can be found elsewhere (Lytle and Schock, 2000; Crank, 1975; Kuch and Wagner, 1983; Schlünder, 1975) and are summarized in Section S7. The theoretical curve was fitted to the measured profile data using the Solver™ routine incorporated in Microsoft Excel (Redmond, Washington) to minimize the sum of squared residuals between actual and diffusion model predicted Pb levels. A regression analysis of the best fit model gave an r^2 value of 0.935 (Fig. 3). The model fit showed that 55% of the maximum equilibrium Pb concentration eventually released from the LSL was already present in the water after only 2 h of stagnation. Pb concentrations in the LSL rapidly increased to elevated levels. These observations reinforced previous findings (e.g., Doré et al., 2018) regarding the limitation of tap flushing programs designed to minimize Pb exposure from LSLs, unless flushing occurs regularly throughout a given day.

3.3. Sequential profile sampling

Faucets 1, 2, 3, and 4 were 578 in. (14.6 m or approximately 1.86 L of water), 510 in. (13.0 m or 1.64 L), 409 in. (10.4 m or 1.32 L), and 294 in. (7.5 m or 0.95 L) away from the LSL, respectively. Sequential Pb profiles of water collected at each faucet characteristically displayed an asymmetric distribution described by a relatively rapid increase in Pb concentration to a peak, followed by a tailing of Pb reflective of longitudinal dispersion (Fig. 4). Peak Pb concentrations appeared reasonably close to where expected within the sequential Pb profile, based on the measured distance and corresponding water volume between the faucet and LSL (Fig. 4).

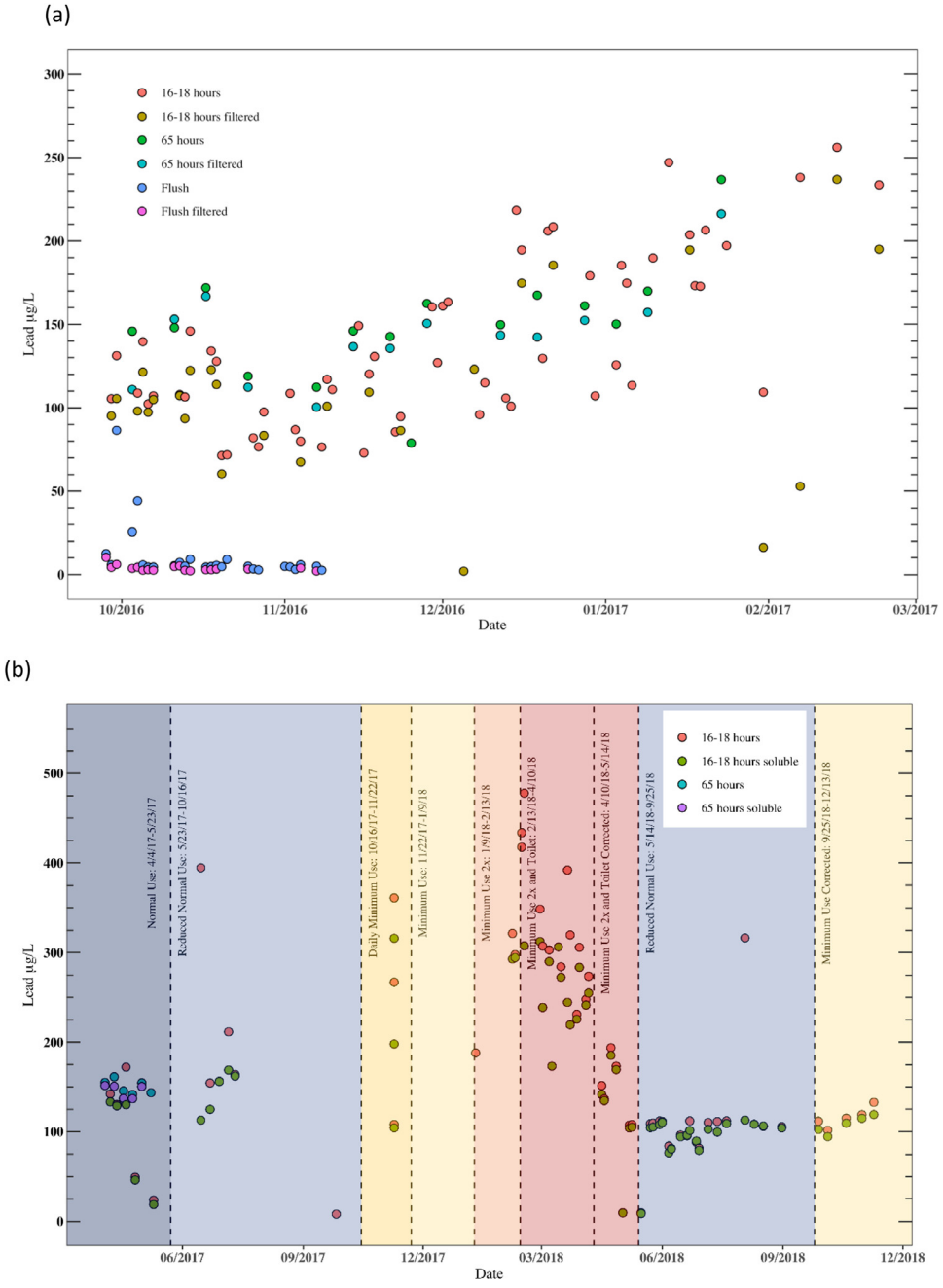


Fig. 2. Total and soluble lead samples collected directly from the LSL after various stagnation times: (a) shows direct LSL results during the LSL conditioning period (flow bypassed HPS), and (b) direct LSL results during the period that the LSL was in line with the entire HPS.

Peak Pb concentrations in the profiles were considerably lower than direct LSL total Pb stagnation samples (144–155 µg/L) collected within 2 weeks of the profiles, and the difference increased as the distance between the faucet and LSL increased. The maximum Pb concentrations in profiles from faucets 1, 2, 3, and 4 (from farthest to closest to LSL) were 26, 30, 40, and 60 µg/L, respectively (Fig. 4). Peak Pb profile concentrations measured from faucets 1, 2, 3, and 4 were only 17%, 21%, 28%, and 42% of the soluble Pb levels measured directly from the LSL during the same time period. These observations demonstrated that maximum Pb levels measured in sequential profiles taken at a faucet will be lower than Pb levels of water directly in contact with the LSL (unless particle Pb release is important) because of longitudinal dispersion effects, and the difference becomes greater as the distance from the LSL increases. As a result, associations between peak Pb concentration and solubility-controlling minerals on an LSL surface cannot be gained from sequential profiles, unless a sampling port in a home is directly near the LSL like it was in the HPS. Factors not considered in this work, including sequential sample volume and sample flowrate, will likely further impact the peak Pb sample concentration.

In theory given that the majority of the Pb originates from the LSL, the total mass of Pb across the profile should be similar to the total mass of Pb within the smaller volume of water in direct contact with the LSL. During the timeframe when profile sampling was performed, the total mass of Pb released directly from the LSL averaged 38 µg. The calculation was based on the LSL pipe volume and Pb concentration measured directly from the LSL. In com-

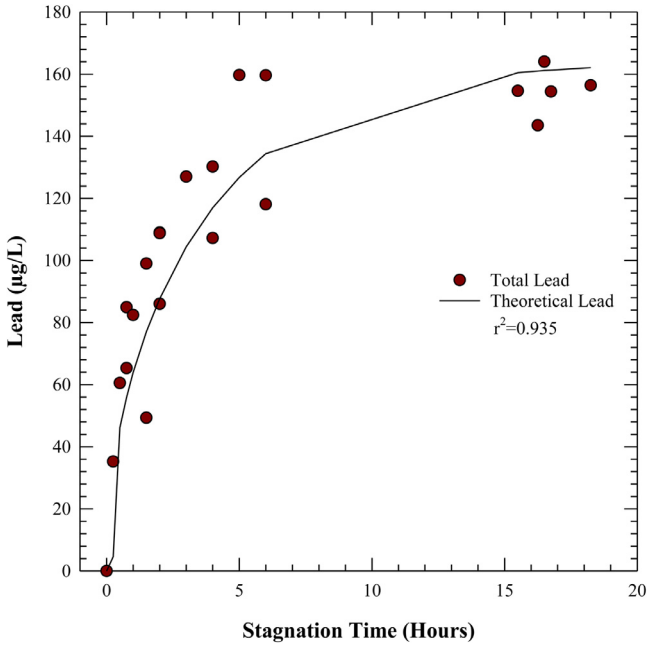


Fig. 3. Results of stagnation profile sampling directly from the lead service line. Direct LSL samples were collected in June 2017 (see Section S4). Lead results followed a logarithmic growth pattern with increased stagnation time (15 min-18 h). The theoretical curve (Kuch and Wagner, 1983; Lytle and Schock, 2000) was modeled to fit with measured lead data.

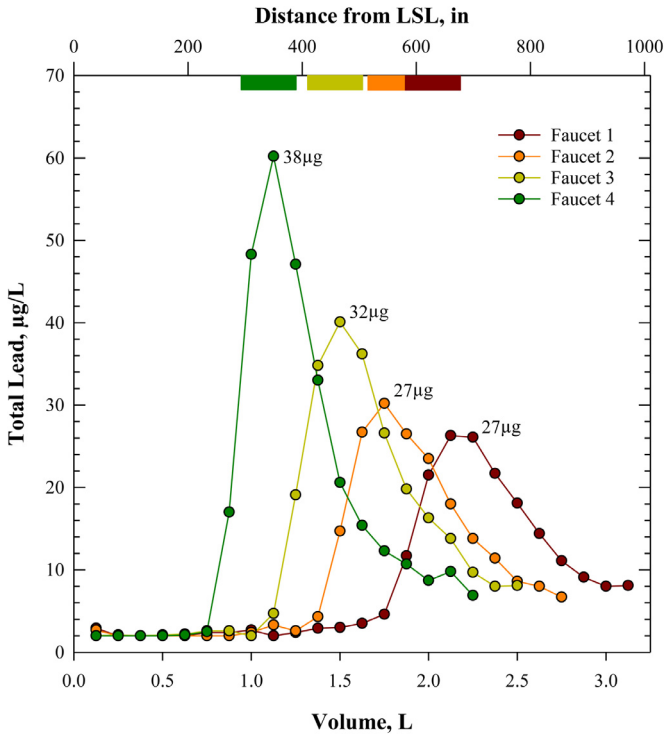


Fig. 4. Sequential profile of total lead in water collected from all four taps in the home plumbing system, collected on separate days in May and June 2017 (see Section S4). Color-coded boxes at the top of the graph represent the location of the lead service line in relation to the profile in matching color. The calculated total lead mass (µg) is given for each profile. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

parison, the total accumulated mass of Pb in water in the profile from faucets 1, 2, 3, and 4 was calculated at 27, 27, 32, and 38 µg, respectively (Fig. 4). Expressed in terms of mass balances, faucets 1, 2, 3, and 4 saw 69, 70, 83, and 99%, respectively, of the original Pb mass built up on the LSL. Pb recovery decreased as faucet distance from the LSL increased. This is because faucets with the greatest distance from the LSL had a Pb concentration dispersion tail that extended beyond the full sequential series volume and dispersion of Pb beyond the LSL volume boundary during stagnation. Additional sequential samples would have improved the balances.

Sequential profile data were also used to calculate equivalent first draw 1 L samples and average Pb concentrations across all sequential samples at each faucet. Equivalent first draw samples from faucets 1, 2, 3, and 4 (from farthest to closest to LSL) were 2.3, 2.1, 2.2, and 9.7 µg/L, respectively. Faucet 4 had the highest equivalent first draw Pb concentration because it was closest to the LSL and therefore capable of capturing direct Pb concentrations from the LSL. When compared to the respective peak Pb concentrations, Pb in first draw samples were 11, 14, 18 and 6 times lower for faucets 1, 2, 3 and 4, respectively. The results were variable within the HPS, greater than observations calculated from profiles reported by others (Del Toral et al., 2013; Triantafyllidou et al., 2015; Sandvig et al., 2008; Cartier et al., 2011) and impacted by distance between the sampling location and LSL (and other factors not considered here such as sequential sample volume). Average Pb concentrations in profiles from faucets 1, 2, 3, and 4 were 8.5, 9.8, 12.7, and 16.8 µg/L, respectively.

3.4. First draw, RDT and manual composite sampling comparisons

First draw, RDT and manual composite samples collected from the designated kitchen faucet (i.e., faucet 3) were compared under the following water use scenarios (1) whole house *normal use* (120 gpd), (2) whole house *reduced normal use* (60 gpd), (3) single faucet *daily minimum use* (1.4 gpd all days), (4) corrected single faucet *minimum use* (1.4 gpd on sampling days), (5) single faucet *minimum use (2x)* (2.8 gpd, on sampling days), and (6) single faucet *minimum use 2x and toilet* (6.6 gpd, on sampling days), (Figs. 2, 6, 7 and Section S4). All three sampling approaches were performed within 24 h of each other, and all data is summarized in Table S2. On non-sampling days, the water was used according to the *normal use* protocol when normal use was being evaluated or *reduced normal use* protocol on all other flow evaluation periods except daily minimum use, where all days were operated under *minimum use* conditions.

Under the normal (whole house) water usage pattern (Fig. 5), first draw sampling resulted in the smallest average total Pb concentration of 2.9 ± 2.9 µg/L (n = 14) (range of 0.7–12.8 µg/L) although there was one spike at 12.8 µg/L (Fig. 5). RDT sampling resulted in the greatest average total Pb concentration of 4.9 ± 3.4 µg/L (n = 14) (range of 2.2–14.3 µg/L), and the highest single observed concentration of 14.3 µg/L, for samples during normal use. Manual composite sampling produced an average total Pb concentration of 3.7 ± 1.1 µg/L (n = 13) and the tightest range (least variability in results) for samples during normal (whole house) use. Sample comparisons were statistically different (H = 12.1, p = 0.002). Dunn's pairwise comparison found the first draw sample to be different from both the RDT and manual composite samples during normal use.

A statistically significant increase in first draw (U = 90.5, p = 0.01) and manual composite samples (U = 88, p = 0.012) to average Pb of 4.6 ± 3.1 µg/L (n = 26) (range of 2.0–14.3 µg/L) and 7.9 ± 8.7 µg/L (n = 27) (range of 2.0–44.6 µg/L), respectively, was observed after reducing the daily water usage by 50% (whole house use). The Pb concentration of RDT samples during this period av-

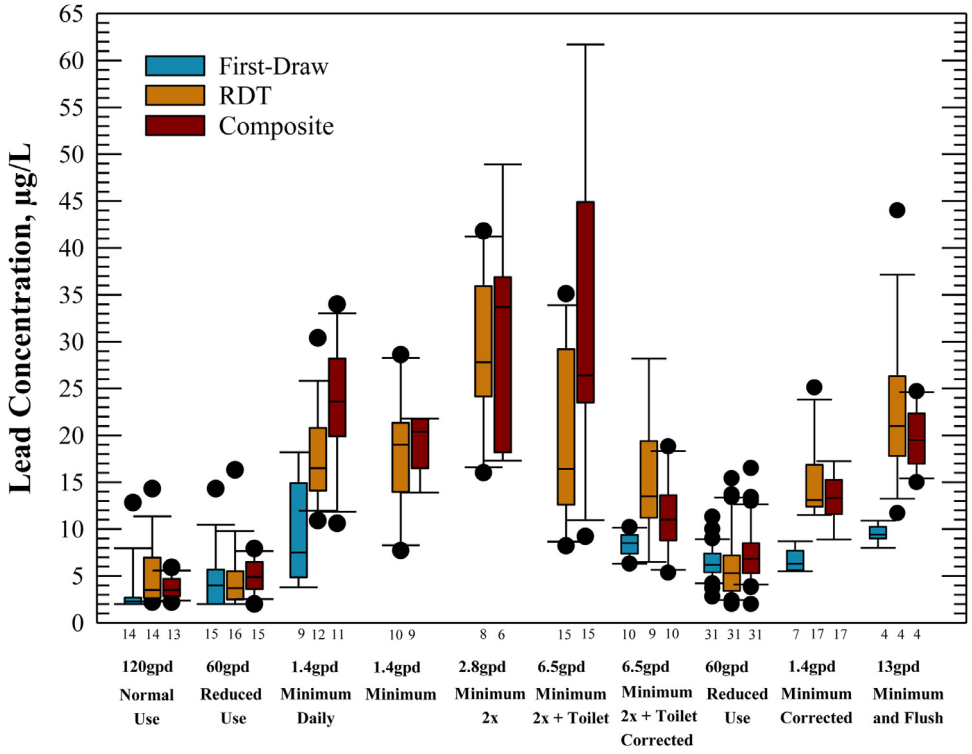


Fig. 5. Lead concentrations at Faucet 3 resulting from different sampling methods as a function of daily water usage. The number above each water usage label represents the amount of water, in gallons, flowing through the system on sampling days. Boxes span the range of the 25th percentile and the 75th percentile data value. The solid line within the box represents the median (50th percentile). The whiskers represent the maximum and minimum data values and each black point represents and outlier. The number of samples in each condition is indicated below the x-axis. First-draw samples were not included in this plot during periods where sampling occurred on the incorrect sampling days. The usage patterns clarified as "corrected" have first-draw data that was collected on the correct sampling days. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

eraged 6.5 ± 8.4 µg/L (n = 27) (range of 2.0–45.8 µg/L), and the increase from normal use was not statistically significant (Mann-Whitney rank sum test, U = 148.5, p = 0.461). Of note, the release of Pb from the LSL increased between the sampling periods from an average of 150 to 182 µg/L (see Table S2) likely due to more contact time between the water and the Pb scale with increased release possibly due to water quality changes (e.g., pH) near the LSL wall. Consequently, this increased Pb release from the LSL may have influenced the Pb increase observed in tap samples along with any dispersion issues. The three samples were statistically different (H = 16.6, P < 0.001) from each other during reduced normal use with the first draw sample being the lowest and the composite sample the highest.

Pb concentrations in samples collected using all three sampling approaches increased when a daily minimum use schedule (only the kitchen faucet used on all days) was adopted. The average first draw sample increased to 9.8 ± 5.5 µg/L (n = 9) (range of 3.8–18.2 µg/L), RDT increased to 18.0 ± 5.2 µg/L (n = 12) (range of 10.9 µg/L-30.4 µg/L), and manual composite increased to 23.5 ± 6.6 µg/L (n = 11) (range of 10.6–34 µg/L). First draw (U = 11, p < 0.001), RDT (U = 15, p < 0.001), and manual composite samples (U = 22, p < 0.001) all had statistically significant increases in Pb compared to reduced normal use samples. The increase could not strictly be attributed to the change in water use pattern, however, because the Pb release from the LSL increased on average to 245 µg/L from 182 µg/L during the daily minimum use sampling period.

When reduced normal use samples were compared to minimum use (i.e., only the kitchen faucet) only on sampling days (Tuesday and Thursday), Pb concentrations increased significantly for RDT (U = 15, p < 0.001) and manual composite samples (U = 22, p < 0.001). RDT and manual composite samples averaged 18.5 ± 5.9 µg/L (n = 10) (range of 7.7–28.6 µg/L) and 19.1 ± 3.0 µg/L (n = 9) (range of 13.9–21.8 µg/L), respectively. There was no statistical difference between the two sampling approaches (U = 36, p = 0.487) during minimum use. The role of LSL contributions could not be considered because direct LSL samples were not collected during this time. Although first draw samples could not be compared to other sampling approaches during this period because they reflected the reduced flow use the day prior to minimum use conditions, they were used to illustrate changes in Pb release from LSL. First draw samples statistically increased (U = 11, p < 0.001) to an average 12.8 ± 3.0 µg/L (n = 9) (range of 8.4–16.9 µg/L). When "daily minimum use" samples were compared to "minimum use" samples (only on sampling days), no statistical difference was found for first draw (U = 27.5, p = 0.269), RDT (U = 54, p = 0.972), or manual composite samples (U = 29, p = 0.128).

Doubling the minimum daily use to 2.8 gpd (only the kitchen faucet) significantly increased Pb concentrations for RDT (U = 13.5, p = 0.006) and manual composite sample (U = 13, p = 0.03) Pb levels while Pb release from the LSL slightly increased to 255 µg/L on average. RDT and manual composite samples averaged 28.7 ± 7.6 µg/L (n = 10) (range of 16.0–41.8 µg/L) and 31.4 ± 10.5 µg/L (n = 8) (range of 17.3–48.9 µg/L), respectively. There was not a statistical difference between the two sampling approaches (U = 32, p = 0.505). First draw samples could not be compared to other sampling approaches during this period because they reflected the reduced flow use the day prior to minimum use conditions. First draw samples did not statistically change (U = 27, p = 0.251) and averaged 11.4 ± 3.8 µg/L (n = 9) (range of 8.4–20.6 µg/L) during the period.

When toilet flushing was added to minimum flow conditions to bring the total daily flow to 6.6 gpd, Pb concentrations significantly decreased for RDT ($U = 41$, $p = 0.063$) but not manual composite samples ($U = 55$, $p = 0.771$). RDT and manual composite samples averaged 20.9 ± 9.1 μg/L ($n = 15$) (range of 8.2–35.1 μg/L) and 32.8 ± 17.7 μg/L ($n = 15$) (range of 9.2–73.4 μg/L), respectively. There was a statistical difference between the two sampling approaches ($U = 68$, $p = 0.068$). The increase could not strictly be attributed to the change in water use pattern because the Pb release from the LSL increased on average to 347 μg/L during the sampling period. Although first draw samples could not be compared to other sampling approaches during this period because they reflected the reduced flow use the day prior to minimum use conditions, they were used to assess changes in Pb release from LSL. First draw samples did not statistically change ($U = 49.5$, $p = 0.296$). First draw samples averaged 11.8 ± 2.8 μg/L ($n = 15$) (range of 9.3–20.8 μg/L).

The first draw sampling protocol was corrected (i.e., samples were collected on the day following reduced usage) on April 10, 2018 so that samples were collected the morning following minimum use with three toilet flushes. The corrected method continued throughout the rest of the study so first draw samples could be compared to manual composite and RDT samples (see Fig. 5). There was a statistically significant decrease in Pb from the incorrect first-draw sampling approach to the corrected one ($U = 7$, $p < 0.001$); however, the decrease in Pb was likely caused by the LSL, which saw the average Pb release decrease to 126 μg/L. The average concentrations of RDT and manual composite samples decreased to 15.5 ± 6.5 μg/L ($n = 10$) (range of 6.5–28.2 μg/L) and 11.5 ± 3.7 μg/L ($n = 10$) (range of 5.3–18.8 μg/L), respectively. A statistically significant difference between the three samples (Kruskal-Wallis test, $H = 12.2$, $p = 0.002$) was noted and further pairwise comparison clarified that the difference was between the first draw sample and the RDT sample.

After returning to reduced normal use, Pb levels in first draw samples decreased to an average of 6.4 ± 1.8 μg/L ($n = 31$) (range of 2.8–11.3 μg/L), and the decrease was statistically significant ($U = 52$, $p = 0.002$). There was also a statistically significant Pb concentration decrease in RDT ($U = 23$, $p < 0.001$) and manual composite samples ($U = 54$, $p = 0.002$) which averaged 5.9 ± 3.6 μg/L ($n = 31$) (range of 2–15.4 μg/L) and 7.4 ± 3.1 μg/L ($n = 31$) (range of 2–16.5 μg/L), respectively. There was a statistically significant difference between the three sample types (Kruskal-Wallis test, $H = 6.68$, $p = 0.035$), and further pairwise comparisons using a Dunn's test showed that the difference occurred between RDT and composite samples. During this time, the direct LSL samples showed Pb release only slightly decreased to an average of 114.0 ± 51.8 μg/L ($n = 18$) (range of 81–316 μg/L) indicating that the change of Pb levels in water samples was attributed to water use change.

After water usage returned to minimum use only on sampling days, the first draw samples remained low with an average of 6.8 ± 1.3 μg/L ($n = 6$) (range of 5.5–8.7 μg/L), and the change was not statistically significant ($U = 81$, $p = 0.635$). There were statistically significant Pb increases in RDT ($U = 41.5$, $p < 0.001$) and manual composite samples ($U = 41.5$, $p < 0.001$), to averages of 14.6 ± 4.1 μg/L ($n = 16$) (range of 11.5–25.1 μg/L) and 13.1 ± 2.8 μg/L ($n = 16$) (range of 8.8–18.6 μg/L), respectively. There was a statistically significant difference between the three samples ($H = 15.1$, $P < 0.001$) and pairwise comparisons yielded a significant difference between the first draw sample compared to both RDT and manual composite samples. Pb release from the LSL was within the same ranges as the previous (reduced normal use) period, indicating that the change of Pb levels in water samples was attributed to water use change.

3.5. Manual composite sample sequence

The 60 mL side-sample of water was collected and poured-off into the manual composite sample bottle after a glass of water was taken in preceding data analysis. Throughout most of the study, multiple manual composite samples were collected to observe any impact the collection sequence might have on results. Results through multiple water usage phases (9–31 samples per usage period) were compared (Figure S3). There were no Kruskal-Wallis statistical differences between the sample sequence during minimum use and toilet ($H = 3.774$, $p = 0.151$), reduced normal use ($H = 1.297$, $p = 0.523$), and during minimum use for the first ($H = 3.261$, $p = 0.196$) or second occurrence ($H = 4.350$, $p = 0.114$). A statistical difference was observed after minimum use was doubled ($H = 6.504$, $p = 0.039$). Further pairwise comparison during this period showed the difference occurred when composite sample 2 was lower than composite samples 1 and 3 (Figure S3).

4. Discussion

The overarching conclusion from this work is that a single Pb level representing the HPS did not exist. A wide range of Pb values were measured in water depending on sampling approach, water usage, sampling tap, water quality, presence of Pb particles, and other factors not specified in this work. There was also inherent temporal variability within each sampling approach for similar reasons. Lead release from the primary source of Pb, the LSL, increased rapidly with stagnation time and plateaued after 7–18.5 h of stagnation, consistent with the solubility of Pb(II) carbonate minerals (Schock et al., 1996). Approximately 55% of the equilibrium Pb concentration was reached within 2 h of stagnation. Soluble Pb was the dominant form of Pb released from the LSL accounting for an average of 89.2% ± 10.3% of total Pb in water. More importantly, Pb released from the LSL had a large, direct impact on all samples measured at the HPS kitchen tap. Exposure-based manual composite samples were particularly sensitive. Subtle pH changes (particularly between pH 7.5–8) likely associated with inconsistent operation of the pH feed system or chemical changes within the LSL during stagnation likely had an important impact on the Pb levels of water directly in contact with the LSL. Thus, measuring water Pb levels from an LSL following stagnation is important to understanding corrosion control mechanisms and effectiveness as well as Pb solubility and pipe scale relationships.

Service lines connecting the home to the water main are not easily accessible by the homeowner and drinking water taps are usually not close enough to the service line to collect a representative sample. Taps are typically installed many feet past the service line, and water movement through the plumbing creates diffusion features that impact measured Pb (Lytle et al., 2019). Therefore, it is impossible to collect a tap sample that represents the Pb concentrations of water directly in contact with an LSL.

Water usage greatly impacted total Pb levels in all water sampling approaches, although the degree of sensitivity was greater in manual composite and RDT samples. The lowest and highest mean total Pb levels of first draw, RDT, and manual composite samples differed by factors of 1.5, 5.8, and 8.9, respectively. The average manual composite and RDT samples were equal to or greater than first draw samples throughout the study (Fig. 6). The differences grew larger as water usage decreased likely due to water quality issues as water ages, including axial differences in water quality with the water at, and within, the Pb scale being the most important. Measured Pb in RDT samples ranged between 0.9–2.6 (average 1.8) times greater than corresponding first draw samples. Measured Pb in manual composite samples ranged between 1.2–2.8 (average 1.9) times greater than corresponding first draw samples. On average, manual composite and RDT samples

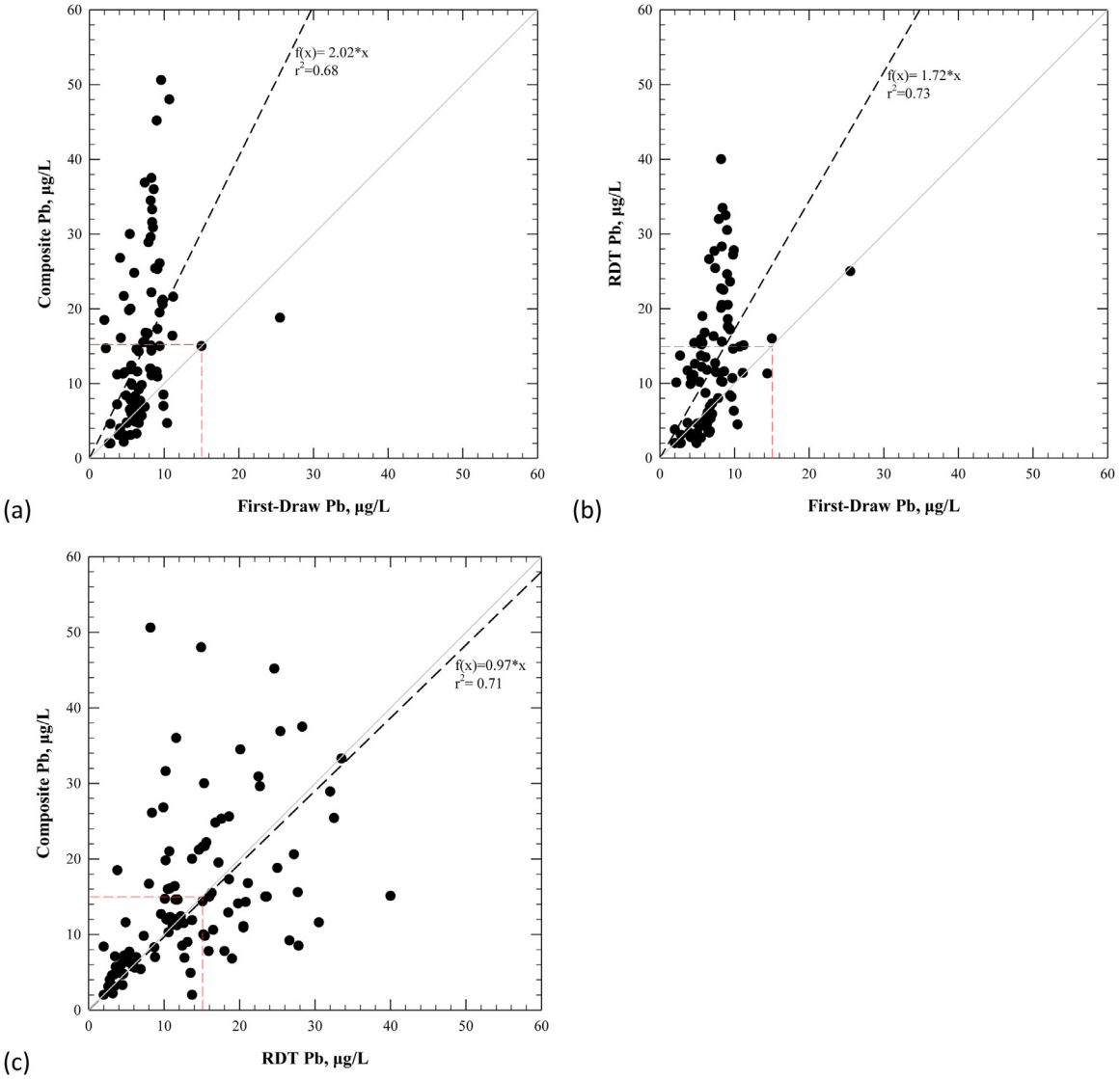


Fig. 6. Comparison of soluble lead results for each sampling approach. Black dots each represent samples of different methods collected on the same day. Solid gray lines represent a 1:1 ratio. The red boxes show 15 µg/L of lead. The black dashed lines represent the projected trend of sample ratios based on real data. Data is compiled from all water usage patterns. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

were similar throughout the study (Fig. 5) which was consistent with what Riblet et al. (2019) found when comparing RDT samples with a proportional composite sampling method.

Manual composite, RDT, and first draw samples were greater than 15 µg/L (EPA's action level for Pb) in 36%, 34%, and 5% of samples (Fig. 6). Lead in tap samples were mostly soluble with the average soluble Pb being 85.1% ± 10.6% for manual composite, 90.8% ± 10.6% for RDT and 86.1% ± 18.2% for first draw samples. The sampling approaches most reflective of exposure (manual composite and RDT) were most impacted by water usage patterns (Riblet et al., 2019). Lastly, Pb measured directly from the LSL ranged between 8 and 40 (average 13) times greater than corresponding manual composite samples, between 8 and 30 (average 14) times greater than RDT samples and between 15 and 52 (average 24) times greater than first draw samples.

5. Conclusions

- Pb levels in water samples collected directly from the LSL increased to the maximum equilibrium concentration after 7–18.5 h of stagnation. Pb levels increased most rapidly early in the stagnation profile; after only 2 h, approximately 55% of the equilibrium Pb concentration was reached.
- Lead released from the LSL during this work was dominantly in the soluble form. Measured soluble Pb levels were consistent with Pb levels predicted by Pb(II) carbonate mineral (hydrocerussite and cerussite) solubility throughout the study.
- Sequential sample profiles accurately identified the location of the LSL based on Pb peak position. Maximum profile Pb concentrations were a fraction of the equilibrium Pb concentration measured directly from the LSL. The fractions decreased as the plumbing distance between the faucet and LSL increased due to dispersion effects. As a result, conclusions regarding Pb solubility associated with LSL pipe scale minerals cannot be directly drawn from sequential profiles.
- Water usage greatly impacted total Pb levels across all sampling methods, although the sensitivity was greatest in manual composite and RDT samples. Manual composite and RDT samples were equal to or greater than first draw samples throughout the study. Pb differences between the sample grew larger as water usage decreased, demonstrating the impact of water age on water quality.

- Pb levels for manual composite and RDT samples were not statistically different, on average. Provided enough samples are collected, RDT samples resembled Pb exposure sampling (ie. composite sampling strategies).
- Different water sampling approaches produced different Pb results. The comparisons clearly demonstrated the importance of selecting the appropriate sampling method to answer the desired question. For example, based on this work, sequential sampling is useful to identify Pb plumbing sources including LSLs and assess corrosion control treatment effectiveness. Manual composite sampling provides a reasonable estimate of exposure whereas RDT samples, if collected in statistically appropriate numbers, may also provide exposure insights. Water samples collected directly from the LSL provides total Pb release details and Pb solubility information.

Notice

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2021.117071.

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