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Exposure of young children to household water lead in the Montreal area (Canada): The potential influence of winter-to-summer changes in water lead levels on children's blood lead concentration

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ABSTRACT

Drinking water represents a potential source of lead exposure. The purpose of the present study was to estimate the magnitude of winter-to-summer changes in household water lead levels (WLLs), and to predict the impact of these variations on BLLs in young children. A study was conducted from September, 2009 to March, 2010 in 305 homes, with a follow-up survey carried out from June to September 2011 in a subsample of 100 homes randomly selected. The first 1-L sample was drawn after 5 min of flushing, followed by a further 4 consecutive 1-L samples after 30 min of stagnation. Non-linear regression and general linear mixed models were used for modelling seasonal effects on WLL. The batchrun mode of Integrated Exposure Uptake Biokinetic (IEUBK) model was used to predict the impact of changes in WLL on children's blood lead levels (BLLs). The magnitude of winter-to-summer changes in average concentrations of lead corresponded to 6.55 μg/L in homes served by lead service lines (LSL+ homes) and merely 0.30 μg/L in homes without lead service lines. For stagnant samples, the value reached 10.55 μg/L in 'LSL+ homes' and remained very low (0.36 μg/L) in 'LSL− homes'. The change in the probability of BLLs ≥5 μg/dL due to winter-to-summer changes in WLL was increased from <5% (in winter) to about 20% (in summer) in children aged 0.5–2 years. The likelihood of having BLLs ≥5 μg/dL in young children during warm months was reduced by at least 40% by flushing tap-water.

1. Introduction

Exposure to lead is a public health concern, particularly as it can affect the neurodevelopment of young children. Previous studies suggested that intellectual deficits in children occurred even at relatively low blood lead levels (BLLs) (Lanphear et al., 2005; US NTP, 2011), leading to the revision of the 10 μg/dL threshold set previously by the CDC (U.S. CDC, 2012).

Drinking water represents a potential source of lead exposure for children. From 2000 to 2007, the proportion of elevated BLLs (≥10 μg/dL) among children aged ≤30 months in Washington, D.C. was strongly correlated to lead in water and the presence of lead service lines (LSLs) (Edwards et al., 2009). Young children living in homes with LSLs are especially at risk of elevated BLLs (Brown et al., 2011). LSLs are still numerous in North America. Furthermore, because of the shared ownership of the LSLs, they are often only partially replaced and continue to contribute to lead at the tap. Health Canada set a Maximum Acceptable Concentration (MAC) of 10 μg/L and requires for residential sites an annual sampling between May and October (Health Canada, 2009). The sampling protocol applied for the sample collection varies from one province to another: in Ontario, 2 consecutive litres are collected after 30 min of stagnation. In Quebec, the regulation applies to a sample collected after 5 min of flushing but since 2013, 30 min of stagnation samples were added to the protocol for subsequent detailed characterization of the LSL.

The contribution of LSLs to total lead in water is reported to be up to 50–75% (Sandvig et al., 2008). Lead released from LSLs may be produced by both chemical and physical factors. Previous studies showed that lead solubility increases with water temperature (Britton and Richards, 1981; Schock, 1990). However, little is known about the seasonal patterns of household water lead levels (WLLs). In the Montreal area (Canada), Deshommes et al. (2013) reported significant differences in WLL over the year. However, the seasonal results used were mostly not from the same households, and other characteristics unrelated to seasons could explain the variations of WLL observed in the study. There are few follow-up studies assessing seasonal changes in WLL.

Abbreviations: BLLs, blood lead levels; IEUBK, Integrated Exposure Uptake Biokinetic model; LSLs, lead service lines; WLL, water lead level.

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Karalekas et al. (1983) reported variations of WLL across seasons in homes with LSL in Boston. From January 1979 to August 1981, the water temperature varied from 5 °C to 20 °C, and the mean WLL increased from 15–20 μg/L during winter to 36–43 μg/L in summer. de Mora et al. (1987) observed similar results in Glasgow. Both studies reported higher lead in summer, and the higher temperature is a possible cause. The two prior studies suffer from small sample size and did not report longitudinal effects of seasonality on WLL. As a result, the extent of seasonal variations remains unknown.

Finally, seasonal variations of children's BLLs have been reported, BLLs being generally higher in summer as compared to winter (Baghurst et al., 1992; U.S. EPA, 1995; Yiin et al., 2000). In most of the studies, the observed BLL variations were attributed to the fact that children are higher exposed to soil and dust in summer through outdoor exposure. Also, in these studies, the lead concentrations were well characterized in soil and dust, but not or poorly characterized in tap water. Therefore, the contribution of drinking water on seasonal changes in BLLs remains unknown. This study aims to estimate the magnitude of winter-to-summer changes in WLL through separate analyses of homes with and without LSLs in Montreal, and to predict the impact of these variations on BLLs in young children.

2. Materials and methods

2.1. General methodology

A cross-sectional study followed by a follow-up was undertaken in Montreal (Canada) to characterize lead exposure in young children (water, dust, paint). The recruitment process and detailed criteria for selecting homes are fully described elsewhere (Levallois et al., 2014). In the framework of this study, a total of 305 private homes were analysed between September 10, 2009 and March 27, 2010, including 177 with a LSL (LSL+ homes), and 128 without a LSL (LSL− homes), and 7 for which no conclusion could be drawn (Deshommes et al., 2013). Over that period (first campaign), the water temperature varied between 1.4 °C and 21.7 °C (ambient temperature: −15.3 °C to 23.8 °C). Then, a stratified random sampling of 100 homes drawn from the initial sample was selected for a quantification of WLL in summer, including 80 homes with a LSL and 20 without a LSL. A second water sampling was performed from June 22, 2011 to September 06, 2011 (second campaign) in this subset of homes using the same protocol. The water temperature during that second campaign varied between 16.0 °C and 24.1 °C (ambient temperature: 20–28 °C). The whole sample of 305 households was useful to characterize the basic water chemistry of the system under study and to estimate the marginal distribution of WLL. The follow-up analyses were carried out using the subset sample of 100 households with repeated measures of WLL during both campaigns.

2.2. Water sampling and system information

The four neighbourhoods considered in this study were served by 2 water treatment plants drawing from the same water source, and water physicochemical parameters were relatively stable. Available data in the period of the study are reported in Table 1. pH and alkalinity data of distributed water were available from January 01, 2003 to December 31, 2010 (provided by the utility). It is assumed that these data are representative of tap level data. Indeed, relatively modest pH variations have been previously reported within the distribution system (Cartier et al., 2011), and were not a significant factor affecting lead concentrations, unlike the length of lead pipes and the presence of particles. In every participating household, the following sampling protocol was applied for tap water. First, a 1 L sample was collected after 5 min of flushing (fully flushed) at the kitchen tap. Then a stagnation of 30 min was carried out in the house and 4 consecutive 1 L samples were collected (namely first-, second-, third- and fourth-draw sample). All samples were collected at a 5 to 7 L/min flow rate in pre-acidified bottles, without removing the tap aerator. The water temperature was measured after approximately 3 min during the flushing of 5 min and flow rate was also measured. Finally, for every home visited, information was collected on the neighbourhood, type of residence, construction year, total number of people living in the home, and the floor level where the sampled tap was located.

2.3. Lead analyses

During the first campaign, the samples acidified at pH < 2 were kept at 4 °C until ICP-MS analysis. Total lead was analysed according to the US EPA method 200.8, but using a 24-hour digestion time for the sample instead of 16 h. This digestion method was tested and compared successfully to stronger digestion methods for the estimation of total and dissolved lead in water samples collected after short stagnation in homes with a LSL in Montreal (Cartier et al., 2011). The lead analyses and QA/QC procedures are summarized in the Supplemental material, Table S1.

For the second campaign, total lead analyses were carried out using a very similar method to the previously described modified US EPA method, including a digestion period of 24-h. The detection and quantification limits were also comparable.

2.4. Variables under study

The response variable was the water lead concentration. Fully flushed and first-draw 1 L samples were considered separately. In addition, a 30MS2-4 variable was created, which refers to the arithmetic mean of the second, third and fourth-draw samples. This stratified modelling was motivated by the fact that a fully flushed sample can provide the home's signature (LSL and plumbing system combined), and the first-draw one provides the specific household tap signature, while the change profiles of lead concentration from the second to the fourth-draw sample differ markedly according to the presence or absence of LSL (Supplemental material, Fig. S1).

For longitudinal analyses, the selected independent variables included flow rate (litre/s), interval time between two measurements, neighbourhood, type of residence, construction year, total number of people living in the home, and tap level (floor level where the tap was

Table 1
Median values (25th percentile–75th percentile) for water physicochemical parameters observed for different seasons.

	LSL− homes[a]			LSL+ homes[b]		
	Autumn	Winter	Summer	Autumn	Winter	Summer
n	38	70	20	32	65	80
WT[c]	12.3 (10.5–15.6)	3.4 (2.6–4.4)	21.0 (19.9–22.9)	12.7 (10.1–14.7)	3.1 (2.4–4.3)	22.4 (21.3–23.4)
pH	8.2 (8.1–8.3)	8.1 (8.1–8.2)	8.2 (8.1–8.3)	8.2 (8.2–8.3)	8.2 (8.1–8.2)	8.2 (8.1–8.2)
Alkalinity	89 (87–89)	89 (88–90)	89 (88–90)	89 (88–89)	89 (88–89)	89 (88–90)

Alkalinity is expressed in mg/L $CaCO_3$.
[a] LSL− homes refer to homes without lead service lines.
[b] LSL+ homes refer to homes with a high probability of lead service lines.
[c] WT refers to water temperature after 3 min of flushing (°C).

located). Tap level was used as an ordinal variable (0 = basement, 1 = first-floor, 2 = second-floor, 3 = third floor). The neighbourhood was used as a nominal variable. The construction year was coded 1 for homes built before 1950 and 0 otherwise. The choice of this cut-off was motivated by the fact that we had very little residence built after 1975 (n = 21). Dichotomization to 1950 was the only categorization ensuring a parsimonious regression model because it allowed having enough residences in each stratum to the regression. The 'type of residence' was stratified before analysis into two categories: 1 = single-houses (n = 24) and 2 = others (row-houses (n = 71) and multi-level houses (n = 5) were combined in this category). The total number of people living in the home was used as an ordinal variable. The period from September 10 to December 15 was defined as "fall", based on water temperature cut-off below 10 °C. 'Winter' was from December 16 to March 27 and 'summer' from June 22 to September 06. Before analyses, two additional dichotomous variables were created: the variable 'TIME' referred to the lead concentration observed in different campaigns (1 = WLL observed in the second campaign (summer), 0 = WLL observed in the first campaign (fall or winter)), and the variable 'REFERENCE PERIOD' referred to the period of first sampling (homes whose first sampling was from September 10 to December 15 were coded as 0 and those with a first sampling from December 16 to March 27 were coded as 1). In the absence of data on water usage, the 'type of residence' and the 'number of persons living at home' were included into the regression models in order to control the influence of water usage on seasonal changes in WLL.

2.5. Statistical analysis

The Kruskal–Wallis test was used to compare mean WLL across seasons, and the Shapiro–Wilk test to assess Gaussian distribution of outcome variables. When the assumption of normality was rejected, a log transformation was then undertaken. Analyses of covariance (ANCOVA) were used to obtain geometric means of WLL and to test the differences across dwelling types (or number of residents).

The influence of seasonality on household WLL was assessed using two approaches. The first approach was based on the cross-sectional sample (n = 305). In the case of the 100 homes for which a follow-up analysis was carried out, only summer data were considered in this analysis, leading to 70 fall data, 135 winter data, and 100 summer data. The variation of WLL throughout the year was modelled using a parametric sine function. This approach was very useful to estimate the daily changes in marginal mean of WLL throughout the year, as predicted by the model. It was assumed that temporal fluctuation in WLL could be well described by the following function: $WLL = A * Sin[(2\pi / 365.2) * (Days - B)] + C$. The variable 'Days' was numbered from 0 to 360 with the null value assigned to the first day of recruitment (September 10, 2009). The non-linear regression of the sine function was used to estimate the parameters (A, B and C). 'A' represents half of the difference between the highest and lowest WLL values. 'B' is the horizontal shift reflecting changes in the 'Days' variable. It is determined by calculating how many days the starting point of a standard sine curve has moved to the right (positive value) or left (negative value). Parameter 'C' gives the vertical shift, by showing how many units the modelled function is moved up (or down). Smoothing analyses (non-parametric) were also performed using GAM and TRANSREG procedures. For assessing the adequacy of fit of models, results from the parametric and non-parametric modelling were compared by using mean absolute percentage error (MAPE). The pseudo-R^2 was also reported.

The second approach was conducted as a longitudinal analysis on the subset sample with a follow-up measure of WLL during summer (n = 100). For this analysis, each home had two values of WLL: a first value (baseline) from the first campaign (fall or winter, depending on the date of the first visit) and a second value from the second campaign (summer). The magnitude of seasonal changes in WLL, as well as the temporal change profile of the 4-L samples collected after stagnation, was estimated by using the general linear mixed model with REPEATED statement. The mixed model was preferred over the ordinary least squares model since the latter fails to control for random effects and has limitations for assessing regression estimates from longitudinal data (Ugrinowitsch et al., 2004). For all the follow-up analyses, the restricted maximum likelihood method was used to choose the appropriate variance/covariance matrix. The beta coefficient obtained for the variable 'TIME' indicated the marginal difference in WLL between the first and the second campaign when REFERENCE PERIOD is equal to zero (i.e. in homes sampled during 'fall'). On the other hand, the beta coefficient obtained for the variable 'REFERENCE PERIOD' indicated the marginal difference in WLL between 'fall' and 'winter', when TIME is equal to zero (i.e. during the first campaign). All analyses were performed using SAS software (version 9.3 SAS Institute Inc., Cary, NC). For estimating the effect of seasonality on WLL, separate analyses for fully flushed, first-draw, and 30MS2-4 were carried out. As part of each analysis, a separate stratified analysis was conducted for LSL+ homes and LSL− homes.

2.6. IEUBK modelling

The modelled WLL was applied in the Integrated Exposure Uptake Biokinetic model (IEUBK win1_1 Build11) in order to predict the seasonal changes of BLL. The IEUBK model allows estimating the predicted mean of BLLs at each point, as well as the expected percentage of exposed children's population exceeding the BLL threshold of 5 μg/dL. Background values of exposure to lead from soil, dust, air, and diet were set according to Deshommes et al. (2013). These values were validated as reflecting well the background exposure in Montreal young children background exposure. The batchrun mode of IEUBK was used according to Deshommes et al. (2013). The WLL entered in the batchrun mode was changed every week of the children life from 0 to 84 months of age, according to the WLL model predictions. Finally, children drinking tap water were considered for the modelling, therefore 0.741–1.0 L/d water intakes were considered as detailed in Deshommes et al. (2013). Based on the previous study reporting the marked influence of the type of residence on the lead concentration in tap-water, and given the small number of single-houses in our dataset, we performed IEUBK on both the whole sample (n = 305) and the subset sample without single-houses (n = 240).

3. Results and discussion

3.1. Characteristics of homes sampled

Several factors have been shown to influence WLL, the type of residence being overwhelming. In the presence of a LSL, higher WLL is generally measured in single-family homes compared to other dwelling types (Deshommes et al., 2013). Such differences are explained by the fact that the LSL is shared between several families in the case of multiple dwelling types (duplex, triplex, etc.) as compared to single-family homes. In terms of exposure, the higher number of users lowers the probability to be exposed to high WLL after stagnation. In addition, the configuration, length, and volume of premise plumbing and LSL are more variable than in single-family homes.

The characteristics of the pools of households monitored in this study are shown in the supplemental material, Table S2. The homes under study were mostly row-homes (66.9%), including paired-duplexes, paired-triplexes, and townhouses. Also, single-family homes with a LSL were few represented as only 30 were sampled during the first campaign and 24 of them during the second campaign. Therefore, the type of residence sampled was quite uniform and consisted mostly of multiple family dwellings. In addition, the households not sampled in the second campaign did not differ from the 100 households sampled in the second campaign for the other parameters studied except temperature. Indeed, as shown in the Supplemental material, Table S3, except for the neighbourhood, the distribution of other fixed variables (e.g., flow-rate, type of residence, floor where tap was located, total number of residents) did not markedly differ when homes sampled in the second campaign were compared with those with no repeated data.

3.2. Water lead concentrations

From September 10 (2009) to March 27 (2010), the geometric mean of WLL after 5 min of flushing was 0.22 μg/L (95% CI: 0.06–0.78) in LSL− homes versus 2.19 μg/L (95% CI: 0.55–8.81) in LSL+ homes. During the summer months (from June 22 (2011) to September 06 (2011)), flushed samples showed a geometric mean of 0.26 μg/L (95% CI: 0.11–0.63) in LSL− homes and 3.03 μg/L (95% CI: 1.06–8.69) in LSL+ homes. The distribution of lead concentration in stagnant samples is fully described in Table 2.

The unadjusted comparison of WLL in different samples showed a significant difference in median WLL across seasons, regardless of the presence/absence of LSLs (Supplemental material, Table S4). Summer data indicated that average lead concentration from the first and second-draw samples exceeded the Health Canada's MAC in 45.5% of LSL+ homes (versus 0.0% in LSL− homes) (data not shown). In addition, the lead concentration after 5 min of flushing remained ≥10 μg/L in 28.6% of LSL+ homes. Therefore, these differences highlight the importance of further study of BLLs in link with WLLs. The wide seasonal variations of lead concentrations highlight the need for predicting the seasonal exposure through tap water, especially during summer and early fall corresponding to higher water temperature and maximum lead dissolution. Moreover, the wider differences between lead concentrations measured by different sampling protocols in warm water suggest that the selection of sampling protocol may be critical to the evaluation of the exposure through tap water during peak exposure periods. Therefore a predictive seasonal model was developed and the impact of sampling protocol on the exposure and resulting BLL was investigated.

3.3. Influence of seasonality on household water lead levels

3.3.1. Results from cross-sectional data

The seasonal patterns of changes in fully flushed and composite stagnant samples (average concentration of the four samples collected after 30 min of stagnation) are shown in Fig. 1 for both LSL− homes (n = 128) and LSL+ homes (n = 177). The estimated coefficients corresponding to the sine non-linear regression (and describing the predicted values of WLL over the year) are presented in the Supplemental material, Table S5. Water lead concentrations vary substantially according to temperature in LSL+ houses and to a much lesser extent in LSL− houses. The nonlinear regression model shows average concentrations after 5 min of flushing ranging from 1.67 to 8.22 μg/L in LSL+ homes (Fig. 1b). The magnitude of winter-to-summer changes in average concentrations corresponded to 6.55 μg/L in LSL+ homes and merely 0.30 μg/L in LSL− homes. Predicted values from smoothed curves and nonlinear regression are fairly similar with excellent MAPEs (0.09 for LSL+ homes, 0.18 for LSL− homes). The MAPE value of 0.09 indicates that the predicted water lead concentration from our model would be similar to the one obtained from the smoothed curve, in 91% of cases. Therefore, the average changes in water lead in LSL+ homes can be well-predicted from our model. For the composite sample, predicted largest average lead concentrations in summer reached 1.10 μg/L in LSL− homes (Supplemental material, Fig. S2a) and 14.32 μg/L in LSL+ (Supplemental material, Fig. S2b). The magnitude of average changes in water lead concentrations from winter to summer reached 10.55 μg/L in 'LSL+ homes' and remained very low (0.36 μg/L) in 'LSL− homes'. MAPEs comparing smooth and sine functions were 0.13 and 0.14 for 'LSL+ homes' and 'LSL− homes', respectively. It has previously been shown that single-houses impacted lead concentration in drinking water (Deshommes et al., 2013). In the presence of LSL, higher WLL is generally measured in single-family homes (compared with row- or multi-level homes). This is mainly due to (1) the configuration of the service lines and; (2) the consumption patterns of water usage since the service line only serves a single household. In single-homes, it is relatively simple to control water usage and apply reference stagnation. Because of the presence of multiple households on a single service line, sampling with controlled stagnation is only possible if all residents restrain from any water use, which is difficult to verify. Therefore stagnation in samples in multiple family dwelling may vary from 0 to 30 min. In terms of exposure, this translates for the residents as there is lower probability to be exposed to high WLL. We noted that our sample included a very small number of single-homes, which make it difficult to obtain a valid statistical analysis in this subset of homes. Because of these important features, we performed the same analyses after excluding the subset of single-homes from the dataset (sensitivity analyses). Fig. 1c and d and the related non-linear regression show that the regression coefficients remain largely unchanged but the model fit is improved significantly (Supplemental material, Table S6). Similar figures for stagnant samples are shown in Supplemental materials, Fig. S2c and S2d. It is likely that the true seasonal changes in WLL are more important in single-homes. The fact that these houses represent a low proportion of our sample (<21%) may explain the lack of change

Table 2
Percentiles and geometric mean (GM) for lead concentration in different water samples in μg/L during the first and the second campaigns.

	First campaign[a] (n = 305)						Second campaign[b] (n = 100)					
	p10	p25	p50	p75	p90	GM (95% CI)	p10	p25	p50	p75	p90	GM (95% CI)
LSL− homes[c]												
5MF[d]	0.10	0.16	0.22	0.32	0.51	0.22 (0.06–0.78)	0.14	0.18	0.28	0.34	0.42	0.26 (0.11–0.63)
30MS1[e]	0.28	0.44	0.70	1.22	1.51	0.71 (0.16–3.19)	0.27	0.43	0.64	0.98	1.32	0.64 (0.21–1.95)
30MS2	0.21	0.31	0.55	0.80	1.19	0.50 (0.12–2.05)	0.21	0.27	0.46	0.77	1.06	0.47 (0.14–1.56)
30MS3	0.16	0.24	0.40	0.59	1.02	0.40 (0.10–1.60)	0.20	0.26	0.39	0.66	0.81	0.41 (0.15–1.60)
30MS4	0.14	0.25	0.36	0.51	0.94	0.37 (0.09–0.47)	0.21	0.27	0.35	0.56	0.78	0.39 (0.13–1.16)
LSL+ homes[f]												
5MF	1.00	1.46	2.19	3.12	5.33	2.19 (0.55–8.81)	1.58	2.09	2.90	4.16	5.95	3.03 (1.06–8.69)
30MS1	1.83	2.61	3.42	5.37	7.05	3.60 (1.07–12.12)	2.69	3.49	4.73	7.13	8.63	4.86 (1.87–12.64)
30MS2	1.64	2.57	3.17	5.35	7.98	3.53 (0.93–13.45)	2.55	3.56	4.28	7.07	10.35	4.78 (1.68–13.65)
30MS3	1.74	2.44	3.64	5.96	8.31	3.77 (0.85–16.67)	2.48	3.14	4.48	7.45	11.41	5.01 (1.44–17.39)
30MS4	1.30	2.25	3.29	6.89	13.98	3.83 (0.67–22.07)	2.34	3.18	5.01	8.95	15.57	5.51 (1.38–22.01)

[a] Refers to home samplings from September 10, 2009 to March 27, 2010.
[b] Refers to home samplings from June 22, 2011 to September 06, 2011.
[c] LSL− homes refers to homes without lead service lines.
[d] 5MF refers to the flushed sample.
[e] 30MSj refers to j^{ième} sample collected after a stagnation time of 30 min.
[f] LSL+ homes refer to homes with a high probability of lead service lines.

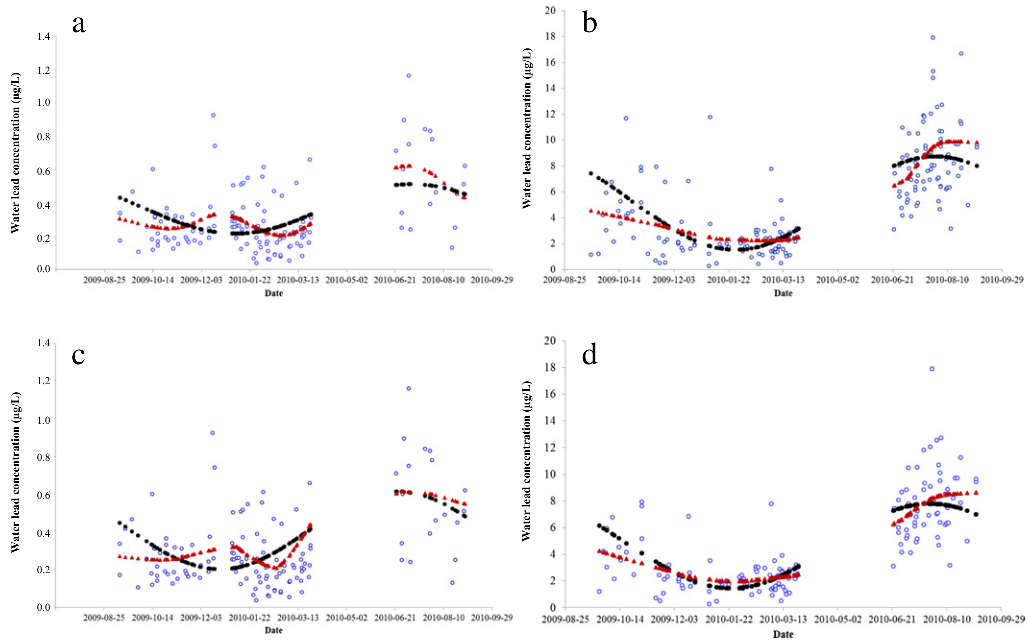

Fig. 1. Temporal variations in water lead concentrations in the flushed sample in homes without (left) and with lead service lines (right), based on the whole sample (above) and after excluding single-houses (below). Observed values (blue circles), smooth curve (red triangle) and predicted values from non-linear regression (black dot).

in regression coefficient in the sensitivity analysis. According to the 2001 Census data, compared with row-houses and apartment buildings, single-homes are a low fraction of total residences in Canada's metropolitan areas, including Montreal (<33%)(Statistics Canada, 2008). Therefore, the distribution of 'type of residence' in our sample is representative of homes in Montreal.

3.3.2. Repeated sampling results

The cross-sectional part allows the estimation of the predictive values of WLL as expected day after day. We could then deduce mean differences in WLL between minimum 'fall/winter' and maximum summer concentrations. However, the characteristics of the households used for transversal sampling conducted during fall, winter and summer may influence the results or trends observed. Indeed, homes included at different seasons may differ in some characteristics unrelated to seasons and which could markedly influence the WLL. In this case, the effect of seasonality may be biased. The follow-up analyses were useful to validate the previous model.

Results from the mixed linear regression are presented in Table 3. The cross-sectional effect at baseline (which represents the mean difference in WLL between 'fall' and 'winter') was very small in LSL− homes. The value of the regression estimate was 0.04 µg/L for the flushed sample ($p = 0.63$). Over the warm months, flushed sample showed a mean increase of 3.60 µg/L ($p < 0.01$) and 0.36 µg/L ($p < 0.01$) in LSL+ and LSL− homes, respectively.

In LSL+ homes, the magnitude of the summer effect on the flushed sample was more marked in homes previously sampled in 'winter'. They showed a discernible additional increase of 2.35 µg/L ($p < 0.01$) in the mean of lead concentrations when compared with homes first sampled in the 'fall' and after adjusting for flow rate, the interval time between two measurements, neighbourhood, construction year, type of residence, the total number of people living in the home, and the floor level where the tap was located. The total increase from winter to summer was then 5.95 µg/L. This winter-to-summer change is very similar to results obtained from the cross-sectional study. Obtaining the same results from both the cross-sectional and follow-up study designs increases the likelihood that these findings are valid, and confirms the adequacy of our non-linear regression. This similarity also suggests that there was no strong selection bias operating in our cross-sectional data. The observed summer effect represents an increase of approximately 150%, given that the mean 5MF observed in LSL+ homes during the winter months was 2.40 µg/L. Some authors argued that lead concentration in the 1 L sample drawn after 5 min of flushing is primarily influenced by the length and the diameter of the LSL (Cartier et al., 2011). However, these parameters are not varying seasonally and the results obtained in our follow-up study support that seasonal-dependent factors play an important role independent of length or diameter of pipes. Given the follow-up design of our study, we believe that the observed lead concentration changes can be mainly attributed to the change in water temperature.

In LSL− homes, the increase of lead concentration in summer in the flushed sample was very low but statistically significant ($\beta = 0.36$ µg/L; $p < 0.01$). In addition, the pattern of changes from baseline (fall or winter) to summer was similar when comparing homes first sampled in the 'fall' and those with first sampled in the 'winter', irrespective of the sample considered. Moreover, the cross-sectional effect at baseline was very small. The slight increase we observed may be due to lead released from solders. Although the use of LSLs for drinking water systems was prohibited in 1975 under the National Plumbing Code of Canada (NPCC), it should be noted that lead solder was allowed to be used in household plumbing until 1986 under the NPCC (Health Canada, 2009).

The summer mean increase of lead in the first-draw samples was observed in both LSL− ($\beta = 0.49$ µg/L, $p < 0.01$) and LSL+ homes ($\beta = 3.28$ µg/L, $p < 0.01$). However, the increase observed in LSL− homes was very low. Moreover, compared with the fall-to-summer change, the winter-to-summer change was more important only in the LSL+ homes. Homes previously sampled over the winter showed an additional increase of 2.52 µg/L ($p < 0.01$), when compared with those previously sampled in the fall. Assuming an absence of contamination at the water source, the changes in WLL may indicate potential contribution of tap itself or lead-based materials contained in the portion of plumbing (fittings, valves, components) close to the tap. The marginal summer

Table 3
Adjusted[a] summer effect on water lead concentration (in μg/L) in the follow-up study (n = 100).

	'LSL−' homes[b] (n = 20)			'LSL+' homes[c] (n = 80)		
	Estimate	Standard error	p-Value	Estimate	Standard error	p-Value
5MF[d]						
Intercept	0.22	0.13	0.1156	8.64	1.51	<0.0001
Time[e]	0.36	0.06	<0.0001	3.60	0.48	<0.0001
Reference period[f]	0.04	0.08	0.6300	−0.76	0.68	0.2624
Reference period ∗ time	−0.11	0.10	0.2803	2.35	0.59	0.0002
R-square	76.84%			71.16%		
30MS1						
Intercept	−0.25	0.89	0.7818	5.09	2.84	0.0773
Time	0.49	0.13	0.0017	3.28	0.70	<0.0001
Reference period	0.15	0.36	0.6978	−1.17	1.19	0.3267
Reference period ∗ time	−0.26	0.17	0.1543	2.52	0.85	0.0042
R-square	48.20%			51.68%		
30MS2-4[g]						
Intercept	0.03	0.49	0.9491	12.73	3.58	0.0007
Time	0.45	0.12	0.0017	3.21	0.77	<0.0001
Reference period	0.15	0.18	0.4129	−1.09	1.60	0.4966
Reference period ∗ time	−0.10	0.17	0.5510	4.21	0.94	<0.0001
R-square	54.96%			49.17%		

[a] Adjusted for flow rate, interval time between two measurements, neighbourhood, type of residence, construction year, total number of people living in home, and floor where tap was located.
[b] Refers to homes without lead service lines.
[c] Refers to homes with a high probability of lead service lines.
[d] Refers to the first 1-L sample collected after 5 min of flushing.
[e] This variable was coded as 0 and 1 for water lead concentration observed in the first and second campaigns respectively.
[f] This variable refers to the period of first sampling: homes whose first sampling was from September 10 to December 15 were coded as 0 and those with a first sampling from December 16 to March 27 were coded as 1.
[g] Refers to the arithmetic mean from the second, third and fourth 1-L samples collected after a stagnation time of 30 min.

effect on lead concentration in the first-draw samples was statistically significant (β = 3.28 μg/L; $p < 0.01$), thereby suggesting the contribution of premise plumbing sources (faucet and interior plumbing) on seasonal changes in WLL. A previous study reported that premise plumbing can contribute 20–35% of the total lead measured at tap (Sandvig et al., 2008). Older homes are more likely to have LSLs, but also leaded solder and other lead bearing materials. The increase in water temperature and the higher water consumption observed during warm months are associated with an increase in lead released from the LSLs, the faucet, the leaded solder and other lead bearing materials. It is more likely that the increase of lead concentration in the first-draw samples probably reflects the direct contribution of premise plumbing and/or LSL.

3.4. Water temperature and lead concentration in flushed sample

We observed a positive linear relation between baseline-to-summer change in lead concentration in flushed sample and baseline-to-summer change in water temperature (Supplemental material, Fig. S3). Our model suggests that 34% of changes observed in lead concentrations are explained by changes in water temperature. We expect an increase of 0.25 μg/L in lead concentration for each increase of 1 °C in water temperature. In a previous study conducted in Montreal (Canada), Cartier et al. (2011) reported similar association between water temperature and lead concentration in flushed sample.

We also observed that water temperature at household level was highly correlated with the water temperature at treatment plant (Supplemental material, Fig. S4). The latter is then a good approximation of the water temperature as observed at tap-level after 3 min of flushing, with a slight bias of 2.64 °C. Detailed investigation on a subset of 34 houses included in the study showed that the total volume in the premise plumbing varied from 2.7 to 9.9 L (outlier of 23 L) with an average of 6.3 L. In all cases, a 3 minute flush prior to measuring temperature was sufficient to measure the temperature of water entering the service line or in the service line.

3.5. Impact of seasonal changes in water lead levels on blood lead levels of young children: results from IEUBK model

In May 2012, the BLL of concern (i.e. 10 μg/dL) has been revised, and the Advisory Committee on Childhood Lead Poisoning Prevention established the 'reference value', currently 5 μg/dL (U.S. CDC, 2012), to identify U.S. children needing more clinical attention and public health follow-up. The estimated BLLs following IEUBK model runs are shown in Fig. 2. The seasonal change in BLLs is evident from our results. If the stagnant water is routinely consumed (we herein considered the mean of four 1 L-samples collected after 30 min of stagnation), the resulting mean BLLs predicted during the summer months (June, July and August) are 3.4 μg/dL in children aged 0.5–1 year and decrease with increasing child's age (2.4 μg/dL for those aged 6–7 years). The magnitude of change in BLLs (i.e. difference between the summer peak and the lowest value) did not strictly differ with child's age, regardless of the exposure scenario (from 0.88 μg/dL in children aged 6–7 years to 1.02 μg/dL in those aged below 1 year). However, the general trend was towards a decrease of both maximum and minimum BLLs with age. Given that all other external sources of lead were kept stable across seasons, the observed changes in BLLs are mainly attributable to changes in WLL.

Although the magnitude of change appears to be independent of child's age, the change in the percentage of children with BLLs ≥5 μg/dL from colder to warmer months was largest in children aged below 2 years, and progressively declines with increasing child's age (Fig. 3). Not surprisingly, the 'best' scenario (lowest exposure) was observed by assuming that children consumed flushed water exclusively. The consumption of stagnant water represented the worst exposure scenario. By assuming that children aged below 2 years routinely consume stagnant water, at least 20% of children aged 0.5–2 years were predicted to develop BLLs ≥5 μg/dL during warm months (the value was 12% by assuming that children consume exclusively flushed water). This result suggests that, by flushing the water for at least 5 min before consumption, we can reduce by about 40% (from 1/5 to 1/8) the proportion of young children with BLLs exceeding the threshold of 5 μg/dL during summer (assuming that all other sources of lead exposure are kept constant across seasons). The difference between the best and the worst scenario was attenuated with age, suggesting that flushing habits would be more effective to prevent elevated BLLs in younger children. In an intervention study examining the effect of flushing, Fertmann and co-workers showed that flushing water before consumption and using bottled water could lower the BLLs in young women by about 0.82 μg/dL and 1.23 μg/dL, respectively (Fertmann et al., 2004). This corresponded to a reduction of 21% and 37% of the initial BLLs, respectively. However, their study included young women rather than children, and the duration of flushing was not determined. We are unaware of previous studies reporting the impact of changes in household WLL on BLLs using IEUBK model (except for study from Deshommes et al., 2013). A recent study suggests that the percentage of students predicted to exceed BLLs of 5 μg/dL could drop from 11.2% to 4.8% due to remediation by flushing at school level (Triantafyllidou et al., 2014). This corresponds to a reduction of 42.9% in the proportion of student with BLLs ≥5 μg/dL, which is fairly similar to the reduction computed in this study.

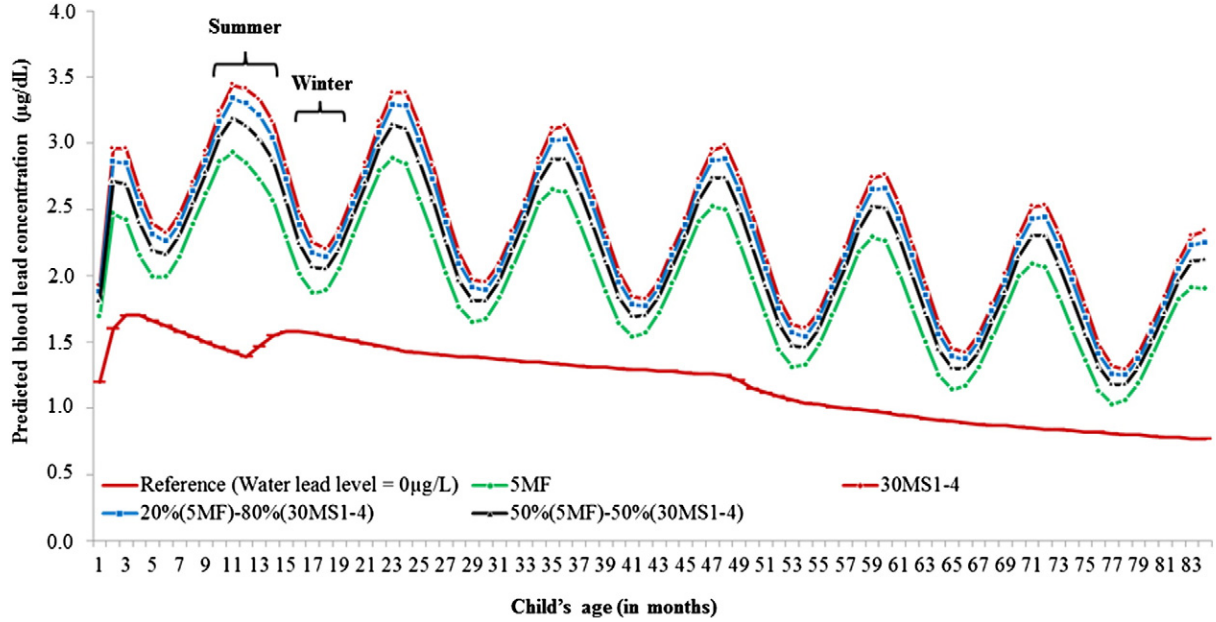

Fig. 2. Predicted distribution of blood lead concentration by child's age and for different exposure scenarios. 5MF: the child is exclusively exposed to water collected after 5 min of flushing; 30MS1-4: the child is exclusively exposed to the mean of the four 1-L samples collected after 30 min of stagnation; 20%(5MF)–80%(30MS1-4): the child consumes 20% of flushed sample and 80% of stagnant sample; 50%(5MF)–50%(30MS1-4): the child consumes 50% of flushed sample and 50% of stagnant sample.

Sensitivity analyses performed after excluding single-homes indicate that children living in row- or multi-level houses are less likely to show BLLs ≥ 5 μg/dL during summer months (Supplemental material, Fig. S5). Results suggest that, by flushing water at least 5 min before consumption, the proportion of children exceeding 5 μg/dL during summer will still remain under 10.5% whatever the child's age is. Moreover, flushing habits could reduce the likelihood of having BLLs ≥ 5 μg/dL in warm months by at least 40% in children aged 0.5–2 years. Finally, if the WLL was set to a constant value of 0 μg/L, the resulting percent of children exceeding the threshold of 5 μg/dL was very low (1% for children aged 0.5–2 years and less than 1% for those aged above 2 years). The resulting mean BLLs predicted between June and August were 1.5 μg/dL in children aged 0.5–2 year and decrease with child's age (0.8 μg/dL for those aged 6–7 years). This represents BLLs from other lead sources.

As a whole, the summer increase in household WLL as observed in this study could impact children's BLLs. Another noteworthy implication is linked to the health of developing foetus and pregnant women living in cities with certain water lead problems. Previous studies indicated that pregnancy and lactating period are associated with marked

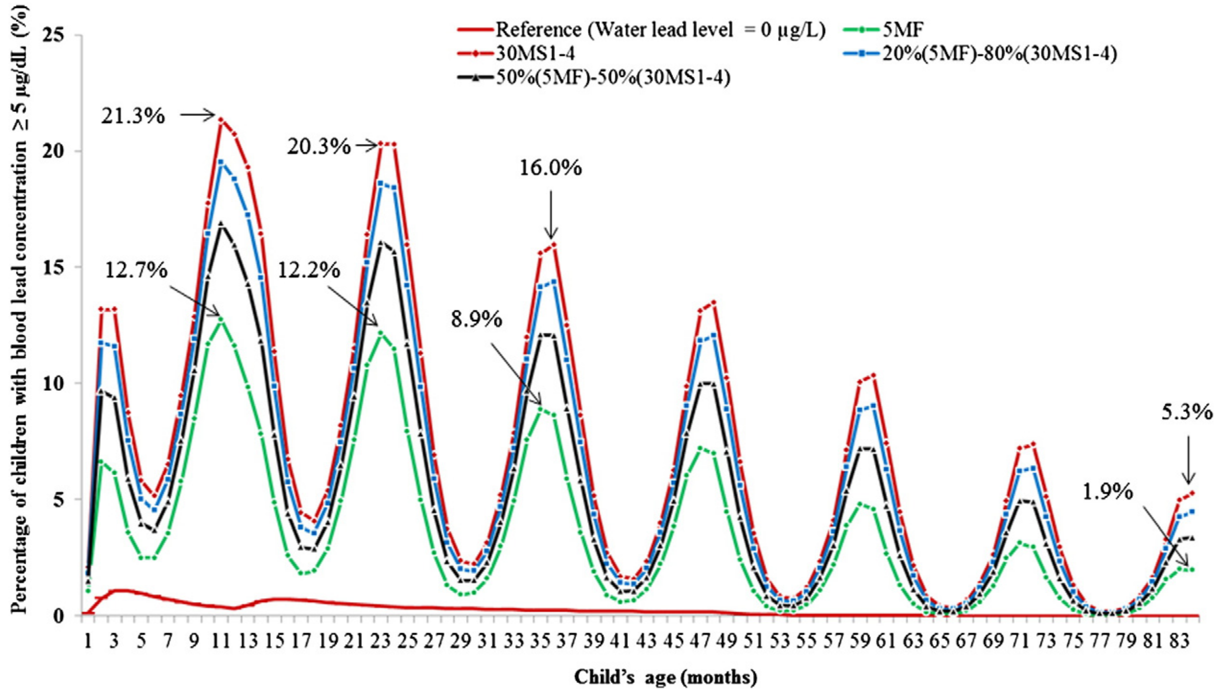

Fig. 3. Predicted distribution of percentage of children with blood lead concentration exceeding 5 μg/dL by child's age and for different exposure scenarios. 5MF: the child is exclusively exposed to water collected after 5 min of flushing; 30MS1-4: the child is exclusively exposed to the mean of the four 1-L samples collected after 30 min of stagnation; 20%(5MF)–80%(30MS1-4): the child consumes 20% of flushed sample and 80% of stagnant sample; 50%(5MF)–50%(30MS1-4): the child consumes 50% of flushed sample and 50% of stagnant sample.

increase in water intake (Ershow et al., 1991; Moya et al., 2014), and that water lead is an important determinant to pregnant women's blood lead (Baghurst et al., 1987; Fertmann et al., 2004). Given the important impact of blood lead to pregnant women and foetus in terms of preterm delivery (Taylor et al., 2014), low birth weight (Andrews et al., 1994; Nishioka et al., 2014) and early neurobehavioral development (U.S. NTP, 2011), even at low maternal blood lead, extension of trends observed in this work to pregnant and lactating women population would be surely interesting for protecting the health of that specific population.

Some study limitations need to be considered when interpreting our results: First, in the multi-level residences/row-houses, although faucets were closed during the stagnation time, we were unable to verify that water was not used in related residences. However, by adjusting for the "type of residence", we believe we corrected for the bias, if it exists. Second, we are aware that there may be other uncontrolled parameters strongly associated with WLL. For example, actual data on water usage were not available. The increased water consumption generally observed during the warm months will reduce the contact time of water with the water distribution pipe surfaces. This can play a major role on the observed lead levels at the tap. Water usage patterns are, at least partly, determined by the number of persons living at home, the type of residence, and possibly other factors (Gregory and Di Leo, 2003). By adjusting for the type of residence and the number of persons living at home, we limited the potential bias due to the absence of data on water usage. Moreover, in the Montreal area, changes in water usage patterns over the year are not evident in the row- or multi-level houses, which represent 90% of homes included in this study. Although the marked difference in WLL was observed between single-homes and row-homes at each campaign, additional analyses indicated that the change in WLL from the baseline to summer is influenced by neither type of residence nor number of persons living at home (Supplemental materials, Tables S7 & S8). The production flow rates at the drinking water plants serving the study area do not support a seasonal change in household water usages. Actually, demand values in hydraulic modelling of these areas are not adjusted for season as industrial demand and leakage are significant in this specific system. It is certainly plausible that increased household usage in summer could lead to lower stagnation times in the service lines and inner plumbing sections, but there is no data to suggest such seasonal trends in the study area. We believe that several features of our study design control for actual drinking water consumption in the household, which is the most important aspect to consider for exposure. Other household demands do not vary significantly in the types of buildings i.e. external uses for pools, gardens etc. are minimal in this dense urban area. Third, data reported here applies to a single distribution system with its proper water quality parameters and hydraulic conditions. They may not fully be generalizable to other distribution system with different characteristics. Finally, IEUBK modelling does not take into account the influence of nutritional intake which could markedly attenuate the changes in BLLs from season to season. The complex interaction between lead ingested from drinking water and nutrients makes it difficult to predict changes in BLLs with high accuracy. Moreover, this model does not consider the time spent outside (specifically at the daycare) and then assumes that the child consumes the same water (either flushed or stagnant), at the same quantity every day.

4. Conclusion

The influence of seasonality on lead concentration in household water is substantial in homes connected to lead service lines. The winter-to-summer change could reach 6 μg/L in flushed samples and 10.55 μg/L in stagnant samples. IEUBK child blood modelling predicts that such changes would lead to a slight increase of about 1 μg/dL in 0.5–7 year-old children. Although such a little increase, the percentage of children aged 1–2 years exceeding the 5 μg/dL BLL threshold could increase by at least 15% due to winter-to-summer changes in water lead levels. Flushing habits reduce the probability for elevated BLLs, especially in children aged below 2 years.

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2014.07.005.

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