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Article

Modeled Impacts of Drinking Water Pb Reduction Scenarios on Children's Exposures and Blood Lead Levels

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ABSTRACT: In recent years, environmental lead (Pb) exposure through drinking water has resulted in community public health concerns. To understand potential impacts on blood Pb levels (BLLs) from drinking water Pb reduction actions (i.e., combinations of lead service lines [LSL] and corrosion control treatment [CCT] scenarios), EPA's Stochastic Human Exposure and Dose Simulation (SHEDS)-Multimedia/Integrated Exposure Uptake and Biokinetic (IEUBK) model was applied for U.S. children aged 0 to <6 years. The results utilizing a large drinking water sequential sampling data set from 15 cities to estimate model input concentration distributions demonstrated lowest predicted BLLs for the "no LSLs" with "combined CCT" scenario and highest predicted BLLs for the "yes LSLs" and "no CCT" scenario. Modeled contribution to BLLs from ingestion of residential drinking water ranged from ∼10 to 80%, with the highest estimated for formula-fed infants (age 0 to <1 year). Further analysis using a "bounding" data set spanning a range of realistic water Pb concentrations and variabilities showed BLL predictions consistent with the sequential sampling-derived inputs. Our study illustrates (1) effectiveness of LSL replacement coupled with CCT for reducing Pb in drinking water and children's BLLs, and (2) in some age groups, under realistic local and residential water use conditions, drinking water can be the dominant exposure pathway.

■ INTRODUCTION

High-profile drinking water distribution system chemistry upsets, like those that occurred in the District of Columbia (2003)[1,2] and Flint, Michigan (2016),[3−5] brought attention to potential lead (Pb) exposure and health risks posed to children consuming drinking water from these systems. But beyond the obvious highly visible Pb in water contamination events, a 2017 report on reducing childhood Pb exposure[6] listed full lead service line (LSL) replacement and improved corrosion control treatment (CCT) requirements for drinking water as recommended Pb exposure reduction actions for children.

Any households with sources of Pb-containing plumbing materials (e.g., Pb service lines, leaded solders, brasses, and accumulation on pipe scale materials) and corrosive drinking water quality characteristics (e.g., low pH, high salt content, and lack of or inadequate CCT) in some or all of the drinking water system are at some level of risk. Changes in the solubility of pipe scales, their physical characteristics, or both, could result in a significant release of Pb to water and subsequent direct and indirect waterborne exposure.[7] Physical disturbances can also cause the release of Pb from Pb-containing pipe scales or newly exposed surfaces.[8]

There are many physical and chemical factors that impact the concentration of Pb in drinking water coming out of a tap.[9−11] When present, Pb piping (such as service lines) comprises the highest potential reservoir for Pb release into the water at those sites.[8,12−15] Many laboratory and field studies have been published on the equilibrium and nonequilibrium relationships among varying background and treatment-related water quality parameters, including corrosion control inhibitors (such as orthophosphate, blended phosphates, or silicates), the rate and processes of Pb leaching from new Pb pipes, the solubility of pipe scales on Pb release, and some factors such as duration of stagnation in the plumbing and oxidant demand.[9−11,15−22] These studies help our understanding of chemistry in the pipes, but they were not designed to represent tap water Pb concentrations ingested by humans.

Because the sources of Pb in municipal drinking water are not homogeneously distributed along the water mains, service lines, and premise plumbing to the consumers' taps, the instantaneous concentration of Pb coming out of the tap will

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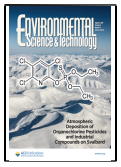

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continuously vary, from the minimum level observed with prolonged flushing to maximum levels that are a nonlinear combination of soluble Pb released during stagnation as equilibrium levels are ultimately achieved and particulate Pb existing as stable colloidal material or eroded from the pipe scales during the water sampling event itself. The maximum levels are a one-time "snapshot" that depend on many factors including the exact location of the Pb sources, the plumbing configuration of the building/dwelling, the extent of the particulate release, stagnation time, and the pattern of water usage at all water-using outlets in that building, relative to the time a sample is taken from a given tap. This may represent the worst-case potential exposure at a given time and tap within a residence but will not properly represent exposure at that tap over time or by individuals. Conversely, the minimum levels, typically the "fully flushed" samples, may represent the "best case" minimum soluble Pb concentration snapshot, but the exposure will fall somewhere between the two boundaries at any given time. Thus, characterizing Pb levels from household drinking water is highly complex.[19,23−31] Any single water sample will be unable to estimate the exposure at that house or any other residences in that water system.

There is a lack of epidemiologic studies focused on drinking water exposure and children's blood lead levels (BLL) that adequately characterize the variability of Pb in household drinking water. In addition, very few Pb modeling exposure analyses with water ingestion as the main exposure pathway have been conducted[32−36] and most studies to date have almost universally used various single-sample inputs for drinking water concentration including first-draw samples,[33,34] flushed samples,[34,35,37] and the combination of flushing followed by stagnation.[36,38]

In this study, a Pb concentration data set based on sequential profile sampling (i.e., water samples collected immediately one after the other to capture the variation of the concentration of Pb from the tap of interest to the water main[8,10,12,15,18,19,24,25,30] using consistent or varying volumes of water) data from taps in 15 cities in the U.S. and Canada (Tables S1−S3) was used to develop estimates of household drinking water concentrations. These drinking water concentrations (i.e., "sequential sampling data set") then served as inputs for the EPA's Stochastic Human Exposure and Dose Simulation (SHEDS)-Multimedia model and Integrated Exposure Uptake and Biokinetic (IEUBK) modeling system[39] to better estimate potential exposures and resultant BLLs for children aged 0 to <6 years under various scenarios. These scenarios were comprised of different combinations of the presence or absence of lead service lines (LSLs) and corrosion control treatment (CCT) status.

We conducted a second modeling analysis using a range of potential water Pb concentrations along with a matrix of variabilities (i.e., "bounding data set"). The purpose of this additional analysis was to establish a range of possible BLLs associated with a realistic range of drinking water Pb concentrations to provide insight on model sensitivity for drinking water Pb inputs.

Together, these two approaches improve the understanding of how drinking water could contribute to children's Pb exposures. The study also sheds light on how Pb exposures and resultant BLLs could be reduced through changing drinking water chemistry and removing the sources of Pb in premise plumbing and the distribution networks.

■ MATERIALS AND METHODS

SHEDS-IEUBK Modeling Approach. The SHEDS-Multimedia model is a probabilistic Monte Carlo exposure model that simulates aggregate or cumulative exposures over time from dietary and residential pathways using observations from human activity diaries, exposure factors, and chemical concentrations in multiple environmental media.[40−42] EPA's IEUBK model predicts childhood BLLs based on an exposure scenario that is provided through specific Pb concentrations and exposure factors used as inputs to the model.[43−45] For the exposure and BLL modeling analysis, the SHEDS-IEUBK modeling approach[39] was used with water Pb concentration distributional inputs described in Tables 1 and 2. For all other Pb media concentrations and exposure inputs, national-scale data were used, consistent with Zartarian et al.[39] Briefly, bioavailability values were provided from the IEUBK model defaults of 30% for soil and dust and 50% for water and food. Simulations were conducted using a 30-day averaging time, with a simulated population sample size of 3000 for each scenario and age group.

Table 1. Lead Service Line (LSL) and Corrosion Control Treatment (CCT) Scenarios and Corresponding Water Pb Concentrations Used as SHEDS-IEUBK Modeling Inputs for the Sequential Sampling Data Set

LSL:CCT scenario	Simulated Geometric Mean (Pb μg/L)	Simulated Geometric SD
Yes LSL:No CCT	18.08	3.78
Yes LSL:Some CCT	9.92	3.78
Yes LSL:Representative CCT	5.48	3.77
Partial LSL:No CCT	8.43	3.77
Partial LSL:Some CCT	4.72	3.75
Partial LSL:Representative CCT	2.64	3.76
No LSL:Combined CCT	0.82	3.86

Daily water consumption values used in SHEDS-IEUBK modeling were obtained from NHANES 2005−2012 data (Table S6). These values represent both direct and indirect (e.g., coffee, tea, infant formula, and food processing) drinking water intake. Consumption of bottled water was not included. Within the SHEDS-Multimedia model, for any modeled individual on a given day, one person with drinking water information is randomly selected from the NHANES data pool based on similar socioeconomic characteristics (i.e., age, gender), season, and day of the week (i.e., weekday or weekend).[41] For this application, 30 days of Pb exposure was simulated to link with the IEUBK model, rather than simulating exposure on a yearly basis. Thus, we modified the 8-diary longitudinal algorithm in SHEDS-Multimedia used to simulate annual exposure,[46] by ignoring season and using four personal days rather than eight, for each person. To obtain Pb drinking water exposure for a simulated day and person, total drinking water consumed for a given day is multiplied by a single water Pb concentration that is randomly selected from the Monte Carlo simulation.

The results graphically displayed in the subsequent section used standard deviations of water Pb concentrations reflecting among-sampling event data variability. Additional analysis to evaluate the sensitivity of the inputs was conducted to reflect simulated "among-site" variability and "among-city" variability (Tables S7 and S8).

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Environ. Sci. Technol. 2020, 54, 9474−9482

Table 2. Simulated Geometric Mean and Geometric Standard Deviation Modeling Inputs for the Bounding Data Set

	Standard Deviation				
Water Pb Concentration (μg/L)	20%	50%	75%	100%	200%
1	(0.98 ± 1.22)	(0.89 ± 1.60)	(0.80 ± 1.95)	(0.71 ± 2.30)	(0.45 ± 3.56)
2	(1.96 ± 1.22)	(1.79 ± 1.60)	(1.60 ± 1.95)	(1.41 ± 2.30)	(0.89 ± 3.56)
3	(2.94 ± 1.22)	(2.68 ± 1.60)	(2.40 ± 1.95)	(2.12 ± 2.30)	(1.34 ± 3.56)
4	(3.92 ± 1.22)	(3.58 ± 1.60)	(3.20 ± 1.95)	(2.83 ± 2.30)	(1.79 ± 3.56)
6	(5.88 ± 1.22)	(5.37 ± 1.60)	(4.80 ± 1.95)	(4.24 ± 2.30)	(2.68 ± 3.56)
8	(7.84 ± 1.22)	(7.16 ± 1.60)	(6.40 ± 1.95)	(5.66 ± 2.30)	(3.58 ± 3.56)
10	(9.81 ± 1.22)	(8.94 ± 1.60)	(8.00 ± 1.95)	(7.07 ± 2.30)	(4.47 ± 3.56)
15	(14.71 ± 1.22)	(13.42 ± 1.60)	(12.00 ± 1.95)	(10.61 ± 2.30)	(6.71 ± 3.56)
20	(19.61 ± 1.22)	(17.89 ± 1.60)	(16.00 ± 1.95)	(14.14 ± 2.30)	(8.94 ± 3.56)
25	(24.51 ± 1.22)	(22.36 ± 1.60)	(20.00 ± 1.95)	(17.68 ± 2.30)	(11.18 ± 3.56)
30	(29.42 ± 1.22)	(26.83 ± 1.60)	(24.00 ± 1.95)	(21.21 ± 2.30)	(13.42 ± 3.56)
40	(39.22 ± 1.22)	(35.78 ± 1.60)	(32.00 ± 1.95)	(28.28 ± 2.30)	(17.89 ± 3.56)

Sequential Sampling Data Set Analysis. For the sequential sampling analysis, the effects of two explanatory variables on concentrations of Pb in drinking water were considered: the presence of LSLs and CCT status. To estimate drinking water Pb concentrations at the tap under each scenario, available sequential tap water sampling data were solicited and obtained from utilities, EPA branch offices, and authors of published journal articles. These data represented 18 039 samples from 3,102 sequential sampling events from 1690 sites in 15 cities across the U.S. and Canada (Tables S1−S3). The number of sequential samples per sampling event varied from 1 to 26. Data included Pb concentrations and information regarding LSL status, location, and date of sample collected between 1998 and 2016. The data set contained samples from eight cities in the northern Midwest, mid-Atlantic, and northeastern U.S. and seven cities in southern Canada; most cities sampled had populations between 100 000 and one million (Table S1). Most data sources contained a series of samples, or profiles, taken after periods of stagnation of varying length, with identifiers for sampling volume and position in series ("profile liter"). Corrosion control status was defined using records of CCT practices and implementation dates, as well as data obtained from water quality samples (Tables S2 and S3). Because these measurements were collected for different purposes and obtained using several sampling designs over many years, a statistical model was developed to account for this variation.

Multiple, nested, mixed-effects models were used (Supporting Information Eqs 1−5 and Table S5) to log-transform the measured Pb concentration with predictor variables for LSL presence or absence, CCT, and profile liter, the cumulative volume interval a sample represented within a sampling series and included nested random effects of the city, site, and sampling event. Using the fitted model and the original data, the data set was simulated, then the geometric mean and standard deviation of simulated Pb concentrations in water after their respective stagnation intervals obtained at a point analogous to the fifth liter taken from the tap were calculated. This was based on the fitted regression and bootstrapped 95% confidence intervals that showed the highest Pb concentrations after the last stagnation period occurring roughly 5 liters from the sampling tap in homes with LSLs (Figure S1). We fitted models and produced simulated data sets using R statistical software (R Core Team 2016) with the *lme*4 package.[47] Details of this analysis are in the Supporting Information.

Seven combinations of LSL and CCT scenarios produced statistically distinct predictions (see the Supporting Information) of drinking water concentrations for use in SHEDS-IEUBK modeling. Table 1 shows the seven scenarios and corresponding estimated geometric mean and geometric standard deviation drinking water Pb concentrations used as inputs in SHEDS-IEUBK. For LSL scenarios, "yes" indicates Pb service lines; "partial" indicates some presence of Pb in service lines (some portion of the LSL remains after a section of the line was removed); and "no" indicates no Pb in service lines. For CCT scenarios, no indicates no CCT, "some" represents systems that have some CCT in place, but not optimized, and "representative" indicates a water chemistry that exemplifies the best CCT currently in use (which can include some combination of higher phosphate values and/or optimized pH levels). The simulated predictions overlapped completely for all CCT scenarios in homes with no LSL (see the Supporting Information), so "combined" indicates pooled CCT estimates representing all three states of CCT in these cases. This is not meant to imply that benefits would not be gained under actual circumstances with implementation of CCT in homes without LSLs.

Bounding Data Set Analysis. The second approach for deriving water Pb concentration for inputs into SHEDS-IEUBK was based on decades of US EPA Office of Research and Development research and sampling to identify Pb sources or characterize corrosion control effectiveness in drinking water systems across highly different water chemistries and treatments, as well as in differently configured houses and buildings. This allowed bounds to be placed around the range of Pb levels that water consumers would be likely to ingest. Therefore, a "bounding" data set was generated, consisting of a matrix of arithmetic mean Pb concentrations and their potential standard deviations to cover the expected range of residential drinking water Pb concentrations and their variability.

Water Pb concentrations from 1 to 40 μg/L were used, for a total of 12 concentrations, and for each water Pb concentration, five standard deviations, 20, 50, 75, 100, and 200%, were evaluated (Table 2). Lognormal distributions were assumed, thus the values in the table were what was used as SHEDS-IEUBK model inputs. Sixty total runs were conducted for each age group to evaluate BLL and the contribution of water Pb to overall BLL.

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Environ. Sci. Technol. 2020, 54, 9474−9482

RESULTS AND DISCUSSION

Sequential Sampling Data Set Modeling Analysis. Figure 1 shows the SHEDS-IEUBK modeling results of the average and geometric mean BLL estimates for ages 0 to <6 years combined for the seven LSL:CCT scenarios. The scenario of yes LSL:no CCT resulted in the highest BLL for all age groups, with the no LSL:combined CCT scenario having the lowest BLLs for all age groups. This is as expected, since the yes LSL:no CCT scenario is comprised of LSL and no CCT and has the highest water Pb concentrations. The opposite is true for the no LSL:combined CCT scenario. The BLLs for all of the other scenarios fall in between yes LSL:no CCT and no LSL:combined CCT. When a *t*-test was applied to compare either the arithmetic or geometric means between any two given scenarios, the differences between all scenarios were highly statistically significant ($p < 0.001$). Given that the standard output for BLL from the IEUBK model is geometric mean, this is generally what is presented in the figures and tables that follow. The Supporting Information provides additional results.

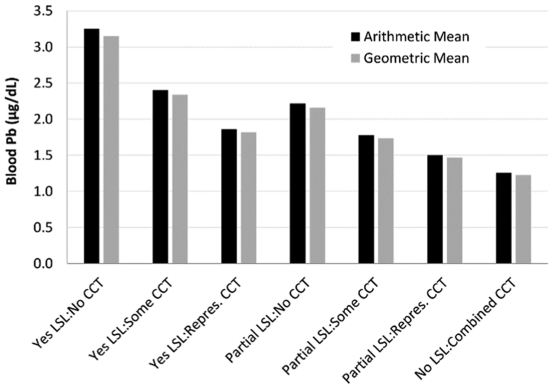

Figure 1. Arithmetic mean and geometric mean BLL predicted by SHEDS-IEUBK for ages 0 to <6 years by the LSL:CCT scenario.

As shown in Figure 2, the highest geometric mean BLLs among all scenarios and age groups were from the yes LSL:no CCT scenario for age 0 to <1 year (3.6 μg/dL). These results can be generally explained by both the water consumption values assumed for formula feeding combined with the highest water Pb concentrations for the youngest age category (i.e., 0 to <1 years). Predictions of the geometric mean BLLs for ages 1 through <6 years were lower than that observed for age 0 for all scenarios except no LSL:combined CCT. For the no LSL:combined CCT scenario, BLL slightly increases from age 0 to <1 to age 1 to <2. The range in the BLL geometric means between ages for a given scenario was lowest for the no LSL:combined CCT scenario (0.2 μg/dL) and greatest for the yes LSL:no CCT scenario (1.1 μg/dL). A similar pattern emerged for all scenarios with relatively consistent BLLs across ages 1 through <6 years. Within a given age group looking across scenarios, the range in 95th percentile BLL was greatest for age 0 to <1 years (9.0 μg/dL) and the least for age 4 to <5 years (3.8 μg/dL) (data not shown).

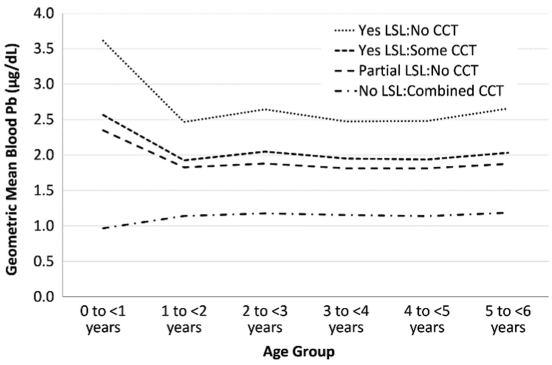

Figure 2. Geometric mean BLL predicted by SHEDS-IEUBK by age and LSL:CCT scenario.

The contribution of Pb exposure from major pathways to estimated BLL is shown in Figure 3. These pathways included food ingestion, nondietary (dust and soil) ingestion, inhalation, and water ingestion. The focus is on age 0 to <1 year (infants), as generally the highest BLLs were observed for this age group due to the assumption that most of the ingestion for children <6 months was formula made with drinking water. For drinking water scenarios that characterize larger Pb source material (LSLs) and less use of corrosion inhibitors (CCT), the water ingestion pathway was the major contributor to BLL, especially at the highest BLL percentiles (i.e., 50th percentile and above; Figures 3A and S3). Total Pb exposure approaches 10 μg/day around the 60th percentile for the yes LSL:no CCT scenario. At the highest BLLs for the yes LSL:no CCT scenario, total Pb exposure was over 44 μg/day, with water ingestion comprising 96% of the total contribution under this scenario. The scenarios for which there was less Pb present in drinking water resulted in total Pb exposures around 25 μg/day for the uppermost percentile of BLLs (Figure S3). Under the no LSL:combined CCT scenario, where water Pb

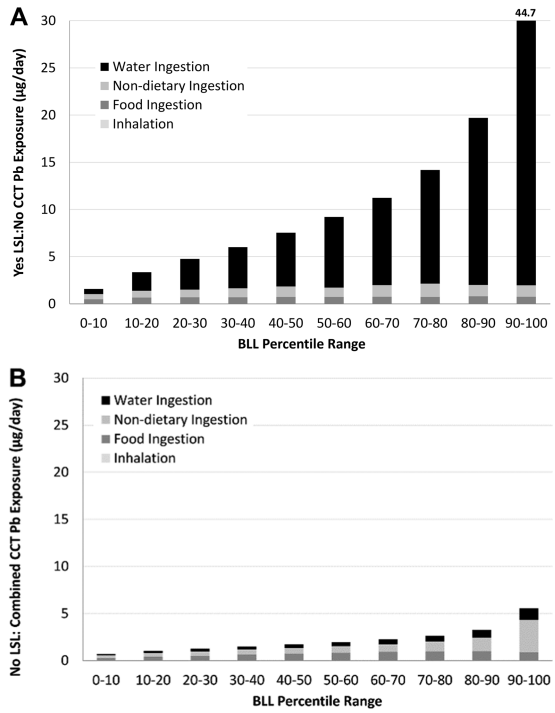

Figure 3. Contribution of Pb exposure to BLL from exposure pathways as predicted by SHEDS-IEUBK by yes LSL:no CCT (A) and no LSL:combined CCT (B) scenarios for age 0 to <1 years. Note that the inhalation pathway was an insignificant contributor to total Pb exposure under these scenarios.

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concentrations are much lower than the other scenarios, soil and dust ingestion was the most important contributor to total Pb exposure, especially for the upper percentiles (i.e., highest BLL), as shown in Figure 3B. For all percentiles in this scenario, the water ingestion pathway consistently contributed approximately 20% to total Pb exposure. The modeled estimates showed that the inhalation pathway was an insignificant contributor to total Pb exposure at the national scale.

Zartarian et al.[39] also reported a national-scale analysis using SHEDS-IEUBK modeling, but focused on different maximum daily average household tap water Pb concentrations that could keep children's BLLs below specified values, rather than community-specific drinking water Pb concentration profiles used in this current analysis. As noted in that paper, "the household tap water monitoring scheme is a factor that influences estimated drinking water Pb concentrations and related exposures" and "contributions from pathways are highly dependent on scenarios being considered". The relative exposure pathway contribution bar charts differ in the two analyses due to different drinking water Pb concentration scenarios and concentration profiles. Zartarian et al.[39] reported, "Analyses revealed relative importance of soil and dust ingestion exposure pathways and associated Pb intake rates; water ingestion was also a main pathway, especially for infants". In this follow-up paper, model analyses show that water ingestion can be the dominant pathway if local community drinking water Pb concentrations are elevated (e.g., a geometric mean of 11.3 μg/L or greater).

Bounding Data Set Modeling Analysis. As expected, with increasing water Pb concentrations from the bounding analysis, increasing BLLs were predicted for all age groups. Focusing specifically on ages 0 to <1 year, the impact of differing standard deviations for a given water Pb concentration on geometric mean BLLs was minimal (Figure S4). When examining the 95th percentile for BLL, the standard deviation of water Pb concentrations was of greater influence. For example, at 30 μg/L, the predicted BLL was 7.6 and 9.1 μg/dL for standard deviations of 20 and 200%, respectively. Furthermore, the higher the standard deviation for a given water Pb concentration, the higher the contribution from the water pathway for the 95th percentile BLL (Figure S4).

This analysis is valuable in demonstrating the relative impact of Pb from drinking water to BLLs and can be expanded to show the potential benefit of CCT, LSL removal, and other strategies to reduce Pb in drinking water. The changes in BLL for each 1 μg/L change in drinking water concentration under the 75th standard deviation case were calculated as 0.11 μg/dL for the 50th percentile BLL and 0.20 μg/dL for the 95th percentile BLL. These relationships were used to illustrate the change in BLLs for a given percent reduction in drinking water Pb concentration as a function of initial drinking water concentration for 50th and 95th percentile BLLs. For example, if a utility that has an average concentration of 20 μg/L and sets a goal to reduce the average Pb level to 10 μg/L (50% reduction), these data suggest that the 50th and 95th percentile BLL will decrease by approximately 1.0 μg/dL (Figure 4A) and 2.0 μg/dL (Figure 4B), respectively. These results are in the range of the sequential sampling data set analysis discussed above. If approximately 80% of the Pb entering the house with an LSL comes from the LSL itself, a large reduction in average water Pb concentration can be achieved by full LSL removal. For example, if an LSL is removed for a house that regularly has an average Pb concentration of 20 μg/L, the final Pb concentration may be in the range of 4 μg/L, and the reduction in BLL could be estimated to be 1.8 and 3.2 μg/dL, for the 50th and 95th percentile examples, respectively.

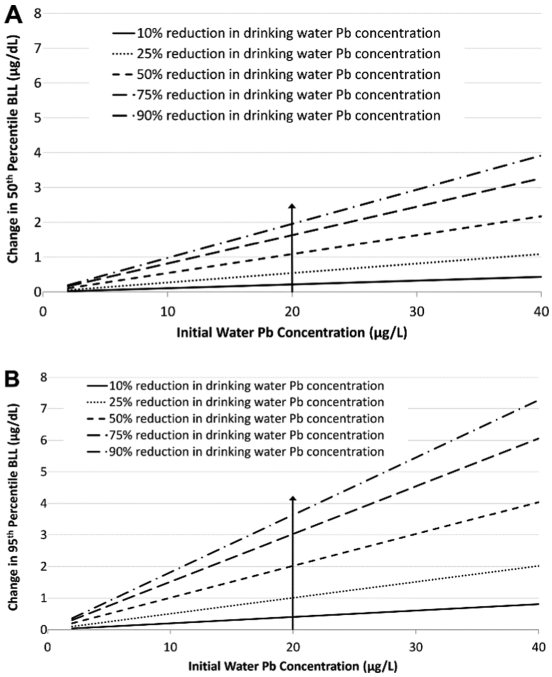

Figure 4. Change in 50th (A) and 95th (B) BLL percentiles by drinking water concentration for the bounding data set 75% standard deviation case for various drinking water reduction scenarios.

Table 3 shows the comparison between BLL modeling results from the sequential sampling data set analysis and the bounding data set analysis with select water Pb concentrations and standard deviations for age 0 to <1 year. Predicted BLLs were comparable between the no LSL:combined CCT scenario and bounding data set results for 2 μg/L water Pb concentration and a standard deviation of 200%. Predicted BLLs were comparable between the yes LSL:no CCT scenario and the bounding data set results for 40 μg/L water Pb concentration and a standard deviation of 200%.

This study provides valuable new exposure and health impact data, which relates residential drinking water Pb concentration explanatory variables to the presence/absence of Pb sources in the drinking water distribution system (e.g., LSL) and water chemistry through CCT. A recent study in Canada used multiple water sampling techniques and multimedia assessments to show that even relatively low water Pb concentrations could cause statistically significant measurable increases in BLLs in infants and small children.[37,48] Despite the uncertainties and limitations in this study, there are two potential public health benefits of controlling Pb release from drinking water. First, drinking water can contribute a large fraction to total Pb exposure, especially to infants and children. Second, there is a substantial benefit to the full removal of LSLs, in terms of reduction of Pb exposure risk (both dissolved and particulate).[6,20,49,50]

As demonstrated in the two approaches for deriving drinking water Pb concentrations, there can be high variability in the measured samples. The scenarios from the sequential sampling analyses aligned well with the bounding modeling results,

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Environ. Sci. Technol. 2020, 54, 9474−9482

Table 3. Comparison Between Sequential Sampling Data Set and Bounding Data Set Water Pb Concentrations and the BLL Modeling Results for Age 0 to <1 years

Approach	Scenario	Geometirc Mean Water Pb (μg/L)	GSD water Pb	Arithmetic Mean BLL (μg/dL)	SD BLL	50th percentile BLL (μg/dL)	75th percentile BLL (μg/dL)	95th percentile BLL (μg/dL)	99th percentile BLL (μg/dL)	Geometric Mean BLL (μg/dL)	GSD BLL
Sequential Sampling	No LSL: Combined CCT	0.82	3.86	1.13	0.70	0.98	1.36	2.41	3.58	0.97	1.74
Bounding	Water Pb = 2 μg/L, SD = 200%	0.89	3.56	1.14	0.76	0.98	1.36	2.44	4.06	0.97	1.75
Sequential Sampling	Yes LSL:No CCT	18.08	3.78	4.72	3.61	3.89	5.92	11.44	18.45	3.61	2.17
Bounding	Water Pb = 40 μg/L, SD = 200%	17.89	3.56	4.68	3.39	3.86	6.14	11.49	15.87	3.59	2.18

providing confidence that the projections from the bounding analysis are a reasonable representation of real-world exposure conditions. The bounding data set allowed for a comprehensive set of simulations using the SHEDS-IEUBK model that provided a range of water Pb concentration information that could be utilized for prioritization of protective actions (such as full LSL replacement or CCT optimization) to potentially reduce children's BLL to a target value. Better exposure-estimation sampling in the future can make the predicted relationships of water Pb concentrations to BLL impacts more precise and accurate.

The relative contribution of exposure pathway to BLLs is highly sensitive to drinking water Pb concentration inputs in both the sequential sampling and bounding data set analyses. Drinking water ingestion dominates in comparison to other exposure pathways for the highest water Pb concentrations (e.g., with LSL and minimal CCT scenarios). Significantly, over the range of input concentrations, representing plausible drinking water Pb exposures, the contribution to BLLs from residential drinking water ranges from approximately 10 to 80%. This is higher than has generally been concluded in many papers, indicating that the science to date has typically under-represented the contribution of drinking water concentrations to children's Pb exposure.

There are limitations and uncertainties associated with this national-scale analysis related to the SHEDS-IEUBK modeling approach. For the youngest children (aged 0 to <6 months), empirical BLL data used in the evaluation of the IEUBK model[45] and SHEDS-IEUBK model[39] was limited. To overcome uncertainties for this lowest age category, ages 0 to <1 year were grouped together. This analysis accounted for residential sources but did not account for other places where children can spend a significant amount of time (e.g., schools and daycare centers). Drinking water from schools and daycares could be a major contributor to Pb exposure.[36] In addition, this analysis did not consider dietary ingestion of food cooked with drinking water containing Pb, which could be particularly important for populations that consume starch food staples.[51] To be consistent with the modeling inputs used for the SHEDS-IEUBK model evaluation, the same inputs were used to represent residential housing and the general U.S. population, except for drinking water Pb concentrations. It is recognized that there are likely uncertainties in some inputs, including soil Pb concentration, dust Pb concentration, soil/dust ingestion, and human activity patterns (including household plumbing configuration and water consumption). However, since these inputs were consistent across the four scenarios, it supports that the differences in predicted BLLs by scenario would be due to differences in water Pb concentration.

Limitations as to the national representativeness of the sequential sampling analysis approach include: (1) data derived from a small fraction of the utilities in North America that must comply with Pb regulations (approximately 68 000 out of 146 000 total public water systems in the U.S. as of 2019 and approximately 2200 water treatment facilities owned by municipal and regional governments in Canada as of 2016); (2) utilization of differing sampling techniques (e.g., differing stagnation times, differing number of samples in profiling, different flow rates, etc.) that made equivalent comparisons difficult; (3) lack of evaluation of piping configurations and water usage (length of service lines, type of plumbing materials and piping lines, patterns of daily use, etc.) in the sampled housing stock; (4) limitations on data for differing source water chemistries, drinking water treatment practices, CCT practices, or the mineralogy of Pb scales across North America; and (5) potential over-representation of utilities that have done large studies.

Analysis of the profile liter data set suggested that first, Pb concentrations came largely from LSLs when they were present and, additionally, that Pb concentrations in homes with LSLs peak when water from the LSL passes through the tap. The first liter collected after stagnation without any prior flushing has historically been used by U.S. water systems to identify water quality issues. Collecting additional samples after stagnation would likely improve the identification of water chemistry issues that lead to high Pb concentrations. Based on our sequential sampling data set, the fifth liter on average may represent water that resided in the service line during the stagnation period, although the peak Pb concentration for any given residence will vary based on plumbing characteristics.

While the specific field observation data set used in this study was comprised only of large water systems, the results of this study are applicable to water systems of all sizes, including private supplies. The same variables apply at the individual house level in small systems as large systems. The water quality, plumbing layout, plumbing materials, and water usage patterns that impact exposure are the same, regardless of the system size. For smaller systems, the consistency and sophistication of CCT to reduce and control Pb release, which often requires the removal of interfering substances and implementing the addition of one or more chemicals to achieve precise water chemistry targets, will be much more economically and physically challenging.[10,11] Thus, the removal of LSL and other Pb sources may be much more advantageous for smaller water systems and private water supplies.

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Environ. Sci. Technol. 2020, 54, 9474−9482

For larger water systems, occasional interruptions of service caused by natural disasters, and ongoing need for changes in the overall treatment, are inevitable. Thus, fluctuations in water chemistry will trigger a risk of elevated exposure to short-term Pb release that will not necessarily be detected by monitoring programs. Permanently removing exposed Pb surfaces that are known to consist of pipe scales that range from a few percent by weight, to nearly ninety percent by weight,[9,15,21,22] adds a considerable measure for preventing Pb exposure from drinking water.

Private water supplies are rarely able to apply any complicated treatment beyond some sort of filtration. Because they are not covered by state, Federal, or provincial water corrosivity standards, they are more likely to have water corrosiveness that exceeds the range of community water systems,[52,53] as well as having a Pb in drinking water risk that may challenge the limits of plumbing materials standards[54] (such as NSF/ANSI/CAN 61).[55]

Since most drinking water sampling protocols are based on regulatory sampling objectives and are not designed to reflect human exposures from drinking water consumption, there would be great value in substituting proportional or composite sampling protocols or developing novel sampling approaches that are better designed to characterize Pb ingestion. Sampling research could focus on refining existing general protocols, such as proportional sampling or manual composite sampling for better implementation and precision for individuals most at risk, or it could investigate new technologies to obtain integrated measurement and data logging of metal concentrations coming out of the tap over time, as a function of water use to better approximate exposure.

Overall, there is a need for additional research that better pairs concurrent Pb tap sample concentrations with household (and other relevant) consumption patterns and BLLs to provide better estimates of human exposure. Data collection efforts are also needed to account for variations in geographic location and extent of leaded materials in the service line and premise plumbing/building water delivery systems. Future studies should incorporate the most appropriate drinking water sampling protocols for the intended research question, better information on household Pb sources and plumbing configurations, and blood lead measurements to enable accurate Pb exposure characterization.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c00479.

> Drinking water sampling approaches, sequential sampling data set characteristics and derivation of scenario concentrations, results from the sensitivity analyses, and additional figures of the blood lead modeling results using SHEDS-IEUBK (PDF)

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Notes

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