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Flint biosolids monitoring data demonstrate a sustained decline in total lead release to potable water from plumbing since the 2014–2015 Flint Water Crisis (FWC), due to enhanced corrosion control treatment (3 mg L⁻¹ orthophosphate as PO₄) and removing of ~80% of lead and galvanized iron service lines through early 2020. The official 90th percentile water lead levels, which have now met the federal Lead and Copper Rule threshold of 15 µg L⁻¹ for the last four years, are in agreement with those predicted by a previously established biosolids regression model. There is also no longer a correlation between the percentage of children under 6 years of age with blood lead \geq 5 µg dL⁻¹ and biosolids lead mass in the 44 months post-FWC (Nov 2015–Jun 2019), nor are there continued correlations between plumbing-related metals in the biosolids, with the exception of Cu:Zn found in brass alloys that remain installed in homes. After Flint achieves 100% replacement of lead and galvanized service line pipes, a biosolids data analysis predicts that the remaining sources of waterborne lead including leaded brass, lead solder and legacy lead in pipe scale, will still release about 16–28% of the pre-FWC lead mass to potable water. The efficacy of enhanced corrosion control and replacement of service lines that contain lead is, therefore, on the order of 72–84% effective at reducing citywide lead exposure, yet some significant water lead sources will still remain even after pipe replacement is complete.

Introduction

When the City of Flint, Michigan switched water sources from Lake Huron to the Flint River in April 2014 and stopped adding orthophosphate corrosion control,^{1–3} higher levels of lead, iron, chlorine decay, deaths from Legionnaire's disease and blood lead in children resulted.^{3–9} After the scope of the water lead problem became apparent, public outcry caused

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Efficacy of corrosion control and pipe replacement in reducing citywide lead exposure during the Flint, MI water system recovery†

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Water impact

In the aftermath of the Flint, Michigan Water Crisis (FWC) and a new proposed US Environmental Protection Agency (EPA) Lead and Copper Rule (LCR), the benefits of replacing lead service lines and implementing enhanced corrosion control to reduce water lead exposure are of high interest. Here we provide a novel analysis of routine Flint biosolids monitoring data, demonstrating a substantial 72–84% citywide reduction in the mass of lead released to potable water *vis-à-vis* pre-FWC 2013 year. Biosolids monitoring has certain advantages in tracking overall source reductions compared to traditional first draw sampling in a subset of homes with lead service lines.

the city to switch back to Lake Huron water in October 2015 and the orthophosphate dose was tripled in December 2015.^{2,5}

A federal emergency was declared in January 2016, and water lead levels (WLLs) have consistently measured below federal Lead and Copper Rule (LCR) standards since early 2017.^{4–6,10} WLLs have even met the more rigorous provisionally adopted Michigan LCR standards since early 2019.^{11,12} The officially reported WLL data is consistent with second, third and even fourth party independent analyses of lead in Flint water.^{4,13,14} The city has also replaced thousands of leaded brass faucets, and 9516 service lines representing about ~80% of the city's total lead and galvanized iron pipes between March 2016 and March 2020.^{15–18} Officials were even having trouble finding enough homes with verified lead service lines to successfully conduct the late 2019 LCR sampling event.¹⁹

Many Flint residents still do not trust the safety of tap water for a variety of reasons, including:

1) Actual cheating on official water lead testing before the crisis was exposed by the authors of this paper late 2015, and resulting betrayal of the public trust due to proven inaction, apathy and/or cover-ups by local, state and federal government agencies,^{5,20–22}

2) In post federal emergency Flint (2016–19), some residents engaged in improper sampling, and in one case



lead fishing sinkers were even discovered in a consumer's plumbing, producing water samples with very high WLLs ($>12\,000\ \mu\text{g L}^{-1}$) and suggestions of an ongoing health threat,²³

3) Social media postings and investigative reports, by a "political reporter" from December 2016 to present, allege an ongoing conspiracy by government agencies and independent lead sampling programs to cover-up water lead problems,^{23–26}

4) Widespread misinformation on the effectiveness of state-distributed lead filters,²⁷ and speculation by academics that the filters were causing Shigellosis or consumer deaths,^{23,28–32}

5) Warnings that vibrations and other disturbances arising during pipe replacements, might also be causing massive release of lead from the Flint pipe network,^{33,34} and unfounded assertions by some media, celebrities and politicians who continue to claim that Flint remains mired in a water lead crisis.^{32,35–37}

We recently utilized data on the monthly lead mass captured in sewage sludge (or, biosolids) at the Flint wastewater treatment plant from 2010–17, to establish that biosolids lead reliably tracked lead release from plumbing to potable water before, during and in the immediate aftermath of the Flint Water Crisis.³⁸ This biosolids data has important advantages compared to official WLL monitoring data collected under the LCR, including: 1) biosolids samples represent a composite of all lead released to Flint's potable water over a several week time period, 2) the sampling methodology and location have remained the same for over a decade, and 3) this data has been collected by entities who are independent of those engaged in measuring water lead in homes.

In contrast, the official 90th percentile WLL only measures lead in the first liter from the tap (*i.e.*, "first draw"), has used first draw sampling protocols that have changed substantially in the last few years, is calculated from sampling pool of only 60–200 "high risk" homes with lead pipe that has been changing as lead service lines (LSLs) are replaced.⁴ The official 90th percentile data is therefore designed to infrequently (once every three years to twice a year) identify a characteristic level of water lead in "worst case" homes, and does not reflect average or total lead release to water across the entire city. Thus, analysis and monitoring of the lead mass in Flint biosolids is complementary, and in some ways superior to traditional in home monitoring to track progress as the Flint system continues to heal from enhanced corrosion control and LSLs are replaced.

Herein, we apply our novel approach³⁸ to the most recent data on biosolids monitoring and elevated blood lead in children (January 2018–June 2019), which reflects a time period of unprecedented replacement of lead bearing (*i.e.*, lead and galvanized iron) service line pipe replacements. The tested hypotheses included the following: a) the State of Michigan, the US Environmental Protection Agency, and others, are providing a false sense of progress in terms of

improving Flint WLLs and decreasing childhood lead exposure, b) the combination of pipe and faucet replacements, and corrosion control are reducing overall release of lead to water, and c) replacing lead pipes will greatly reduce (but not eliminate) lead release to drinking water due to remaining sources of lead from brass and solder in consumers' homes.

Materials and methods

Metals in biosolids

Monthly metal concentrations in biosolids (lead, cadmium, copper, nickel and zinc; mg kg^{-1} on a dried weight basis) measured per Standard Method SW 6020A³⁹ and total monthly biosolids production (kg) for January 2018–June 2019 were obtained from City of Flint's Water Pollution Control Plant *via* Freedom of Information Act (FOIA) requests. The monthly mass of metal in biosolids was calculated by multiplying the metal biosolids concentration by the total biosolids production. Metal mass in biosolids data for prior years (2010–17) for comparison were sourced from another study.³⁸

Modeled relationship between biosolids lead and water lead levels

A regression model between lead in biosolids and in water for the City of Flint (eqn (1)) was used to estimate water lead levels (WLLs) from obtained biosolids lead data.³⁸

$$\text{Biosolids-Pb (kg per month)} = 0.37 \times \text{WLL}_{90} (\mu\text{g L}^{-1}) + 1.41 \quad (1)$$

where, biosolids-Pb = biosolids lead mass in kg; WLL_{90} = composite 90th percentile water lead level in $\mu\text{g L}^{-1}$, estimated as a 50:50 weighted average of "first draw" and "second draw" WLLs.

The model assumes a one-month offset between biosolids-Pb and WLL_{90} (*i.e.*, biosolids-Pb measured in February 2018 is paired with WLL_{90} for January 2018) to account for a few weeks of biosolids and activated sludge detention times. The model also relies on the assumption that 90th percentile first draw lead ranges between 1.6 to 4.0 times the 90th percentile second draw lead in Flint, based on WLL data from five Virginia Tech citywide water sampling rounds between 2015–17.³⁸ This relationship was used to estimate a 90th percentile first draw range from the biosolids-predicted WLL_{90} values and compared against that calculated from official LCR testing for six month periods for 2016–19.¹¹ The only exception was 2019 where biosolids data corresponding to first five month period of WLLs (Jan–May 2019) was available.

Elevated blood lead levels

The de-identified aggregated monthly data on percentage of children under six with elevated blood lead $5\ \mu\text{g dL}^{-1}$ (*i.e.*, % EBL5) during January 2018–June 2019, and from prior years



(2010–17) for comparison, were obtained from Hurley Medical Center's Dr. Mona Hanna-Attisha⁴⁰ and a previous study³⁸ respectively.

Statistical analyses

All statistical analyses were conducted in Microsoft® Excel® (version 2016) and IBM® SPSS (version 25). A p value of <0.05 with an alpha value (α) of 0.05 was selected to determine statistical significance. The coefficient of determination (R^2) was calculated to examine the associations between monthly biosolids metal masses for Pb, Cu and Zn.

Results and discussion

After examining trends on plumbing-related metals captured in biosolids from January 2018–June 2019, we use the established regression model between biosolids and water lead in Flint to estimate WLLs 2018–19 for comparison to reported (official and independent) 90th percentile WLLs. We then examine whether lead levels spiked or declined across the city, during implementation of Flint's unprecedented lead and galvanized service line pipe replacement program, and attempt to quantify the benefits and limitations of service line replacement in reducing WLLs.

Trends in plumbing metals captured in biosolids

Over 95% of treated wastewater in Flint is domestic in origin and most of the lead comes from corrosion of lead pipes and lead-bearing plumbing.^{41–43} The lead mass in biosolids has continued to drop in the last few years (Fig. 1A) and reached another historic low in 2019. From January–June 2019, average lead was 5 kg per month (range = 1.2–8.5 kg per month), *versus* 10.2 kg per month (range = 5.2–24.5 kg per month) for the 18 months of FWC (April 2014–October 2015) and 9.3 kg per month (range = 5.4–15.7 kg per month) for a comparable period pre-FWC (April 2012–October 2013).

Biosolids masses for all five plumbing related metals (Cd, Cu, Ni, Pb, and Zn) during 2018–2019 also dropped to between 35–76% of that measured during the FWC, due to enhanced corrosion control, service line and lead faucet replacement (Fig. S1 in ESI†). While all three plumbing-related metals (Pb, Cu and Zn) in biosolids correlated before (2011–14), during (2014–15), and after the FWC (2015–17),³⁸ the only correlation that remained significant in this latest period of enhanced corrosion control and plumbing material replacement during 2018–19 was Cu vs. Zn ($R^2 = 0.30$; $p < 0.05$). Copper and zinc are both present in the new brass alloy valves, faucets and fixtures that are still being installed throughout the Flint water system. Correlations between Pb:

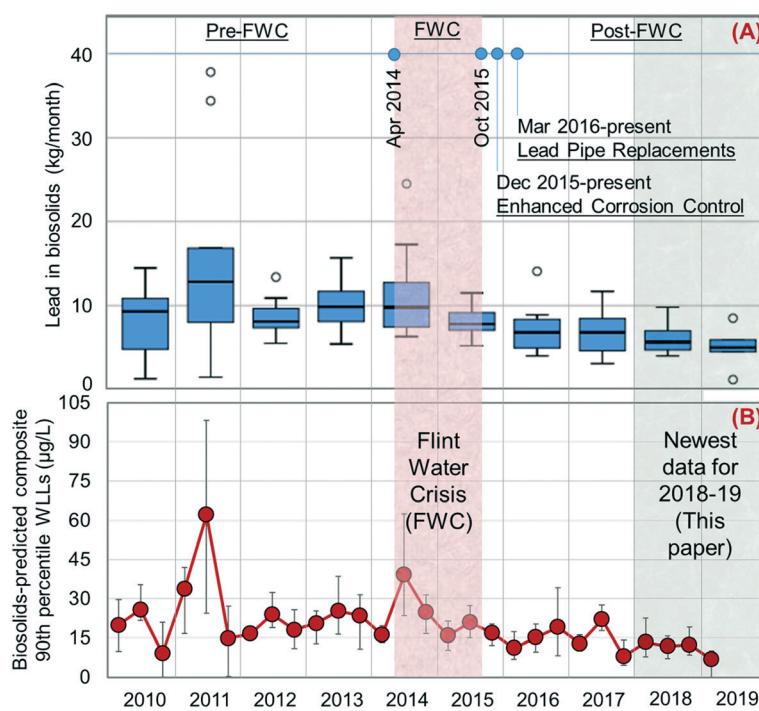


Fig. 1 (A) Box-and-whiskers plot of total monthly biosolids lead mass captured at the Flint wastewater plant, 2010–19. The open circles indicate outlier lead mass values. The plot summarizes data for January–December for 2010–18 and January–June for 2019. Enhanced corrosion control refers to tripling of orthophosphate corrosion inhibitor dose starting Dec 9, 2015. Lead pipe replacements began in March 2016 under the Flint FAST start program. Faucet replacements began in January 2017 with funding from State of Michigan. (B) Composite 90th percentile water lead levels (WLL_{90}) for Flint, MI predicted by a regression model (eqn (1)). The WLL_{90} values are derived from biosolids lead mass averaged over 4 month intervals (Dec–Mar, Apr–Jul, and Aug–Nov) for December 2009–March 2019. Error bars indicate the entire range of predicted WLL_{90} values (i.e., minimum and maximum). Future models prospectively applying our approach, should also consider including a sensitivity analysis and error estimation, to further increase the statistical accuracy of the estimated WLL_{90} range.



Cu or Pb:Zn were no longer statistically significant ($p > 0.05$) (Table S1†).

All of the above is consistent with expectations based on the ongoing removal of lead pipes and the effectiveness of improved corrosion control.^{18,38,44} Enhanced corrosion control has also essentially “decoupled” metal release from most of the different alloys that comprise the Flint water distribution system, including lead pipe, galvanized iron pipe, copper pipe, lead solder and brass (*i.e.*, copper, zinc, and lead).

Declining lead levels in Flint's potable water

Using the lead in biosolids data, the predicted composite WLL₉₀ dropped below 10 ppb in 2019 for the first time in a decade, and was about 90% lower than that seen in the worst year pre-FWC (2011) and 83% lower than during the FWC (2014) (Fig. 1B). Moreover, the official 90th percentile first draw WLLs for samples collected under the federal LCR, are in or within 1 ppb of the predicted WLL₉₀ range, based on the calibration using data collected from July 2016–June 2019 (Fig. 2). For instance, the official 90th percentile first draw WLL for July–December 2018 of 4 $\mu\text{g L}^{-1}$ and the independent result of 4 $\mu\text{g L}^{-1}$ from third-party testing led by Michigan State University,¹⁴ is at lower range of 4–8 $\mu\text{g L}^{-1}$ predicted by the model. This finding indicates that the conventional biosolids lead sampling is sometimes consistent with much more complicated first draw LCR sampling events in high risk homes with lead pipe.

The consistently decreasing water lead trends in Fig. 1 and 2, also demonstrate that concerns about WLL spikes following lead pipe replacements, did not overwhelm overall benefits from enhanced corrosion control and replacements of faucets, galvanized steel pipes and lead pipes.

Flint is about to enter a post-lead pipe era

The complete removal of LSLs will not eliminate lead release to drinking water. Lead solder, leaded brass, and premise plumbing that was “seeded” or coated with lead from LSLs in the preceding decades remain as significant lead sources. Roughly 95% of Flint homes were built in the pre-1986 time

period when high lead content solder and brass was commonplace.^{44,46,47}

To further highlight the importance of this issue, Flint resident X, who participated in five sampling rounds with the authors of this paper between August 2015–August 2017, had the highest WLL (1051 $\mu\text{g L}^{-1}$) in the August 2015 pool of 269 homes during the height of the water crisis.⁴ We did a special investigation of this home, by paying to replace its entire home plumbing system except for the last few inches of pipe before the kitchen faucet, and we also examined the entire service line replaced by the city on the same day (March 9, 2016).⁴⁸ To our surprise, this worst case home did not have any pure lead or galvanized iron pipe—it had only lead solder and leaded brass. Sampling in four subsequent rounds determined Resident X's flushed sample WLLs to be consistently below 10 $\mu\text{g L}^{-1}$, whereas first draw WLL (Fig. 3) originating from the very short section of indoor plumbing continued to be as high as 230 ppb.⁴

Analysis of random tap samples that the authors of this paper analyzed from 138 Flint homes (Pieper *et al.*, 2018) in July 2016 before any lead or galvanized iron service line replacement, and again in August 2017 when ~30% (*i.e.*, 3624 homes) of service pipes were replaced, indicate that mean WLLs had dropped by just 6% at that time (Table S2†).⁴ This raises the question, as to how effective the lead pipe and galvanized iron service line replacement program will be, in terms of reducing the mass of total lead released to water across the city.

Assuming that the total lead in biosolids is a summation of lead release from lead bearing service line pipes (lead and galvanized), indoor plumbing (*e.g.*, brass, solder, and lead coated onto indoor pipes from service lines), and non-plumbing sources, the biosolids lead can be described:

$$\text{Biosolids-Pb} = (\% \text{ services remaining}) \times \text{Pb}_{\text{services}} + \text{Pb}_{\text{indoor}} + \text{Pb}_{\text{other}} \quad (2)$$

Lead from other non-plumbing sources (*i.e.*, Pb_{other}) was estimated as 1.41–1.79 kg per month in our prior analysis.³⁸ We then solved for the remaining variables of lead from service pipes (or, Pb_{services}) and indoor plumbing (or, Pb_{indoor})

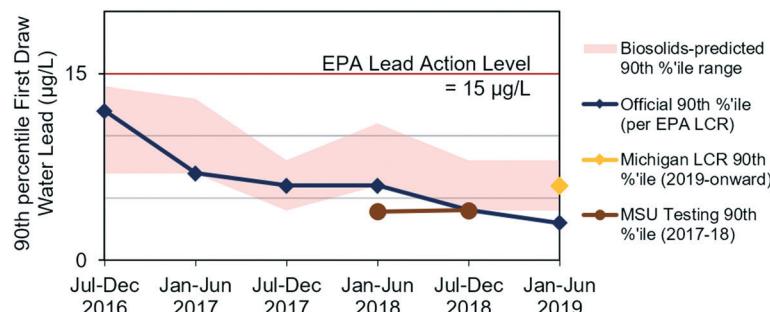


Fig. 2 Comparison in post-federal emergency Flint of 90th percentile first draw lead from official Lead and Copper Rule (LCR) sampling¹¹ against that predicted by the biosolids model (this paper), independent sampling led by Michigan State University or MSU,¹⁴ and the new Michigan LCR that uses the highest WLLs of either first or fifth draw.⁴⁵ The 90th percentile first draw water lead “range” was calculated from the biosolids-predicted WLL₉₀ using the first-draw-to-second-draw ratio of 1.6 (minimum) to 4.0 (maximum) observed in five water sampling rounds 2015–17.



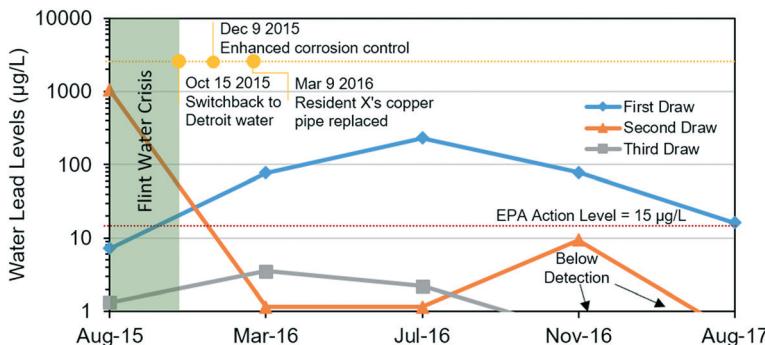


Fig. 3 Water lead levels (first draw, second draw, and third draw) in five sampling rounds during 2015–17 for resident X, who had a copper service line that was removed and replaced with a new copper pipe on March 9, 2016.

in eqn (2), during comparable periods of treated Lake Huron water as Flint's water source and stable phosphate corrosion control, for the years 2013, 2017 and 2018 when the percentage of service pipes in Flint's distribution system were 100%, 48%, and 34%, respectively (Fig. S3†) (see Text S1† for full solution). Projecting results to late 2020 when Flint will have replaced 100% of its lead and galvanized service pipes (*i.e.*, 0% of leaded service pipes remain), we calculate that that remaining sources of lead to water (*i.e.*, leaded brass, lead solder and also legacy lead in pipe scale) will still release about 16–28% of the 2013 pre-water crisis lead mass (Text S1:† Tables I [row G] and II [row G]) as expected.^{13,46,49,50} The characteristic composite WLL due to remaining lead in plumbing would be 5.3–7.4 µg L⁻¹ (Text S1:† Table I [row H]). While this is a 67–77% improvement from pre-FWC 2013 year and an 82–87% reduction from the height of the FWC (Text S1:† Table I [row J]), it illustrates that the post-lead pipe era will not result in completely lead free drinking water.

Historically low incidence of elevated blood lead in children

In terms of childhood lead exposure in Flint, the mean % EBL5 levels for post-FWC months 1–18 (Nov 2015–Apr 2017) and months 19–44 (May 2017–Jun 2019), respectively dropped 55% and 63% below that seen in summer 2014 (Fig. S2†). There was no longer a relationship between biosolids lead and % EBL5, in the any post-FWC period ($p > 0.05$), as would be expected due to near elimination of exposure to waterborne lead from widespread use of bottled water and filters.^{38,51,52}

A general survey of Flint residents ($n = 1913$) in December 2017 confirmed they were following recommendations of public health agencies to not drink unfiltered tap water, as 96% of respondents were using bottled water for cooking, 91.2% were even using it to brushing their teeth and 58.7% were even using it for bathing.⁵³ However, there has been a drop in overall household stress and fear regarding drinking, cooking, bathing, and brushing teeth with unfiltered tap water.⁵⁴ Overall, the analysis strongly supports continued reductions in release of lead to water in post-federal emergency Flint, Michigan.

Conclusions

This research supports the following conclusions about the City of Flint's recovery from high water lead levels that was first revealed through the collaborative work by the authors of this paper and Flint residents in August 2015:⁴

- Lead in biosolids reached a historical low in 2019, due to enhanced corrosion control and replacement of 80% of the lead and galvanized iron service pipes in Flint.
- Estimated composite water lead levels (*i.e.*, equally weighted first draw and second draw or service line WLLs) in 2019 have dropped 90% and 83% from worst levels seen before (2011) and during the Flint Water Crisis (2014), respectively.
- Official LCR 90th percentile first draw WLLs and independent WLLs, in 2016–19, were in good agreement with those predicted using a previously calibrated regression model relying on independent biosolids lead.
- The mean percentage of children ≤ 6 yo with elevated blood lead (% EBL5) in the latest months (May 2017–Jun 2019) is at a historic low, and is 63% lower than that observed during the height of the FWC in summer 2014.
- There is no correlation between % EBL5 and biosolids lead mass in the 44 months post-FWC (Nov 2015–Jun 2019), supporting the reasonable expectation of low consumer water lead exposure during this time of bottled water and lead filters.
- The biosolids data support official data, that lead levels in Flint water are dropping, and do not support unfounded assertions that Flint water still has “crisis” levels of lead in water.
- As Flint approaches 100% lead pipe elimination, building plumbing (*i.e.*, brass and solder) sources of water lead will become dominant, and are estimated to represent 16–28% of the lead released to water in 2013 when all lead sources including service lines were present. The post-lead pipe era in Flint (or anywhere in America) will not result in lead free drinking water.

Conflicts of interest

Aside from our work with Flint residents exposing the water crisis in the first place, our data and testimony have been subpoenaed in several Flint water-related lawsuits. We are



not party to any of the lawsuits. Dr. Edwards has been subpoenaed as a fact witness in many of the lawsuits, but he has refused all financial compensation for time spent on those activities.

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Communication

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Environmental Science: Water Research & Technology

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