



Evaluation of exposure to lead from drinking water in large buildings



Elise Deshommès^{a,*}, Robert C. Andrews^b, Graham Gagnon^c, Tim McCluskey^d,
Brad McIlwain^c, Evelyne Doré^a, Shokoufeh Nour^a, Michèle Prévost^e

^a Polytechnique Montreal, 2900 Boulevard Edouard-Montpetit, Montreal, QC, H3T 1J4, Canada

^b University of Toronto, 35 St George Street, Toronto, ON, M5S 1A4, Canada

^c Dalhousie University, 1360, Barrington St., Halifax, NS, B3H 4R2, Canada

^d Department of Education and Early Childhood Development, 2450 King St., PO Box 6000, Fredericton, NB, E3B 5H1, Canada

^e Polytechnique Montreal, NSERC Industrial Chair in Drinking Water, 2900 Boulevard Edouard-Montpetit, Montreal, QC, H3T 1J4, Canada

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ABSTRACT

Lead results from 78,971 water samples collected in four Canadian provinces from elementary schools, daycares, and other large buildings using regulatory and investigative sampling protocols were analyzed to provide lead concentration distributions. Maximum concentrations reached 13,200 and 3890 $\mu\text{g/L}$ following long and short stagnation periods respectively. High lead levels were persistent in some large buildings, reflected by high median values considering all taps, or specific to a few taps in the building. Simulations using the Integrated Uptake Biokinetic (IEUBK) model and lead concentrations after 30 min of stagnation in the dataset showed that, for most buildings, exposure to lead at the tap does not increase children's blood lead levels (BLLs). However, buildings or taps with extreme concentrations represent a significant health risk to young children attending school or daycare, as the estimated BLL far exceeded the 5 $\mu\text{g/dL}$ threshold. Ingestion of water from specific taps could lead to acute exposure. Finally, for a few taps, the total daily lead intake reached the former World Health Organization (WHO) tolerable level for adults, suggesting potential health risks.

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1. Introduction

Extreme lead concentrations have been reported in large buildings. These elevated concentrations result from the combination of three factors: water quality which favours lead corrosion, long stagnation times, and the presence of lead-bearing components. Leaded solders, brass fittings, fountains, and taps are typically the sources of lead in tap water in large buildings (Cartier et al., 2012; McIlwain et al., 2015). These can contribute to significantly higher lead levels when compared to those observed in households with lead service lines. Up to 1600 $\mu\text{g/L}$ of lead was measured in Seattle schools (Boyd et al., 2008), up to 1987 $\mu\text{g/L}$ in Washington DC schools (Triantafyllidou et al., 2009), and up to 1000 $\mu\text{g/L}$ at taps used for consumption in Canadian penitentiary

complexes (Deshommès et al., 2012). This situation is not restricted to old buildings. Elfland et al. (2010) reported lead concentrations of 350 $\mu\text{g/L}$ at fountains in a new building and identified brass fittings as the main source.

Lead is neurotoxic for young children and fetuses and is associated with intellectual deficit even at low blood lead levels (BLLs) previously considered to be safe (Canfield et al., 2003; CDC, 2012). Considering these adverse effects and the lack of a safe threshold, specific guidelines have been published for schools. Since 1994, the USEPA has formulated guidance to support sampling and remediation actions to lower lead concentrations in schools (USEPA, 2006). Recently, new regulations were introduced to reduce the maximum acceptable total lead content in brass fixtures from 8% to 0.25%. In Ontario (Canada), regulatory monitoring was implemented in 2007, as well as flushing in schools and daycares (Government of Ontario, 2007). In collaboration with public health services, New-Brunswick school boards have completed comprehensive lead sampling at every tap of every school, along with remediation actions (The Canadian Press, 2012).

The contribution of lead in tap water in households to the BLLs of children has been demonstrated in Washington DC (US),

* Corresponding author.

E-mail addresses: e.deshommès@polymtl.ca (E. Deshommès), andrews@ecf.utoronto.ca (R.C. Andrews), graham.gagnon@dal.ca (G. Gagnon), tim.mccluskey@gnb.ca (T. McCluskey), bmclwain@cbcl.ca (B. McIlwain), evelyne.dore@polymtl.ca (E. Doré), shokoufeh.nour@polymtl.ca (S. Nour), michele.prevost@polymtl.ca (M. Prévost).

Montreal (Canada), Glasgow (UK), France, and recently in Flint, Michigan (Brown et al., 2011; Deshommès et al., 2013; Hanna-Attisha et al., 2016; Levallois et al., 2013; Oulhote et al., 2013; Watt et al., 2000). Information is however scarce regarding the exposure of young children to lead in the tap water of schools and daycares. When compared to residential households with lead service lines, lead release in non-residential large buildings is mostly in the particulate form and flushing is not always effective for reduction due to the high volume of piping and low water usage (Deshommès et al., 2012; Elfland et al., 2010). Lead concentrations can vary significantly in the same building, depending on the components of the tap sampled and upstream fixtures. Corrosive water and intermittent use also contribute to increased lead levels (Barn et al., 2014; Elfland et al., 2010; McIlwain et al., 2015). By applying the United States Environmental Protection Agency (USEPA) Integrated Exposure Uptake Biokinetic model (IEUBK), Sathyanarayana et al. (2006) showed that exposure to lead in tap water in Seattle public schools resulted overall in a geometric mean BLLs below the 5 µg/dL threshold set by the CDC (CDC, 2012). Deshommès and Prévost (2012) estimated that large buildings with high particulate lead concentrations can contribute to BLL exceedances in young children. Moreover, when considering pre-flushed lead concentration results from 5 schools in British Columbia, Canada, Barn et al. (2014) estimated that the total lead intake of children increased 2-fold when compared to Health Canada estimates. Finally, limited benefits of lead remediation efforts (flushing, pipes/fountains/bubbler heads replacement) on the exposure of children in schools were reported for two systems served by distinct water qualities, both of which met the federal lead regulation of 10 µg/L (90th percentile) at household taps (Triantafyllidou et al., 2014).

It is estimated that for children between 7 and 10 years old, lead absorption rates decrease from about 50% to 10%, and then remain stable (Mushak, 1991). Most studies focusing on children's exposure consider high absorption rates and low body weights when compared to adults. Exposure of adults has been limited to specific cases of occupational exposure. Nonetheless, adverse impacts of lead for adults and at BLLs below 10 µg/dL have been documented, notably with respect to cardiovascular effects and renal effects (Ekong et al., 2006; Menke et al., 2006). As a consequence, the WHO provisional tolerable weekly intake (PTWI) of 25 µg Pb/kg body weight/week (µg Pb/kg bw/week) was put off (WHO, 2011). Moreover, the USEPA developed the All Ages Lead Biokinetic Model (AALM) and is currently updating its 2005 version (US EPA, 2005).

In this study, results were gathered from regulatory and investigative lead sampling campaigns in large buildings in Canada, including schools, daycares, and public large buildings. These lead concentrations were used to estimate the exposure of children and adults to lead resulting from the consumption of tap water from these locations.

2. Materials and methods

2.1. Lead sampling data

Data were gathered from 8530 large buildings (defined as non-residential buildings) in four Canadian provinces, including elementary schools, secondary and high schools, universities, hospitals, and penitentiaries. Most of the data originates from sampling campaigns conducted by large buildings' staff for regulatory purposes ($n = 70,709$ samples) or remediation purposes ($n = 7332$ samples) in three provinces using Health Canada's guideline for non-residential buildings (2009). Data from additional investigative sampling in three provinces to determine the source of lead and the impact of sampling protocols ($n = 930$

samples) were also included (Cartier et al., 2012; Deshommès et al., 2012; Doré et al., 2013; McIlwain et al., 2015). All samples were taken from cold water taps used for consumption, including fountains, classroom taps, kitchen or cafeteria taps, and bathroom taps.

Depending on the data subset, first flush results alone or combined with other sampling protocols were available for all taps sampled in the buildings (see Supporting Information SI). First flush sampling consisted of collecting the initial volume of tap water after overnight stagnation, consisting of at least 6 h but no more than 24 h (6hS-1), except for buildings where stagnation could not be controlled due to usage patterns (hospitals, universities, penitentiaries) or for which taps were not systematically pre-flushed the day before sampling (penitentiaries). The volume collected varied between 125 mL, 250 mL, and 1 L depending on the sampling protocol used, although 1 L samples represented the majority of the dataset (85%). Second flush sampling (6hS-2) consisted of 1750 mL water samples collected immediately following the first sampling. This type of sampling was limited to 57 taps in the dataset. Other samples included those collected after flushing the tap for 30 sec (30sF, 125–250 mL) and 5 min (5minF, 250 mL) following the collection of first draw samples (6hS-1, or 6hS-1 and 6hS-2). Finally, 30 min stagnation samples of 250 mL or 1 L in the dataset were collected after flushing the tap for 5 min followed by 30 min of stagnation, with 1 L samples representing >95% of the dataset (30minS).

All samples were collected in polypropylene bottles and acidified to pH < 2 with nitric acid for at least 16 h. The percentage of acid addition by volume varied between 0.15% and 2% depending on the dataset. Total lead concentrations were analyzed according to EPA 200.8 method by accredited laboratories and academic research laboratories, using inductively coupled plasma mass spectrometry (ICP-MS). Detection limits varied between 0.02 and 0.5 µg/L depending on the laboratory. For one dataset containing 51% of all 30sF data and 6% of all 6hS-1 data, only values above the quantification limit (1.0 µg/L) were available. Values below the detection or quantification limit were considered equal to 0.01 µg/L.

Data were segregated according to the age of the main users in the large buildings. To estimate young children's exposure, daycares and elementary schools were grouped into one dataset and categorized as '0–7 yrs dataset' (children). Similarly, to estimate older children and adult exposure data from other large buildings were grouped into a second dataset classified as '7–99 yrs dataset'. The distribution of the data as well as the types of samples collected for each dataset are shown in Table 1.

2.2. Estimation of children's exposure in elementary schools

The USEPA IEUBK model (version win1_1 Build11) was used to analyze the impact of lead on young children's (0–7 yrs) BLLs. Background exposure from sources other than tap water in the model (soil, dust, air, and food) was selected according to recent Canadian values (Table 2; see Table S1 for additional details). These parameters were validated by Deshommès et al. (2013) as representing children's background exposure to lead in urban areas, as the modelled BLLs were very close to the BLLs measured in 306 children (0–5 yrs) living in households without a lead service line in an epidemiological study (Levallois et al., 2013). The batchrun mode of IEUBK was used as described by Deshommès et al. (2013) to include varying exposure of children to water lead levels before and after starting school at approximately 5 years of age. From 0 to 5 years old, it was assumed that children drank 100% tap water containing 2 µg/L lead, which is representative of concentrations in a household with no lead service line according to previous sampling studies (Deshommès et al., 2013). For 5–7 years age range (age limit for IEUBK simulations), it was considered that children

Table 1
Distribution of tap water samplings carried out in large buildings from the '0–7 yrs dataset' (children) and the '7–99 yrs dataset' (older children and adults).

	Provinces	Types of large buildings	Approximate number of buildings	Types of taps sampled	Sampling types	N samples
0–7 YRS DATASET	A, B, C	- Elementary schools - Daycares	4010	Fountains, classroom taps	6hS-1	31,679
					6hS-2	57
					30sF	1260
					5minF	57
					30minS	31,061
				TOTAL	64,114	
7–99 YRS DATASET	A, B, C, D	- Other schools - Universities - Penitentiary complexes - Hospital	4520	Fountains, kitchen taps, bathroom taps, classroom taps	6hS-1	6998
					30sF	1747
					5minF	1318
					30minS	4794
						TOTAL

Table 2
Parameters applied for the estimation of exposure of children and adults in this study.

Parameter	Children 0–7 yrs	Adults
Water concentration — µg/L	Variable	Variable
Drinking water intake — L/d	0.742 to 1	1.5 ^a
Gastro-intestinal (GI) absorption — %	50%	10% ^b
Air concentration (indoor/outdoor) — µg Pb/m ³	0.0015	0.0015 ^b
Ventilation rate — m ³ /d	2 to 7	20 ^c
Lung absorption — %	32%	32% ^d
Daily intake from air — µg Pb/d	0.0030 to 0.0105	0.0300
Daily uptake from air — µg Pb/d	0.0010 to 0.0034	0.0096
Dust intake — g/d	0.051 to 0.081	0.040 ^e
Soil intake — g/d	0.034 to 0.054	0.000 ^{c, e}
Soil concentration — µg Pb/g	33.78	—
Dust concentration — µg Pb/g	101.61	101.61
Soil absorption fraction — %	30%	—
Dust absorption fraction — %	26%	10% ^b
Soil/dust daily intake — µg Pb/d	6.33 to 10.05	4.06
Soil/dust daily uptake — µg Pb/d	1.69 to 2.69	0.41
Dietary absorption fraction	50%	10% ^b
Dietary daily intake — µg Pb/d	1.95 to 2.26	16.1^f
Dietary daily uptake — µg Pb/d	0.975 to 1.13	1.61
Background daily intake (without water) — µg Pb/d	8.6 to 12.2	20.2
Background daily uptake (without water) — µg Pb/d	2.8 to 3.8	2.0

Justifications for adult's exposure scenario parameters: ^a Roche et al. (2012); ^b Health Canada (2013); ^c Health Canada (1992); ^d value from IEUBK model and indicated similar in adults and children in Health Canada (1992) and US EPA (2005); ^e US EPA (2005); ^f 0.23 µg Pb/bw/d (µg Pb/body weight/d) for a 70 kg bw adult (Health Canada, 2013). Other parameters (for children) are detailed and justified in Table S1 in SI.

drank 70% of water from their home (2 µg/L) and 30% of water from their school (X µg/L). This breakdown of water consumption was chosen considering that water usage patterns usually increase at specific times of the day, including breakfast (home), lunch (school) and dinner (home). Moreover, considering the daily water intake applied in IEUBK (SI, Table 2), the fraction of 30% corresponds to about one cup of water ingested at school, as used by Sathyanarayana et al. (2006). Considering that the particulate lead fraction of the samples was not evaluated (no filtration), total lead is considered 100% soluble in the IEUBK simulations. The weighted mean of home (70%, 2 µg/L) and school (30%, X µg/L) water lead concentrations was computed as following:

$$Pb_{\text{water, } \mu\text{g/L}} = 1.4 + 0.3 \cdot X$$

Lead concentration for a given school (X) was calculated based on 30minS values obtained from the Children (0–7 yrs) dataset. The 30minS sampling was selected since it has been proposed as representative of tap water inter-use time and therefore indicative of typical exposure at the tap (van den Hoven and Slaats, 2006). Median and 90th percentile values from 30minS samples in the total dataset, or from worst case buildings or worst case taps were used in the model to evaluate the overall and site-specific effects of large

building tap water. Worst case buildings were identified based on the tail of the 30minS concentrations distribution of the 0–7 yrs dataset, according to the flow chart presented in SI (Figure S3).

2.3. Estimation of children exposure in daycares

BLLs were estimated using IEUBK for very young children attending daycares (1–5 years old). It was considered that from 1 to 5 years old, the child was going to daycare, drinking 30% of his daily water from the daycare large building (X µg/L) and 70% at his home (2 µg/L). Simulations were stopped at 5 years old, when the child was expected to start elementary school.

2.4. Estimation of adult exposure in large buildings

Considering that updates of the AALM model to estimate adults BLLs are in process, the impact of lead in the tap water of large buildings on adult's exposure was investigated by estimating the total daily lead intake and uptake. Canadian values for soil, food, dust, and air were used for the calculation of background exposure (Health Canada, 1992, 2013; US EPA, 2005). Table 2 compares parameters applied for background exposure to those applied for the estimation of children exposure in the IEUBK model. A tap water

consumption of 1.5 L per day was selected according to the 75th percentile of tap water consumption for North-American in Roche et al. (2012) review. As for children BLL modelling, the consumption was divided between home and large building, with 1 L from households at 2 µg/L and 0.5 L from large buildings at X µg/L. Lead concentrations for the large buildings (X) were calculated based on the 7–99 yrs dataset, similar to the estimation of young children exposure (overall and worst case buildings concentrations).

3. Results and discussion

3.1. Lead concentrations in the tap water of large buildings

Lead concentrations were analyzed for each dataset considering the type of sampling and their probability of occurrence at the tap. Fig. 1 presents the distribution of lead concentrations observed in Canadian elementary schools and daycares (0–7 yrs dataset) per sampling type. Fig. 2 presents the distribution obtained for large buildings serving adults (7–99 yrs dataset). Overall, lead concentrations in the 0–7 yrs dataset were below the 10 µg/L threshold. 90th percentile values ranged from 3.0 to 11 µg/L except for 6hS-2 samples (20 µg/L) which were less representative when considering sample size ($n = 57$). Also, for the 57 taps sampled for 6hS-1 and 6hS-2 successively, 6hS-1 concentrations were generally higher (Doré et al., 2013). Lead concentrations in the 7–99 yrs dataset were comparable to those in the 0–7 yrs dataset, except 6hS-1 concentrations that were consistently higher with 75th and 90th percentiles at 8.2 and 26 µg/L respectively. This can be explained by the fact that 18% of the 6hS-1 samples in the Adults dataset are from penitentiaries where lead concentrations were significantly higher when compared to the other sites sampled (secondary and high schools, universities, etc). Indeed, in those complexes, median and 90th percentile lead concentrations for first flush samples reached 10 and 97 µg/L respectively ($n = 1291$), as compared to 2.3 and 15 µg/L for the remaining of the 6hS-1 samples in the dataset ($n = 5707$). Such differences are attributable to two characteristics of penitentiaries dataset, namely: (i) the absence of a pre-flush the day before collecting samples (stagnation may have exceeded 24 h for some taps), and (ii) the low water usage caused by a large number of taps attributed to the prisons restricted environment which increases stagnation times (Deshommès et al., 2012).

For the two datasets, median values are low and comparable for all types of sampling (0.01–2.9 µg/L). Differences between the different types of sampling are systematic with higher concentrations after long stagnations and concentrations exceeding 10 µg/L occurring at the 90th percentile. Indeed, 11–22% of the lead concentrations exceeded 10 µg/L after long stagnations (6hS-1, 6hS-2) when compared to 0–5% following long flushing or short stagnation (5minF, 30minS) in agreement with prior studies (Barn et al., 2014; Doré et al., 2013; McIlwain et al., 2015). Short flushing of 30 sec was less effective in reducing lead levels as 10–13% of the samples still exceeded 10 µg/L. This is consistent with profiling sampling showing a progressive decrease of lead release, concentrations being the highest in the first 250–500 mL of water (Cartier et al., 2012; McIlwain et al., 2015).

When considering all types of samples, extreme concentrations were measured. Up to 13,200 µg/L was detected in a day nursery at first flush, up to 3890 µg/L after 30 min of stagnation in a public school, and up to 930 µg/L after 5 min of flushing in a penitentiary (Figs. 1 and 2, and SI). Such concentrations are extremely high when compared to the lead concentrations reported in households with a lead service line using similar sampling protocols (Del Toral et al., 2013; Deshommès et al., 2010). These levels are however comparable to lead levels measured in large buildings reported in various studies and indicative of high particulate lead fractions in the samples (Boyd et al., 2008; Deshommès et al., 2012; Elfland et al., 2010; McIlwain et al., 2015; Triantafyllidou et al., 2009). The causes for such extreme lead concentrations have been investigated in several studies. Corrosive water, low usage patterns, and the presence of lead bearing components in and/or upstream of the tap were identified as the causes of elevated lead levels. Elfland et al. (2010) attributed the prevalence of high lead levels at new fountains to lead-bearing brass fittings and to low water demand. McIlwain et al. (2015) sampled all fountains in a large building and showed that those with a lead-lined tank or with very low water usage had systematically high lead concentrations when compared to lead-soldered fountains. The highest lead levels generally occurred in the first 500 mL collected, although for infrequently used fountains, an increase of lead concentrations was reported after flushing the tap due to scale detachment and resuspension. Copper risers with leaded solders, faucets and brass meters have also been linked to high lead release in large buildings (Cartier et al., 2012; Deshommès et al., 2012). Finally, lower lead release was

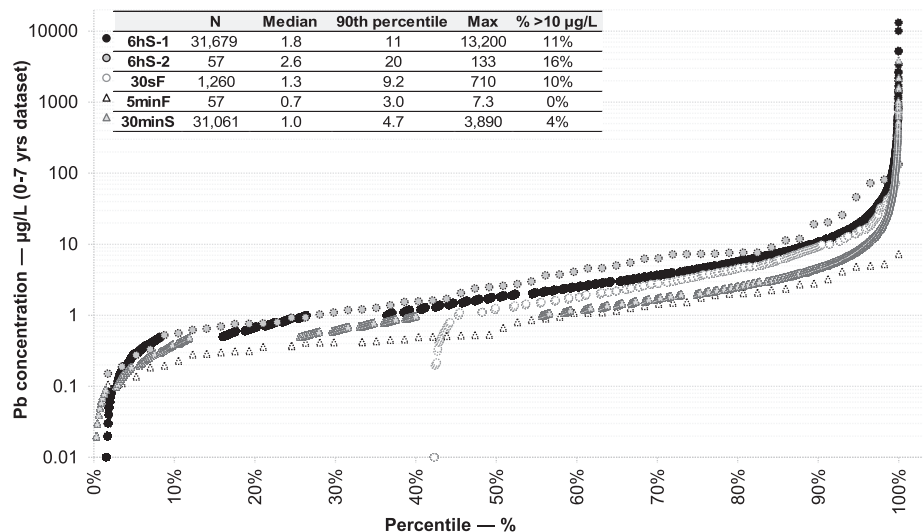


Fig. 1. Distribution of lead concentrations in the tap water of large buildings - Children (0–7 yrs) dataset.

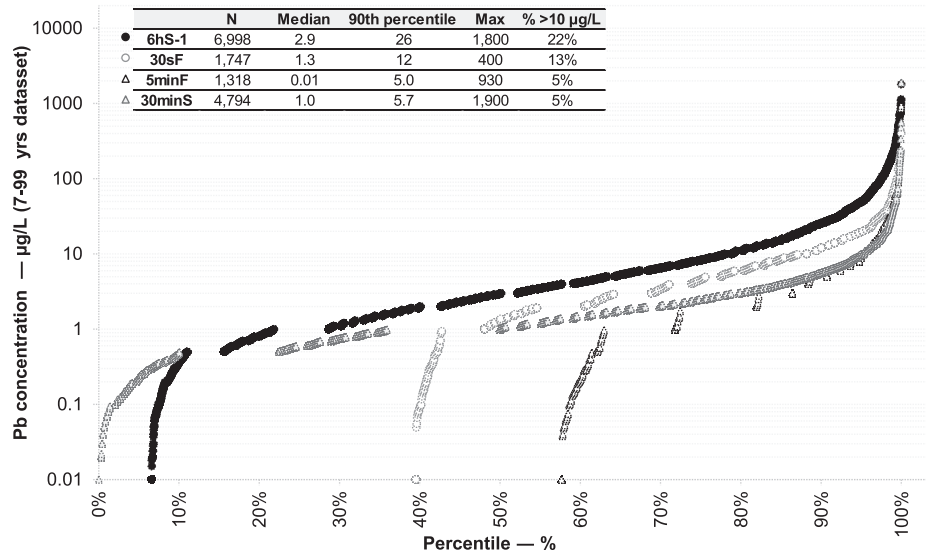


Fig. 2. Distribution of lead concentrations in the tap water of large buildings – Older children and adults (7–99 yrs) dataset.

measured in large buildings supplied by water treated by ortho-phosphates or pH adjustment. An opposite trend was measured when increasing the chloride to sulfate mass ratio (Cartier et al., 2012; Doré et al., 2013). In this study, corrosion control was mandatory in one of the provinces, suggesting that some of the buildings sampled received less aggressive water. Nonetheless, as mentioned by Triantafyllidou et al. (2014), corrosion control regulations aiming to reduce lead at home taps may not be always efficient to reduce lead at large buildings taps. For most of the data in this study no detailed information was available regarding the type of tap (e.g., fountain, classroom tap), water demand, plumbing materials and renovation work in the buildings, and water quality. However, it can be considered that all of these factors contributed to the wide range of lead concentrations observed.

The variability of lead concentrations was studied using subsets of data from taps in the same large building (same water quality) or in large buildings from different municipalities/schools (different water quality). As shown in Fig. 3a) and b), for a given building, lead concentrations vary by a factor of 10–2000 between taps (Kruskal-Wallis test, $p < 0.001$). Extremely high lead concentrations of 10,100 µg/L (6hS-1) and 3890 µg/L (30minS) were measured at some taps, while concentrations typically < 10 µg/L were measured at other taps (Fig. 3a). Considering this, sampling only one tap in a large building is not indicative of the exposure risk to lead in tap water since extreme lead levels are unlikely to be captured by a one tap/one event sampling. Moreover, this reflects that children's exposure can vary considerably depending on usage patterns in the building. Lead concentrations also varied with respect to water quality (utility/district) as shown by Fig. 3c) and d) (Kruskal-Wallis tests, $p < 0.001$). Nonetheless, concentrations could vary from < 0.05 µg/L to 500 µg/L or more depending on the utility/district. This clearly indicates that lead concentrations are explained by factors other than water quality, such as the age and type of plumbing components and water usage patterns.

In order to identify the extent and the occurrence of extreme lead concentrations in a building and to estimate resulting children BLLs, a flow chart presented in SI (Figure S3) was developed. First, the 30minS samples from the Children dataset were analyzed. Ninety three schools/daycares presenting at least one sample with an elevated lead concentration after 30 min of stagnation (> 99 th percentile of 30minS distribution, which corresponds to a range of

81–3890 µg/L) were identified. Then, the median and 90th percentile concentrations were calculated for each of the 93 buildings identified, using all of the 30minS sample results for each building (0–100% of the distribution). The buildings were sorted in ascending order of median values. Of the three buildings with the highest median values the one with the highest 90th percentile value was designated as “worst case building”. Fig. 4 presents 30minS concentrations from the Children dataset for 10 buildings (3 daycares, 7 schools) with the highest 30minS median values in the subset of 93 buildings having at least one 30minS sample result with elevated concentration (> 99 th percentile). The daycare A and elementary school D were identified as worst cases based on the flow chart. High lead levels were observed following a short stagnation of 30 min. Median and 90th percentile concentrations for the worst case daycare reached 109 µg/L and 254 µg/L respectively ($n = 5$), representing 10 to 20 times the maximum reference level of 10 µg/L. The 30minS median concentration in the worst case elementary school D remained high (24 µg/L) while 90th percentile reached 412 µg/L ($n = 13$). Daycare B had overall lead levels > 10 µg/L whereas lead concentrations in Daycare C varied between 0.05 and 257 µg/L. Similarly, lead levels measured at the taps of schools D, E, and to a lesser degree F were generally higher when compared to schools G to I. As such, lead concentrations can be persistently high for specific schools. Conversely, and as shown by elementary school data presented in Fig. 3a), extreme levels of up to 3890 µg/L were observed following short stagnations while median concentrations (3.0 µg/L) in the building did not reflect a lead issue ($n = 180$). For this school, 90th percentile reached 54 µg/L, indicating that some areas in the school may be more susceptible to high lead release due to specific usage patterns or plumbing materials. Besides, median value for the 93 buildings with elevated 30minS concentrations (1.5 µg/L, $n = 2003$) is comparable to the median value from all buildings in the Children dataset (1.0 µg/L, $n = 31,061$). However, 90th percentile value is 10 times higher (47 µg/L versus 4.7 µg/L) showing that lead concentrations can vary widely between different taps for a given school. In this particular situation, the data indicate that relying on results from a limited number of taps sampled in a school building can be misleading, which may lead to a false sense of compliance. Broad variation of lead concentration between taps and extreme concentrations support the need to sample all taps used by children for a specific

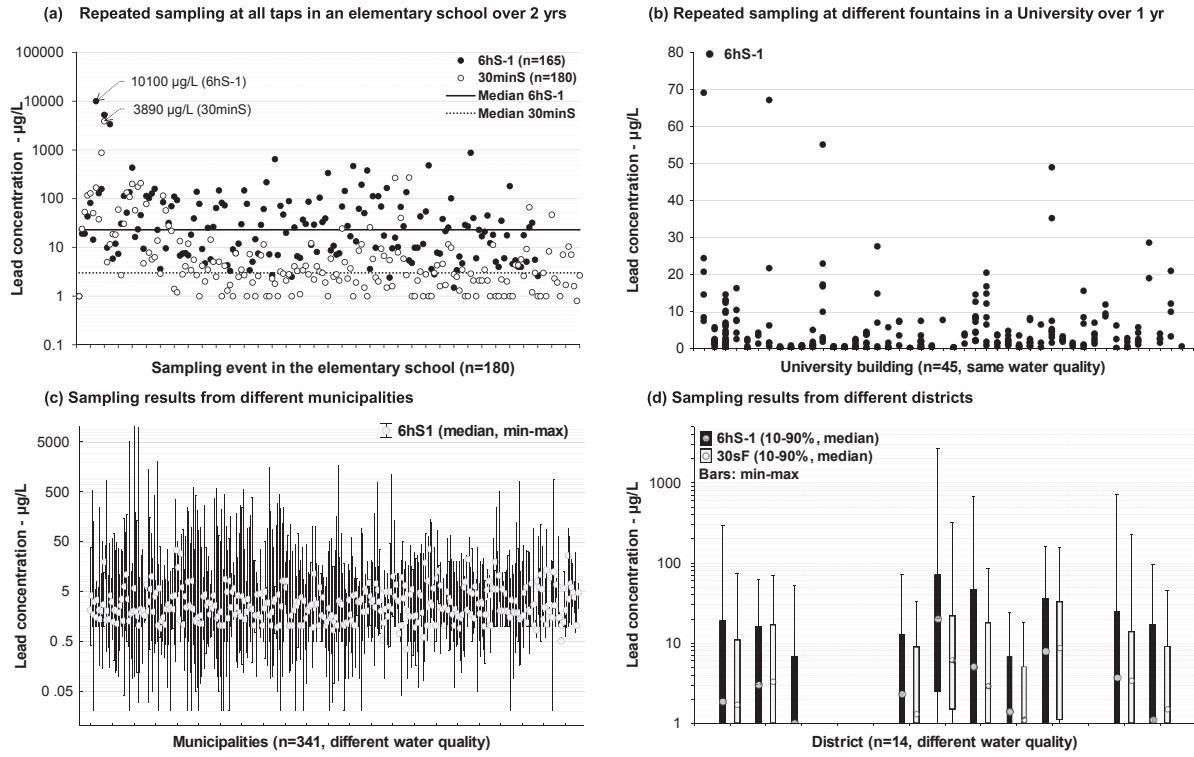


Fig. 3. Variability of lead concentrations using 4 subsets of data from the whole dataset (a, b) for different taps and sampling events for the same water quality, and (c, d) for different buildings locations (municipality or district).

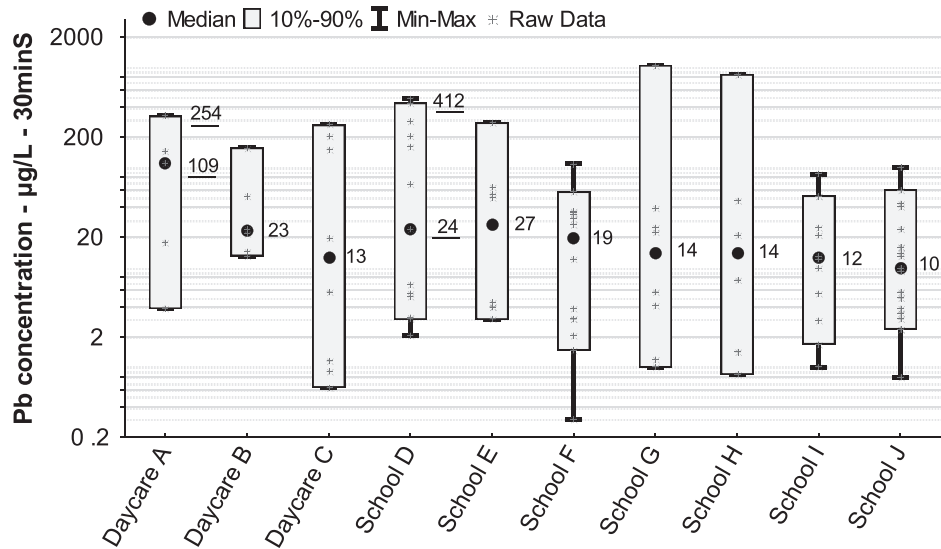


Fig. 4. Lead concentrations after 30 min of stagnation for the Children (0–7 yrs) dataset in worst case daycares and schools. Underlined characters represent values used for IEUBK modelling (Daycare A and School D).

school (e.g., classroom taps). The significant and sometimes acute concentrations at specific taps may warrant immediate corrective action.

A worst case large building based on 30minS concentrations was also identified in the Adults (7–99 yrs) dataset. Maximum concentration following 30 min of stagnation reached 1900 µg/L in this dataset. For the worst building, the median concentration was 6 µg/L and the 90th percentile 145 µg/L.

3.2. IEUBK simulations

Median and 90th percentile of 30minS concentrations from the Children (0–7 yrs) dataset, from worst case school and worst case daycare were applied in IEUBK to evaluate overall and site specific risk of high BLLs in young children. Simulations using overall 30minS concentrations in the 0–7 yrs dataset show similar trends: geometric mean (GM) BLLs are comparable to those estimated without the contribution of tap water (0.8–1.3 µg/dL), and remain

stable in the range of 1.3–2.1 $\mu\text{g}/\text{dL}$ or increase slightly (+0.3 $\mu\text{g}/\text{dL}$) following the transition to school or daycare (Fig. 5). As well, the estimated fraction of children with a BLL exceeding 5 $\mu\text{g}/\text{dL}$ remains below 1.3% (Fig. 6). These results are consistent with the 1.0–1.7 $\mu\text{g}/\text{dL}$ GM BLL range reported in 0–6 yrs old children in Montreal and Nunavik (Quebec) and St John's (Newfoundland and Labrador) between 2006 and 2010, and lower when compared to the GM BLL of 2.75 and 5.1–5.6 $\mu\text{g}/\text{dL}$ reported in Flin Flon (Manitoba) and Trail (British Columbia) respectively (Health Canada, 2013). The increase from 1.8 to 2.1 $\mu\text{g}/\text{dL}$ (+17%) in estimated BLLs for all daycares (90th percentile) is lower but consistent with NGueta et al. (2016) results showing a 35% increase of low BLLs per increment of 1 $\mu\text{g}/\text{L}$ at home taps in Montreal. Therefore, in the majority of schools and taps sampled in this study, the modelled BLLs which were based on measured lead concentrations suggest that tap water was not an important source of exposure to lead for the children attending these schools and daycares.

When considering the lead concentrations measured in specific schools with confirmed lead issues in IEUBK, predictions of the impact of drinking water while in school vary significantly depending on the sub-group of schools and taps considered. Using the median levels from the worst case elementary school (24 $\mu\text{g}/\text{L}$), modelled GM BLLs at the age of 5 increase from 1.3 to 2.3 $\mu\text{g}/\text{dL}$, and approximately 3% of children exceed 5 $\mu\text{g}/\text{dL}$ as opposed to 1% prior to starting school (Figs. 5a and 6a). Using the 90th percentile value (412 $\mu\text{g}/\text{L}$) allows estimation of the exposure of children repeatedly using high lead releasing taps. For those cases GM BLLs increased to 15 $\mu\text{g}/\text{dL}$ and almost 100% of children drinking that water are at risk of exceeding the 5 $\mu\text{g}/\text{dL}$ BLL threshold value. Therefore, for the worst case school, a child drinking water from multiple taps in the school (as represented by simulations using median values), or in specific areas of the school with high content taps (as represented by simulations using 90th percentile value) will be at a higher exposure than at home and could present BLLs exceeding 5 $\mu\text{g}/\text{dL}$.

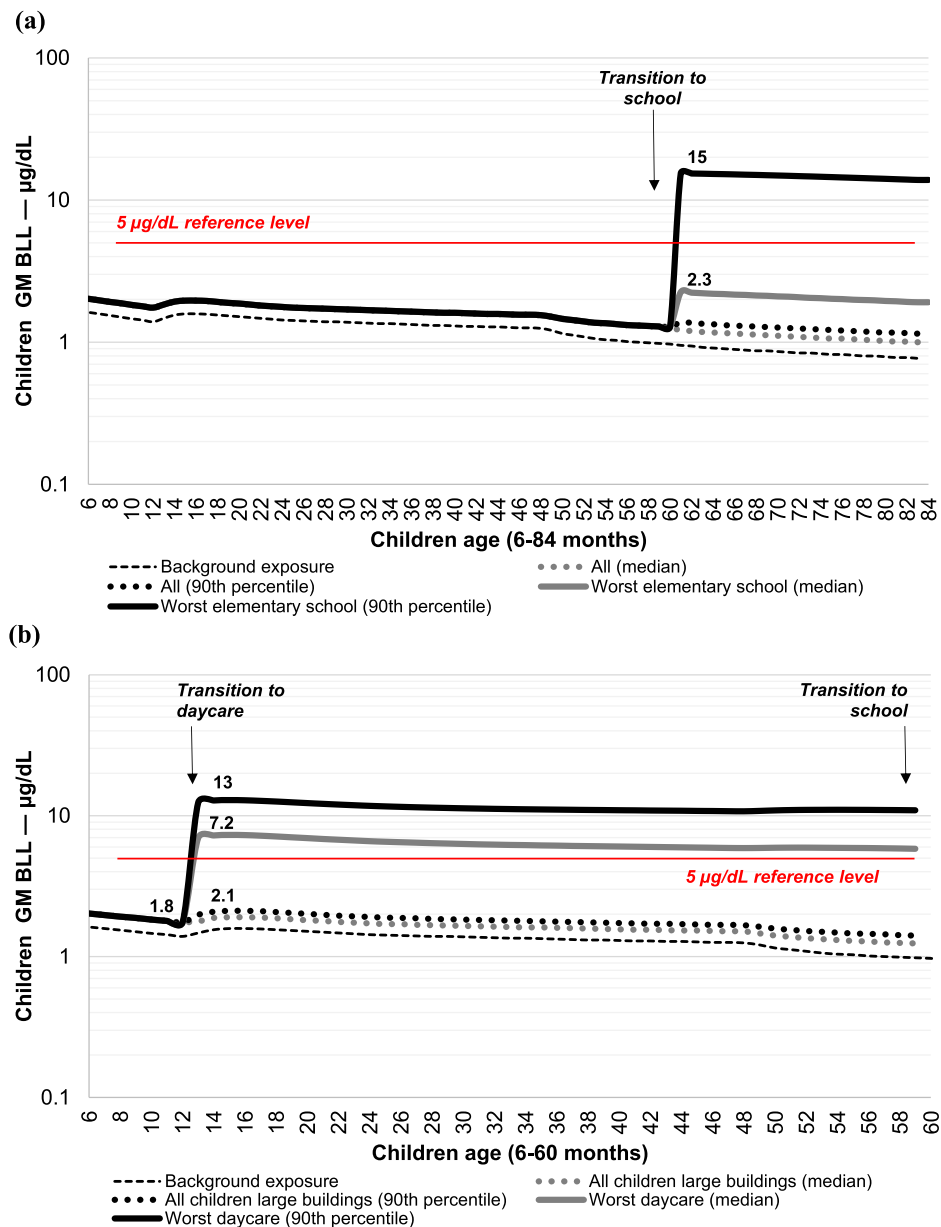


Fig. 5. Geometric means estimated from IEUBK simulations in batchrun mode for young children (a) starting school at the age of 5 years old (simulations from 6 to 84 months), and (b) starting daycare at the age of 1 (simulations from 6 to 60 months).

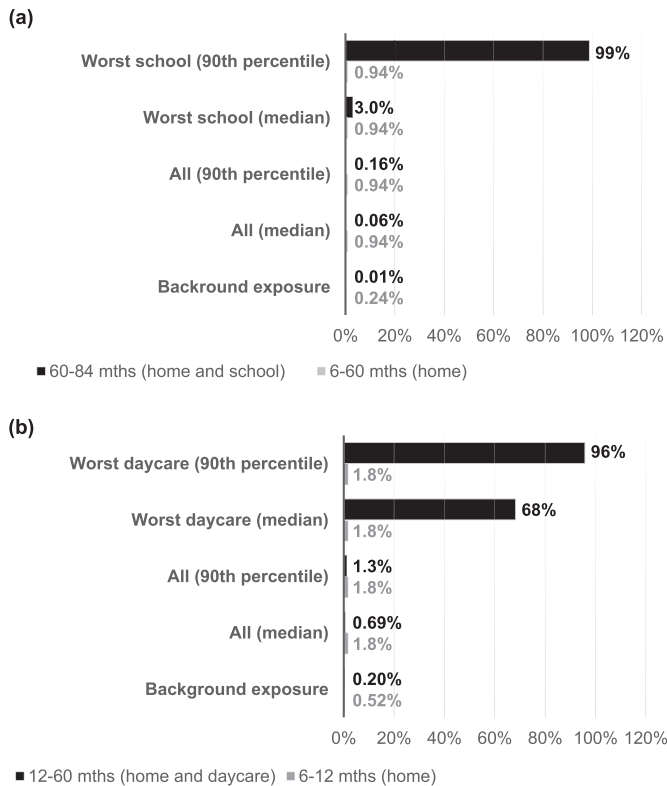


Fig. 6. Fraction of children exceeding the BLL of 5 µg/dL estimated from IEUBK simulations in batchrun mode for young children (a) starting school at the age of 5 years old (60–84 mths), and (b) starting daycare at the age of 1 (12–60 mths).

To place the potential impact of this exposure in context, the modelled GM BLL estimated for these children far exceeds the modelled GM BLL estimated for children living in households with up to 30 m of lead service line in Montreal during summer (5 µg/dL) (Deshommès et al., 2013).

Simulations for worst case daycares result in even higher estimated BLLs considering the significantly high lead concentrations measured in tap water (median and 90th percentile of 109 and 254 µg/L respectively). GM BLLs increase from 1.8 µg/dL to 7.2–13 µg/dL after the child starts attending daycare at the age of 1, and these levels persist until the age of 5 assuming that the child remains in the same daycare (Fig. 5b). For this daycare, 68–96% of the children could exceed the 5 µg/dL threshold (Fig. 6b), which is far over the 10.6% fraction estimated to date for high lead levels areas in Flint, Michigan (Hanna-Attisha et al., 2016).

Although only 4% of the 30minS samples exceeded 10 µg/L in schools/daycares, 93 buildings (2.3%) showed at least one sample containing lead concentrations within 81–3890 µg/L after only 30 min of stagnation based on the decision flow chart (SI). The wide differences observed between and within daycares and elementary schools show that some buildings (or specific locations in the buildings) can contribute to increase BLLs in early childhood. These buildings should be identified and corrective actions be taken to limit potential health risks.

For the evaluation of the impact of extreme concentrations (such as 3890 µg/L after 30 min of stagnation) the frequency of the occurrence of these extreme concentrations must be considered. The probability of a child being repetitively exposed to such extreme concentrations seems low considering the variability of lead levels in a building depending on the tap and stagnation prior to water consumption (Figs. 1–3, Figure S2 in SI). Therefore, a

simple estimation of the impact of repeatedly consuming these extreme concentrations using the IEUBK model is not possible nor representative and would exceed the IEUBK maximum of reliable estimation of 30 µg/dL. Drinking a glass of water containing extreme lead concentration would be considered an acute exposure. The U. S. Consumer Product Safety Commission Washington (USCPSC) defines the acute health risk as one dose ingestion of 175 µg lead from lead-contaminated toys (USCPSC, 2005). As demonstrated by Triantafyllidou and Edwards (2009), the ingestion of one dose at a level of 175 µg or above would represent an acute exposure that could induce a temporary risk of the child's BLL exceeding 10 µg/dL. Assuming the consumption of a 300 mL of tap water, lead concentrations above 580 µg/L would therefore represent an acute health risk since they correspond to a dose of 175 µg or more. When considering all types of samples (6hS-1, 6hS-2, 30sF, 5minF, 30minS) from the 0–7 yrs old children dataset in this study, 40 samples (0.06%) would cause an acute health risk. This method of estimation depends however on the volume of samples and the fraction of lead particles in it. In this study particulate lead was not investigated however concentrations over 100–200 µg/L may contain a high fraction of lead particles (Deshommès et al., 2012). Moreover, as compared to soluble lead, its occurrence is more sporadic and the concentrations would vary depending on the volume of sample collected (Deshommès et al., 2010). Specifically, lead particles collected in a large volume of water (eg. 1 L), which represent 85% of our dataset can be underestimated if a volume of 300 mL is considered. Nonetheless, collecting a large volume of water can increase the probability of capturing lead particles in the sample, provided that acidification took place in the bottle used for sample collection (Triantafyllidou et al., 2013). Considering these factors, the 0.06% occurrence of acute health risk estimated in this study may vary. In order to avoid any case of acute exposure especially in very young children, taps presenting high lead levels should be identified and prohibited for water consumption use until an investigation is completed and corrective actions implemented.

3.3. Estimation of the total daily intake for adults

Total daily lead intake and total daily uptake values for older children and adults were calculated using assumed background levels associated with exposure from diet, air, and dust/soil as presented in Table 2, and by adding the contribution of tap water from home and large buildings. Without the contribution of tap water, total lead intake is 20 µg Pb/d while uptake is 2.0 µg Pb/d representing about 10% of the intake. When the contribution of tap water from large buildings (7–99 yrs dataset) is included, and considering the median or 90th percentile values, the total intake increases slightly to 23–25 µg Pb/d (Fig. 7). When considering 30minS concentrations from the worst case large building identified in the 7–99 yrs dataset, the estimated total intake does not increase using median value, but does increase from 20 to 95 µg Pb/d using 90th percentile value (Fig. 7). As previously mentioned, the worst case building for the 7–99 yrs dataset presents lower lead concentrations when compared to worst case buildings in the 0–7 yrs dataset due to differences in building types, usage patterns, and water quality. Considering the worst case concentration after 30 min of stagnation (1900 µg/L), and assuming the consumption of 2 cups of this water (500 mL), the total lead intake is about 72 times (1448 µg Pb/d) the background intake for adults and exceeds by a factor of 5.8 the former WHO provisional tolerable weekly intake of 25 µg Pb/kg bw/week for a 70 kg bw adult (equivalent to 250 µg Pb/day). Overall, considering all types of samples in the 7–99 yrs dataset (6hS-1, 30sF, 30minS, 5minF), 33 samples exceeded 455 µg/L resulting in a total daily intake over the former WHO tolerable

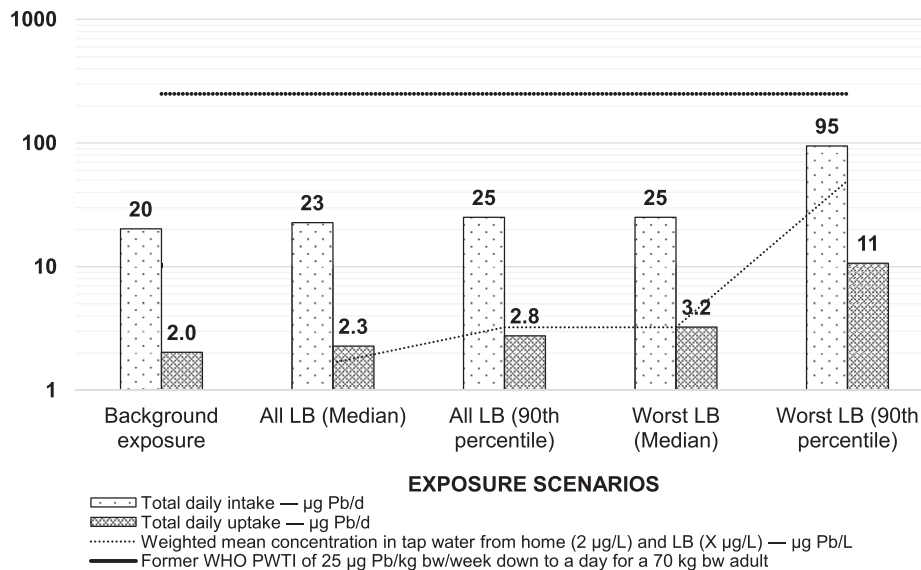


Fig. 7. Average daily intake and average daily uptake estimated for adults considering different scenarios of exposure using 30minS concentrations in the tap water of adult's large buildings (LB, 7–99 yrs dataset).

intake (assuming the consumption of 500 mL of water). Again, as for the Children dataset, this estimation will vary with the sample volume and particulate lead fraction. It is not clear as to how these levels would impact adults BLLs, but in light of the findings on the health effects of lead on adults even at low BLLs (Ekong et al., 2006; Menke et al., 2006), as a precaution, consumption should be avoided at such taps.

4. Conclusion

Lead concentrations measured at the tap of 8530 elementary schools, daycares, and large buildings across a range of water qualities and building types in four Canadian provinces were generally low. Based on extensive monitoring and investigative data as input to biokinetic modelling, the authors anticipate that:

- Lead at the tap would not contribute to elevated BLLs in young children and adults at the majority of the taps monitored.

However, the analysis of the data also reveals concerning observations:

- Some daycares and elementary schools present system-wide lead release and are likely to cause elevated BLLs in young children.
- Some taps with extreme lead concentrations could cause rare but acute risk of elevated BLL in young children.

As public health initiatives promote drinking water over other beverages for children, it appears critical to prevent lead exposure in daycares and schools. Furthermore, in light of the short lived benefits of flushing to reduce lead concentrations at the tap, guidance that relies on weekly and daily flushing should be re-examined.

It is important to underline that the presented biokinetic simulations were all based on lead concentrations after a short stagnation (30 min) which are considered representative of consumers' exposure. Using concentrations after extended stagnation would provide more conservative estimates. Basing the estimation of exposure on concentrations after extensive flushing is neither

justified nor ethically acceptable given the usage patterns in large buildings. Although rare, lead concentrations in tap water corresponding to ≥ 175 µg lead dose can be considered an acute exposure. In this study, at least 40 samples were estimated to cause acute health risks according to the USCPSC. Finally, although adults absorb less lead than children, the total daily lead intake at some taps in large buildings can approach or exceed former WHO tolerable intake.

In conclusion, the analysis of a very large dataset of samples in large buildings confirms that lead concentrations at a given tap in a building cannot predict the concentrations at other taps in the same building. As a consequence, this study confirms the need for mandatory sampling at each consumption tap in elementary schools and daycares to identify problematic fountains and faucets. Corrective actions should then be taken in order to prevent high risk exposure to lead in children. In all cases, on-site analysis of the samples should be prioritized as it can provide a quick and low-cost response for each tap and elevated lead levels could then be confirmed by ICP-MS measurements.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.watres.2016.04.050>.

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