Environmental Science & Technology

pubs.acs.org/est

Article

Reductions in Children's Blood Lead Levels from a Drinking-Water Intervention in Madagascar, Sub-Saharan Africa

Published as part of the Environmental Science & Technology virtual special issue "Accelerating Environmental Research to Achieve Sustainable Development Goals".

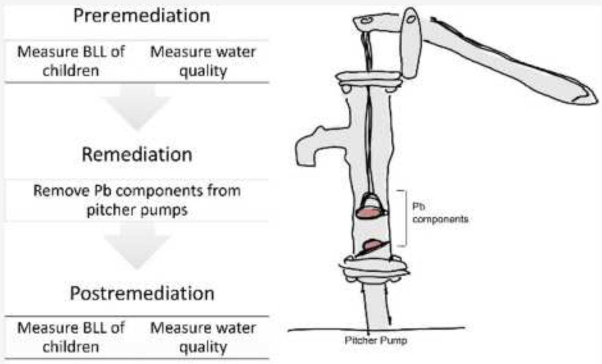
Adaline M. Buerck,* Mahmooda Khaliq, Katherine Alfredo, Jeffrey A. Cunningham, Luke John Paul Barrett, Rinah Rakotondrazaka, Lova Rakotoarisoa, Wyatt M. Champion, and James R. Mihelcic

Cite This: *Environ. Sci. Technol.* 2023, 57, 16309−16316

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: One in three children globally is estimated to have blood lead levels (BLL) at or above the BLL reference value of 5 μg/dL with increased burden falling on low- and middle-income countries (LMIC). Within developed countries, aqueous lead is the predominant exposure route. However, aqueous lead exposure is rarely examined in the LMIC, leaving a gap in the literature that ignores a potentially significant route of exposure. Furthermore, limited lead-based remediation efforts around consumer products have been examined. This study investigates the importance of lead exposure from the water supply through a case study in Toamasina, Madagascar. The project measured aqueous lead and BLL of children pre- and postremediation efforts (i.e., removal of leaded pump components in hand pumps) to verify the impact of aqueous lead exposure within this community. Removal of the leaded pump components (i.e., piston and foot valves) and replacement with nonleaded components decreased aqueous lead levels below the World Health Organization provisional guideline of 10 μg/L in all but 4% of pumps tested. Measured BLL concentrations indicated a statistically significant decrease in BLL from pre- to postremediation. Furthermore, the remediation resulted in a decrease in BLL for 87% of children with the greatest changes in BLL observed for children with the highest preremediation concentrations. These findings point to a need for greater consideration of lead in drinking and cooking waters as an important exposure route in LMIC.



KEYWORDS: *drinking water, groundwater, prevention, children's health, toxic metals, sustainable development goals*

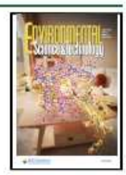
1. INTRODUCTION

Societal lead (Pb) regulation (e.g., phase-out of leaded gasoline, Lead and Copper Rule in U.S., E.U. Drinking Water Directive) has resulted in substantial decreases in elevated blood lead levels (BLL) in high-income countries (HIC).[1−3] For example, the phase-out of leaded gasoline in the U.S. was associated with a decrease in the average BLL (37% or 5.4 μg/dL) in a cross-sectional survey representative of the U.S. civilian noninstitutionalized population.[4] Unfortunately, lead is still a major global environmental pollutant estimated to account for over one million annual premature deaths.[5]

Children are most at risk of elevated BLL, with a recent study reporting approximately 800 million children globally (1 in 3) with BLL at or above 5 μg/dL.[6,7] Most of these children reside in low- and middle-income countries (LMIC) where there is a lack of regulations and understanding of the harmful effects of lead, as well as other comorbidities, such as improper nutrition, that can increase childhood lead absorption.[8−10] All ages have the potential for negative health implications from lead exposure, but children are at the highest risk. Examples of health implications include, but are not limited to, idiopathic developmental intellectual disability (IQ loss), gastrointestinal disturbances, delayed growth, and hematological effects.[7] Currently, there is no identified threshold below which lead is no longer associated with adverse effects.

Because of advances in reducing nonwater-related routes of exposure, a predominant source of childhood lead exposure in HIC is now piped drinking water, as noted with high-profile

Received: May 18, 2023
Revised: September 26, 2023
Accepted: September 27, 2023
Published: October 18, 2023



cases such the District of Columbia (USA),[11] Glasgow (UK),[12] and Flint, Michigan (USA).[13] Many studies have looked at the relationship between aqueous lead concentrations and BLL in children within HIC.[11,14−16] Remediation efforts such as corrosion control, service-line replacements, and point-of-use filters have been effective in reducing BLL and in improving public-health outcomes.[17−20]

In LMIC, however, there is lack of data and limited discussion, if any at all, on the importance of aqueous concentrations on elevated BLL.[21] For example, a 2021 systematic review of 520 lead studies in LMIC identified zero studies classifying aqueous lead as a possible exposure route.[6] The major sources of lead exposure identified in that review included informal lead-acid battery manufacturing and recycling, metal mining and processing, and electronic waste.[6] However, a study by Fisher et al.[22] reported that 9% (24/216) of rural community water system samples (collected from handpumps and community taps) in Mali, Ghana, and Niger had elevated total (dissolved plus particulate) lead concentrations in excess of the World Health Organization (WHO) drinking water guideline of 10 μg/L.[23] This suggests the need to evaluate and address the presence of lead in potable water in LMIC.

Consistent with these recent findings by Fisher et al.,[22] investigations into lead exposure in Madagascar identified a source of aqueous lead exposure in locally manufactured pitcher pumps (i.e., handpumps).[24,25] These handpumps are a common primary or supplemental water source that account for approximately three-quarters (i.e., ∼62%−77%) of water systems in Madagascar according to reports from the Ministry of Water, Sanitation, and Hygiene of Madagascar.[26,27] In Toamasina, there are estimated to be upward of 9000 pumps that service ∼60% of the 280,000 persons locally, as centralized piped water is not always affordable or reliably available.[28] A recent study by Champion et al.[21] indicated that 75% (265/354 respondents) of pump users in the study report that their child uses the pump water for drinking, cooking, or both. Dissolved aqueous lead concentrations from these pump systems were observed to often exceed the WHO guideline of 10 μg/L, with some pump water samples reaching concentrations >100 μg/L.[24,25,28] The major contributors to the aqueous lead were two leaded pump components—the piston and foot valves, made with lead from recycled batteries. Using the dissolved aqueous lead measurement data, children's BLL were projected with the U.S. Environmental Protection Agency (EPA) Integrated Exposure Uptake Biokinetic (IEUBK) model,[29] estimating that up to 90% of children could exhibit BLL > 5 μg/dL.[25] Furthermore, IEUBK modeling outputs predicted that remediation of pumps (i.e., removal and replacement of lead components) would result in a reduction in BLL.[30] Given the fact that nutrition deficiencies can increase childhood lead absorption in LMIC, it is unknown how reducing lead exposure through a source of water would affect measured BLL. In addition, we were unable to identify a study specific to Sub-Saharan Africa or other LMIC, measuring childhood BLL before and after an intervention to replace a known source of lead in a water system to track the resulting decrease in measured childhood BLL.

Therefore, the main objective of this study is to quantify the reduction in children's BLL when leaded components are removed (i.e., remediated) from a water-supply system in a low-income country. By doing so, this study quantifies the importance of lead exposure from drinking water as an important pathway for elevated BLL in an LMIC. Specifically, this study uses Toamasina, Madagascar, and the locally manufactured pitcher pumps as a case study. By examining pitcher pumps that serve as the primary water supply for a large segment of the population, we demonstrate the impact that lead exposure from water supply systems may have on childhood BLL in LMIC. We utilize a methodology transferable to other LMIC where similarly constructed handpumps and components of private and public distribution systems may also contain lead.

2. MATERIALS AND METHODS

2.1. Sample Selection and Distribution. This observational pre/postremediation (i.e., replacement of lead pump components with nonleaded pump components) study evaluated aqueous lead concentrations of pumps and BLL measurements of children under six years of age in Toamasina, Madagascar (refer to Supporting Information for more details on the remediation process). This assessment is part of a larger project that took place November 2020 to February 2021, which remediated 500 pumps, coupled with the implementation of a social marketing campaign (i.e., behavior change campaign) aimed at phasing out the use of lead components in the manufacturing and repair of pitcher pumps.[21,31] BLL were measured at two points: at the beginning of the project for over 300 children in Toamasina prior to pump remediation and the social marketing campaign efforts, and after remediation and the social marketing campaign for a subset of the original sample. Recruitment for the larger project employed a cluster sampling approach and used nine local health clinics across five arrondissements (or city districts) in Toamasina. Families seeking care at the clinic were recruited based on willingness to participate and having a child between six months and six years old. All data were collected at the clinic (BLL data) or in the field (aqueous data) by trained local staff, transported to the local project partner's office, and then put into an online application, verified by a second staff member for correctness, and shared with the study team for review and analysis. All study protocols were approved by the Biomedical Research Ethics Committee of the Ministry of Public Health of Madagascar and the USF Institutional Review Board (STUDY000143).

For this study, a sample size of 34 paired samples was required based on an assumed medium effect size of 0.05, 80% power, and 95% confidence interval. In total, this study collected and analyzed paired data (i.e., data collected pre- and postremediation of the two pump check valves) for 55 children 6 months to 6 years (31 males and 24 females) and 48 associated primary water source pumps. All pumps were associated with at least one unique child. Six of the pumps were associated with more than one child in our study (one pump was associated with three children, and five pumps were associated with two different children each). BLL readings for each child were taken prior to the pump remediation, and at least 1 month (37−67 days) after pump remediation to allow for approximately one-half-life of lead in the blood to occur (half-life of lead in blood is reported as 1−2 months).[32] Further breakdown of sample population, including geographic location, age, and gender, can be found in the Supporting Information.

2.2. Data Collection Methods. *2.2.1. Blood Lead Level (BLL) Measurements.* Following parental consent, a 50 μL blood sample was collected from the child's finger by trained

https://doi.org/10.1021/acs.est.3c03774
Environ. Sci. Technol. 2023, 57, 16309−16316

medical personnel and analyzed using a Magellan LeadCare II device (North Billerica, MA). The LeadCare II testing instrument is a portable device for testing the amount of lead in whole blood by using anodic stripping voltammetry. Our test kits were not affected by the 2021 FDA recall.[33]

To limit contamination, the child's hand was washed with soap and water, and the finger was cleaned with alcohol prior to sampling. The LeadCare II system displayed measured BLL readings as either a value between 3.3–65.0 μg/dL to one decimal place (i.e., 0.1 ug/dL, consistent with the manufacturer's reported accuracy at lower BLL ranges[34]): "Low" if below the lower detection limit of 3.3 μg/dL or "High" if above the upper limit of 65.0 μg/dL. Values below the detection limit were replaced with 2.33 μg/dL in our analysis, calculated as the lower detection limit divided by the square root of 2, consistent with previous studies.[35] No values above 65 μg/dL were measured in the study sample.

BLL reference values from the Centers for Disease Control and Prevention (CDC) were updated in 2021 moving the reference value from 5 to 3.5 μg/dL, though it is found that there is no safe level of lead exposure.[36−38] This decrease in the reference value is based on U.S. data, which are from a high-income country where many sources of exposure have been addressed. Due to this work taking place in a LMIC where similar regulations reducing exposure are not in place as well as to be consistent with past efforts in the region, this analysis used a reference value of 5 μg/dL.

2.2.2. Analysis of Aqueous Lead Concentrations in Pumped Water. Following the methodology outlined by Akers et al.,[24] water samples were collected both prior to and following the remediation of the pump. The remediation process consisted of removing and replacing the lead-containing piston and foot valves with nonleaded components. Logistically, given that these pumps are the only sources of water for households, controlling all sampling with specified stagnation intervals was not employed. Instead, a flushed, composite sample sampling approach was adopted that consisted of wasting 15 L of the pitcher pump system and then collecting another 15 L of sample for analysis. The choice of 15 L as the collection volume for the composite sample was based on the prevalent use of 15 L buckets to collect water by community members.[24] A 10 mL glass pipet was rinsed using the sample water, and a 5 mL aliquot sample was collected to represent a flushed composite sample. Samples were analyzed for dissolved lead using a Palintest Scanning Model 1100 Analyzer (SA1100) that measures dissolved lead by the process of anodic stripping voltammetry with a lower limit of detection of 2 μg/L of lead. Readings under the detection limit (12.5% (n = 6/48) pre- and 60.4% (n = 29/48) postremediation) were recorded as 1.41 μg/L for analysis (calculated as the lower detection limit divided by the square root of 2[35]).

2.2.3. Statistical Analysis. Paired analysis was conducted to determine if there were significant differences in measured aqueous lead levels and BLL pre- to postremediation. Pumps with aqueous lead concentration at detection or under detection prior to the remediation were removed (n = 12) from this analysis, as it was not possible to quantify a change in aqueous lead concentration. The 12 pumps removed from this analysis remained at or below detection postremediation. A significance level (alpha) of 0.05 was used for this test. Appropriateness of the Wilcoxon signed rank test was confirmed visually and by a Shapiro–Wilk normality test (refer to Supporting Information for additional detail).

Further analysis of paired samples included examining correlations between the aqueous lead and BLL via Pearson correlations. Following review of variables, multiple linear regression analyses were performed based on significance while also controlling for demographic variables such as age, gender, and weight. Variables were added to analysis keeping in mind significance and ability to gather data (e.g., aqueous Pb measurements are less invasive and easier to collect than BLL data). A value of $p < 0.001$ was used as the cutoff for statistical significance in all Wilcoxon tests. For correlation and regression analyses, we consider $p < 0.001$ (***) as highly significant, $p < 0.05$ (**) as great significance, and $p < 0.05$ (*) as significant.

3. RESULTS AND DISCUSSION

3.1. Blood Lead Level (BLL) Measurements. Prior to the pump remediation, 96% of children (n = 53) had measured BLL over 5 μg/dL (the BLL reference value used for this study). Following the remediation, only 65% of children (n = 36) had measured BLL over 5 μg/dL. In total, the BLL of 48 children (87%) decreased, with post-BLL ranging in magnitude from decreases of 0.2–24.8 μg/dL (mean = 4.91 μg/dL), while six children had an increase of 0.9 μg/dL on average. With the half-life of BLL estimated to be 1–2 months, and an average of 45 days between the remediation and the post-BLL test, it is likely that BLL will further decrease in the future.[32]

Analyzing the entire population of participants, the pre- and postremediation BLL are plotted in Figure 1 and Figure S.3. A Wilcoxon signed rank test conducted on the BLL values showed a significant difference ($p < 0.001$, n = 55) between pre- and postremediation BLL. The median pre-BLL was 8.6 μg/dL compared to 6.3 μg/dL for post-BLL. Given the short duration between remediation and post-BLL sampling and the statistically significant decrease witnessed already, it is expected that aqueous lead was a significant portion of lead intake for most study participants and that BLL will continue to decrease with increased time postremediation.

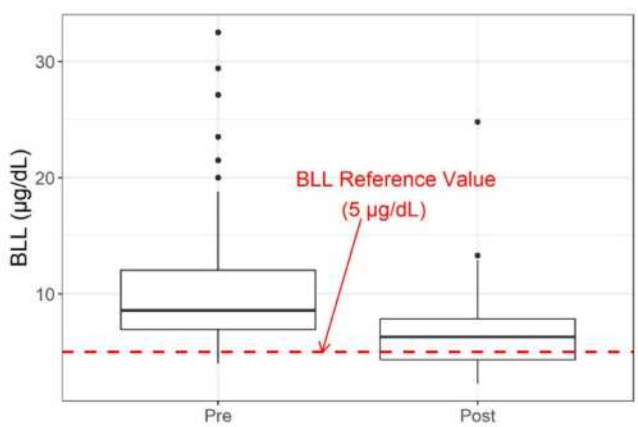


Figure 1. Box plots of BLL concentrations (μg/dL) of children aged six months to six years pre- and postremediation of pumps (i.e., removal and subsequent replacement of leaded components) (n = 55). Preremediation BLL concentrations (mean = 10.85, SD = 6.30) and postremediation BLL concentrations (mean = 6.68, SD = 3.70). Box hinges represent the first and third quartiles, and the middle represents the median. Whiskers extend 1.5 times the interquartile range of the hinge. Points beyond the whiskers are outliers.

https://doi.org/10.1021/acs.est.3c03774
Environ. Sci. Technol. 2023, 57, 16309−16316

3.2. Aqueous Lead Concentrations in Pumped Water. Prior to the remediation, 27% ($n = 13$) of the pumps sampled using the flushed sampling protocol of this study contained aqueous lead concentrations that exceeded the WHO provisional guideline of 10 μg/L,[23] suggesting that total lead concentrations in samples collected immediately after a stagnation period could be far higher; this is concerning. The concentrations measured in this study were higher than other studies examining aqueous lead in Sub-Saharan Africa, such as Fisher et al.,[22] where 9.2% (24/261) of samples exceeded the WHO provisional guideline. Fisher et al.[22] measured total lead and sampled using a 1 h stagnation time, compared to our flushed sampling protocol and analysis of dissolved aqueous lead concentrations.

All pumps with quantifiable changes in aqueous lead levels ($n = 36$) observed a decrease in aqueous lead concentration ranging from 1 to 97 μg/L. The aqueous lead concentration of pump water prior to and following the removal of lead is shown in Figure 2, Figure S.3, and Table S.2. This decrease in lead concentrations across our pump study sample was statistically significant (Wilcoxon signed rank test, $p < 0.001$, $n = 36$). It is worth noting that even using a flushed, composite sample and analyzing for only dissolved lead, statistically significant decreases are measured. While our aqueous sampling method might not provide the highest lead concentration possible, the contribution from the two replaced valves were great enough to observe these changes. The impact of our remediation on total lead intake is likely much greater.

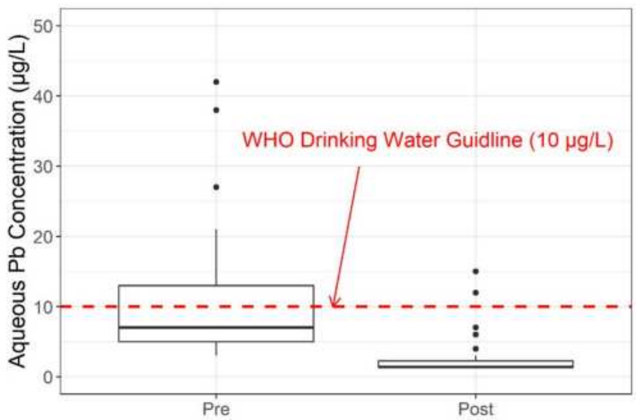


Figure 2. Box plots of the aqueous lead concentrations (μg/L) pre- and postremediation of pumps (i.e., removal and subsequent replacement of leaded components) ($n = 36$). Preremediation levels (median = 9.0, mean = 17.31, SD = 23.97) and postremediation levels (median = 1.41, mean = 2.72, SD = 2.98). Box hinges represent first and third quartiles, and middle represents the median. Whiskers extend 1.5 times the interquartile range of the hinge. Points beyond the whiskers are outliers. Two outliers for the preremediation were over 50 μg/L and are not represented in the figure.

Following remediation, a small number of pumps (2 out of 48 total pumps) continued to exceed the provisional guideline of 10 μg/L (postremediation levels of 12 and 15 μg/L). While one pump experienced a greater than 50% decrease in lead concentrations, one essentially remained unchanged. This was unexpected, yet a similar result was observed during the 2018 remediation campaign where 2% of 418 pumps measured postremediation exceeded the guideline. Additionally, of the 418 pumps examined postremediation, 244 had a preremediation measurement. Comparison of the paired pumps within the 2018 remediation had groupings of pumps that had measurable decreases (86%, $n = 194/244$), had no measurable change (11%, $n = 24/244$), and had small increases in aqueous lead concentrations (3%; $n = 7/244$).[30] There are several potential explanations for the continued elevated dissolved lead concentrations postremediation that include unaccounted for lead release from solder,[39−41] brass components used within the pump,[22,42] release of adsorbed lead from other pump components,[43−45] and water quality characteristics.[39,46] Also, the leaded components themselves are likely to have great variation in lead concentrations. Field observations of the pumps being manufactured and installed revealed that technicians melted automobile batteries to make the valve components. Raman spectroscopy of several valves confirm that they primarily consist of lead[47] providing confidence that they are the primary source of lead contamination for the pitcher pumps; however, all the other potential sources of lead contribute to the overall concentrations.

3.3. Determining Correlations and Drivers Associated with the Magnitude of BLL Reductions. Aqueous lead concentrations and BLL both experienced statistically significant decreases in our study sample. Pearson correlations were examined for 10 variables: age, height, weight, pre-BLL, post-BLL, preaqueous Pb, postaqueous Pb, ΔBLL, Δaqueous Pb, and time between remediation and the post-BLL test to further determine key demographics and variables that contribute to the greatest reductions in BLL (see Table 1). Six combinations of variables were found to be highly correlated ($r \geq 0.6$) and statistically significant to the 0.001 level. The highest correlations were among age, height, and weight. These three variables were all highly positively

Table 1. Correlation Matrix of Study Variables

		1	2	3	4	5	6	7	8	9	10
1	Age (years)	1									
2	Height (cm)	0 93[b]	1								
3	Weight (kg)	0.83[b]	0.86[b]	1							
4	Pre-BLL (ug/dL)	0.13	0.17	0.22	1						
5	Preaqueous Pb (ug/L)	−0.04	0.00	−0.15	−0.24	1					
6	Post-BLL (ug/dL)	0.18	0.14	0.28	0.62[b]	−0.33[a]	1				
7	Postaqueous Pb (ug/L)	−0.18	−0.2	−0.23	−0.23	0.31[a]	−0.29	1			
8	ΔBLL (ug/dL)	0.03	0.12	0.07	0.81[b]	−0.06	0.04	−0.07	1		
9	ΔAqueous Pb (ig/L)	−0.02	0.03	−0.13	−0.22	0.99[b]	−0.31[a]	0.2	−0.05	1	
10	Days between remediation and Post-BLL test (days)	−0.028	−0.029	−0.015	−0.011	0.28	0.071	0.11	−0.067	0.28	1

[a] $p < 0.05$ (there are no variables measuring between 0.001 and 0.01). [b] $p < 0.001$; ($N = 45$).

https://doi.org/10.1021/acs.est.3c03774
Environ. Sci. Technol. 2023, 57, 16309−16316

correlated as expected for an average population and suggest that our population would track on an expected growth chart. This is valuable since we did not collect nutrition information nor did we have our medical staff examine the participants for potentially severe malnutrition.

Also, as expected, pre-BLL was correlated with both post-BLL ($r = 0.62$, $p < 0.001$) and the change in BLL ($r = 0.81$, $p < 0.001$). These correlations show that participants with a higher starting BLL tended to have a higher post-BLL but also a greater change in BLL. As there is greater potential for a BLL to decrease if the participant had a high starting BLL, these correlations do follow expected trends in BLL reduction. As expected, there was also a high correlation between preaqueous lead and the change in aqueous lead ($r = 0.99$, $p < 0.001$), with all pumps having a decrease in aqueous lead levels. It stands to reason that the higher the starting aqueous lead level is, the larger the absolute change in aqueous lead witnessed is.

Multiple linear regressions were calculated to predict the change in BLL based on various potential predictors controlling for specific demographics of the population. Table 2 summarizes the results of the final models. All models presented have statistically significant predicting power accounting for up to ∼66% of the variation in change in BLL. Pre-BLL was the strongest predictor of the resulting change in BLL, which reinforces the correlation discussed previously between pre-BLL and change in BLL ($r = 0.81$, $p < 0.001$). Though other predictors were not found to be statistically significant, they did strengthen the regressions' prediction power. When controlling for demographics, we included either age or weight in the regression analysis to avoid collinearity given the strong correlation between the two variables. Neither was statistically significant as a driver for changes in BLL most likely due to the limited ranges of participant weights and ages. Therefore, weight was selected as a controlling variable given the bioaccumulation pathways for lead within the body and the impact of nutrition on lead uptake and accumulation. All ages contained within the study population fall into the category of having exponential changes in the half-life of blood lead.[48] The half-life of blood lead has also been shown to vary based on the peak lead concentration further complicating prediction ability within a population.[49] Additionally, previously identified risk factors that could impact BLL of children in Toamasina, such as proximity to railways or major roadways, type of flooring, and food consumption (e.g., beans, vegetables, and rice)[21] would be of interest for strengthening the prediction power of the regression. However, due to the current sample size, adding in additional variables would result in overfitting in the analysis and was therefore not considered in our final model.

Once demographics were controlled for, the most significant driver for changes in the BLL was the preremediation BLL values. Across all of our models, an increase in 1 μg/dL in preremediation BLL is equated with an additional approximately 0.6 μg/dL decrease in overall postremediation BLL. We expect these numbers will increase as time passes since the remediation was completed. Our study assumes that the only major change in lead exposure were the pump remediation. Given the statistically significant decreases in BLL, it is reasonable to consider this assumption to be true. The lack of significance within our regression of preremediation aqueous lead concentrations highlights the two limitations within our pump sampling campaigns: (1) using a flushed, composite 15 L sample to represent true exposure and (2) our analysis of

Table 2. Multiple Linear Regression Analysis Summary with Changes in BLL Concentrations As the Response Variable

Variable	Model 1		Model 3		Model 4		Model 5		Model 6		Model 7	
	b	SE b	b	SE b	b	SE b	b	SE b	b	SE b	b	SE b
Pre-BLL	0.641[a]	0.07	0.649[a]	0.07	0.660[a]	0.071	0.669[a]	0.071	0.688[a]	0.074	0.695[a]	0.00
Age			−0.275	0.33								
Weight					−0.165	0.13			−0.145	0.134	−0.145	0.133
Preremediation Aqueous Pb							0.034	0.021	0.029	0.022	−0.599	0.55
Female									−0.452	0.993	−0.599	0.995
Days between remediation and Post-BLL test											−0.113	0.093
Intercept	−2.77[b]	0.912	−2.16	1.174	−1.19	1.57	−3.65[b]	1.046	−1.96	1.79	2.99	0.503
F	83.58[a]		41.84[a]		43.07[a]		44.64[a]		22.33[a]		18.38[a]	
Degrees of freedom	43		42		42		42		40		39	
Adj R^2	0.6524		0.6499		0.6566		0.6648		0.6598		0.6639	
N	45		45		45		45		45		45	

[a] $p < 0.001$ as highly significant. [b] $p < 0.01$ as great significance (there are no variables measuring between 0.01 and 0.05).

https://doi.org/10.1021/acs.est.3c03774
Environ. Sci. Technol. 2023, 57, 16309−16316

only dissolved aqueous concentrations. Both of these sampling techniques most likely underestimated lead exposure and not necessarily in a systematic manner; however, these sampling limitations are most likely present in other LMIC that experience similar lead exposure. Ongoing investigations by the research team have determined that particulate lead can account for over 30% of total lead in stagnation samples.[50]

Therefore, it is recommended that future studies account for total (aqueous and particulate) lead in *consumed* water samples. In depth analysis of all pump components by a portable X-ray fluorescence analyzer (pXRF) would allow for greater certainty that lead exposure from all above ground pump components is accounted for. Furthermore, participant home examinations including collecting readings of potential sources of exposure (e.g., soil, dust) are suggested. It is also recommended that future studies use a larger sample size to allow for more in-depth statistical analysis.

3.4. Impacts and Limitations. This study highlights the potential importance of reducing aqueous lead exposure via remediation of lead components in a water supply of an LMIC through the measurement of children blood levels and water lead concentrations (pre- and postremediation). Measurements showed only 4% of pumps had aqueous lead concentrations over the WHO drinking water guideline of 10 μg/L at the completion of the pump remediation campaign. These results are similar to those recorded on the larger pump sample size populations of our previous studies. Subsequently, the BLL of 87% of the children tested decreased and measured childhood BLL showed a statistically significant decrease after the removal of leaded components ($p < 0.001$, $n = 55$). Major limitations of the study include not having detailed dietary information for the participants within the study and the method of water sampling. While using a flushed, composite sample was the most practical methodology given the major logistical challenge of requiring residents to not use a water source to enable stagnation sampling, we realize it might not present the truest account of lead exposure for all residents. Future research should quantify consumed lead concentrations and actual intake rates for ingested water to link aqueous lead consumption with measured BLL. It was out of the scope of this study to track water collection patterns and associated consumption rates to quantify lead intake ratios from all sources. Therefore, we assumed no additional drastic changes to lead exposure occurred during the study and that lead exposure from water was significant. Given this assumption, the statistically significant reduction in BLL was most likely due to the decrease in aqueous exposure, even if a causal link is not possible at this time and despite aqueous lead concentration not being a statistically significant driver of lower BLL postremediation. Thus, small- and large-scale remediations can play an important role in decreasing exposure to lead, even by way of water exposure for residents in this study. The impact on BLL will depend on other sources of lead in the environment and participant diets if expanded to other LMIC. If it is also true in other locations that leaded components in pumps are a major source of lead exposure, then it is possible to have a significant return on investment for every dollar spent[30] remediating pumps. Though not a common lead source addressed in the LMIC, limiting aqueous lead exposure holds the ability to generate positive health benefits through economically feasible remediation efforts.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.3c03774.

> Details on the remediation process (S.1); breakdown of the study population including age, gender, and location (S.2); paired BLL and aqueous lead data for participants (S.3); and statistical and visual check for normality of data (S.4) (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Adaline M. Buerck − *College of Engineering, Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida 33620, United States;* Present Address: Adaline M. Buerck: Department of Environmental and Civil Engineering, Mercer University, 1501 Mercer University Dr., Macon, Georgia 31207, United States; orcid.org/0000-0002-4671-5285; Email: Buerck_am@mercer.edu

Authors

Mahmooda Khaliq − *College of Public Health, University of South Florida, Tampa, Florida 33612, United States;* orcid.org/0000-0002-1265-4795

Katherine Alfredo − *College of Engineering, Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida 33620, United States;* orcid.org/0000-0003-4392-9031

Jeffrey A. Cunningham − *College of Engineering, Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida 33620, United States;* orcid.org/0000-0001-9654-8262

Luke John Paul Barrett − *ONG Ranontsika, Ampasimazava Toamasina 501, Madagascar*

Rinah Rakotondrazaka − *ONG Ranontsika, Ampasimazava Toamasina 501, Madagascar*

Lova Rakotoarisoa − *ONG Ranontsika, Ampasimazava Toamasina 501, Madagascar*

Wyatt M. Champion − *College of Engineering, Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida 33620, United States;* orcid.org/0000-0003-3968-1811

James R. Mihelcic − *College of Engineering, Department of Civil and Environmental Engineering, University of South Florida, Tampa, Florida 33620, United States;* orcid.org/0000-0002-1736-9264

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.3c03774

Funding

This material has been supported a grant from the National Science Foundation under Grant No. 1735320 (Collaborative Research: NRT-INFEWS: Systems Training for Research on Geography-based Costal Food Energy Water Systems), by funding provided by Water Charities Fundraising, the Oak Foundation through Pure Earth, a University of South Florida Strategic Investment Pool (SIP) Award, and a U.S. Department of Education Graduate Assistance in Areas of National Need (GAANN) Fellowship (Project Number P200A180047) awarded to Adaline M. Buerck.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The Raman analysis of the leaded components in the foot and piston values was performed by the laboratory of Dr. Marc Edwards at Virginia Tech.

■ ABBREVIATIONS

BLL = blood lead level
LMIC = low- and middle-income countries
HIC = high-income countries
EPA = U.S. Environmental Protection Agency
IEUBK = integrated exposure uptake biokinetic model

■ REFERENCES

(1) Hayes, C., Hoekstra, E. J. *Lead in Drinking Water*; Technical Digest; Luxembourg, 2010.

(2) Mathee, A.; Street, R.; Teare, J.; Naicker, N. Lead exposure in the home environment: An overview of risks from cottage industries in Africa. *Neurotoxicology* **2020**, *81*, 34−39.

(3) Lead laws and regulations, 2020. *United States Environmental Protection Agency*. https://www.epa.gov/lead/lead-laws-and-regulations.

(4) Annest, J. L.; Pirkle, J. L.; Makuc, D.; Neese, J. W.; Bayse, D. D.; Kovar, M. G. Chronological trend in blood lead levels between 1976 and 1980. *N. Engl. J. Med.* **1983**, *308* (23), 1373−1377.

(5) Lead poisoning and health, 2019. *World Health Organization*. https://www.who.int/news-room/fact-sheets/detail/lead-poisoning-and-health.

(6) Ericson, B.; Hu, H.; Nash, E.; Ferraro, G.; Sinitsky, J.; Taylor, M. P. Blood lead levels in low-income and middle-income countries: A systematic review. *Lancet Planet Heal.* **2021**, *5*, E145−E153.

(7) Rees, N., Fuller, R., 2020. The toxic truth: Children's exposure to lead pollution undermines a generation of future potential.

(8) Kordas, K.; Ravenscroft, J.; Cao, Y.; McLean, E. V. Lead exposure in low and middle-income countries: Perspectives and lessons on patterns, injustices, economics, and politics. *Int. J. Environ. Res. Public Health* **2018**, *15*, 2351.

(9) Mahaffey, K. R. Nutrition and lead: Strategies for public health. *Environ. Health Perspect* **1995**, *103*, 191−196.

(10) *The Sustainable Development Goals Report 2019*; United Nations Publication, 2019.

(11) Edwards, M.; Triantafyllidou, S.; Best, D. Elevated blood lead in young children due to lead-contaminated drinking water: Washington, DC, 2001−2004. *Environ. Sci. Technol.* **2009**, *43*, 1618−1623.

(12) Akoumianaki, I. *Lead in Drinking Water: Public Health, Mitigation and Economic Perspectives*; James Hutton Institute and Crew Scotland's Centre of Expertise for Waters: Craigiebuckler, Scotland, UK, 2017.

(13) Roy, S.; Tang, M.; Edwards, M. A. Lead release to potable water during the Flint, Michigan water crisis as revealed by routine biosolids monitoring data. *Water Res.* **2019**, *160*, 475−483.

(14) Deshommes, E.; Prevost, 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*, 2409−2420.

(15) Levallois, P.; St-Laurent, J.; Gauvin, D.; Courteau, M.; Prévost, M.; Campagna, C.; Lemieux, F.; Nour, S.; D'Amour, M.; Rasmussen, P. E. The impact of drinking water, indoor dust and paint on blood lead levels of children aged 1−5 years in Montréal (Québec, Canada). *J. Expo. Sci. Environ. Epidemiol.* **2014**, *24*, 185−191.

(16) Hanna-Attisha, M.; LaChance, J.; Sadler, R. C.; Champney Schnepp, A. Elevated blood lead levels in children associated with the Flint drinking water crisis: A spatial analysis of risk and public health response. *Am. J. Public Health* **2016**, *106*, 283−290.

(17) Cartier, C.; Dore, E.; Laroche, L.; Nour, S.; Edwards, M.; Prevost, M. Impact of treatment on Pb release from full and partially replaced harvested Lead Service Lines (LSLs). *Water Res.* **2013**, *47* (2), 661−671.

(18) Bosscher, V.; Lytle, D. A.; Schock, M. R.; Porter, A.; Del Toral, M. POU water filters effectively reduce lead in drinking water: a demonstration field study in Flint, Michigan. *J. Environ. Sci. Heal. Part A* **2019**, *54*, 484−493.

(19) Pieper, K. J.; Katner, A.; Kriss, R.; Tang, M.; Edwards, M. A. Understanding lead in water and avoidance strategies : a United States perspective for informed decision-making. *J. Water Health* **2019**, *17*, 540−555.

(20) *Preventing Disease through Healthy Environments: Exposure to Lead: A Major Public Health Concern (2019 revision)*; World Health Organization: Geneva, 2019.

(21) Champion, W. M.; Buerck, A. M.; Nenninger, C.; Yusuf, K.; Barrett, L. J. P.; Rakotoarisoa, L.; Rakotondrazaka, R.; Alfredo, K.; Cunningham, J.; Khaliq, M.; Mihelcic, J. R. Childhood blood lead levels and environmental risk factors in Madagascar. *Environ. Sci. Pollut. Res.* **2022**, *29* (45), 68652−68665.

(22) Fisher, M. B.; Guo, A. Z.; Tracy, J. W.; Prasad, S. K.; Cronk, R. D.; Browning, E. G.; Liang, K. R.; Kelly, E. R.; Bartram, J. K. *Occurrence of lead and other toxic metals derived from drinking-water systems in three west african countries* **2021**, *129*, na.

(23) *Guidelines for Drinking-Water Quality*, 4th ed. incorporating the first addendum; World Health Organization: Geneva, 2011.

(24) Akers, D. B.; Maccarthy, M. F.; Cunningham, J. A.; Annis, J.; Mihelcic, J. R. Lead (Pb) contamination of self-supply groundwater systems in coastal Madagascar and predictions of blood lead levels in exposed children. *Environ. Sci. Technol.* **2015**, *49*, 2685−2693.

(25) Akers, D. B.; Buerck, A.; MacCarthy, M. F.; Cunningham, J. A.; Mihelcic, J. R. Estimates of blood lead levels (BLLs) for children in coastal madagascar: Accounting for dietary uptake of lead (Pb). *Expo. Heal.* **2020**, *12*, 501.

(26) Ryan, P. Madagascar WASH Sector Sustainability Check; UNICEF, 2014.

(27) Ryan, P., Carter, R. WASH Service Provision in Rural Madagascar: Sustainability Check 2; Government of Madagascar, 2016.

(28) MacCarthy, M. F.; Annis, J. E.; Mihelcic, J. R. Unsubsidised self-supply in eastern Madagascar. *Water Altern.* **2013**, *6*, 424−438.

(29) *Integrated Exposure Uptake Biokinetic Model for Lead in Children (IEUBK win v1.1 build 11)*; United States Environmental Protection Agency, 2010. https://www.epa.gov/superfund/lead-superfund-sites-software-and-users-manuals.

(30) Buerck, A. M.; Usowicz, M.; Cunningham, J. A.; Khaliq, M.; Barrett, L. J. P.; Rakotoarisoa, L.; Rakotondrazaka, R.; Alfredo, K.; Sommariva, S.; Mihelcic, J. R. Health and economic consequences of lead exposure associated with products and services provided by the informal economy. *Environ. Sci. Technol.* **2021**, *55*, 8362−8370.

(31) Buerck, A. M.; Khaliq, M.; Rakotondrazaka, R.; Rakotoarisoa, L.; Paul Barrett, L. J.; Sommariva, S.; Mihelcic, J. R. Convergence of Social Marketing and Engineering: A Lead Mitigation Study in Madagascar. *Social Marketing Quarterly* **2023**, *29* (1), 87−104.

(32) Biomonitoring summary, lead, 2017. *National Biomonitoring Program, Centers for Disease Control and Prevention*. https://www.cdc.gov/biomonitoring/Lead_BiomonitoringSummary.html.

(33) Magellan Diagnostics Recalls LeadCare II, LeadCare Plus, and LeadCare Ultra Blood Lead Tests Due to Risk of Falsely Low Results, 2021. *U.S. Food and Drug Administration*. https://www.fda.gov/medical-devices/medical-device-recalls/magellan-diagnostics-recalls-leadcare-ii-leadcare-plus-and-leadcare-ultra-blood-lead-tests-due-risk.

(34) LeadCare II Blood Lead Test Kit Package Insert, 2021. chrome-extension://efaidnbmnnnibpcajpcglclefindmkaj/https://www.magellandx.com/uploads/2021/03/80-0052-v1.09-LeadCare-II-Package-Insert-Rev-05.pdf.

(35) Desai, G.; Anzman-Frasca, S.; Vernarelli, J. A.; Ravenscroft, J.; Yang, J.; Burstein, G.; Kordas, K. Examining links between diet and lead exposure in young children: 2009 to 2014 national health and nutrition examination survey. *Acad. Pediatr.* **2021**, *21*, 471−479.

https://doi.org/10.1021/acs.est.3c03774
Environ. Sci. Technol. 2023, 57, 16309−16316

(36) CDC response to advisory committee on childhood lead poisoning prevention recommendations in "Low level lead exposure harms children: A renewed call of primary prevention, 2012. *ACCLPP*. https://stacks.cdc.gov/view/cdc/37586.

(37) Childhood Lead Poisoning Prevention, 2021. *National Center for Environmental Health, Division of Environmental Health Science and Practice, Centers for Disease Control and Prevention*. https://www.cdc.gov/nceh/lead/prevention/sources/water.htm.

(38) *Guidelines for Drinking-Water Quality*, second ed.; World Health Organization, 1993.

(39) Sandvig, A., Kwan, P., Kirmeyer, G., Maynard, B., Mast, D., Trussell, R. R., Trussell, S., Cantor, A., Prescott, A. *Contribution of Service Line and Plumbing Fixtures to Lead and Copper Rule Compliance Issues*; AWWA Research Foundation: Denver, CO, 2008.

(40) Brown, M. J.; Raymond, J.; Homa, D.; Kennedy, C.; Sinks, T. Association between children's blood lead levels, lead service lines, and water disinfection, Washington, DC, 1998–2006. *Environmental Research* **2011**, *111*, 67–74.

(41) Lee, R. G.; Becker, W. C.; Collins, D. W. Lead at the tap: Sources and control. *J. AWWA* **1989**, *81*, 52–62.

(42) Kimbrough, D. E. Brass corrosion as a source of lead and copper in traditional and all-plastic distribution systems. *J. AWWA* **2007**, *99*, 70–76.

(43) Clark, B. N.; Masters, S. V.; Edwards, M. A. Lead Release to Drinking water from galvanized steel pipe coatings. *Environ. Eng. Sci.* **2015**, *32*, 713–721.

(44) Li, M.; Liu, Z.; Chen, Y.; Korshin, G. V. Effects of varying temperatures and alkalinities on the corrosion and heavy metal release from low-lead galvanized steel. *Environ. Sci. Pollut. Res.* **2020**, *27*, 2412–2422.

(45) Mcfadden, M.; Giani, R.; Kwan, P.; Reiber, S. H. Contributions to drinking water lead from galvanized iron corrosion scales. *J. AWWA* **2011**, *103*, 76–89.

(46) *Guidelines for Canadian Drinking Water Quality: Guideline Technical Document – Lead*; Catalogue No. H144-13/11-2018E-PDF; Water and Air Quality Bureau, Healthy Environments and Consumer Safety Branch, Health Canada: Ottawa, Ontario, 2019.

(47) Shah, N. Understanding the Leaching Mechanism for Lead (Pb) Found in Components of Locally Manufactured Handpumps in Eastern Madagascar. USF Tampa Graduate Theses and Dissertations, 2021. https://digitalcommons.usf.edu/etd/8865.

(48) Specht, A. J.; Weisskopf, M.; Nie, L. H. Childhood lead biokinetics and associations with age among a group of lead-poisoned children in China. *J. Expo Sci. Environ. Epidemiol.* **2019**, *29* (3), 416–423.

(49) Roberts, J. R.; Roberts, J.; Reigart, J. R.; Ebeling, M.; Hulsey, T. C. Time required for blood lead levels to decline in nonchelated children. *Journal of Toxicology: Clinical Toxicology* **2001**, *39* (2), 153–160.

(50) Wilson, M. Laboratory Examination of Lead Weights Harvested from Pitcher Pumps in Eastern Madagascar. USF Tampa Graduate Theses and Dissertations, 2021. https://digitalcommons.usf.edu/etd/9265.

https://doi.org/10.1021/acs.est.3c03774
Environ. Sci. Technol. 2023, 57, 16309–16316