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Analysis and assessment of human lead exposure from drinking water and the influencing factors associated with lead

Zakiyeh Namrotee[a], Zsuzsanna Bufa-Dőrr[b], Viktória Finta[b], Bálint Izsák[b], Ágnes Sebestyén[b], Károly Törő[b], Márta Vargha[b,*]

[a]*Doctoral School of Environmental Sciences, Eötvös Loránd University, Pázmány Péter sétány 1/C, H-1117, Budapest, Hungary, Tel. +970598314086; email: znamrouti@gmail.com (Z. Namrotee), Tel. +3614761173; emails: dorr.zsuzsanna@nnk.gov.hu (Z. Bufa-Dőrr), finta.viktoria@nnk.gov.hu (V. Finta), izsak.balint@nnk.gov.hu (B. Izsák), sebestyen.agnes@nnk.gov.hu (Á. Sebestyén), toro.karoly@nnk.gov.hu (K. Törő)*

[b]*Department of Public Health Laboratories, National Public Health Centre, Albert Flórián út 2-6., H-1097, Budapest, Hungary, Tel. +3614761173; email: vargha.marta@nnk.gov.hu (M. Vargha)*

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ABSTRACT

Lead exposure via drinking water is still a major public health concern, mainly in older buildings serviced by lead pipes. Lead concentration can vary widely both in space and time even within a building, however, generally applied monitoring schemes fail to capture the full extent of this variability. The objective of this study was to identify highest risk points within a 4-storey public building and gain better understanding of the drivers of in-building variations in lead concentration. First draw (RDT) and 1 min flushed (F) samples were taken at each tap (n = 56) in the building in two sampling periods (summer–spring). In total, 220 samples were analyzed. Lead concentration exceeded the regulatory limit value (10 µg/L) in 62% and 32% of the RDT and F samples respectively. Non-compliant samples were found in every storey of the building, indicating the extensive presence of lead pipes. However, lead concentrations were significantly higher on the upper floors flushing reduced lead concentration in the majority of the cases, but was often insufficient for reaching compliance. Other water quality parameters varied in a narrow range and had limited impact on lead leaching. Results confirmed that in-building variability of lead in drinking water can exceed two orders of magnitude. Representative sampling point in large buildings for single-sample monitoring schemes should be designated at a regularly used tap on the upper levels of the building. Sampling in the warmer months, and collecting pairs of first draw and flushed samples also assist reliable estimation of lead exposure via drinking water.

Keywords: Lead; Drinking water; Water safety, Health risk

1. Introduction

Drinking water is one of the most important health determinants. Its constituents and potential contaminants have a positive or negative health effect depending on their characteristics and concentration. The effect of some parameters, including lead, can be influenced by conscious decisions of the consumers. Lead is a toxic heavy metal mainly introduced to drinking water through corrosion of

* Corresponding author.

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lead-containing plumbing materials. High lead levels in drinking water are still a major concern for older homes serviced (at least partially) by lead pipes. Lead has long been recognized as toxic metal; it was one of the first pollutants to receive widespread attention for its health impacts, such as disruption of kidney function, interference with the synthesis of haemoglobin, deterioration of the immune system and miscarriages [1].

According to the Guidelines on Drinking Water Quality published by the World Health Organization (WHO), the recommended provisional limit of lead in drinking water is 10 μg/L (ppb) [2]. This limit is adopted as the drinking water standard in many countries including Canada, Australia, China, and the European Union, while in the United States the intervention value is 15 μg/L [3].

1.1. Lead in drinking water

Water consumption contributes to an estimated 10%–20% of the general population's total lead exposure, but can occasionally be the dominant source of exposure [4].

Source waters generally have relatively low lead content. Rare exceptions may be found, as river waters sometimes contain detectable lead levels from industrial discharges of acidic mine drainage. However, water treatment can efficiently remove lead even in these cases. Groundwaters generally have very low lead concentration. The main source of lead is the water distribution system, which can also affect the water quality. Traditionally water pipes were constructed from lead, ductile iron, cast-iron, and asbestos cement. Lead service pipes were commonly used in many countries up to the 1980s to connect between the water mains and the buildings where drinking water is consumed (houses, apartments, institutional buildings, and industrial premises). The main advantages of lead pipes are resistance to corrosion (compared to iron) and their malleable nature, which minimized fracture and leakage under changing ground conditions.

Lead pipes were also used extensively premise plumbing to convey drinking water to points of use up to the 1950s, when it was superseded by copper piping. In some countries, lead pipes have been gradually replaced by copper or plastic within domestic dwellings during refurbishment, particularly in the modernization of kitchens [5]. More recently, the use of plastic piping (e.g., MDPE, medium density polyethylene) is almost universal in premise plumbing.

Brass fittings, such as elbows, connectors, and valves are also very common in conjunction with copper piping. Brass manifolds have also been used to distribute drinking water to a group of dwellings from a single mains connection.

Galvanic corrosion can induce the leaching of high and often erratic concentrations of lead into drinking water. Low pH and aggressive anion gradients in the proximity of wrapped solder joint galvanic connections can also mobilize locally high concentrations of dissolved and particulate lead. Some plasticizers in plastic pipes (e.g., PVC) also contain lead, and may be a source of lead in drinking water. Such plasticizers are no longer in use and while the problem is still detectable, it is not associated with high-level lead leaching [5].

1.2. Health perspectives

Lead is one of the most intensively studied hazardous compounds of the twentieth century. The continuous exposure of millions of persons worldwide to lead in the environment forces an understanding of the short- and long-term hazards of lead, especially its role in causing or contributing to chronic diseases, including cancer [6].

The best indicator of the concentration of lead in soft tissues that reflect recent and past exposure is the lead in blood, whereas bone lead in vivo reflects long-term uptake and body load. A non-specific biomarker for lead exposure is the inhibition of heme metabolism. Lead in drinking water was shown to correlate with lead level in blood (BLL) by numerous studies and in turn, clinical effects were associated with different BLL, of which neurological effects in infants (reduction of IQ) are the greatest concern [7].

Environmental lead poisoning is still considered the principal environmental health threat to preschool children in industrialized countries, particularly for infants and toddlers. The gender, age, iron and calcium metabolism and the general immune status play an important role [8,9]. Lead has a disproportionate effect in childhood because their behavioral patterns make them more sensitive to lead exposure, their bodies absorb more of the lead they ingest, and they exhibit lead toxicity symptoms at lower levels of exposure than adults do. Many organs of the children are still immature, and thus much more sensitive on toxic effects [10].

1.3. Control of lead in drinking water

Plumbosolvency is the dissolution of lead in drinking water. The extent of lead leaching is influenced by various water quality characteristics, including pH, alkalinity, orthophosphate concentration, corrosion inhibitors [11], water hardness (calcium and magnesium), dissolved oxygen, oxidation–reduction potential, and the concentration of ammonium, chloride and sulphate [12]. Soft acidic water is the most plumbosolvent. The amount of lead dissolved from the plumbing system also depends on the temperature and stagnation time of the water [5].

Lead dissolution from lead pipes is exacerbated by organic matter, particularly humic and fulvic acids, also by iron causing "red-water" discoloration. These may need to be minimized if fully effective plumbosolvency control is to be achieved by corrective water treatment. Lead dissolution in low alkalinity waters can be significantly reduced by elevating the pH of water to between 8.0 and 9.0. This however is insufficient for meeting modern-day standards for lead in drinking water [1].

Actions intended to improve water quality can produce serious unintended consequences – especially in the areas of corrosion, stability of existing pipe scales, and aesthetics. Preventing corrosion of water supply infrastructure is an important objective for the drinking water community [1].

Lead in tap water is usually measured at a single selected sampling point within a building. This, however, does not necessary reflect the water quality of the entire building, especially in large facilities with a complex premise plumbing system. Due to the variation of the factors above, lead concentration and consequently exposure of the consumers can vary significantly even within a building. Lead pipes in premise plumbing often only partially replaced, which can further increase internal variability of lead concentration. This research aims to assess the extent of variation in lead concentration within a building, and identify areas where lead exposure risk is elevated by understanding its relationship to water quality parameters and other characteristics (such as location or water use) that influence the lead content of drinking water and thus affect the distribution of lead in the building. This research was conducted as part of project in the National Public Health Centre aiming to assess lead exposure via drinking water on a national level (EFOP-1.8.0. – VEKOP-17-2017-00001).

2. Methods

The period of the study was planned to cover two seasons with different ambient temperature (summer and spring). The study site was a four storey public building in Budapest, Hungary, built in the 1930s, where the lead pipes of the original water system were partially replaced. Sampling points were selected in every room in the four floors where there was a tap ($n = 112$), including the kitchens where most water is used for drinking and cooking purposes. To determine the lead concentration in water stagnating in the system, two samples were taken from each location, a random daytime (RDT) sample (i.e., taking the first liter of the water upon opening the tap) and a flushed (F) sample, taken after flushing the cold water for 1 min. The following parameters were measured from the samples: pH, specific electrical conductivity, redox potential and temperature.

The determination of the metal parameters is done by an inductively coupled plasma ion source mass spectrometer (ICP-MS) using a standard method (ISO 17294-2:2016). Lower limit of quantification for lead was 1 μg/L. Water quality parameters pH, conductivity and redox potential were measured by a WTW Multi 3430 SET F instrument (WTW, Germany) according to the corresponding national standards (MSZ 1484-22:2009 and MSZ EN 27888:1998, respectively. Temperature was measured by a calibrated thermometer. All analysis was performed at National Public Health Centre. Microsoft Excel™ and Statistica™ programs were used to analyze the data. Statistical analyses were carried out for the collected data to understand the relationship between the lead content and the other factors related to the seasonality, building and the water quality parameters. The impact of sampling method and the season of sampling was analysed by t-test, differences by floors and room types by Kruskal–Wallis non-parametric test and correlation with water quality parameters by Pearson correlation analysis.

3. Results

In total, 220 samples were analyzed in two sampling events (summer and spring) from 56 sampling points (two taps were out of service on the second sampling occasion). Overall, lead concentration exceeded the regulatory limit value (10 μg/L) in almost half (47.2%) of the samples. The median concentration was 8.9 μg/L. Extreme concentrations (>100 μg/L) were detected in 8.6% of the samples. The effect of different variables is analysed below.

3.1. Effect of the sampling method on the lead content

Random daytime (RDT) samples a sample that is taken at a random time of a working day directly from the tap in a property without previous flushing [5]. When the sample is taken, the tap should be fully opened or as open as possible without losing the sample. Stagnation of water in the domestic distribution system influences the concentration of lead in a random manner. It is common practice to select sampling points at random and take 1 L sample volume. Flushed (F) samples are taken after prolonged flushing of the tap so stagnation of water in the domestic distribution system does not influence the concentration of lead. In practice, a sample is taken after flushing at least three times the volume of the plumbing. In large, complex buildings, this might be difficult to determine, so in this study samples were taken after 1 min flushing.

The results of pairwise comparison indicate that in line with previous expectation flushing generally reduced lead content compared to the first flush (Fig. 1). The mean lead concentration was 22.3 μg/L (range <1–2,870) and 4.3 μg/L (range <1–412) in the RDT and F samples, respectively. The difference was significant (dependent t-test, $p < 0.001$). 38% of the sampling points were compliant with the parametric value 10 μg/L, even without flushing. Flushing reduced the lead concentration at almost all taps, and it was sufficient to decrease the lead concentration below parametric value in about half of the non-compliant sites. However, in 3 samples the lead levels increased after 1 min of flushing. This effect was observed in higher (2nd–4th) floors of the building. It is likely that in the complex water distribution system of an old public building 1 min was insufficient to fully flush stagnant water from the system. In some seldom-used taps, flushing might mobilise sediment from the pipes containing high levels of lead, thus resulting in increased lead concentration in the water. Though in the present building the effect of prolonged flushing was not measured, at other sampling sites the effect of 1 min and 10 min flushing was also tested. Results indicated that the first minute of flushing was the most relevant, reducing lead concentration by 90% on average. Further flushing for 10 min did not result in significant additional decrease (1.2–4.0 μg/L). Such extensive flushing is not realistic in everyday use.

Flushing taps is a general good practice before consuming water, but optimal flushing time also depends on the materials in the plumbing systems. For instance, in Australia the current Environmental Health Standing Committee (enHealth) advice recommends flushing kitchen taps for 30 s each morning, because lead can leach into water that has been in contact with brass plumbing fittings for an extended period. A study of 108 Sydney households identified that a five to ten second flush was sufficient to reduce lead concentrations below the Australian Drinking Water Guidelines in all kitchen tap water samples. This finding supports a revision of the enHealth recommendations, as a five to ten second flush not only meets public health requirements but also is also more realistic for customers to achieve and increases water savings. However, as the current study shows, this might be insufficient in other places where lead originates from still existing lead pipes and not only brass components [13].

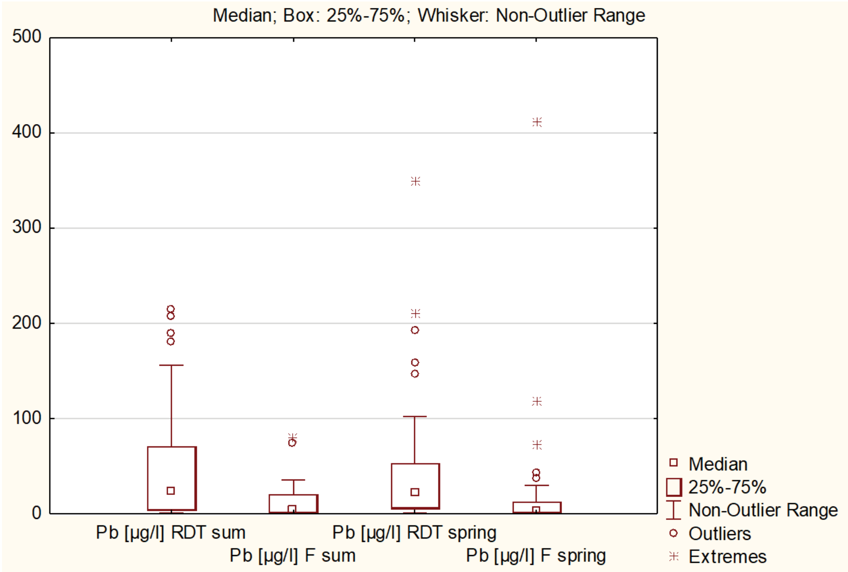

Fig. 1. Effect of the sampling method and the season of sampling on the lead concentration in drinking water. RDT: random daytime sampling, F: fully flushed samples; Sum: August sampling; spring: March sampling; n(RDT) = 110; n(F) = 110.

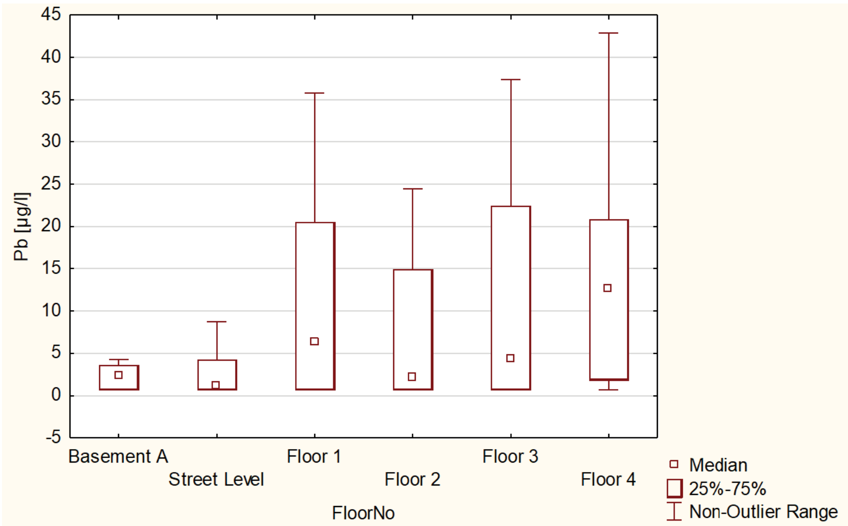

Fig. 2. Effect of the building levels on the lead concentration in drinking water. Flushed samples. n(basement) = 12; n(street level) = 16; n(Floor 1) = 22; n(Floor 2) = 23; n(Floor 3) = 14; n(Floor 4) = 25.

3.2. Effect of the season on the lead content

The mean lead concentration of samples collected in August and March were similar, 8.8 and 8.9 µg/L, respectively (Fig. 1). Water quality in the spring was more favourable, than in the summer: the ratio of stagnant samples with extremely high concentrations was lower (13% vs. 18%) and the rate of compliance after flushing was higher (72% vs. 64%, respectively). However, the difference of lead content in different seasons was statistically not significant, neither in the stagnant or flushed samples. Previous studies also found significantly higher lead leaching in the summer months compared to colder periods [14]. The lack of difference in the present study is most likely to be due to similarity in water temperature (Table 2). Since the sampled building is not fully air-conditioned, in the summer ambient temperature is often above 25°C. However, similar high water temperatures were measured in the spring as well, probably due to the lack of insulation on the pipes. Summer holidays might have contributed to the higher number of extremes in the summer, as many people are on leave during August, and thus water consumption is lower.

3.3. Effect of the floors and room type

The building where the experiment was carried has four floors, ground level and basement. The rooms included in the study were used for different purposes (bathrooms, showers, kitchens, offices, laboratories, etc.). Samples were taken from each tap in the rooms. Results of non-parametric Kruskal–Wallis test indicated significant differences between the different levels of the building (Fig. 2. Table 1), while the room type did not affect the lead concentration (Fig. 3).

Generally, lead concentration is expected to be higher on the upper floors, if lead pipes are present, since the water travels through lead pipes for a longer distance. Flushing is also expected to be more efficient in the lower levels due to the smaller volume of stagnant water. This hypothesis was generally reflected by the results, lead concentration in the basement and street level was lower than on Floors 2–4. However, the difference was only statistically significant for the flushed samples (Table 1). Mean lead levels in Floor 3 were lower than expected. According to the building operator, pipes on Floor 3 were replaced in a recent restoration project.

The type of the room has dual importance: water use can be widely different (e.g., less water is used in the offices, than in the toilets or showers). Also, water is usually consumed in the kitchen, therefore that is where the lead concentration is most relevant. In the present study, no difference was observed between the different room types. Extreme high concentrations were not observed in RDT samples the kitchens and toilets, probably due to the more frequent use of the taps.

3.4. Effect of water quality parameters

Water quality parameters were tested in 220 samples to determine the relationship between the lead content and pH, conductivity, redox potential and temperature.

3.4.1. pH

The pH of the samples varied in a narrow range (between 7.38–7.90). Seasonal differences in the pH were not observed (mean value 7.6 for both spring and summer, Table 2). Statistical analysis by Pearson correlation did not indicate significant association between the lead content and the pH value.

Previous studies indicated strong correlation between the pH and lead leaching. pH strongly affects the solubility of lead containing deposits in water pipes [15]. Significant increase in lead corrosion is usually seen at pH < 7. Kuch and Wagner [15] studied the effect of pH on lead concentrations in drinking water delivered through lead pipes. Particulate lead is the primary contributor to total lead concentration in flowing systems. The adjustment of pH is a common corrosion control treatment strategy. Changes to the water supply that increase the water's pH decrease water lead levels by the formation of mineral scale on the inner surface of older plumbing the prevents lead from leaching into drinking water. In the present study, the lead levels were high in several samples spite of the slightly alkaline pH. The absence of association with the pH is probably due to the low variability of the parameter.

The analytical method measures both dissolved and particulate lead; it is possible that in some cases (especially where extreme values above 100 µg/L were detected) large amounts of deposited particulate lead were flushed into the samples. Though in these samples the ratio of dissolved and particulate lead was not determined, previous data generated in the project indicated that under the circumstances and water quality representative of the sampled building, majority (on average 70%) of lead is in dissolved form.

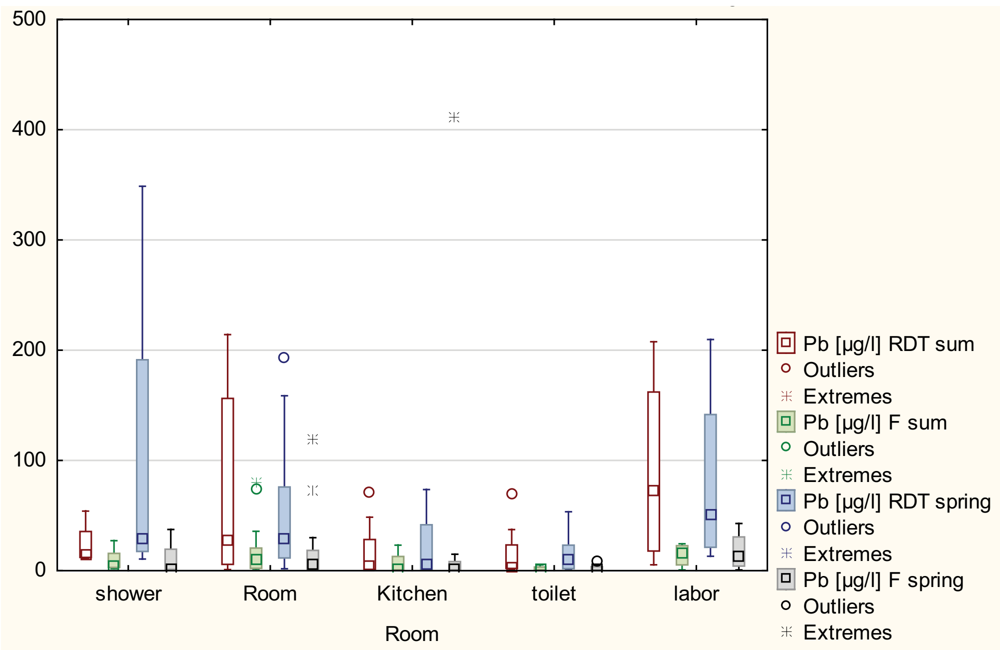

Fig. 3. Effect of the room type on the lead concentration in drinking water, by sampling method and sampling season. RDT: random daytime sampling; F: fully flushed samples; Sum: August sampling; spring: March sampling; n(shower) = 4 n(Room) = 32; n(Kitchen) = 6; n(toilet) = 8; n(laboratory) = 4.

Table 1
Results of the non-parametric Kruskal–Wallis test comparing flushed drinking water samples on different building levels

	Basement	Street level	Floor 1	Floor 2	Floor 3	Floor 4
Basement		1.0000	1.0000	1.0000	1.0000	0.2499
Street level	1.0000		0.2861	1.0000	1.0000	0.0315*
Floor 1	1.0000	0.2861		1.0000	1.0000	1.0000
Floor 2	1.0000	1.0000	1.0000		1.0000	1.0000
Floor 3	1.0000	1.0000	1.0000	1.0000		1.0000
Floor 4	0.2499	0.0315*	1.0000	1.0000	1.0000	

*$p < 0.05$.

Table 2
Water quality parameters (mean [min – max]) by season and sample type

	Summer		Spring	
	RDT	F	RDT	F
pH	7.66 [7.50–7.90]	7.64 [7.40–7.80]	7.61 [7.44–7.84]	7.60 [7.38–7.81]
Conductivity (μS/cm)	440 [424–458]	437 [348–449]	431 [413–450]	431 [410–448]
Temperature (°C)	25.7 [21.5–32.1]	23.4 [20.6–29.1]	19.5 [11.5–31.1]	20.9 [13.3–28.4]
Redox potential (mV)	261.8 [219.9–359.5]	333.2 [243–286.3]	371.4 [266.1–487]	382.6 [266.3–534]

RDT: random daytime, F: flushed.

3.4.2. Conductivity

The conductivity values ranged from 348 to 458 μS/cm. Neither the sampling season, nor the sampling method (RDT or F) affected the conductivity values. Statistical analysis indicated positive association between the lead concentration and conductivity in the RDT samples. However, this correlation does not necessarily reflect a causal relationship. Conductivity reflects the amount of dissolved ions in the water, dissolved lead also contributes to this value, so increased levels of lead (e.g., due to mobilisation of lead-containing sediments) can increase conductivity.

3.4.3. Temperature

Water temperatures were higher in the summer. The mean temperature of the RDT samples was 25.7°C in the summer and 19.4°C in the spring (Table 2). After flushing, mean water temperature was 23.4°C and 20.9°C in the summer and spring, respectively. The observed temperature values indicate that flushing was not always sufficient in removing stagnant water from the premise plumbing, since the temperature of the supplied water is generally 10°C–15°C. The same conclusion was drawn from the comparison of lead concentration in RDT and F samples.

It is a common finding that lead levels in drinking water are higher in the summer than in winter periods, and this effect is usually associated with warmer temperature. The solubility of lead is higher at warmer temperatures [14,16]. The impact of temperature is often investigated in combination with pH. Mohammadzadeh et al. [3] studied the effects of different temperature conditions (20°C vs. 5°C) on dissolution of hydrocerussite and cerussite, the major lead species in drinking water at bench scale under various pH and alkalinity (moderate vs. low) conditions. Highest solubility was found at 20°C, pH < 7. In the present study, the temperature range was much smaller, and higher values did not lead to significantly increased lead leaching.

3.4.4. Redox potential

The mean redox potential of the samples in the summer varied from 261.8 and 333.15 mV in RDT and F samples, respectively. Values were consistently higher in the spring (mean: 371.4 in RDT, 382.61 mV in F samples). Increasing redox potential is generally expected to increase the formation of PbO_2, which has low solubility in water and thus forms particulate lead in the distribution system [17]. However, in the complex interplay of water quality parameters, high redox potential may also lead to increased corrosion and thus elevated lead concentration [18]. In the present study, the association between redox potential and lead concentration was statistically not significant.

4. Conclusions

Lead intake via drinking water can be detrimental to human health, especially infants and young children. Generally applied sampling schemes relying on selected sampling taps fail to capture the extent of variation in lead exposure within a building. Objective of the present study was to investigate these differences by sampling every water outlet in a selected 4-storey building in Budapest, Hungary, as a model for complex public buildings. The building is dated from 1930s and known to contain lead pipes, which have been partially replaced.

Lead concentration exceeded the regulatory limit value (10 μg/L) in 47% of the samples, in some cases reaching extremely high (>100 μg/L) concentration. Non-compliant samples were found in every storey of the building, but the lead concentration was significantly higher on the upper levels, than on the ground floor. Partial replacement of lead pipes in one storey of the building reduced lead concentrations compared to the adjacent floors.

Study results confirmed that flushing for 1 min reduced lead content compared to the first draw, though not in every instance: in rarely used taps flushing can increase lead concentration through the mobilization of lead-containing deposits. In large, complex, multi-storey buildings 1 min, flushing might not be sufficient in removing stagnant water from the pipes, especially on the upper levels. This result was confirmed by the temperature of the water samples, which was generally higher than that of the supplied water even after flushing. However, extensive flushing (up to 10 min) before consumption is not realistic. Understanding the water quality conditions that impact the release of lead in drinking water provides a foundation for making effective treatment decisions. In the present study site, water quality parameters did not have a significant impact on the lea concentration. Extreme concentrations in stagnant water were associated mainly with seldom used taps. Replacement of all lead pipes is expected to provide a final solution in the studied building. Partial reconstruction only locally reduced lead leaching to some extent, and flushing was not always sufficient in reducing lead concentration below the regulatory limit. Based on the results, monitoring schemes relying on periodic single samples within large buildings should be designed to collect samples in summer and sampling taps should be designated on the highest floor of the building to represent the worst case scenario.

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