

Profile Sampling To Characterize Particulate Lead Risks in Potable Water

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S Supporting Information

ABSTRACT: Traditional lead (Pb) profiling, or collecting sequential liters of water that flow from a consumer tap after a stagnation event, has recently received widespread use in understanding sources of Pb in drinking water and risks to consumer health, but has limitations in quantifying particulate Pb risks. A new profiling protocol was developed in which a series of traditional profiles are collected from the same tap at escalating flow rates. The results revealed marked differences in risks of Pb exposure from one consumer home to another as a function of flow rate, with homes grouped into four risk categories with differing flushing requirements and public education to protect consumers. On average, Pb concentrations detected in water at high flow without stagnation were at least three to four times higher than in first draw samples collected at low flow with stagnation, demonstrating a new “worst case” lead release scenario, contrary to the original regulatory assumption that stagnant, first draw samples contain the highest lead concentrations. Testing also revealed that in some cases water samples with visible particulates had much higher Pb than samples without visible particulates, and tests of different sample handling protocols confirmed that some EPA-allowed methods would not quantify as much as 99.9% of the Pb actually present (avg. 27% of Pb not quantified).

Corrosion Control		Durable Scale	Non-Durable Scale
	Optimized	Low Soluble Pb Low Particulate Pb Low Risk	Low Soluble Pb High Particulate Pb Filter or Bottled Water
Not Optimized		High Soluble Pb Low Particulate Pb Flush 30 sec - 2 min	High Soluble Pb High Particulate Pb Filter or Bottled Water

INTRODUCTION

As the United States strives to reduce levels of elevated blood lead (Pb) in children,¹ accurate quantification of Pb risks from potable water taps in individual homes will become of greater concern.^{2,3} Recent studies have identified significant increases in blood Pb for children^{4,5} and young women⁶ at water lead exposure levels that are common at many homes in older cities with Pb-bearing plumbing. Recent analyses of Pb exposure in U.S. schools,⁷ Washington, D.C.,⁸ and Germany⁶ have confirmed expectations that public health interventions such as filters, flushing, and the use of alternative water sources can markedly reduce blood Pb levels and even incidence of fetal death due to water Pb exposure.

Regulatory Limitations. Pb in drinking water is regulated by the U.S. Environmental Protection Agency (US EPA) under the Lead and Copper Rule (LCR). The LCR sets a Pb action level (AL) of 15 ppb in first draw samples collected from consumer taps after 6 h of stagnation.⁹ If the 90th percentile Pb concentration in these samples exceeds the 15 ppb AL, then the rule requires the utility to take measures that can include adding treatment to control plumbing corrosion, adding source water treatment, replacing lead service lines, and implementing consumer education on avoiding water lead risks.⁹ Research by EPA scientists and others has confirmed that existing EPA sampling methods and analysis protocols can sometimes substantially underestimate Pb in drinking water, especially if

Pb is present in particulate form or if Pb service lines are present.^{2,3,10–12} The extent to which established EPA-approved analysis protocols can miss particulate Pb was recently verified,^{10,13} in that for some worst-case samples as much as 81% of the Pb was missed using the standard EPA sample handling methods, which involve acidification to pH 2 for 16 h before removing an aliquot for analysis.¹⁰ Furthermore, up to 98% of the Pb was missed if samples were not thoroughly mixed upon acidification, which is not explicitly required by some methods that are accepted by EPA for analysis.¹⁰

The LCR was also intended to provide a system-wide, community-based reduction in water Pb exposure, and it is acknowledged that it does not control or account for harmful exposures at individual taps.¹⁴ Dependent on factors such as plumbing material, water use patterns, prior disturbances, and galvanic corrosion, Pb levels in individual homes can vary by 2 orders of magnitude or even more.^{7,8,12,15,16} As public health programs begin to more carefully scrutinize individual cases of Pb poisoning and Pb exposure, it is important to identify and assess Pb in water risks at individual taps.^{4,7}

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Traditional Pb Profiling. A tool that has recently been used to assess risks at individual taps is Pb profiling, or collecting a series of samples of Pb in water as it flows from the tap to ensure that water in contact with all types of Pb-bearing plumbing is sampled.^{12,15–19} Typical profiling produces graphs of Pb concentration versus either flushing time or flushing volume.^{12,16–20} Compared to collection of a standard EPA single “first-draw” sample, this method has the advantage of providing more detailed information about the sources of Pb present in the home’s water supply and can often detect Pb hazards from the service line that first draw sampling can completely miss.^{12,15,18,19} Profiles have also been used to better estimate the required flushing time for consumers to protect public health^{15,17} and to identify homes with high risk lead service lines for additional scrutiny and public health protection.²¹ However, many of these profiles have been collected at a very low flow rate of less than 2 L per minute,^{2,17,18,22} while other studies did not report the flow rate.¹⁹

Importance of Flow Rate to Pb-in-Water Risks. Several studies have investigated the link between higher flow rate and mobilization of Pb particulates to drinking water.^{20,22–25} If there is a reservoir of nonadherent particulate Pb on plumbing surfaces that is relatively easy to mobilize via hydraulic scour, then much more Pb can be released at higher flow rate.^{2,22–24} However, if nonadherent particulate Pb reservoirs are absent within the pipe, higher flow rates during sampling will tend to have no effect or would even decrease soluble Pb concentrations due to dilution. This can explain why Britton reported that the concentration of Pb as a function of flow rate is minimized at intermediate flow rates in some systems.²⁰ Conceptually, using lower flow rates during sampling will either have no effect on or increase the concentration of soluble Pb dissolved from Pb-bearing plumbing into the water,²⁵ whereas lower flow rates minimize mobilization of particulate Pb.^{20,22–24} The release of total Pb (the sum of soluble Pb and particulate Pb) at various flow rates is highly dependent on the water chemistry, types and durability of Pb deposits, Pb solubility and dissolution rates, and specifics of the premise plumbing system, including pipe materials and pipe diameters.^{23,24} Flow rates at individual taps also vary according to design and consumer behaviors, which in turn can profoundly affect the concentration of Pb detected or the risk of consumer exposure.^{2,23} Thus, the tap flow rate during water sampling or consumer use represents an important dimension in assessing both the effectiveness of corrosion control programs across a system and public health risks in individual homes. After the link between flow rate and particulate lead release was discovered,²⁶ recent work adopted a high flow rate profiling approach, which was used to reveal significant problems with elevated lead in Chicago.¹²

An Updated Profiling Method. To quantify the release of Pb to drinking water as an explicit function of flow rate, a new Pb profiling method was conceptualized and tested. By collecting a conventional Pb profile at very low flow rate after a long stagnation event, and immediately thereafter collecting additional profiles at moderate and then high flow rates, risks from particulate Pb release can be explicitly quantified with appropriate analytical methods.^{10,11,13} The importance of particulate release mechanisms has been highlighted in several recent studies, which linked spikes in particulate Pb release to water to physical pipe disturbances,^{8,12} galvanic corrosion,²⁷ and instances of childhood Pb poisoning from tap water.¹¹

Compared to traditional profiling conducted at low flow rates not representative of normal consumer use, this profiling approach can better quantify Pb exposures during a range of home water use scenarios, and results can be used to both provide improved public health advice to consumers and directly evaluate effectiveness of corrosion control for particulate Pb. The approach would also have potential to characterize health hazards from other inorganic contaminants in tap water such as arsenic^{28,29} and radionuclides,^{30–32} which can be concentrated in pipe scale.

■ MATERIALS AND METHODS

Site Selection. Samples were collected in the summer of 2011 in two cities, Washington, D.C. and Providence, RI. These cities were selected due to a recent history of lead action level exceedence and partial lead service line replacement. Washington, D.C. exceeded the lead action level after switching from chlorine to chloramine disinfectant in 2001¹⁵ and now controls lead release by dosing an orthophosphate corrosion inhibitor.²⁷ Providence, RI exceeded the lead action level in 2006 after changing pH from 10.3 to 9.7, and has since reversed the pH change.³³ Detailed recent water quality information for both cities has been recently reported elsewhere.^{27,34} At the time of sampling, Providence had not yet adjusted pH back to 10.3, and corrosion control was not optimized; in Washington, D.C., traditional optimized corrosion control practices (orthophosphate) were already in place at the time of sampling. Individual homes were selected on a volunteer basis from a pool of homes with known or suspected lead service lines. Twelve sites were selected each city, and a breakdown of the sites into “full lead service line,” “partial lead service line,” or “unknown” is given in the Supporting Information (SI) (Table S1). Due to inadequate representation of partial lead service lines in Washington, D.C. and full lead services lines in Providence, RI, a comparison of lead release from full versus partial lead service lines was not possible.

Flow Rate Selection. Flow rates were primarily selected to compare different consumer exposure scenarios. The low flow rate represents traditional profile sampling at 1 L/min, the medium flow rate represents the highest flow rate achieved during typical consumer use (kitchen tap fully open), and the high flow rate represents the worst-case scenario of the maximum achievable flow rate at the tap without an aerator. The latter flow rate is similar to that expected from some nondrinking-water taps such as Roman bath spouts and laundry room faucets. Differences in achievable flow rates at a given tap are responsible for the wide range of flow rates for the medium and high flow categories (Figure 1); flow rates used for sampling at each site are summarized in the SI (Table S1). These different flow rates also have mechanistic significance in terms of a transition between laminar and turbulent flow. Although precise Reynolds numbers (Re) at each site cannot be calculated due to lack of pipe diameter information, the low flow sample is expected to be in the laminar region (Re = 994 for a 3/4” service line).³⁵ However, flow in a 3/4” service line can be expected to become turbulent (Re > 4000)³⁵ once the flow rate exceeds 4 L/min, which was exceeded at all sites for the high flow sample and at all but one site for the medium flow sample.

Sample Collection. Profiles of Pb concentration in water were collected using new, wide-mouth, one-liter HDPE bottles (Fisher Scientific). Three sequential profiles were collected after a minimum of 6 h stagnation in each home (Figure 1). For

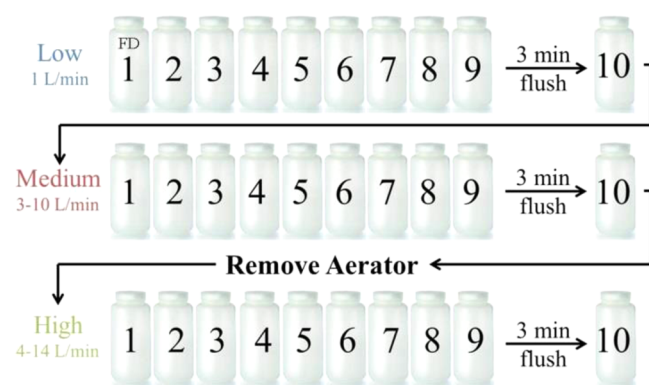


Figure 1. Illustration of the sequential sampling protocol used in each home. One-liter water samples are collected in the sequence shown at low, medium, and high flow rates after at least 6 h of stagnation. The first liter of the low flow profile represents the first draw (FD) after stagnation.

the first profile, nine sequential liters of water were collected at a low flow rate of approximately 1 L/min. The tap was then allowed to run at the same low flow rate for 3 min before collecting a tenth one-liter sample. Without allowing additional stagnation time, the tap was then fully opened (3–10 L/min) and the second profile, reported here as “medium flow,” was collected in the same manner. The faucet aerator was then removed, and two one-liter samples were collected at a low (1 L/min) flow rate with 3 min of flushing between the first and second sample. These samples represent particles released due to aerator removal, and in some cases contain very high (more than 10 mg/L) lead levels as a result. The lead concentration in each of these samples is available in Table S2 in the SI. These samples are not included in this analysis because they do not reflect lead levels resulting from normal water use patterns. For the final profile, reported here as “high flow,” the tap was fully opened with the aerator off (4–14 L/min), and 10 L were collected in the same way as for the first two profiles. An aliquot from liters 3 and 5 from each of the three profiles (Figure 1) was filtered in the field through a 0.45 μm nylon syringe filter (Whatman). These samples were selected for filtering because the third to fifth liters of a profile often contain the water (and soluble or particulate Pb) derived from service lines,²¹ either by dissolution during stagnation or via particulate detachment caused by abrupt changes in flow.

Sample Analysis. This part of the study explored whether observations from previous work regarding the limitations of EPA-approved methods¹⁰ hold true for field samples designed to capture particulate Pb risks.

General Sample Analysis. Before analysis, samples were screened by eye for visible particles (SI Figure S1). Samples without visible particles, including all field-filtered aliquots, were acidified in the sampling container to 1% nitric acid (trace metal grade, Fisher Scientific) by volume. An aliquot was removed from each sample and adjusted to 2% nitric acid by volume for analysis a minimum of 30 h after acidification. For samples containing visible particles, most samples were acidified in the sampling container to 2% nitric acid by volume immediately after particulate screening. An aliquot was removed from each sample for analysis a minimum of 48 h after acidification.

Detailed Sample Analysis. A subset of the samples with visible particulates was subjected to a more detailed sequential sample preparation procedure (SI Figure S2). Only samples

with accompanying field-filtered aliquots were chosen for this detailed analysis. Five samples were collected with increasingly rigorous sample handling methods to detect particulate Pb. The first and second samples were taken directly from the unacidified bottle without and with shaking, respectively, as 20 mL aliquots. These aliquots were then acidified with 2% nitric acid by volume for 48 h before analysis. After taking Samples 1 and 2, the original sampling container was acidified to pH <2 with nitric acid. After 16 h, Samples 3 and 4 were taken without and with shaking, respectively. These aliquots were also adjusted to 2% nitric acid by volume for analysis. After taking the third and fourth sample, the original sampling container was adjusted to 2% nitric acid by volume. The fifth and final sample was taken 48 h after acidification with shaking and is deemed representative of “total Pb” herein, even though other research has indicated that, in rare instances, hydrochloric acid, heat, and/or days of digestion can be required to dissolve all Pb present in drinking water.^{11,13} All samples were analyzed for total metals by inductively coupled plasma–mass spectrometry (ICP-MS; Thermo Scientific Thermo Electric X Series) using Standard Method 3125B.³⁶

RESULTS AND DISCUSSION

Pb Profiling and Consumer Risk. The concentration of Pb in water from a consumer tap as a function of time and flow rate as measured using the new profiling method (Figure 1) was variable and site-specific. However, homes could be grouped into four general categories of behavior, which also fit into current conceptualizations of behavior based on effectiveness of corrosion control as measured by levels of soluble lead in each system (Figure 2). Traditional corrosion control is based on lead solubility, and can include changes in alkalinity or pH or the addition of orthophosphate to form insoluble lead phosphate scales.³⁷ As we gain appreciation for the important role of particulate lead as a key route of exposure in modern premise plumbing systems, the concept of “corrosion control” must be expanded to explicitly consider aspects of scale durability and adherence as well as lead solubility. Ultimately, it is likely that particular mineralogies and cofactors such as iron and manganese will play critical roles in creating reservoirs of nonadherent lead as has been suggested in prior research.^{11,27,33,37–40}

In the first group of homes (Case A), the Pb concentration remained low in all samples at all flow rates (Figure 2A). For the example profile shown, Pb was less than 2 ppb in all samples. This type of Pb profile is expected in homes in which traditional corrosion control is optimized, leading to low soluble Pb, and also for which pipe scales are durable and adherent, leading to a low incidence of particulate Pb release. At these taps, the risk of Pb exposure was low regardless of flow rate.

In a second group of homes (Case B), the Pb concentration at low flow rate increased over the first few liters, reached a maximum value when collecting water that had been sitting in the Pb service line, and then decreased (Figure 2B). For these homes, the resulting profile was consistent with conventional wisdom that the highest risk from soluble lead derives from the water sitting in the lead service line.^{12,17,18} In the example shown, the Pb concentration reaches a maximum value of nearly 100 ppb in the fourth liter, which is the water contacting the lead service line, but is reduced to 15 ppb by the sixth liter as the water in the service line mixes with water from the main. At taps with a Pb release pattern dominated by soluble lead,

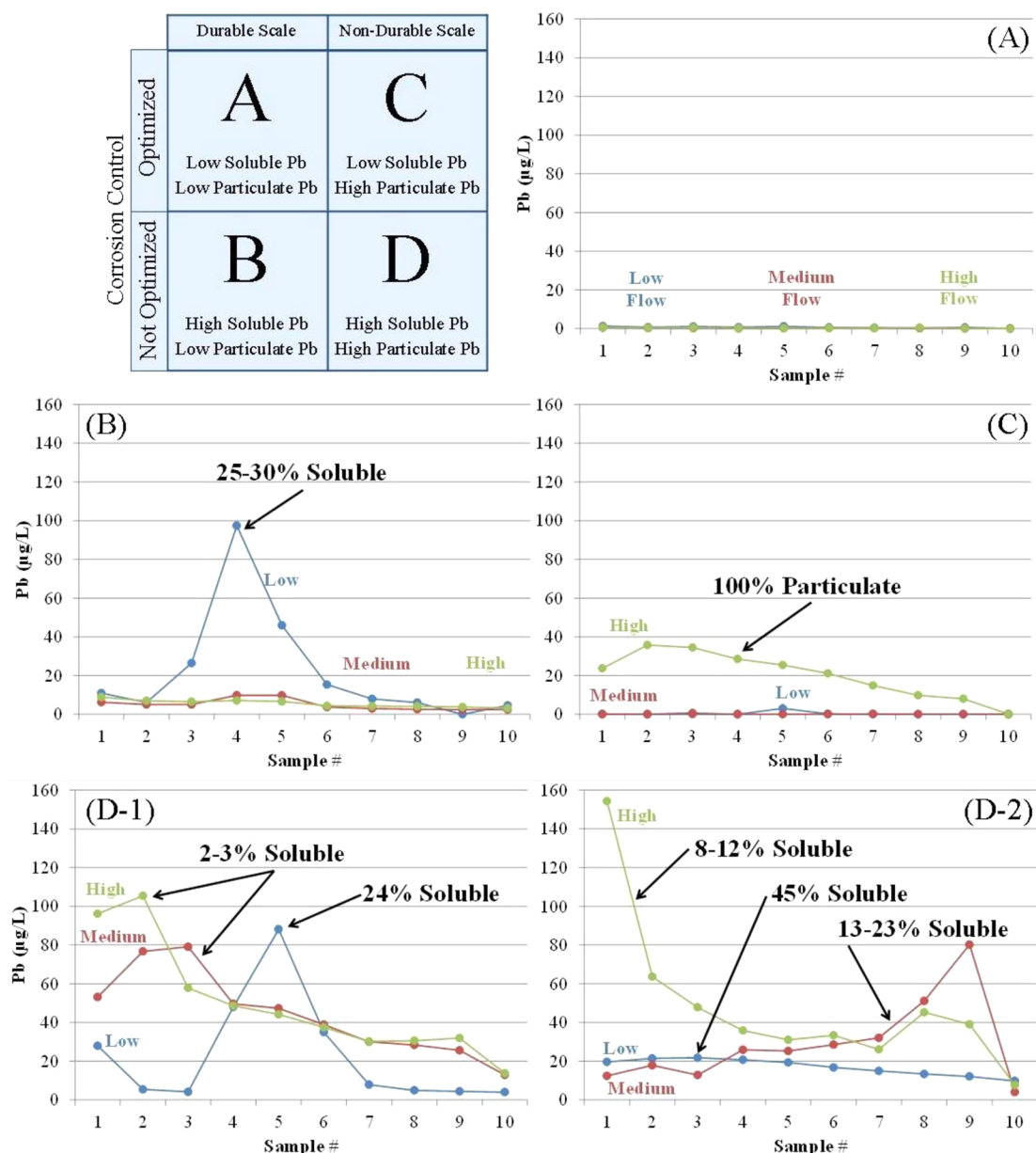


Figure 2. Upper left indicates four theoretical lead (Pb) release patterns and representative lead profiles based on patterns of release for (A) Washington, D.C. (Site 24); (B) Providence (Site 9); (C) Washington, D.C. (Site 4); (D-1) Providence (Site 16); (D-2) Providence (Site 12). “Soluble Pb” is defined as the Pb measured in the field-filtered aliquot taken from the 3rd and 5th liter in each profile, and is given as a percentage of the total lead measured in the sample.

very little additional Pb was released when the flow rate was increased to moderate and high flow rates without additional stagnation. This type of release is expected to occur when soluble Pb dominates (nonoptimized traditional corrosion control) and there are few reservoirs of detachable particulate Pb on plumbing surfaces (durable scale).²⁵

In the third group of homes (Case C), profiles collected at low flow showed very little Pb release, as in Case A; however, increases in flow rate caused spikes in Pb concentration due to particulate Pb detachment. In an exemplary profile (Figure 2C), the Pb concentration is less than 3 ppb for all samples collected at both low and medium flow rates, but spikes as high as 35 ppb were present at the highest flow rate. This release pattern is representative of a case in which traditional corrosion control is optimized, resulting in low soluble Pb concentrations, but the scales present on the lead-bearing plumbing are not durable,

resulting in erratic release of particulate Pb. If this home had been sampled using LCR methods or even a traditional lead profile, then consumption of water in this home would have been erroneously classified as presenting little or no risk to consumers, when, in fact, a relatively high risk of particulate Pb exposure risk exists.

In the fourth and final group of homes (Case D), high soluble Pb concentrations were observed as in Case B; however, particulate Pb spikes at higher flow rates also occurred that were equal to or worse than the peak Pb concentration from the service line after long stagnation (Figure 2D-1,2). These homes represent the worst case for exposure, since traditional corrosion control for soluble lead is not optimized and nondurable scales are present, resulting in release of high levels of both soluble and particulate Pb. In one of these homes (Case D-1; Figure 2D-1), the Pb profile at low

flow was similar to the Case B profile (Figure 2B), but when flow was increased, particulate Pb was released at high levels: up to 79 ppb at moderate flow and up to 105 ppb at high flow. In total, 17 min of cumulative flushing (12.5 min low flow; 4.6 min moderate flow) were required before Pb dropped below 15 ppb at a moderate flow rate. When the flow rate was increased, an additional 11 min of flushing were required to reach 15 ppb again. This demonstrates that in homes of this type, routine advice to flush lines for 30 s to 2 min is not adequately protective of public health. As with Case C, if this home had been sampled using typical Pb profiling, then the markedly different health risk to consumers in homes corresponding to Case B and Case D-1 would not be distinguishable. In some Case D homes, particulate Pb release was less predictable. For one home (Case D-2; Figure 2D-2), the Pb concentration was only slightly elevated at low flow, but higher flows were typified with two relatively large spikes of 80 and 154 ppb. It is believed that this type of behavior is caused by sporadic release of larger Pb particulates. As a rule the Pb levels did not systematically decrease with increased flushing time in Case D-2 situations.

Public health advice for consumers could be tailored depending on which of these four categories a given home falls into (Table 1). This case-specific advice may be especially

Table 1. Potential Public Health Advice for Individual Homes in Each Category Represented in Figure 2a

case	potential flushing advice
A	No special precautions needed.
B	Flush tap at high flow rate for 30 s–2 min before using water for cooking or drinking.
C, D	Flush tap at high flow rate for AT LEAST 10 min before using water for cooking or drinking ^a

^aFlushing time is likely unrealistic, and use of filters certified to remove Pb particles is recommended as an alternative.

important in homes with particulate Pb contamination, which in some cases can increase with flushing and is relatively unaffected by stagnation time,⁴¹ contrary to conventional wisdom and current consumer guidance based on soluble Pb release. Case A has low Pb regardless of flow rate or flushing time, while Case B can be considered dominated by soluble Pb and risks are expected to be mitigated after standard EPA advice to flush Pb service lines for 30 s to 2 min. However, problems associated with Cases C and D are dominated by particulate Pb, and in such situations consumers should probably use filters certified to remove Pb because risks cannot be adequately mitigated by realistic flushing times.

Sampling the Worst Case. Another goal of this work was to illustrate the extent to which sampling conditions could detect worst-case exposure. To do this, samples were separated into categories as follows: Case A = no samples above 15 ppb; Case B = at least one sample above 15 ppb at low flow, but no samples above 15 ppb at high flow; Case C = no samples above 15 ppb at low flow, but at least one sample above 15 ppb at medium or high flow; Case D = at least one sample at low flow and one sample at medium or high flow is above 15 ppb. Using these criteria produced a fairly even split, assigning 8 homes to Case A, 5 homes to Case B, 6 homes to Case C, and 5 homes to Case D. Boxplots for each category at each flow rate (Figure 3) demonstrate that C and D homes have greater variability in lead release, especially at high flow rates, which is expected based on the sporadic nature of particulate lead release. To

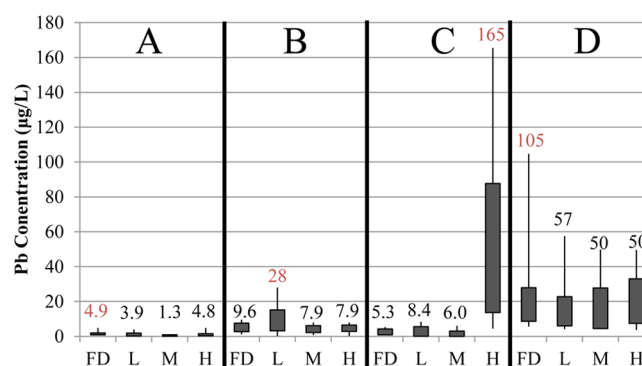


Figure 3. Box plot showing the lead concentrations for first draw (FD), low flow (L), medium flow (M), and high flow (H) samples for lead release categories A–D. The top and bottom of the box represent the 75th and 25th percentiles, respectively. The top and bottom whiskers represent the 90th and 10th percentiles, respectively. 90th percentile values are shown on the chart, with the highest value for each lead release category shown in red.

determine the worst case, 90th percentile values for each category and flow rate were compared (Figure 3; SI Table S3). As might be expected, the flow rate required to sample the “worst case” Pb concentration was dependent on the Pb release category. For the best and worst homes (Cases A and D), the first draw sample contained the highest 90th percentile Pb concentration, and the EPA LCR sampling protocol generally detected the risks to public health. For Case B, the highest Pb concentrations were observed at low flow, but not in the first draw sample, consistent with expectations based on a traditional profiling approach.¹² For Case C, the highest Pb concentration was observed at the highest flow rate, reflecting a particulate Pb hazard.

When all sites are considered together, samples collected at the highest flow rate had a 90th percentile value more than 4× higher than the first draw and 3.5× higher than samples collected by typical profiling at low flow rate (SI Table S3). Since the samples collected at the highest flow rate had been previously flushed for a cumulative 20 min at low and moderate flow rates and had no stagnation time, whereas samples collected at low flow rates had a minimum of 6 h stagnation, it is clear that flow rate can have a much greater impact on compliance with the EPA 90th percentile action level of 15 ppb than stagnation time in the systems studied herein. This confirms a previous research report that noted compliance with EPA standards can be “all in the wrist,” and recommended LCR sampling rates of less than 1 L/min in order to meet EPA standards.⁴² The obvious downside of this approach is that the actual lead in water when it is consumed can be much higher than when it is sampled, and the public would not be alerted to the true health risk that is present or instructed as to how to avoid it. Worst case sampling protocols designed to detect the health risk would require sampling of first draw after stagnation at a realistic (moderate to high) flow rate and also collection of a second draw sample that is representative of soluble and particulate lead from the service line. If a single profile is used to determine health risks and/or provide flushing advice, then samples should be collected at flow rates consistent with the highest flow rate realistically expected to be used by consumers.

Analytical Method and Pb Recovery. Field samples from this profiling study were also used to determine the extent to which Pb concentrations may be underestimated by some

accepted sample handling methods and acidification procedures.^{10,13}

Trends in Visible Particle Release. In the first part of this analysis, the idea that samples with high Pb could be identified visually, contrary to conventional wisdom and regulatory assumptions⁴³ derived from experiences with dissolved lead, was investigated. Samples with visible particles identified before sample preparation and analysis had higher Pb concentrations, on average, than those without visible particles (Table 2). In

Table 2. Summary of the Lead Concentration and Number of Samples (N) with and without Visible Particles^a

		N	average (ppb)	median (ppb)	range (ppb)
Washington, D.C.	WITH visible particles	182	32.3	3.2	ND–1801
	NO visible particles	158	2.2	1.2	ND–12.7
Providence, RI	WITH visible particles	255	13.5	5.0	ND–155.8
	NO visible particles	105	10.9	4.3	ND–110.1

^aConcentrations are reported for the most rigorous digestion method used in each case (1% nitric acid without visible particles, 2% nitric acid with visible particles). On average, the lead concentration is nearly 4× higher when particles are visible.

Washington, D.C., this difference was large and statistically significant (two-sided *t* test, *p* = 0.0045), with samples containing visible particulates having nearly 15× higher Pb on average when compared to samples without visible particles. However, both in Providence samples and another recent study in Chicago,¹² there were no significant differences in Pb concentration for samples with and without visible particles. One possible reason for this finding in Providence is a large number of nonlead particles due to a substantial iron corrosion problem at the time of sampling.³³ Similarly, in Chicago, some sampling took place during the fire department's annual valve exercising activity, resulting in high levels of nonlead particulates.¹²

The number of samples collected in this study with visible particles was large, with over two-thirds of all samples collected containing visible particles. Of these samples, approximately 40% were collected at high flow, 35% at medium flow, and 25% at low flow. Although no direct isolation and characterization of particles was undertaken during this study, some further information about the type of particles released can be gained by examining the concentrations of nonlead elements in each sample (SI Tables S4–S5). For example, in Providence, lead was often found to be correlated with iron, a trend which was further explored with follow-up sampling and reported elsewhere.³³ In Washington, D.C., lead particles were sometimes associated with phosphate, implying that lead phosphate solids were released, but at other times associated with tin or zinc, implying that brass or solder sources existed within the home.

Effect of Digestion Procedure. When samples were subjected to the more detailed sequential analysis, the standard EPA method gave similar results, on average, to the 2% acid digestion (Figure 4). However, if these samples were not shaken, then 0–99.9% (27% average) of the Pb was missed. If samples are not digested in the original bottle, as occurs in some customer service sampling by utilities,¹⁰ then very large

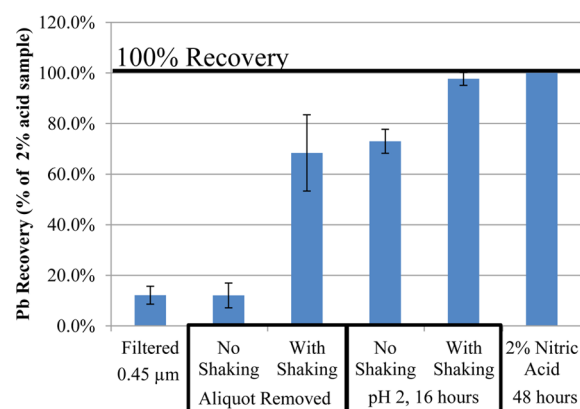


Figure 4. Plot of average lead recovery for each analytical procedure, calculated by dividing the concentration in the aliquots for each digestion procedure by the concentration of the most rigorously digested sample (2% nitric acid for 48 h). Error bars represent 95% confidence. Moving from left to right on the chart, the digestion procedure becomes more rigorous, and the average lead recovery increases. A total of *N* = 88 particulate-containing samples were used for this analysis; at least one sample from each of the 24 sites was used.

errors in quantification can occur. In this study, digesting samples outside of the original bottle missed 0–100% (88% average) and 0–100% (32% average) of the Pb without and with shaking before aliquot removal, respectively.

Regulatory Implications. As the EPA continues to consider changes to be made in the long term Revisions to the LCR, revising site selection and sampling protocols will be a key consideration.⁴⁴ This work demonstrates that Pb release/exposure can be a strong function of sample flow rate and site selection. In some cases, sampling at flow rates typical during normal use (“medium” flow) detected lead levels 4× higher than that in a low flow first draw sample, even without any stagnation time. In general, medium flow samples gave 60% of the average and 40% of the peak lead concentrations determined at low flow, demonstrating that flow rate is almost as important as stagnation in detecting worst-case consumer exposures. Furthermore, samples collected at realistic flow rates in homes believed to have lead service lines, which would be considered “worst case” sites under the current site selection criteria, ranged from nondetect to 180 ppb, demonstrating that the true “worst case” is not simply a function of service line material and is probably related to total water use, presence of disturbed service lines, longer service lines, particulate iron, and other factors.^{12,21,22,33,40,45}

■ ASSOCIATED CONTENT

⑤ Supporting Information

Site-specific information, concentrations of nonlead elements, and illustrations for sampling protocols. This material is available free of charge via the Internet at <http://pubs.acs.org/>.

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Notes

The authors declare no competing financial interest.

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REFERENCES

- (1) U.S. Department of Health and Human Services. *Healthy People 2020 Summary of Objectives*. [cited Sept 3, 2012; Available from: <http://healthypeople.gov/2020/topicsobjectives2020/pdfs/EnvironmentalHealth.pdf>].
- (2) Triantafyllidou, S.; Edwards, M. Lead (Pb) in tap water and in blood: Implications for lead exposure in the United States. *Crit. Rev. Environ. Sci. Technol.* **2011**, *42* (13), 1297–1352.
- (3) Brown, M. J. Margolis, S. Lead in drinking water and human blood lead levels in the United States. *Morbid. Mortal. Weekly Rep. (MMWR) Suppl.* **2012**, *61* (4).
- (4) Triantafyllidou, S.; Gallagher, D.; Edwards, M. Assessing risk with increasingly stringent public health goals: The case of water lead and blood lead in children. *J. Water Health* **2014**, *12* (1), 57–68.
- (5) Deshommes, E.; Prévost, M.; Levallois, P.; Lemieux, F.; Nour, S. Application of lead monitoring results to predict 0–7 year old children's exposure at the tap. *Water Res.* **2013**, *47* (7), 2409–2420.
- (6) Fertmann, R.; Hentschel, S.; Dengler, D.; Janßen, U.; Lommel, A. Lead exposure by drinking water: An epidemiological study in Hamburg, Germany. *Int. J. Hyg. Environ. Health* **2004**, *207* (3), 235–244.
- (7) Triantafyllidou, S.; Le, T.; Gallagher, D.; Edwards, M. Reduced risk estimations after remediation of lead (Pb) in drinking water at two US school districts. *Sci. Total Environ.* **2014**, *466–467* (0), 1011–1021.
- (8) Edwards, M. Fetal death and reduced birth rates associated with exposure to lead-contaminated drinking water. *Environ. Sci. Technol.* **2013**, *48* (1), 739–746.
- (9) United States Environmental Protection Agency. Maximum Contaminant Level Goals and National Primary Drinking Water Regulations for Lead and Copper. *Fed. Regist.* **1991**, *56* (110), 26460–26564.
- (10) Triantafyllidou, S.; Nguyen, C.; Zhang, Y.; Edwards, M. Lead (Pb) quantification in potable water samples: Implications for regulatory compliance and assessment of human exposure. *Environ. Monit. Assess.* **2012**, 1–11.
- (11) Triantafyllidou, S.; Parks, J.; Edwards, M. Lead particles in potable water. *J. AWWA* **2007**, *99* (6), 107–117.
- (12) Del Toral, M. A.; Porter, A.; Schock, M. R. Detection and evaluation of elevated lead release from service lines: A field study. *Environ. Sci. Technol.* **2013**, *47* (16), 9300–9307.
- (13) Haas, C.; Koch, L.; Kelty, K.; Triantafyllidou, S.; Lytle, D. Effectiveness of the Preservation Protocol within EPA Method 200.8 for Soluble and Particulate Lead Recovery in Drinking Water (EPA/600/R-13/222); U.S. Environmental Protection Agency, Ed., 2013.
- (14) United States Environmental Protection Agency. *Is There Lead in my Drinking Water?* (EPA 816-F-05-001). 2005 [cited 2014 Feb 28]; Available from: <http://water.epa.gov/drink/info/lead/leadfactsheet.cfm>.
- (15) Edwards, M.; Dudi, A. Role of chlorine and chloramine in corrosion of lead-bearing plumbing materials. *J. AWWA* **2004**, *96* (10), 69–81.
- (16) Cartier, C.; Nour, S.; Richer, B.; Deshommes, E.; Prévost, M. Impact of water treatment on the contribution of faucets to dissolved and particulate lead release at the tap. *Water Res.* **2012**, *46* (16), 5205–5216.
- (17) Giani, R.; Edwards, M.; Chung, C.; Wujek, J. *Use of Lead Profiles to Determine Source of Action Level Exceedences from Residential Homes in Washington, D.C.*; presented at Water Quality Technology Conference: San Antonio, TX, 2004.
- (18) HDR Engineering Inc., *An Analysis of the Correlation Between Lead Released from Galvanized Iron Piping and the Contents of Lead in Drinking Water*; HDR Engineering Inc.: Bellevue, WA, 2009.
- (19) Sandvig, A.; Maynard, B.; Mast, D.; Trussel, R. R.; Trussel, S.; Cantor, A.; Prescott, A. *Contribution of Service Line and Plumbing Fixtures to Lead and Copper Rule Compliance Issues*; American Water Works Association Research Foundation: Denver, CO, 2008.
- (20) Britton, A.; Richards, W. N. Factors influencing plumbosolvency in Scotland. *J. Inst. Water Eng. Sci.* **1981**, *35*, 349–364.
- (21) Cartier, C.; Bannier, A.; Pirog, M.; Nour, S.; Prévost, M. A rapid method for lead service line detection. *J. Am. Water Works Assoc.* **2012**, *104* (11), E596–E607.
- (22) Cartier, C.; Arnold, R. B., Jr.; Triantafyllidou, S.; Prévost, M.; Edwards, M. Effect of flow rate and lead/copper pipe sequence on lead release from service lines. *Water Res.* **2012**, *46* (13), 4142–4152.
- (23) Schock, M. R. Causes of temporal variability of lead in domestic plumbing systems. *Environ. Monit. Assess.* **1990**, *15* (1), 59–82.
- (24) Deshommes, E.; Laroche, L.; Nour, S.; Cartier, C.; Prévost, M. Source and occurrence of particulate lead in tap water. *Water Res.* **2010**, *44* (12), 3734–3744.
- (25) Cardew, P. T. Development of a convective diffusion model for lead pipe rigs operating in laminar flow. *Water Res.* **2006**, *40* (11), 2190–2200.
- (26) Clark, B.; Masters, S.; Edwards, M. *3-D Lead Profiling Detects Particulate Lead-in-Water Risks as a Function of Flow Rate*; presented at Water Quality Technology Conference: Toronto, ON, 2012.
- (27) Wang, Y.; Mehta, V.; Welter, G. J.; Giammar, D. E. Effect of connection methods on lead release from galvanic corrosion (PDF). *J. Am. Water Works Assoc.* **2013**, *105* (7), E337–E351.
- (28) Lytle, D. A.; Sorg, T. J.; Frietch, C. Accumulation of arsenic in drinking water distribution systems. *Environ. Sci. Technol.* **2004**, *38* (20), 5365–5372.
- (29) Lytle, D. A.; Sorg, T. J.; Muhlen, C.; Lili, W. Particulate arsenic release in a drinking water distribution system. *J. Am. Water Works Assoc.* **2010**, *102* (3), 87–98.
- (30) Schock, M. R.; Hyland, R. N.; Welch, M. M. Occurrence of contaminant accumulation in lead pipe scales from domestic drinking-water distribution systems. *Environ. Sci. Technol.* **2008**, *42* (12), 4285–4291.
- (31) Valentine, R. L.; Stearns, S. W. Radon release from water distribution system deposits. *Environ. Sci. Technol.* **1994**, *28* (3), 534–537.
- (32) Lytle, D. A.; Sorg, T.; Wang, L.; Chen, A. The accumulation of radioactive contaminants in drinking water distribution systems. *Water Res.* **2014**, *50* (0), 396–407.
- (33) Masters, S.; Edwards, M. *Holistic Examination of Lead Corrosion Control in Low Alkalinity High pH Water Distributed Through Unlined Iron Mains*; presented at Water Quality Technology Conference: Long Beach, CA, 2013.
- (34) Wang, Y.; Jing, H.; Mehta, V.; Welter, G. J.; Giammar, D. E. Impact of galvanic corrosion on lead release from aged lead service lines. *Water Res.* **2012**, *46*, 5049–5060.
- (35) Young, D.; Munson, B.; Okiishi, T.; Huebsch, W. *A Brief Introduction to Fluid Mechanics*, 4th ed; John Wiley & Sons: Hoboken, NJ, 2007.
- (36) APHA. *Standard Methods for the Examination of Water and Wastewater*, 20th ed; APHA, AWWA, WEF, 1998.
- (37) Brown, R.; McTigue, N.; Cornwell, D. Strategies for assessing optimized corrosion control treatment of lead and copper. *J. AWWA* **2013**, *105* (5), 62–75.

- (38) Cantor, A.; Denig-Chakroff, D.; Vela, R.; Oleinik, M.; Lynch, D. Use of polyphosphate in corrosion control. *J. AWWA* **2000**, 92 (2), 95–102.
- (39) Camara, E.; Gagnon, G. A. *The Relationship between Iron Particles from Water Mains and Lead Release*; presented at AWWA Annual Conference & Exposition, Dallas, TX, 2012.
- (40) Camara, E.; Montreuil, K.; Knowles, A.; Gagnon, G. Role of the water main in lead service line replacement: A utility case study. *J. AWWA* **2013**, 105 (8), E423–E431.
- (41) De Rosa, S.; Williams, S. M. *Particulate Lead in Water Supplies (TMU 9024). Final Report to the Department of the Environment*; Water Research Center: Swindon, England, 1992.
- (42) Reiber, S.; Poulsom, S.; Perry, S. A. L.; Edwards, M.; Patel, S.; Dodrill, D. M. *A General Framework for Corrosion Control Based on Utility Experience*; AWWA Research Foundation and American Water Works Association: Denver, CO, 1997.
- (43) U.S. Environmental Protection Agency. *Long-Term Lead and Copper Rule Revision Public Meeting, October 14–15, 2008*.
- (44) U.S. Environmental Protection Agency. *Lead and Copper Rule*, [cited 2014 May 23]; Available from: <http://water.epa.gov/lawsregs/rulesregs/sdwa/lcr>.
- (45) Arnold, R.; Edwards, M. Potential reversal and the effects of flow pattern on galvanic corrosion of lead. *Environ. Sci. Technol.* **2012**, 46 (20), 10941–10947.