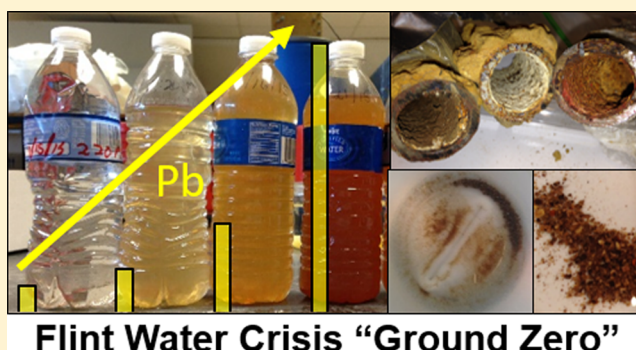# ENVIRONMENTAL Science & Technology

Flint Water Crisis Caused By Interrupted Corrosion Control: Investigating "Ground Zero" Home

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S *Supporting Information*

ABSTRACT: Flint, Michigan switched to the Flint River as a temporary drinking water source without implementing corrosion control in April 2014. Ten months later, water samples collected from a Flint residence revealed progressively rising water lead levels (104, 397, and 707 μg/L) coinciding with increasing water discoloration. An intensive follow-up monitoring event at this home investigated patterns of lead release by flow rate—all water samples contained lead above 15 μg/L and several exceeded hazardous waste levels (>5000 μg/L). Forensic evaluation of exhumed service line pipes compared to water contamination "fingerprint" analysis of trace elements, revealed that the immediate cause of the high water lead levels was the destabilization of lead-bearing corrosion rust layers that accumulated over decades on a galvanized iron pipe downstream of a lead pipe. After analysis of blood lead data revealed spiking lead in blood of Flint children in September 2015, a state of emergency was declared and public health interventions (distribution of filters and bottled water) likely averted an even worse exposure event due to rising water lead levels.

1. INTRODUCTION

As sources of lead exposure are steadily reduced and attention turns to eliminating all cases of children with elevated blood lead levels (BLLs), exposure to lead in potable water is of increasing concern.[1−6] The existing regulation controlling waterborne lead exposure is the U.S. Environmental Protection Agency (EPA) Lead and Copper Rule (LCR), which allows up to 10% of first draw samples (i.e., samples collected after 6+ hours of stagnation) from high-risk households to exceed the lead action level of 15 μg/L.[7] Water utilities typically implement and maintain corrosion control strategies (e.g., pH/alkalinity adjustments or corrosion inhibitors) to achieve LCR compliance. One popular strategy is dosing orthophosphate (PO_4^{3-}) inhibitors to promote formation of insoluble corrosion scales on pipe walls that reduce lead release (Figure 1a).[8−12]

Several studies have shown that the destabilization of corrosion scales can cause lead and iron problems if deposits begin to dissolve or detach into the water (Figure 1b).[1,4,13−15] For instance, in November 2000, the District of Columbia Water and Sewer Authority changed secondary disinfection chemicals from free chlorine to chloramine in order to comply with new disinfection byproduct regulations.[13] This change in disinfectants destabilized insoluble lead scale layers on pipe surfaces and increased the incidence of dissolved and particulate lead.[13,16,17] Likewise, Providence Water reduced the pH of their finished water in November 2005 from 10.3 to 9.7 in an attempt to minimize lead solubility, but this chemistry modification resulted in the breakdown of lead-bearing iron
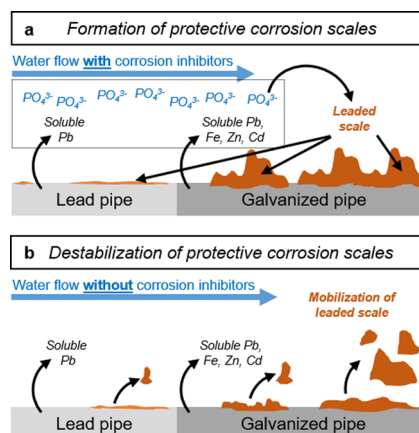

Figure 1. Formation and destabilization of protective scales: (a) corrosion inhibitors (e.g., PO_4^{3-}) control the release of metals from plumbing through the formation of protective scales and (b) without corrosion inhibitors previously formed protective scales can become unstable and deteriorate resulting in high lead and iron in water. Pb = lead; Fe = iron; Zn = zinc; and Cd = cadmium.[14,20]

rust layers and increased total lead and iron in the drinking water, even though it likely decreased soluble lead as

planned.[14,18] As a result, the municipality failed to meet the LCR and experienced high levels of red water complaints.[14,19] Thus, changes in water chemistry that once seemed innocuous, can greatly disrupt and impact lead and iron scale, and increase the likelihood of iron and lead particulates in water.

1.1. Source Water Change in Flint, Michigan. On April 25, 2014, Flint, Michigan discontinued purchase of treated water from the Detroit Water and Sewer Department (DWSD) in preparation for the switch to Karegnondi Water Authority in 2016.[21,22] In the interim two-year period before the pipeline was completed, water from an existing treatment plant treating Flint River water was used. Although research and engineering studies were conducted,[22] the water utility did not implement corrosion control (e.g., continue orthophosphate dosing) to meet the provisions of the LCR (Table 1).[7] In addition, to

Table 1. CSMR and Corrosion Inhibitors Levels in Flint, Michigan before and after April 25, 2014

parameter	before[32] 2014	after[33] Dec. 2014
phosphate (mg/L as P)	1.07 (0.67–1.69)	absent[21]
chloride (mg/L)	11.4 (4.2–39.8)	80 (74–87)
sulfate (mg/L)	25.2 (11.9–33.9)	39.2 (24.3–45.0)[b]
CSMR[c]	0.45	2.04

[a]Average (minimum-maximum) concentrations reported. [b]Sulfate measured in January 2015 reported in this study (Table SI-2). [c]CSMR calculated using average chloride and sulfate values. CSMR > 0.5 indicates high corrosion potential.[23,24]

control total trihalomethanes, the utility used ferric chloride (FeCl$_3$) as a coagulant,[21] further increasing the chloride-to-sulfate mass ratio (CSMR) which is a measure of water corrosivity for galvanic connections (e.g., partial lead service lines and lead solder.)[23–25] Since the LCR monitoring pool in Flint did not have 50%$^+$ homes with lead service lines (LSLs) as required and other sampling methods known to reduce the detection of lead in water were used, officials claimed that the city was in compliance with the LCR throughout 2014 and 2015.[21,26–28] After a proper sampling pool and protocol without preflushing was implemented, Flint was acknowledged to have exceeded the action level at least through June 2016.[29–31]

A Flint resident whose child was having health problems participated in the 2015 LCR water testing and subsequent follow-up tests, revealing first draw lead concentrations of 104 (February), 397 (March), and 707 μg/L (April; Figure 2) suggesting steadily worsening conditions.[21,27,34] However, samples from this home were excluded from LCR reporting.[31] Immediately after a city official instructed the resident to not use the water for drinking or cooking, one of two twin children living in the home had an elevated BLL (6.5 μg/dL), and an EPA inspection revealed no other lead sources in the home.[21,35] On the basis of the consumers experience and revelation that there was no corrosion control plan, Virginia Tech conducted an intensive follow-up monitoring effort to assess the nature of metal release in the home of this resident (referred to as "Resident Zero") including:[34] (1) 3-D lead profiling as a function of flow volume and flow rate;[15] (2) forensic evaluation of lead leaching from plumbing and destabilization of leaded scales; and (3) association between high lead and other constituents of pipe scale exhumed from the property.

2. METHODS

2.1. Water Samples Collected in January 2015. Resident Zero collected six discolored water samples under normal water use conditions between January 15−30, 2015, to demonstrate to city and state officials that the aesthetic characteristics of the drinking water were deteriorating since the switch to treated Flint River water (Figure 2). The resident later sent these samples stored in 500 mL plastic bottles to Virginia Tech. For each of these samples, a 10 mL aliquot was collected for analysis after thoroughly shaking the bottles (Supporting Information, SI, Section SI-1). Aliquots were acidified with 2% nitric acid and 2% hydroxylamine, and heated at 50 °C for a minimum of 24 h. Metals were analyzed using Inductively Coupled Plasma−Mass Spectrometry (ICP-MS) per method 3125 B.[36]

2.2. 3-D Lead Profiling Protocol. The water supply to the home of Resident Zero was shut off by city officials on April 3, 2015 due to the high lead in water (Figure 2).[21,34,35] As a result, water had remained stagnant for more than 3 weeks (not uncommon in Flint due to high rates of vacancy and water shut-offs) before flow was restored for the sampling effort on April 28, 2015. The night before sampling, the tap was
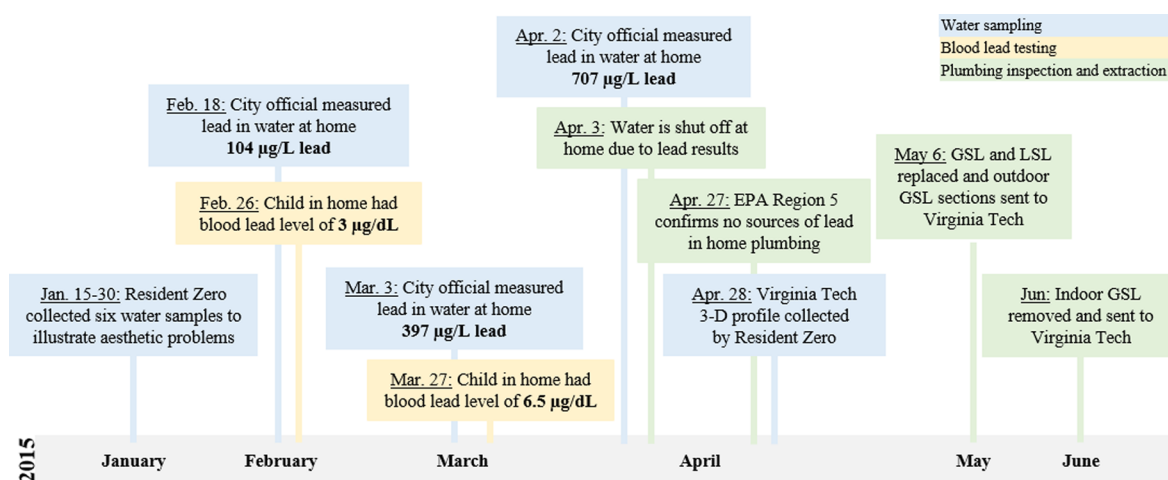

Figure 2. Chronology of water sampling (blue), child BLL testing (yellow), and plumbing system characterization (green) at the home of Flint Resident Zero between January and June 2015. GSL = galvanized iron service line; LSL = lead service line.[21,34,35]

preflushed and precleaned for 25 min to bring fresh water into the system and to ensure that all soluble lead was displaced from the line. The 3-D lead profiling used in this study (Figure SI-1) was a modified version of the protocol developed in Clark et al. (2014),[15] which collected 1 L sequential samples and 250 mL flushed samples from the kitchen cold-water tap at low (1.3 L/min), moderate (4.2 L/min), and high flow rates (7.5 L/min). Specifically, after a 6 h stagnation period, seven 1 L sequential samples were collected at low flow (1.3 L/min) followed by three 250 mL flushed samples collected at 2 min intervals. Immediately afterward and with no stagnation, the flow rate was adjusted to moderate flow (4.2 L/min), and the sample collection was repeated. The water was then turned off and the aerator removed for a final sequence of sampling. At high flow (7.5 L/min), six 1 L sequential samples were collected, and six 250 mL flushed samples were collected at 1 min intervals.

Upon express mail delivery to Virginia Tech, samples were visually inspected to assess water clarity, cloudiness, and color. To quantify the relative fraction of particulate lead in water after shipment to the lab (lead may have adhered to the container walls and/or particulate lead may have dissolved), a 10 mL aliquot was collected from the fifth bottle of each flow rate (bottles 5, 15, and 25) and filtered (0.45 μm). All samples were acidified with 2% nitric acid and 2% hydroxylamine in bottle, and heated at 50 °C for a minimum of 24 h before analysis on the ICP-MS.[36]

2.3. Characterization of Service Line and Leaded Scale. The EPA Region 5 Ground Water and Drinking Water Branch determined that the lead contamination was not associated with the interior household plumbing as it was primarily plastic plumbing with several "lead-free" fittings and fixtures.[21] On May 6, 2015, two 0.6–0.9 m (2–3 ft.) outdoor sections of the 58.5 m (192 ft.) galvanized iron service line (GSL; iron pipe with a protective "galvanized" surface coating composed of zinc, lead, and cadmium) were exhumed by a representative of EPA Region 5 and sent to Virginia Tech for analysis (Figures 2 and 3, #1–2). In June 2015, Resident Zero
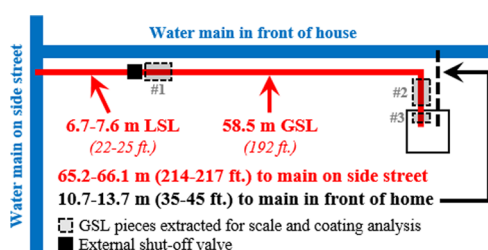

Figure 3. Schematic of galvanized iron service line (GSL) and lead service line (LSL) at home of Resident Zero. Data derived was provided by EPA Region 5.[21,37]

extracted a 1.5 cm (6 in.) section of the indoor GSL for analysis of the original galvanized coating content as revealed by the uncorroded outer surface of the pipe (Figure 3, #3). Scrapings of corrosion rust scales from the outdoor and indoor GSLs and galvanized coating of the indoor GSL were dissolved in 100 mL of water and digested as previously described for analysis via ICP-MS. The pipe surface of the indoor GSL was also analyzed via X-ray fluorescence spectrometry (XRF; Innov-X Alpha Series). In addition, depth profiles of corrosion scales were performed using an environmental scanning electron microscope with an attached X-ray energy dispersive system (ESEM/

EDS). Scale formed along the inner surfaces of GSL #1 and #3 were analyzed at various depths (6 depths of GSL #1 and 5 for GSL #3), with each depth examined in triplicate. The EPA later confirmed the presence of a 6.7–7.6 m (22–25 ft.) LSL between the water main and external shut-off valve (i.e., before the GSL; Figure 3);[37] however, this pipe was not recovered during the service line replacement.

2.4. Statistical Analysis. Nonparametric statistics were used in this study due to the non-normal distribution of the lead data (Shapiro-Wilk; $p < 0.05$). Spearman's rank correlation (ρ) was used to evaluate the associations between lead and other water quality parameters (e.g., copper and zinc). The Kruskal–Wallis test was used to compare lead and iron concentrations based on flow rate during the profiling effort.

3. RESULTS

3.1. 3-D Lead and Iron Profiling. On April 28, 2015, 32 water samples were collected from the kitchen cold-water tap as a function of time and flow rate at a home with known lead in water problems. All 32 samples contained lead above the EPA action level of 15 μg/L, with a minimum concentration of 217 μg/L (Figure 4; Table SI-1). The average and median lead concentrations were 2393 and 1747 μg/L, respectively. A sample collected after more than 20 min of flushing (>50 L) contained 13 200 μg/L lead, which was almost three times the EPA's hazardous waste threshold of 5000 μg/L.[38] In total, four of the 32 samples contained lead above 5000 μg/L, occurring sporadically throughout the profile. Furthermore, lead levels increased in the last five samples of this profiling effort, indicating that lead concentrations were not subsiding. Thus, flushing would not protect the occupants from high lead in water, even after following instructions of city officials to flush the line for 25 min before water use.[39]

3.1.1. Lead Release As a Function of Flow Rate. Although low "pencil-thin" flow rates (~1 L/min) are known to underestimate lead in water concentrations during typical flow because the mobilization of particulate lead is greatly reduced,[15,40−42] there was no significant difference in median lead concentrations based on flow rate (Kruskal−Wallis Test, $p = 0.30$; Figure 4) from these samples collected at progressively higher flow rate. Specifically, average and median lead concentrations were 2384 and 2014 μg/L at low flow (1.3 L/min), 1687 and 1119 μg/L at moderate flow (4.3 L/min), and 2988 and 1747 μg/L at high flow (7.5 L/min). This consistent presence of elevated lead in water at all flow rates was attributed to the mobilization of particulate lead (Figure 4, photos A−C). Three filtered aliquots (0.45 μm) collected from sample bottles 5, 15, and 25 indicated that ≥99% of the lead was in the particulate form after receipt at the laboratory.

With continued water use at low flow, there appeared to be a downward trend in lead concentrations, indicating that long flushing times did progressively "clean" the pipes by flushing the lead built up during the overnight stagnation. When the tap was adjusted to moderate flow, two large spikes in lead (3655 and 6048 μg/L) were observed during the sequential sampling, which was attributed to the sporadic mobilization of lead bearing sediment from the pipe at an increased flow rate, but lead levels again decreased somewhat. When the aerator was removed and water was set to high flow, lead levels became highly sporadic. The first sample contained 5702 μg/L, which was 20 times higher than the previous sample of 292 μg/L. Lead concentrations decreased in the next four sequential
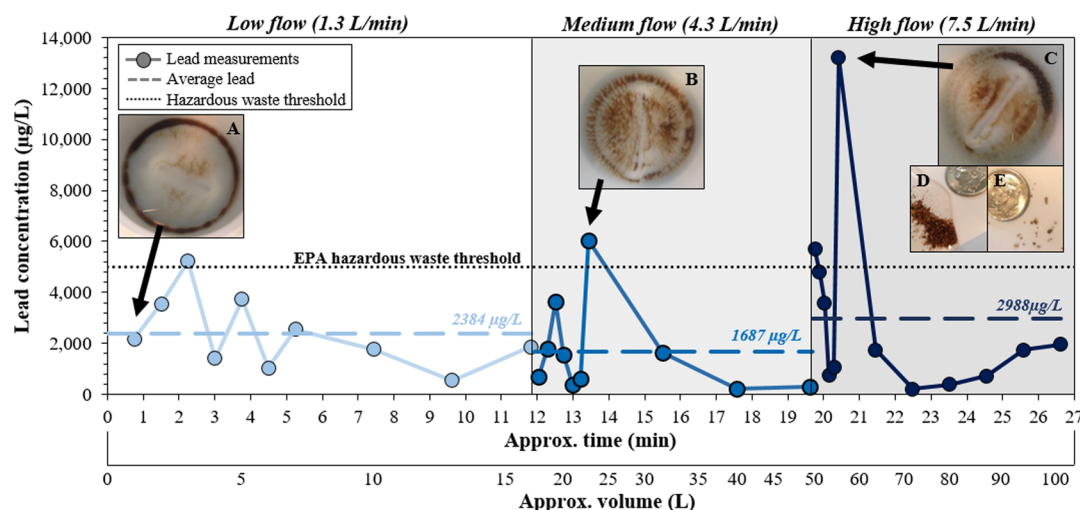
Figure 4. Lead in water concentrations collected at low, moderate, and high flow. The dashed blue lines indicate the average lead measured at each flow rate, and the dotted black line is the EPA hazardous waste threshold of 5000 μg/L. Photos A−C were taken after the water samples settled in their bottles overnight. Photos D and E illustrate the size of the particles shown in photo C.

Table 2. Average Ratios of Metals ($\pm$ Standard Deviation) in GSL Coating and Water Samples

| | | indoor GSL surface coating | | | | water samples | | | |
| | | XRF analysis ($n = 6$) | | ICP-MS analysis ($n = 6$) | | ICP-MS January sample ($n = 6$) | | ICM-MS April profile ($n = 32$) | |
ratio	units								
Cd:Sn[a]	μg/L:μg/L			1.0	$< \pm 0.1$	1.1	$< \pm 0.1$	0.9	± 0.1
Cd:Zn[a]	μg/L:mg/L			2.6	± 0.2	3.0	± 0.1	2.6	± 0.3
Fe:Pb	mg/L:mg/L	1.4	± 0.3	NS[b]	NS[b]	39	± 11	1.7	± 0.7
Fe:PO$_4$	mg/L:mg/L			NS[b]	NS[b]	4.1	± 0.6	1.3	± 0.3
Pb:Cd[a]	μg/L:μg/L			7.0	± 0.9	336	± 37	744	± 283
Pb:Zn	μg/L:mg/L	17.1	± 5.4	18.2	± 1.4	989	± 134	1824	± 747
PO$_4$:Pb	mg/L:mg/L			1.0	$< \pm 0.1$	9.4	± 1.7	1.2	± 0.3

[a]Sample sizes varied due to nondetectable Cd and Sn concentrations: January ($n = 5$) and April ($n = 30$). [b]NS: no sample. Iron concentrations were not consistent with ASTM standards, indicating scrapings of the underlying iron pipe influenced the analysis.

samples, but the sixth sample contained 13 200 μg/L. Lead concentrations in the last five water samples (i.e., flushing interval) continued to increase, indicating that water from the kitchen tap was still not safe for consumption despite flushing the line for more than 26 min (>100 L).

3.1.2. Iron Release As a Function of Flow Rate. All 32 water samples collected during the profile contained iron concentrations above the EPA secondary maximum contamination level (SMCL) of 0.3 mg/L (Figure SI-2; Table SI-1). Concentrations ranged from 0.7 to 6.5 mg/L, with average and median concentrations of 2.7 and 2.8 mg/L. There was no significant difference in iron concentrations based on flow rate during this effort (Kruskal−Wallis Test, $p = 0.50$). Specifically, average and median iron concentrations were 2.8 and 3.1 mg/L at low flow, 2.2 and 1.9 mg/L at moderate flow, and 3.1 and 3.1 mg/L at high flow. As iron in water concentrations were strongly correlated (Spearman's, $p < 0.05$) with lead ($\rho = 0.91$) and phosphate ($\rho = 0.94$), it appeared that the high lead and iron levels were a result of the destabilization of lead-bearing corrosion rust layers.

3.2. Sources of Lead in Water at Home of Resident Zero. *3.2.1. Destabilization of Leaded Corrosion Scale.* Drinking water supplied by the DWSD contained approximately 1.07 mg/L of orthophosphate as P,[32] which would be expected to form insoluble phosphate scales with soluble metals in water on the inside pipe surfaces over decades of water use

(Figure 1a). When Flint switched to the Flint River as a water source, the practice of adding orthophosphate inhibitors was discontinued,[21,27] and phosphate corrosion scales began deteriorating (Figure 1b). This was evident during the profiling effort as all 32 samples contained phosphate (0.4−9.9 mg/L as P; Figure SI-1), even though the distributed water had undetectable phosphate for 11 months by this time.[21] The average and median phosphate concentrations were 2.4 and 2.1 mg/L as P, and concentrations were strongly correlated with lead ($\rho = 0.98$), demonstrating that the scales previously formed in DWSD water were deteriorating and falling into the water supply at high levels. In addition, two short-term laboratory experiments conducted at Virginia Tech demonstrated that addition of orthophosphate inhibitors to the Flint River water would have markedly reduced iron and lead release rates (Figure SI-3).[43−45] Although the increase in water corrosivity due to higher CSMR promoted corrosion at galvanic connections (e.g., partial LSL and lead−copper soldered joints), the continuation of orthophosphate corrosion control would have reduced the breakdown of the lead-bearing and other corrosion scales. Scrapings of the interior scale from the outdoor and indoor GSLs (Figure 3, #1−3) were dissolved in water for analysis via ICP-MS. The composition varied considerably based on the section of scale analyzed, as the scales contained 2−47% iron (Fe), 3−11% zinc (Zn), 0.11−

0.37% Pb, 0.005−0.02% tin (Sn), 0.005−0.02% cadmium (Cd), and 0.6−4% phosphate as P ($PO_4^{3−}$).

3.2.2. Lead Content in Corrosion Scales. One 1.5 cm (6 in.) section of indoor GSL was extracted and sent to Virginia Tech (Figure 3, #3). The outer surface of this GSL, which had been relatively uncorroded by the decades of use, contained 94.2 ± 1.2% Zn, 2.2 ± 0.4% Fe, and 1.6 ± 0.5% Pb as identified by XRF, which was consistent with galvanized iron pipe composition.[20] To detect trace metals, scrapings of the coating were digested and analyzed via ICP-MS, and were found to contain 92.0 ± 0.6% Zn, 1.7 ± 0.1% Pb, 0.2 ± 0.02% Cd, and 0.2 ± 0.02% Sn. When evaluating metals in the tap water during the profiling effort, lead was strongly correlated with Cd (ρ = 0.98), Zn (ρ = 0.95), and Sn (ρ = 0.95). This indicated that zinc, lead, cadmium, and tin likely accumulated in the GSL scale and released when the pipe of this home was exposed to the corrosive Flint River water without corrosion inhibitors.

To attempt to evaluate the lead contribution from the GSL versus the pure LSL, cadmium was used as a GSL fingerprint, since it is in the GSL coating and is rarely present in other plumbing.[20] The average ratio of Cd:Zn in the original relatively uncorroded coating of the indoor GSL pipe wall was 2.6 (±0.2; μg/L:mg/L; Table 2), consistent with the average ratio observed in water samples during all of the profiles (2.6 ± 0.3). Thus, the presence of zinc and cadmium released to water was almost certainly due to the disrupted GSL scale, and the zinc could also be used as a fingerprint for GSL lead release for this home with all PVC interior pipe (minimal influence of brass corrosion). The average ratio of Pb:Zn in the original GSL coating as determined by ICP-MS was 18.2 (±1.4; μg/L:mg/L). However, the average ratio observed during the profiling effort was 1824 (±747; μg/L:mg/L), two magnitudes higher than that observed in the original coating. We hypothesize that this indicates that lead had previously leached from the upstream LSL and sorbed to the iron rust layers forming on the GSL when the home was supplied by DWSD, and that this lead enriched scale was being released due to the change in source water (Figure 1). This is problematic given that the lead enriched iron rust layers present on the GSL can serve as a large lead in water source, even after the original LSLs are removed.[14,20,46]

The breakdown of these lead-rich iron rust layers was also evident during the January sampling effort. The Cd:Zn ratio (3.0 ± 0.1) in water samples was consistent with the original GSL coating composition, and the Pb:Zn ratio was higher (989 ± 134) than the coating ratio. However, there was a large difference in the Fe:Pb ratio between the January sampling (39 ± 11) and April profile (1.7 ± 0.7). We speculate that different types of scale containing iron and phosphate were disrupted during periods of water use (Figure 3, #1−3). Specifically, the average iron content in the interior scale was 5.1 ± 4.6% for GSL #1, 17.9 ± 2.3% for GSL #2, and 42.3 ± 4.8% for GSL #3, while the average lead content in the GSL scales remained relatively consistent (0.2−0.3%).

Scale depth profiles were performed to verify that lead-rich iron rust layers formed on the GSL, and scale compositions were nonuniform along the 58.5 m (192 ft.) GSL. A simple visual inspection and ESEM/EDS analysis confirmed that scales formed on GSL #1 (adjacent to the LSL) and GSL #3 (inside home) varied considerably (Figures 3 and SI-4). White scales approximately 3.6 mm thick formed on GSL #1 while brown scale approximately 1.8 mm thick formed on GSL #3. The composition of GSL #1 scales was relatively homogeneous with

depth, as there was an average fluctuation of 39−48% in oxygen (O), 41−54% in Fe, and 0.07−0.12% in phosphorus (P). The highest lead percentage was in the layers closest to the pipe surface (average 0.43−0.53%) and decreased in layers closer to the water (average 0.06−0.10%). In contrast, GSL #3 scale was heterogeneous, as the percent of O and Fe varied with depth. Layers closer to the pipe surface contained higher average Fe levels (25−41%) compared to the outermost layers with higher average O (55−57%) and P (2.4%). The percent of lead within the scale was sporadic ranging from 0.0 to 0.8%. Thus, with nonuniform composition of the GSL scales in different pipe segments and the semirandom nature of scale detachment, variations in ratios of metals between sampling efforts as the interruption to corrosion control continued, were not unexpected.

3.3. Water Quality in January 2015 Samples. *3.3.1. High Lead and Red Water Complaints.* After the switch to treated Flint River water, there was an increase in "red/rusty water" complaints, which was most likely further exacerbated by additional disruptions to the iron water mains through hydrant flushing.[27,47] While red and rusty water complaints were caused by an increase in iron,[14,46] they were also associated with elevated lead in water levels in this home as there was a strong correlation between lead and iron levels in the January samples (ρ = 1, p < 0.05; Table SI-2). The first water sample collected on January 15 had the least discoloration and had a lead and iron content of 360 μg/L and 9.0 mg/L, respectively (Figure 5). In contrast, the first
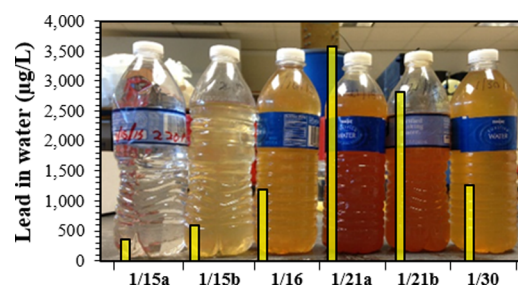

Figure 5. Association between water discoloration and lead concentrations for water samples collected in January 2015. A photo of the original sample bottles is overlaid with a bar chart of lead in water concentrations.

water sample collected on January 21 was highly discolored and had a lead and iron content of 3585 μg/L and 168 mg/L. Water discoloration was predictive of high lead in water in this home with a lead service line, with the average iron-to-lead ratio of 1 mg/L iron to 28 μg/L lead. Thus, statements by authorities that water with red/rusty characteristics was safe for consumption was inaccurate in this instance.[27] However, it is important to note that while water discoloration was strongly associated with high lead in water in the January samples, the lack of discoloration was not an indicator of safe water lead levels, as illustrated by the lack of discoloration observed in the April profiling samples (i.e., samples with no aesthetic issues but Pb ≥ 217 μg/L).

3.3.2. Deficiencies in the LCR Sampling. Lead levels in the January 2015 samples highlighted instructions in the LCR protocol that artificially reduced lead in water during sampling. Specifically, instructions to preflush the night before sample collection and low-flow rates imposed by small bottle openings, are known to minimize particulate lead and produced levels of

104, 397, and 707 μg/L in February, March, and April.[21] These LCR levels were much lower than the average lead level (1635 μg/L) collected by this resident in January during normal water use, which is disconcerting given that the goal of LCR sampling is to detect "worst case" lead. Furthermore, of the 100 LCR samples originally required, officials only collected 71, and all of the high LCR lead results from this home were excluded "conveniently and without adequate investigation".[31] The net result was an "official" 90th percentile lead result below the action level.

4. IMPLICATIONS

While purchasing water from DWSD, Flint's 2013 Consumer Confidence Report documented a 90th percentile lead value of <2 μg/L,[48] indicating reasonable corrosion control. In April 2014, with the disruption of corrosion control, Flint residents started reporting aesthetic water quality issues and the city issued boil water advisories.[31] But the lead in water problem would have likely gone undetected indefinitely without the scientific and investigative work of Resident Zero. The high lead and iron levels observed were consistent with destabilization of lead-rich corrosion rust layers,[1,4,13−15] the nature of which varied considerably with time, and with distance from the LSL. Although this case study might very well be among the worst-cases for lead in Flint, as the service line to this property was unusually long at 65.2−66.1 m (214−217 ft.; Figure 3), the relative increase in lead hints at the likely magnitude of problems that could have occurred in other Flint homes.

Even incidental exposure to drinking water with high lead observed at Ground Zero poses serious health concerns. For chronic, long-term exposure, the EPA estimates a child's BLL will increase by 1 μg Pb/deciliter blood for every 5 μg/L lead in water increase.[49] A single 250 mL drink of water containing even the lowest concentrations observed (217 μg/L) at this home, is estimated to raise the average child's BLL to 0.4 μg/dL.[5,50] Moreover, 250 mLs of water containing 2500 μg/L Pb is predicted capable of raising a child's blood lead from 0 to 5 μg/dL (the Centers for Disease Control and Prevention's BLL of concern),[5] and roughly one-third of samples collected (10 of 32) during the profiling effort exceeded this threshold. At the highest observed lead level of 13 200 μg/L at this home, a child's BLL can be elevated from 0 to >5 μg/dL via consumption of a single swallow of water (50 mLs).[21]

Assuming a uniform lead content of 0.15% by weight, the 58.5 m (192 ft.) GSL would contain an estimated 10.4 g of lead within its corrosion scales. As a person in the U.S. typically uses between 80 and 100 gallons of water per day,[51] the continued breakdown and mobilization of the leaded GSL scales could have contaminated every drop of water used by this family of five to 15 μg/L for 12−15 months if this lead were all released uniformly. The kinetics of lead release are more complex, but this calculation illustrates that there can be significant long-term problems associated with lead sorbed to galvanized iron, as noted by others.[14,20,46] This is particularly problematic for the estimated 50% of homes with full or partial lead piping.[52,53]

On the basis of the absence of corrosion control treatment and related observations of Resident Zero,[21,27] a system-wide sampling effort was initiated by our team in August 2015 in direct collaboration with Flint residents which revealed a city wide problem.[54] Preliminary results indicated that the 90th percentile was 25 μg/L, based on 252 randomly sampled homes throughout the city. After an analysis of blood lead data revealed spiking lead in blood of Flint children city wide in late September 2015,[55] a state of emergency was declared by local, state, and federal health officials which included distribution of filters and bottled water to avert additional health harm.

■ ASSOCIATED CONTENT

⑤ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b04034.

Additional water quality measurements and figures collected during the profile and January sampling efforts and information regarding the 3-D lead profiling protocol(PDF)

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Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

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