Accumulation on and Extraction of Lead from Point-of-use Filters for Evaluating Lead Exposure from Drinking Water

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Revised manuscript submitted to *Environmental Science: Water Research & Technology*

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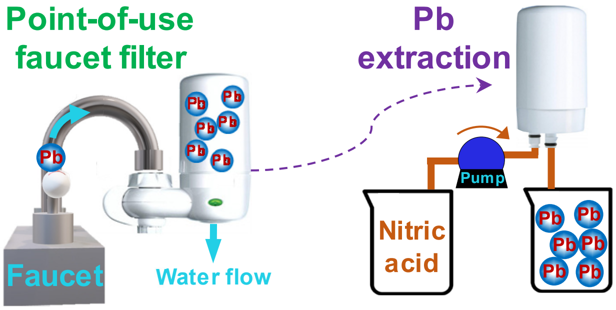

Point-of-use filters can both remove lead from drinking water and also be used for evaluating drinking water lead exposure.

Abstract

The precise quantification of Pb exposure from tap water can help water utilities and public health organizations assess and mitigate elevated Pb concentrations. Several sampling protocols have been developed for this purpose; however, each existing protocol has limitations associated with sampling time, sample sizes, and ease of application. This study confirmed the ability of point-of-use faucet filters to accumulate Pb and then developed an extraction method that can enable quantification of Pb exposure from tap water. Nearly all Pb from both real and synthetic tap water was accumulated on POU filters, and four different methods for extracting the accumulated Pb were evaluated. Approximately 100% Pb recovery was achieved with a single pass flow-through method using a nitric acid solution. This Pb exposure quantification method could potentially be applied to real drinking water systems to provide an effective indication of Pb exposure from tap water.

Water Impact

Better lead exposure data can be used to improve lead control strategies. The ability to extract lead accumulated on point-of-use filters has been a bottleneck in their use for lead exposure assessment. We developed an extraction method that achieves complete recovery of accumulated lead, which will enable its application in assessing potential lead exposure from drinking water in real homes.

Introduction

Lead (Pb) in tap water is mainly contributed by lead service lines and Pb-containing components of premise plumbing.[1–7] Although Pb pipes were banned for potable water supply before the 1990s in the United States, millions of partial or whole lead service lines are still in active use.[8–10] To limit Pb concentrations in tap water in the United States, the Lead and Copper Rule (LCR) set an action level of 0.015 mg/L Pb for the 90th percentile of specific homes' 1 L first draw samples of tap water after a minimum of 6 h of stagnation.[11] Recently, the United States Environmental Protection Agency proposed a new Pb "trigger level" of 10 µg/L in addition to the current 15 µg/L action level.[12] Health Canada lowered the maximum acceptable concentration (MAC) of Pb in the guideline from 10 µg/L to 5 µg/L; this guideline applies to a sample of water taken at the tap and using the appropriate protocol for the type of building being sampled.[13] The sampling approaches used for regulatory compliance are based on single samples and do not provide direct information about Pb exposure associated with actual household water use patterns.

With increasing public concern on Pb contamination in tap water, there is a growing need for rigorous and robust assessments of Pb control and exposure for tap water.[2,14–16] Several sampling protocols have been developed to help water utilities assess and mitigate the risk of Pb in tap water.[5,17–21] In addition to the regulatory compliance sampling protocols noted above, diagnostic sampling may be performed to identify exact sources of Pb, and exposure sampling may be used to provide exposure estimates.[19,22–25] For diagnostic purposes sequential sampling is used to identify sources of Pb from a service line, premise plumbing, and end-use fixtures. Exposure assessment protocols include random daytime sampling (RDT), 30-minute stagnation time (30MS) stagnation, and composite proportional sampling.[13,21,26] Large numbers of RDT samples and 30MS samples may provide a reasonable estimate of the average Pb concentration at the tap.[19] The RDT sampling approach can also provide a good estimate for utilities to know if the corrosion control treatments applied are effective in lowering Pb concentrations at the tap. However, the RDT method provides highly uncertain exposure estimates if insufficient samples are taken. At least 100 samples for each water system are required to provide useful information.[17,27] The information yields a general exposure estimate for a given water system that does not provide specific exposure estimates for individual homes. This approach also does not account for variations in household water use patterns that can influence Pb concentrations in tap water.[28] By collecting a portion of water every time a tap is used to provide water for human consumption, proportional composite sampling gives a more precise measurement of exposure to Pb, but it is only feasible as a research tool since it requires installation of a special tap and the storage and collection of the composite sample from each home.[21,28] Furthermore, residents are still consuming any Pb in the water during these exposure measurements.

NSF-53 certified point-of-use (POU) devices can be a potential Pb exposure assessment tool that can overcome some of the limitations of proportional sampling and random daytime sampling. To quantify cumulative Pb exposure from tap water using POU filters, two requirements must be met. First, the POU filters must remove nearly all of the Pb (both dissolved and particulate) from tap water. Second, it must be possible to extract all of the Pb that had accumulated on the POU filters. The ability to meet the first requirement has been demonstrated by both lab and field studies[29–31] and by the certification process. Among the NSF certifications, NSF-53 focuses on removing health-related contaminants (including Pb) from drinking water.[32] In the NSF-53 protocol, two types of water (pH 6.5 with alkalinity 10–30 mg/L $CaCO_3$ and pH 8.5 with alkalinity 100 mg/L $CaCO_3$) with 150 μg/L added Pb are used to challenge each device. To achieve NSF-53 certification (before 2019), the total Pb concentration in the effluent cannot exceed 0.010 mg/L.

The maximum allowable concentration for this certification has subsequently been lowered to 0.005 mg/L in the latest certification (NSF-53 2019), but the devices used had pre-2019 certification. Deshommes et al. (2010) studied the Pb removal performance of different POU devices and reported that different filter materials had different Pb removal efficiencies.[29] Faucet-mounted and under-the-sink POUs all had adequate Pb removal performance.[29] Different POU filter materials also exhibited different dissolved and particulate Pb removal performances. Cation exchange resin is the most efficient for removing dissolved Pb, and solid block activated carbon is effective for removing both dissolved and particulate Pb.[29] A field study in Flint conducted by Bosscher et al. (2019) found that POU filters can *effectively* remove Pb from the drinking water.[30] However, the ability to meet the second requirement of being able to recover all of the accumulated Pb has not been reported. Cantor et al. (2013) initiated the idea of using POU filters for analyzing Pb exposure from drinking water, but extraction extents of only up to 70% were achieved.[31] Lytle and Schock (2019) have filed a patent on Pb exposure devices with a similar idea, but no data have been publicly reported regarding the ability to recover the accumulated Pb.[33]

Researchers have previously reported using point-of-entry (POE) filtration to monitor particulate Pb release from lead service lines in real homes and compared the results with other Pb sampling approaches.[34,35] Particulate Pb (> 1 μm) accumulated on the POE filters (1-μm polypropylene filter), and the harvested POE filters were acidified with nitric acid to a concentration of 5% v/v for more than 5 days for Pb extraction.[34,35] However, POE filters only remove Pb-containing particles, so such measurements would underestimate total Pb concentrations at the point of entry since they do not include dissolved Pb. The Pb extraction method developed for particulate Pb accumulated on POE filters is not directly transferable for Pb accumulated in POU filters since the filter media (such as polypropylene) used in POE target the removal of only Pb-containing particles, while the solid carbon block filter media in POU devices can remove both Pb-containing particles and dissolved Pb. As a result, there was a need for an extraction method that could achieve complete Pb recovery from POU filters.

There are two objectives in this study. The first one is to evaluate different Pb extraction approaches. The second one is using the best extraction approach to assess the feasibility of using POU filters to quantify potential Pb exposure from consumption of Pb-spiked tap water. Different extraction methods were tested for their ability to recover the Pb from Pb-loaded POU filters. After determining the best extraction method, we applied it to POU filters that were loaded with Pb at realistic drinking water conditions. These experiments demonstrated the feasibility of this method for assessing Pb exposure from drinking tap water.

Materials and Methods

Materials

Reagent grade NaOH and $Pb(NO_3)_2$ and trace metal grade concentrated HNO_3 were purchased from Fisher Scientific. St. Louis tap water was used to provide realistic conditions. NSF-53 2018 certified solid block activated carbon (SBAC) type faucet filters were purchased from Brita®. The structure of the filter is shown in Figure S1. The faucet filter has a bypass that allows the consumer to use unfiltered water as well. Therefore, if only water used for drinking and cooking is filtered, then the POU filter only accumulates Pb that would have been ingested by the consumer. All stock solutions were prepared using ultrapure water (resistivity >18.2 MΩ·cm, Milli-Q, Millipore Corp.).

Method of loading Pb onto POU filter

The Pb was loaded onto POU filters using two different combinations of Pb(II) concentration and water volume. One combination involved feeding each filter once with 10 L of a solution that contains ~2500 μg/L dissolved Pb in ultrapure water. These filters were then harvested for extraction method evaluations. No other water chemistry parameters were adjusted. This solution was supplied to the filter by a peristaltic pump at 2 mL/min, and all the filtered effluent was collected. The Pb concentrations in the influent tank and effluent tank were monitored at the beginning and end of each 10 L flow event, respectively. Each flow event generated 1 filter. This approach was used to generate Pb-loaded filters for the subsequent assessment of four different extraction protocols. The masses of Pb loaded onto the filters are listed in Table 1.

Table 1. Amounts of Pb loaded on filters for extraction experiments.

Extraction experiments	Amount of Pb loaded (mg)[1]
Whole filter in acid	24.41 ± 0.51
Crushed filter in acid	24.74 ± 0.90
Acid recirculation	23.56 ± 0.12
Acid flow-through	25.01 ± 0.30

[1] The uncertainties are the standard deviations from duplicate experiments.

For the other combination of Pb concentration and water volume, each filter was supplied with 100 gallons (379 L) of Pb-spiked St. Louis tap water to reach its certified treatment capacity. The Pb-spiked feedwater was prepared daily by adding 100 mL of $Pb(NO_3)_2$ stock solution (with 19 mg/L Pb(II)) to a mixing tank with 10 gallons (37.9 L) of St. Louis tap water to reach a Pb concentration of 50 μg/L. Before the addition of the stock solution, the Pb concentration in the St. Louis tap water is negligible (< 0.5 μg/L) relative to the amount added, so essentially all Pb loaded onto the filters is from the added Pb(II). Water in the mixing tank was then pumped through the filter at a flow rate of 1.5 L/min. Each filter received 10 gallons of water for 10 days to reach its design capacity. The Pb concentration in the influent tank was monitored every time a new 10-gallon batch was prepared. The Pb concentration in the effluent was measured in two 1-L samples collected during each 10-gallon flow event. The average of these two samples was reported as the effluent concentration. Between each 10 gallons event, 10 L reverse osmosis (RO) water was used to flush the influent tank and tubing to clean the system. After receiving 100 gallons of water, harvested filters were processed to extract loaded Pb. The pressure of the water was not monitored during the experiment. The experimental setup is shown in Figure S2.

The total amount of Pb loaded onto a filter (Pb_{loaded}) in the above two methods was calculated using the difference in concentration (ΔC_{Pb}) between the influent and the effluent and the total volumes of the solution (eq. 1).

$$Pb_{loaded} = \Delta C_{Pb} \times Volume \qquad (1)$$

The chloramine concentration and pH of both the influent and effluent were also monitored. All experiments were conducted at room temperature (21 ± 1 °C) in duplicate.

Pb extraction method

Four extraction methods were evaluated with respect to their ability to recover Pb from Pb-loaded filters (Figure 1). All filters were pretreated with compressed air to remove stagnant water before extractions. The acid flow-through and acid recirculation extraction methods were conducted with intact filters with their filter medias still in original plastic casing. For the whole filter in acid and crushed filter in acid extraction methods, the plastic casing surrounding the filter core was cut open so that the core could be removed. The cores were then dried at 100 °C overnight. For the whole filter in the acid extraction method, the intact filter cores were placed directly into 1 L of 1 M HNO_3 solution for 48 h. For batch acid extraction of crushed filter material, the filter cores were broken into small pieces manually and then further ground to a powder, and all the filter material was then placed into 1 L of 1 M HNO_3 solution for 48 h. All solutions were magnetically stirred at 500 rpm. The acid recirculation extraction method was conducted by recirculating 1 L of 1 M HNO_3 through the filter for 48 h. Acid flow-through extraction was conducted by pumping 2 L of 1 M HNO_3 through the filter in a once-through flow mode. The flow rate for the above two methods was 2 mL/min. For all methods, the mass of Pb extracted was determined from the extractant volumes and Pb concentrations.

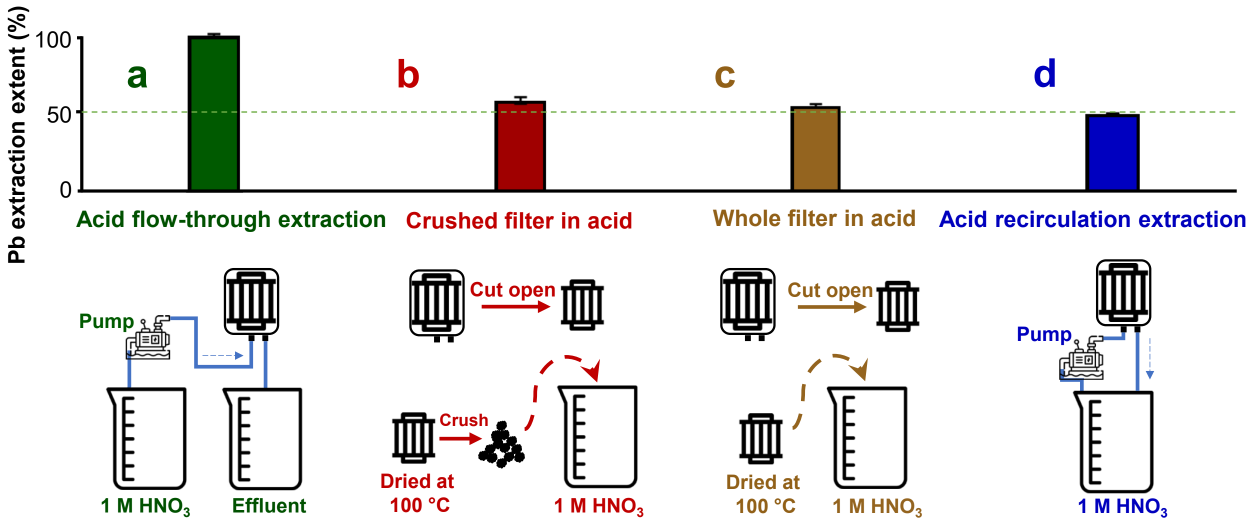

Figure 1. Extraction performance using (a) acid flow-through, (b) crushed filter in acid, (c) whole filter in acid, and (d) acid recirculation approaches. Experiments were conducted with filters loaded with approximately 25 mg Pb. Error bars represent the standard deviations from duplicate experiments.

Analysis methods

Solution pH was measured with a glass pH electrode (TRIS Compatible pH/ATC electrode, Fisher Scientific) and pH meter (Accumet AB150 pH Benchtop Meter, Fisher Scientific). To measure the dissolved Pb concentration, solutions were first filtered through 0.22 μm pore diameter polyethersulfone (PES) syringe filters (Environmental Express) and then acidified to 1% trace metal grade nitric acid. Samples were then analyzed by inductively coupled plasma mass spectrometry (PerkinElmer ELAN DRC II) following EPA Method 200.8 (detection limit is 0.5 μg/L).[36] The residual chloramine concentration was measured with the standard DPD method.[37]

Results and Discussion

Pb extraction methods

The Pb extraction recovery is the mass of Pb in the extracted solution divided by the amount of the Pb that had accumulated in the filter during the loading stage (eq. 2). The acid flow-through extraction achieved 100% Pb recovery (Figure 1a) with 2 L of acid extraction solution. The filter solid-to-acid mass ratio in the acid flow-through extraction method was approximately 1:10. Each filter core has a mass of about 200 g.

$$Pb\ extraction\ extent = \frac{Pb\ in\ the\ extracted\ solution}{Pb\ accumulated\ in\ the\ filter} \times 100\% \qquad (2)$$

Both batch acid extractions (intact filter core and smashed filter) achieved 50-60% Pb recovery (Figure 1b & c). These recoveries are within the range reported for previous experiments involving Pb desorption from similar carbon-based materials (20-100%) with 0.02 – 1 M HNO_3 or HCl.[38–41] Extraction of the crushed filter material achieved 60% recovery, while extraction of the intact filter recovered about 50% of the Pb. This difference indicates that crushing the filter core to a powder did increase the extent of Pb extraction, but it was still not to a level approaching full recovery. Recirculation of acid through an intact filter in the housing also yielded only about 50% recovery (Figure 1d) when operated with the same solid-to-acid ratio (1:5) as used in the batch extraction. Furthermore, no significant difference was observed between duplicates (as indicated by error bars in Figure 1), which suggests the experiments are reproducible. The Pb extraction extents of batch acid and acid recirculation extraction methods were similar compared with the previously reported results.[31] Cantor et al. (2013) tested two extraction methods: (1) acid recirculation extraction with a 2% nitric acid/5% hydrochloric acid solution with intact POU filter in the plastic casing, and (2) batch digestion with a solution of hydrogen peroxide, hydrochloric acid, and nitric acids with 1 gram of crushed POU filter media powder.[31] However, neither approach approached complete recovery (< 70%) of the Pb that had accumulated on the POU filters.[31] The similarity of the recoveries for the batch extractions and the recirculating flow extraction is reasonable because both are closed systems with the same total masses of carbon filter material and volumes of acid; after sufficient recirculation cycles, the recirculating system acts as a batch system. Comparing these four different extraction methods, the acid flow-through extraction method had the highest recovery of Pb, which likely resulted from the continuous supply of Pb-free acid solution to the filter. Because of this continuous supply the concentration of Pb in the acid never reaches a value that would be set by equilibrium adsorption to the filter. Therefore, all of the Pb accumulated in the POU filter can be desorbed as there is always a driving force for transfer of Pb from the filter to the acid. Although particulate Pb was not tested, we hypothesize that a similar recovery extent would be observed and discuss this issue in a later section.

An additional experiment was conducted in which the Pb concentration of the flow-through acid extraction effluent was monitored in 100 mL increments to determine the necessary volume of acid to achieve 100% recovery. More than 97.3% of Pb was extracted in the first 1 L of extractant (Figure 2). This recovery extent is much higher than in the whole filter in acid, the crushed filter in acid, and the acid recycling extraction methods, which were also done with 1 L acid solution. In the flow-through extraction, 100% recovery within uncertainty had been achieved after a cumulative volume of 1.4 L of acid had passed through the filter. As a result, using a total volume of 2 L would provide an additional margin of safety to ensure full recovery of Pb from the filter. We note here that a larger volume of acid may be required to achieve the same recovery extent if a much higher amount (>> 25 mg Pb) of Pb is suspected to be accumulated in the POU filter.

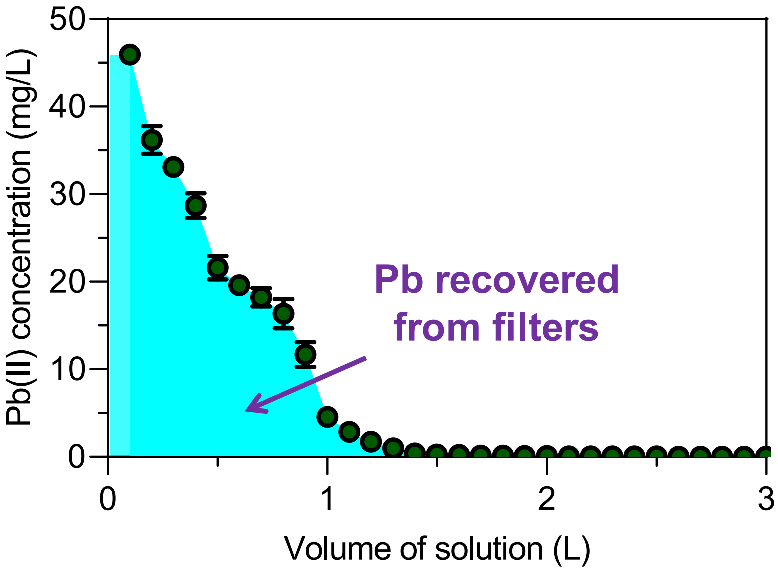

Figure 2. Pb concentrations in the effluent of Pb-loaded POU filters during extraction using 1 M HNO_3 as influent. Measurements were taken at each 100 mL increment. The light-blue shaded area indicates the mass of Pb extracted by the acid solution. The error bars represent the standard deviation from duplicate experiments.

A case study with Pb-spiked St. Louis tap water

Pb-spiked St. Louis tap water was used to represent a realistic drinking water composition. Each POU filter received 100 gallons of Pb-spiked tap water with a 50 µg/L dissolved Pb concentration. The POU faucet filters removed almost all (> 99%, average) of the Pb in the influent (Figure 3a). The Pb concentration in the influent was around 50 µg/L during the whole experiment, and the concentration in the effluent was near or below the detection limit. Although the Pb removal efficiency was slightly lower in the last 10 gallons of water compared with the previous 90 gallons, the Pb concentration in the effluent was still below 10 µg/L, which meets the requirement for NSF-53 2018 certification. The inconsistent Pb removal performance may lead to an underestimation of Pb exposure. Similar POU performance tests conducted by Deshommes et al. (2010) showed

80 - 92% removal of dissolved Pb and 92 - 99% removal of particulate Pb.[29] After reaching the product-specified treatment capacity of 100 gallons, the flow-through acid extraction method was then applied to these Pb-loaded filters. The extraction resulted in 100% recovery of Pb from POU faucet filters that had received 100 gallons of Pb-spiked St. Louis tap water (Figure 3b).

The pH values and chloramine concentration in the influent and effluent were monitored during the experiment. While the effluent pH values were stable during the experiment (Figure S3), an increase in the chloramine concentration was observed in the effluent over the lifetime of the filter (Figure S4). The decrease of chloramine concentration is normal since this POU filter is also certified by NSF-42 for chlorine/chloramine, taste, and odor compound removal. Similar results were observed by Deshommes et al. (2010).[29]

Although particulate Pb was not tested in this study, a similar extraction efficiency could be expected based on lab-scale experiments, field studies, and the particulate Pb extraction results from POE filter studies.[29,30,34,35] Solubility calculations with common Pb(II)-containing solids present in pipe scales (Figure S5) indicate that these Pb solids can be complete dissolved in the acid provided. Reductants (such as ascorbic acid or hydroxylamine) could be added if PbO_2 is present, and slower flow rates would allow more time for Pb solid dissolution.[42] Further tests are needed to determine how low the flow rates would need to be to allow for the dissolution rates of any Pb-containing solids to result in 100% recovery of particulate Pb from a POU filter.

We note that there are limitations to using POU filter for evaluating Pb exposure from drinking water. For example, this method cannot provide information on specific Pb sources, and that information is still best obtained by profile sampling. The approach can only be applied to situations in which the POU devices accumulate all of the Pb from tap water. Recently, the Pb concentration in tap water exceeded the LCR action level in some districts of Newark, New

Jersey.[43,44] POU devices were distributed to reduce the risk of Pb exposure from tap water, but in several cases they were unable to lower lead concentrations to expected levels.[45] This poor performance may have been caused by the formation of well-dispersed Pb nanoparticles at this particular water chemistry condition that were then able to pass through the POU devices.[7,46–48] While the situation in Newark was unusual since most other studies observed effective lead removal in real homes, it does warrant further attention with respect to using POU filters for evaluating Pb exposure from drinking water.

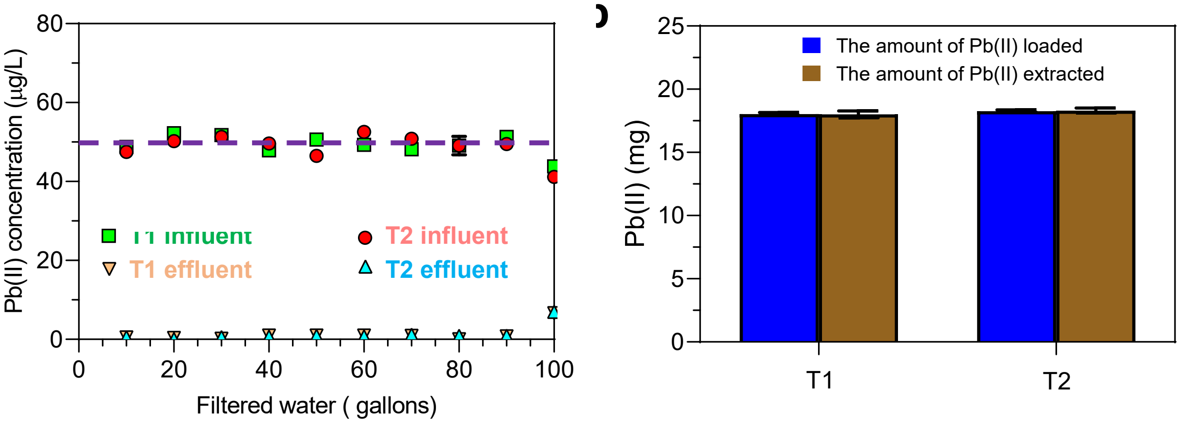

Figure 3. Evaluation of (a) removal of dissolved Pb from Pb-spiked St. Louis tap water to create Pb-loaded point-of-use filters and (b) recovery of Pb from the filters using a flow-through acid extraction approach. T1 and T2 are two independent experiments. The influent is the Pb concentration in the Pb-spiked St. Louis tap water, and the effluent is the Pb concentration after that water had passed through the POU filters. The error bars represent the standard deviation from duplicate experiments.

Conclusion

We demonstrated a simple but effective method for extracting dissolved Pb that had accumulated on POU filters. Based on this extraction method and earlier work that examined POU devices for assessing Pb exposure, we conclude that POU filters can be used to evaluate Pb exposure from drinking water in real residences.[31,33] The exposure evaluation method in this study is built on earlier research. The method involves easy installation of the POU filter is convenient, and no further modification (such as opening the plastic casing or pulverizing the filter) to the filter is needed for the Pb extraction protocol. Residents could mail their filters to a laboratory analysis after they reach their certified treatment capacity, which is monitored automatically by an indicator on the faucet mount of most POU filter system. Utilities that distribute filters after a Pb service line replacement could use this measurement of Pb accumulated on the POU filters to both confirm the efficacy of the replacement in decreasing Pb concentrations and to determine when the distribution of filters is no longer needed. We note that Brita® POU filter User's Guide recommends a 5-second flush before each use and a 30-second flush if the tap has not been used in a few days. Flushed water would still accumulate Pb in the filter but would not have been consumed by a user. Because of the small volumes involved, the effect on the exposure estimation will be small and the impact on either overestimating or underestimating exposure will depend on how the Pb concentration in that flushed water compares with the Pb concentration in the water flowing through the filter after the flush. The method can be adapted for evaluation of exposures to other inorganic and organic contaminants, given the excellent adsorption ability of the NSF-53 certified POU filters.[29,30,49–55]

Further research can extend the water chemistry conditions evaluated. In the NSF-53 certification protocol, the total Pb concentration in the influent challenge water is 150 µg/L with 30 ± 10% total particulate Pb, and at least 20% of that fraction must be 0.1-2.0 µm in size. Additional experiments are needed regarding different tap water conditions and PbO_2 solids. Field studies that compare this exposure measurement method with other methods (e.g. RDT, 30MS, and composite proportional sampling) would help researchers and water utilities select appropriate methods for assessing Pb exposure and corrosion control effectiveness.

Acknowledgments

This research was supported by the U.S. National Science Foundation (CBET 1603717, CHE 1709484). W.P. acknowledges the fellowship support through the McDonnell International Scholars Academy. This work was performed in part using the Nanoscale Research Facility at Washington University in St. Louis, a member of the National Nanotechnology Infrastructure Network (NNIN), which was supported by the National Science Foundation under Grant No. ECCS-0335765. We thank Anushka Mishrra, Anshuman Satpathy, Neha Sharma, Guiwei Li, and Yeunook Bae for assistance with experimental activities and helpful discussions.

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