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Intake of lead (Pb) from tap water of homes with leaded and low lead plumbing systems

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Abstract

Methods of quantifying consumer exposure to lead in drinking water are increasingly of interest worldwide, especially those that account for consumer drinking habits and the semi-random nature of water lead release from plumbing systems. A duplicate intake protocol was developed in which individuals took a sub-sample from each measured drink they consumed in the home over three days in both winter and summer. The protocol was applied in two different water company regional areas (WC1 and WC2), selected to represent high risk situations in England, with the presence or absence of lead service pipes or phosphate corrosion control. Consumer exposure to lead was highest in properties with lead service pipes, served by water without P dosing. The protocol indicated that a small number of individuals in the study, all from homes with lead service pipes, consumed lead at levels that exceeded current guidance from the European Food Standards Agency. Children's potential blood lead levels (BLLs) were estimated using the Internal Exposure Uptake Biokinetic model (IEUBK). The IEUBK model predicted that up to 46% of children aged 0-7 years old may have elevated BLLs (>5 μg/dL) when consuming the worst case drinking water quality (>99%ile). Estimating blood lead levels using the IEUBK model for more typical lead concentrations in drinking water identified in this study (between 0.1-7.1 μg/L), predicts that elevated BLLs may affect a small proportion of children between 0-7 years old.

KEYWORDS: Drinking water, intake, lead, phosphate, plumbosolvency

1. Introduction

Lead is a metal that is known to be neurotoxic to humans, and to have many other deleterious health effects at high levels of exposure. At lower exposure levels, children appear to be particularly vulnerable to environmental lead effects, with an association with intellectual and cognitive outcomes observed at blood lead levels below <10 μg/dL (Tong et al., 2000; Lanphear et al., 2005). Humans are exposed to lead through ingestion and inhalation. The main sources of lead for humans are leaded paint, water contacting lead bearing plumbing, diet, soil, dust and dirt. Although the potential routes for lead entering into the body are relatively well documented, there is still much to understand about the factors determining uptake, particularly around how interactions and genetic factors influence lead absorption (Larsen et al. 2002; Whitfield at al., 2007).

Drinking water has been established as a significant contributor to an individual's overall lead burden, with estimates being that it accounts for on average between 1 and 20% of the total (European Food Standards Agency (EFSA), 2010). For individuals living in properties known to be served by lead plumbing, exposure may be much greater than 20% (Triantafyllidou et al., 2009). Links have been established between high concentrations of lead in drinking water and raised blood lead levels (BLLs) by studies conducted in the US (Edwards et al., 2009; Clark et al., 2014), Canada (Deshommes et al., 2013) and the UK (Moore et al., 1977; Sherlock et al., 1984). Epidemiological and modelling studies have evidenced the importance of drinking water to young children's BLLs, particularly in recent well publicized water quality incidents, for example in Washington DC (Brown et al., Montreal (Levallois et al, 2014) and Flint (Hanna-Attisha et al., 2017). Lead in drinking water has been identified as a problem in other parts of the world, including Australia (Handley et al., 2016) and Hong Kong (Lee et al., 2016).

Lead enters drinking water through leaching from lead pipes and other plumbing fittings, and fittings that contain lead such as solder and brass. In the UK, lead can be prevalent in properties built before 1970 when lead was the preferred material for small diameter water supply pipes and lead based solders were used extensively to join copper pipes in drinking water systems. There are an estimated 9 million homes in the UK that are affected by lead pipes (Hayes, 2010). In Europe, anywhere from <5 to 50% of households are estimated to be supplied with water via a lead pipe (Hayes, 2010). In the US, it has been estimated that approximately 9.7 million houses are supplied by either lead pipes or leaded connection pipes. Furthermore, up to 81 million homes in the US are believed to contain plumbing with lead solder joints (Triantafyllidou and Edwards, 2012). While lead pipes and leaded solder are no longer allowed to be used, homes built in the US up to 2014 can legally contain brass with up to 8% lead.

In many parts of the world, water is treated to minimize the release of lead from pipes, fixtures and fittings. Traditionally, this was through raising the pH of the water to 8.5-9.0 to reduce the solubility of lead. This was effective for meeting historic lead drinking water quality standards (DWQS) of 50 μg/L, but has not usually been adequate for current regulatory standards in Europe and North America. In Europe, the lead standard is 10 μg/L, however recent proposals by the European Union are to reduce this to 5 μg/L (EU, 2018). Similar standards have been proposed in Canada (Health Canada, 2017). In the USA, the lead and copper rule (LCR) stipulates an action level of 15 μg/L for lead based on 90th percentile tap water samples from buildings identified to be at highest risk of elevated lead (Edwards et al., 2009). To meet these more challenging requirements, a combined approach of pH adjustment and dosing orthophosphate (usually in the form of monosodium phosphate (MSP) or orthophosphoric acid) is usually applied. These chemical changes to the water encourage the formation of a very insoluble scale layer on the internal diameter of the pipe, reducing lead leaching into the water and often reducing detachment and release of particulate lead as well.

The approach of pH adjustment and/or orthophosphate dosing has had a significant beneficial effect on lead concentrations in drinking water and is now standard practice for water treatment in Europe and North America. For example, in the UK and US over 95 and 50% of the water supplies dose orthophosphate, respectively (Hayes, 2010; McNeill and Edwards, 2002). While effort has been made to reduce lead in drinking water and link BLLs with water concentrations, there is little knowledge on how much lead is actually being consumed from tap water for different risk groupings. For example, a significant minority of consumers living in properties with lead piping are drinking tap water where there is no orthophosphate dosing. In the UK alone this may be some 450,000 homes out of 9 million properties containing some lead plumbing.

Further, few studies employ accurate methods to estimate individual tap water intake, relating this directly to the lead concentration of the consumed water, instead relying on statistical numerical distributions or consumer retrospective recall to determine individuals' intakes (Gofti-Laroche et al., 2001 and Hynds et al. 2012), and single point estimates of lead concentration. Gillies and Paulin (1983) concluded that, 'the only reliable way to determine mineral intakes from this source is to analyze representative samples of the water actually drunk by the subjects'. Few, if any, studies have done that, particularly in relation to lead intake. The aim of this research was, therefore, to show, through a comprehensive intake study, how much lead was ingested from drinking water by consumers in properties with and without lead plumbing, and for homes receiving water from various sources with and without orthophosphate dosing. A further aim was to compare individual's intake levels in both winter and summer. These intakes were then compared with published guideline lead exposures.

2. Materials and methods

The duplicate intake study was carried out to determine lead consumption from tap water in two different water company operational areas with different water quality in England (WC1 and 2). Locations were selected following discussions with the water company to help identify known risk areas for properties with lead pipes. The water companies and area locations have been made anonymous at their request. Within each regional area, the objective was to select consumers in properties that fell into one of the following four risk categories: (1) Leaded properties (with lead pipes) & non-phosphate (non-P) dosed – these were properties where there was lead plumbing supplying water to, and/or within, the home and received water where the supplying utility did not add phosphate to the water as a plumbosolvency inhibitor; (2) Leaded properties & phosphate (P) dosed – as above but these properties received water where the supplying utility added a chemical phosphate inhibitor to the water to reduce lead dissolution to the water. In all cases, phosphate was dosed into treated water as orthophosphoric acid; (3) Unleaded, control properties (no lead pipes) receiving a water supply that non-P dosed; and (4) Unleaded, control properties (no lead pipes) receiving a water supply that was P dosed. It is acknowledged that these properties may have contained lead in the water from other sources, such as the brass in water meters and fixtures and fittings and solder containing lead. Suitable properties were identified within in each region and risk grouping based on water company knowledge of water supplies that were and were not phosphate dosed, known and probable locations of leaded plumbing and new build areas where no leaded plumbing was present. The same properties were visited in both summer (Jul-Aug) and winter (Oct-Dec). At the time of the study, 1.0 and 0.7 mg/L as P orthophosphate was being added to the water in WC1 and WC2 respectively.

The participating water companies provided street level information with respect to high and low risk areas where leaded and unleaded properties were known to be present for water supplies with and without phosphate dosing. A targeted recruitment campaign was then carried out by the project team in these areas through delivery of an introductory letter, outlining the aims and objectives of the study. A follow up visit was made the following day to provide further information, confirm the suitability of the occupant(s) and property for taking part in the study and sign-up interested householders. Those successfully completing the study were incentivised with a shopping voucher. Approximately, 10-20% of those receiving a letter were recruited to the study. On successful recruitment, participants were given a set of written sampling instructions, recording sheets and a sampling kit comprising sample bottles, a measuring jug for recording drink volumes and a funnel to aid transfer of water to the sample bottles. To help ensure the sampling was carried out correctly, the sampling instructions were also verbally explained by one of the project team and any questions raised were addressed. Each individual in the household was asked to take part in the study, and was asked to collect samples and record the volume of water drunk for each drinking water 'event', over a three-day period, to include at least one weekend day. From this, each participant's water intake was assessed by measuring the volume of water they consumed from each drink, and the total lead concentration in the water in each drink. A water sample was also taken from each household for measurement of routine water quality parameters (pH, UV_{254}, dissolved organic carbon, chloride, sulfate, nitrate, alkalinity).

Sampling was achieved following a duplicate water intake protocol, whereby a duplicate water sample was taken from each drink the participant of the study was about to consume.

Participants filled the cup or glass with the amount of water used for making the drink. If the drink used boiled water, the sample was taken after the water had boiled. The water from the drinking vessel was poured into a measuring jug, and the volume of water was recorded. 125 mL Azlon sample bottles were then filled with water from the measuring jug. The rest of the water from the measuring jug was then returned to the cup or glass, and topped up from the tap or kettle and the drink prepared as usual. Samples were analyzed for lead in laboratories that were United Kingdom Accreditation Service (UKAS) accredited and met the requirements for Drinking Water Testing Specification (DWTS), and compliance with the analytical quality control (AQC) procedures as specified in the Manual on analytical quality control for the water industry (document NS30). For the winter survey, samples were only analysed for total lead. In the revisit sampling in summer, both total and filtrate lead were measured to enable quantification of dissolved and particulate lead. Samples were sent to the laboratory in completely filled 125 mL Azlon bottles as supplied by Severn Trent Services. Samples for total lead were acidified to 1% nitric acid to ensure complete solubilisation of all lead. Samples for dissolved lead were first filtered through a 0.45 μm filter paper prior to acidification. Particulate lead was found from the difference between unfiltered and dissolved lead.

Lead concentrations were measured using inductively coupled plasma mass spectroscopy (ICP-MS). The measurements had a reporting limit of 0.1 μg/L. Samples reported below the limit of detection were recorded with a value of 0.05 μg/L, as is normal practice for such analysis. The instrument was calibrated before each batch of samples were run, using standards in the range 0 to 31.25 μg/l. A calibration check standard was placed at the beginning and end of each run to check for any drift. A control standard and a blank were placed at random intervals throughout the run (at a maximum frequency of every 19 samples).

As children are the most vulnerable group in relation to harmful effects from lead exposure, hypothetical child BLLs were estimated using the US Environmental Protection Agency's Internal Exposure Uptake Biokinetic model (IEUBK), following an approach used in previous studies (Edwards et al., 2009; Akers et al., 2015; Deshommes et al., 2013). The model has been demonstrated to be reasonably accurate at predicting BLLs (Hogan et al., 1998; Mickle, 1998). A number of scenarios were run through the model based on the range of lead concentrations observed in drinking water found in this study. Lead exposures from other sources were kept constant using median values from European surveys and reports (EFSA, 2010) for air 0.005 μg/m^3 and soil 23 mg/kg. IEUBK default values were used for food, between 1.95 and 2.26 μg per day depending on age category. The definition of elevated BLL in children <16 years old is open to some debate. The Centers for Disease Control and Prevention (2012) consider 'elevated' to be 5 μg/dL, while the British Paediatric Surveillance Unit ascribe a value of 10 μg/dL (Ghosh et al., 2014). The World Health Organization state that there is no known safe BLL and that levels as low as 5 μg/dL may impact on the cognitive development of children (WHO, 2015). Levallois et al. (2014) considered elevated BLLs at 1.8 μg/dL. The present study compared the modelled BLLs to these values.

Statistical analysis: Mann-Whitney U tests were carried out for comparisons between data in the sample groupings. Wilcoxon's matched pair tests for differences between winter and summer lead values were carried out for each participant. Kruskal-Wallis tests were carried out for non-parametric comparisons of particulate and soluble lead.

3. Results and Discussion

In total, 48 individuals (7 of these aged under 16) were recruited to the lead study from 23 properties, providing 539 and 570 duplicate water intake samples from drinking water events in winter and summer respectively (see Table 1). Although there was some imbalance in the distribution of properties and individuals recruited to each risk category, enough samples were obtained in each category to enable robust statistical analysis (Fowler and Cohen, 1995).

The volumes of individual drinks consumed were approximately normally distributed, with the highest frequency of drinks consumed being between 0.15-0.30 L, resulting in a median drink volume of 0.26 and 0.25 L for adults in winter and summer respectively. For children the corresponding figures were 0.23 and 0.265 L in winter and summer. The average daily tap water consumption for adults was 1.067 L per day in winter (with a range of 0.17-2.5 L) and 1.32 L in summer (a range of 0.335-4.18 L). For children, the equivalent average figures were 0.48 and 0.46 L. These results compare favorably with results from a water intake study carried out in 2011 (Parsons et al., 2013), which reported a mean tap water intake of 1.29 L for adults and 0.51 L per day for children (0-16 years old).

[Table 1 here]

3.1 Lead in tap water samples

Over the study, 7.4% and 10.1% of all duplicate water intake samples taken in winter and summer respectively, had lead levels that were greater than the current drinking water quality standard in Europe of 10 μg/L Pb. These samples were from three leaded properties sampled in winter and ten leaded properties sampled in summer (23 properties were involved in the study overall).

When these data were split into the different risk groupings for the two different operational regions, the highest lead levels were observed in drinking water in the WC1 region (Figure 1), particularly for the non-P dosed drinking water. The water supplying both the P and non-P dosed properties was acidic and of low alkalinity (see Supporting Information, Table S1 for water quality information), the aggressive nature of which is well known to increase the solubility of lead (Cardew, 2009; Hayes et al., 2010; Liu et al., 2010). It was clear that P dosing had significant benefit on lead concentrations in drinking water from leaded and unleaded properties in WC1 and WC2, and this was particularly the case in WC1 (Figure 1 and Table 2). In WC1, the median lead concentration for the non-P dosed leaded properties was 4.5 and 3.7 μg/L in winter and summer, with more than 38% of samples above the European DWQS. Samples taken from 4 out of 5 properties that fell into this group exceeded the 10 μg/L standard. For the P dosed leaded properties, the median lead level was 0.1 and 0.2 μg/L in winter and summer, with no values >10 μg/L. In WC2, the median lead concentration for non-P dosed properties was 5.7 and 8.5 μg/l respectively, while the equivalent was 1.7 and 2.9 μg/L for P dosed properties. It was apparent that the P dosing was less effective in WC2 than for WC1 with 9.1 and 20.7% of samples being >10 μg/L in winter and summer. In both WC1 and WC2, occasional samples contained very high lead concentrations. In WC1 one sample from a leaded and non-P dosed property collected in summer was 1050 μg/L. The same property provided 14 out of 23 drinks samples that were >40 μg/L. In WC2, the maximum lead concentration observed was 224 μg/L from a leaded and P dosed property, further supporting the view that P dosing into this supply was sub-optimal. In these cases, particulate lead was the dominant fraction present in the sample (>98% particulate lead). These results show higher proportions of lead samples at concentrations >10 μg/L than those seen in tap water from across Europe, obtained from random daytime and fully flushed samples. For example, surveys in the Netherlands and Germany have seen 2 and 3.9-10 % of samples above 10 μg/L (Hayes and Skubala, 2008). In France, 5% of regulatory samples were >10 μg/L (Glorennec et al., 2007). However, it should be noted that in the present case, properties were selected based on lead plumbing being present. It was therefore expected that the lead concentrations would be higher in the tap water from these homes.

Unleaded properties, unsurprisingly, resulted in lower water lead levels than for the leaded properties although there was still benefit from P dosing for reducing lead concentrations. For P dosed properties in WC1 the median lead levels were 0.1 and 0.2 μg/L in winter and summer, while this was 1.9 and 2.7 μg/L for non-P dosed systems. While the levels were much lower than for the leaded properties, these data show that significant lead concentrations can enter into drinking water from sources other than from lead pipes. The likelihood here was that brass fixtures, fittings and water meters, as well as hidden leaded solder, were the source of the lead. While samples were generally low in lead, a significant minority of samples from unleaded properties were >10 μg/L in summer in the non-P dosed properties in both WC1 (7.4%) and WC2 (1.2%). This highlights one of the difficulties that water utilities face in compliance to lead regulations: complete removal of lead piping and plumbing is unlikely to result in complete compliance with lead water quality regulations. One route by which lead can be released into the water when there is no lead plumbing present is from galvanic corrosion, a process whereby lead is released into the water as a result of dissolution from solder or brass (that contain lead) when connected to copper pipes or connections (Nguyen et al., 2011). Galvanic currents form between the two metals, which can lead to a very corrosive environment at this juncture which can then lead to very high levels of metal dissolution, particularly at the anode, which is usually the lead containing material. Lead dissolution rates are enhanced when the relative concentration of chloride to sulphate increases. This increase in the chloride to sulphate mass ratio (CSMR) increases the production of chloride-lead products (such as lead chloride, $PbCl_2$) which are more soluble when compared with sulphate-lead products (such as anglesite, $PbSO_4$). Nguyen et al. (2011) state that waters with a CSMR of <0.2 are of low concern for galvanic corrosion, waters with a CSMR between 0.2-0.5 are of significant concern, and waters with CSMR >0.5 and an alkalinity <50 mg/L as $CaCO_3$ are of serious concern. High CSMR values and higher alkalinities are considered as a significant concern. In this study, apart from in the P dosed area in WC1, all CSMR were significantly higher than 0.2 and were all in the 'significant concern' category. This helps explain some of the relatively high lead levels seen in unleaded properties which were known to have brass water meters in all WC areas, as brass fittings and fixtures have been shown to be a significant source of lead in drinking water (Sandvig et al., 2007).

The data clearly show that higher lead concentrations occur in summer months compared with winter, with all of the very high lead levels (>40 μg/L) observed in summer (Table 2). The largest difference in lead concentration was in WC2 for leaded and non-P dosed properties where the median lead concentration was 1.8 μg/L higher in summer than winter. Within each property category for the different water supply areas, significantly higher lead levels were observed in summer compared to winter (P <0.0001 up to 0.01, Mann-Whitney U test). In nearly every case, the mean water lead concentration in the samples for each participant was higher in summer compared to winter (Figure 2). These differences were highly significant (Wilcoxon's test for matched pairs, p <0.00001) and held true for both leaded and unleaded properties, with and without P dosing. Out of 48 individuals, only 2 consumed water that had lower concentrations in summer than winter, although these were in homes where the mean lead concentration was very low, <1 μg/L. The main causative factor is the increased solubility of lead scales at higher water temperatures (Triantafyllidou and Edwards, 2012). For WC1, the water temperature was 11.7 and 17.2 °C in winter and summer respectively for the P dosed supply and 10.3 and 11.2 °C for the non-P dosed supply. In WC2, the water temperature was 9.0 and 19.5 °C in winter and summer respectively for the P-dosed supply, while it was 12.0 and 13.0 °C for the non-P dosed supply. The non-P dosed water supplies had a significant composition of groundwater, meaning that the temperature differential between summer and winter was much less than for the surface water dominated water sources. Overall, these results are in agreement with a recent study in Canada that identified up to a 6-10.6 μg/L difference between winter and summer lead concentrations in tap water, depending on how the sample was taken (flushed or stagnant) (Ngueta et al., 2014). Experiments involving a full scale pipe rig utilising 'harvested' pipes from Washington and Providence RI, demonstrated the relationship between temperature and lead release from pipes (Masters et al, 2016). There was a correlation of r=0.73 for particulate lead and r=0.70 for dissolved lead, resulting in average particulate lead levels that were six times higher in summer than in winter, and average dissolved lead levels three times higher. It also appeared to indicate that the rate of temperature change affected lead release. These results were not consistent across different experimental pipe loop rigs, or over time, as in some conditions the relationship between temperature and lead release diminished or disappeared after 12 months. The authors speculate that this was due to the formation of insoluble orthophosphate scale. Small scale field sampling in the same study, in eight homes with some lead piping served by the same water supply, did show a correlation between temperature and lead release, but only in half of the homes. This demonstrates the complexity of the relationship of temperature to lead scale dissolution, and that it is only partially understood. Differential thermal contraction and expansion of pipes and scale layers can also lead to fragmentation of scales and the release of particulate lead into the water, but this is likely to give a much more unpredictable output in lead concentration.

The other notable observation from the samples collected from consumer's drinks was the highly variable nature of the lead concentrations in samples taken from the same household (see SI Figure S1 for example distributions from two leaded properties). The largest differences were seen in homes containing leaded plumbing. The household which provided water samples with the highest levels of lead (from WC1 and non-P dosed) contained lead concentrations that were quite stable in winter (range from 13 to 19 μg/L in 20 samples), but much more variable in summer (ranging from 25 to 1050 μg/L in summer). Other households supplied by the same water supply had much lower levels of lead, but saw periodic spikes in lead, for example going from 0.3 to 22 μg/L (Figure S1). These between-drinks variations are presumably a reflection of the differences in patterns of household water usage. The very high lead levels in tap water may be explained by factors such as pipe disruption or changes in water pressure. Stagnant water remaining in contact with leaded pipes for extended periods of time can also lead to elevated lead concentrations. It is widely thought that the water first drawn from a tap following overnight stagnation is likely to contain the highest lead concentrations (Cardew, 2009; Hayes and Hydes, 2012).

To investigate whether consumers' first drinks of the day were higher in lead content they were compared with subsequent drinks. This was not a perfect analysis because the activity of one individual in a household will have an influence on the activity of another in the house. Also, consumers may have different habits in properties that are known to contain lead plumbing following a period of water stagnation. For example, some may flush their taps, as advised by water utilities, while others may not. Those that do flush their taps may do this for a length of time that may not be effective to remove all of the stagnant water. In addition, there may have been other extended periods of water stagnation during the day in households (for example, when people go out to work). However, the results indicate that there were no consistent trends with respect to the 'first draw' samples, with differences in lead levels from these drinks compared to subsequent drinks being normally distributed around zero in leaded properties (Figure 3). This is ostensibly a result of the differences in consumer behaviors, for example flushing or not flushing, and the differences in plumbing systems, such as different lengths of lead pipe being present requiring different times of flushing.

[Figure 1 here]

[Table 2 here]

[Figure 2 here]

[Figure 3 here]

There was wide variability in the form of lead found in the tap water (Figures 4 and 5). In leaded and unleaded homes with P dosing, the proportion of particulate lead in the sample was significantly higher than in systems where there was no P dosing (Figure 4). The mean proportion of particulate lead in leaded non-P samples was 0.54, increasing to 0.70 in P dosed water. Equivalent data for unleaded homes were 0.48 and 0.68 for P and non-P households respectively (Leaded homes: Kruskal Wallis p-value <0.00001; Unleaded homes: Kruskal Wallis, p-value <0.00001). These results show that lead reaching the household tap is approximately 50:50 dissolved to particulate lead for homes without P dosing, while this switches to a higher proportion of particulate lead for homes receiving water that is P dosed, irrespective of whether the home contained lead plumbing or not. Interestingly, there was no consistent overall correlation between the total lead concentration in the sample and the form of the lead present in the sample (Figure 5). It was apparent that for some conditions, high lead concentrations were associated with more particulate lead. For example, for leaded properties served by water dosed with P, all samples above 4.5 μg/L the total lead concentration contained >94% particulate lead. For non-P dosed households with leaded plumbing, samples >90 μg/L total lead were dominated by particulate lead (>67%). Below this concentration there were some very high lead concentrations with a much smaller proportion of particulate lead, for example 83 μg/L containing only 21% particulate lead. In the unleaded properties, for both the P and non-P dosing conditions, the higher lead concentrations were much more variable. For example, the samples above 5 μg/L had between 5 and 95% particulate lead. However, within this data it was evident that there were aligned strings of data for the P dosed unleaded and leaded properties, showing increasing proportions of particulate lead as the total lead concentration increased. Inspection of the data showed that these were samples from the same household. This observation shows that within property lead variation was more consistent with respect to more particulate lead as the total lead concentration increased for the P dosed properties, likely as a result of lead-phosphate scales being released from the plumbing systems. Because each property has a bespoke arrangement of pipes and plumbing, there was significant variation in the relationship between the total lead concentration and the amount of particulate lead.

These results are in partial agreement with the observations of other researchers, who have consistently noted that most lead in tap water is in the particulate form as a result of adsorption of lead onto suspended particles and from the release of scales containing lead (Olson et al. 2017; Del Toral et al., 2013). This was particularly the case for water samples containing high concentrations of total lead.

[Figure 4 here]

[Figure 5 here]

3.2 Lead consumption

The lead concentration and water volume consumed for each participant in the study was converted into a lead consumption. This was reported as an average lead load per person (μg

Pb/day) over the three days of the trial. These data have been reported in frequency histogram plots and split by area and lead control strategy (Figures 6). The consumption data have been considered with respect to the benchmark dose lower confidence limit (BMDL) from the European Food Standards Agency (EFSA) guidance (EFSA, 2010). For adults the $BMDL_{10}$ for nephrotoxicity is the relevant comparator and for children this is the $BMDL_{01}$ based on neurotoxicity. The $BMDL_{10}$ (adult, nephrotoxicity) and $BMDL_{01}$ (child, neurotoxicity) have values of 0.64 and 0.5 µg/kg/day respectively. As weight data was not recorded for participants in this study, body weights of light, average and heavy adults (40 kg: 25.6 µg/day, 65 kg: 41.6 µg/day and 90 kg: 57.6 µg/day respectively) have been taken from the National Health Service database (NHS, 2015). Children's lead consumption is dealt with below due to the availability of an effective model to determine BLLs from lead exposures, including tap water (Mickle, 1998; Triantafyllidou et al., 2014).

As was expected from the lead concentration data obtained, lead consumption from tap water was frequently quite low. When considering the whole data set, 3 out of 48 (6%) of the participants in the study were consuming more than 5 µg Pb/day from drinking water in winter (Figure 6). In summer, this increased to 11 out of 48 participants (23%) (see Supporting Information Figure S2 for winter and summer cumulative distribution probability plot of lead consumption). The lead consumption was highest in the leaded and non-P dosed properties in WC1, in line with the highest lead concentrations observed. The maximum lead consumption from drinking water was 20.5 µg/day in winter, while the same participant consumed 129 µg/day in summer. In WC2, the maximum lead consumed for an individual from a leaded non-P dosed household in winter was 5.6 µg/day. In summer, the highest levels of consumption were from a P-dosed property at 99.6 µg/day, while other members of the same household were also consuming high levels of lead, >3.7 µg/day. This was a surprising result given that the median lead concentrations were lower in the P dosed area. However, while the average lead level was less, there were a number of very high lead levels observed for the P-dosed samples from one property (presumably particulate lead detaching semi-randomly) that resulted in high lead consumption from some single drinks which skewed the lead intakes. In addition, individual daily water intake varied by an order of magnitude across the study (from 166-4183 mL per day) such that big differences in intake were observed for water of similar lead concentration.

For unleaded properties in the P dosed areas very low levels of lead consumption were observed in both WC1 and WC2 in winter and summer (<1.52 μg/day). More significant lead consumption was observed in unleaded and non-P dosed properties. In WC1 up to 10 μg/day was consumed in summer, significantly more than for the leaded and P dosed properties in the same supply region. In WC2, one individual was consuming >5 μg/day lead in summer in an unleaded and non-P dosed home, consistent with the high lead concentrations observed in some of the drink samples.

Lead doses received by individuals in the study were a function of both water consumed and lead concentration. In this regard there were some large seasonal differences in lead consumption (Figure S2). Statistical comparison of participant's winter and summer consumption confirmed the difference to be significantly lower in winter than summer ($Z = -5.69$, $P<0.0001$ Wilcoxon's test for matched pairs). Similar significant differences were found in each household risk category. Although individual drink volume did not change much from winter to summer, the frequency of drinking events did increase resulting in an average 24% increase in water consumed from winter to summer for adults (from 1.06 to 1.31L) . This, combined with the higher concentrations of lead found in water in summer, resulted in the increased lead consumption.

There were also some large variations in lead exposure for individuals in the same household drinking water with the same plumbing. To illustrate, in one household the difference between the highest and lowest individual lead consumed was 0.09 and 0.39 μg/day, a difference of a factor of x4.3. The average lead concentration each was exposed to in their drinks differed by less than a factor of x1.2 (0.24 and 0.20 μg/L) while the amount of tap water consumed differed by a factor of x2.3 (0.45 and 1.57 L/day). On the other hand, spikes in tap water lead concentration also had a significant effect on lead consumption. In a household with a relatively high level of lead in the tap water, two individuals were consuming 0.72 and 0.36 L/day of tap water. The individual with the lower water consumption was exposed to a number of very high concentrations of lead in their drink resulting in much higher lead consumption over the duration of the study: 99.6 μg/day compared to 4.3 μg/day for the other individual. These findings demonstrate the importance of taking consumption factors as well as lead concentrations into consideration.

Two individuals involved in the study were consuming levels of lead directly from tap water that exceeded the BMDL. It is therefore likely that only in a relatively small number of cases that tap water alone will be responsible for consumption of lead that may have potentially damaging health effects. However, it must be considered that tap water only represents a proportion of the total lead consumed by an individual. In the UK, average 'non-tap water' lead contributions from food and drink have been estimated to be between 18-40.5 μg/day (EFSA, 2010). When considered in this overall context, and if it is assumed that other lead contributions remain constant, the input of lead from drinking water becomes much more important for individuals in leaded properties in non-P dosed areas. Here the median lead consumption from leaded non-P dosed properties was 5.3 and 3.1 μg/day in WC1 and WC2 respectively. This was between 5 and 21% of the threshold $BMDL_{10}$ for nephrotoxicity effects for a heavy and light adult respectively. Therefore when other food and drink contributions are considered on top of this, the risk of consuming potentially harmful levels of lead significantly increases. In other words, tap water can be an important contributor to an individual's lead burden and for a number of high risk properties will be the dominant contribution. It has also been shown that food prepared in high lead water, has caused elevated blood lead in cases where children were not directly consuming tap water (Triantafyllidou and Edwards, 2012).

[Figure 6 here]

3.3 Predicted blood lead levels in children

Because few children took part in the study (7 participants <16 years old), the IEUBK model was used to estimate corresponding blood lead levels (BLLs) for the water lead concentrations found from this study. Six different drinking water lead concentrations were used in the model based on the range of lead concentrations observed in the study. It should also be noted that the model outputs do not relate specifically to the children who were part of the study, but shows hypothetically how children might be impacted if they were exposed to the range of measured lead concentrations seen in this study.

Simulations run were the median lead concentration for leaded properties with and without P dosing in WC1 and WC2 (leaded and P dosed: WC1: 0.11 μg/L; WC2: 2.23 μg/L; leaded and non-P dosed (WC1: 4.47 μg/L; WC2: 7.14 μg/L) as well as the highest average lead concentration observed for an individual in WC1 and WC2 (WC1: 53.4 μg/L; WC2: 16.5 μg/L). In these high concentration scenarios, the calculated average excluded the highest single lead concentration observed in a drink (1050 μg/L in WC1 and 220 μg/L in WC2). The model predicts a distribution of BLLs estimated for children <7 years (Figure 7). This is because children of different ages and different physiology respond differently in their uptake of lead (Akers et al., 2015). In addition, individuals have different drinking habits (as shown in this study). There is a baseline BLL as a result of intake from other sources (such as food, soil and air), so for tap water containing no lead, which resulted in a modelled median BLL of 0.6 μg/dL.

For the high water lead concentration scenario (53.4 μg/L), 46% of children were modelled to have elevated BLLs >5 μg/dL and 6% >10 μg/dL. For the next highest lead concentration (16.5 μg/L), 5% of the BLLs were predicted to be >5 μg/dL with no levels greater than 8.5 μg/dL. These model predictions need verification from measurement of BLLs, however the results align well with BLL data collected in Canada from children aged between 1-5 years old that identified that the likelihood of a child having an elevated BLL (in this case >1.8 μg/dL) was 4.7x as great when the concentration of water consumed was >3.3 μg/L (Levallois et al., 2014). In this context, at the low BLL threshold, 98% of children were modelled to have elevated BLLs for the highest water lead concentration (53.4 μg/L), while this dropped to less than 1% for the lowest median water lead concentration observed (0.11 μg/L). Predicted increases in BLL were particularly evident for water lead concentrations above 5 μg/L, in the same order as seen by Levalllois et al. (2014). These modelling results indicate, therefore, that tap water from a small minority of properties with significant leaded plumbing and ineffective plumbosolvency control has the potential to cause elevated BLLs in children.

The modelled results also show the benefit P dosing has on estimated BLLs. For example in WC1, the proportion of the child population with elevated BLLs (>1.8 μg/dL) was modelled to increase from <1 to 10% as the median tap water lead concentration increased from 0.11 to

4.47 μg/L (with and without P dosing). In WC2, P dosing reduced the predicted elevated BLLs from 22% to 4% (the median water lead reduced from 7.14 to 2.23 μg/L). In comparison to these results, one of the earliest research into lead exposure in Europe was a duplicate intake diet study in Glasgow (Lacey et al., 1985). This was carried out at a time when lead plumbing was more widespread, and plumbosolvency control less practiced. In this study, regression analysis of measured BLLs showed the mean BLL of children increased from 5 to 44 μg/dL as the water lead concentration increased from 0 to 500 μg/L, much higher than those modelled in the present work. This shows both how lead concentrations in tap water have been significantly reduced and how exposure to lead from other sources has also reduced, given the high historical baseline BLL when there were low levels of lead in tap water.

[Figure 7 here]

3.4 Implications of the study

The results from the study have shown that lead consumption from tap water covered a range from 0.02 to 129 μg Pb/day. These results are hard to compare given the paucity of information on directly measured lead consumption from tap water. Studies have reported tap water concentrations from spot samples, but do not account for the wide variability in lead concentrations in tap water from a single household and the variability in the volume of water consumed by individuals (Ryan et al., 2000). However, reported exposures varied from 0.005 to 18.13 μg Pb/day for 2 minute flushed tap water samples (Ryan et al., 2000). The World Health Organization report daily lead exposures of up to 10 μg Pb/day for adults for tap water containing 5 μg/L. Our study has shown a much broader range of lead exposure from tap water. As expected, lead consumption increased in leaded properties with no, or inadequate, P dosing.

The data suggest that overall lead consumption rates from tap water were generally low for the average home. However, it was evident that in certain properties where there was lead plumbing and no P dosing, consumers were likely to be drinking more than the BMDL for lead just from drinking water. In these properties, vulnerable groups such as children will be at particular risk of having elevated BLLs. It therefore seems sensible to reduce the lead consumed by reducing the lead concentration in tap water by as much as possible. As lead in water is something that can be evidently controlled by P dosing, water companies should therefore try to ensure that all water supplies have effective P dosing or have comparable corrosion control. Other lead avoidance strategies should also be promoted, including use of flushing or filters.

The study has shown that lead consumption varies significantly with both lead concentration and water consumption. There are limitations to the study that should be acknowledged. Only drinks in the home were considered. Some studies suggest that up to 37% of fluid intake may be consumed out of the home (Kaur et al., 2004). This was mitigated in our study by most people who were involved in the study being at home during the day over the duration of the study, and evidenced by the water volumes consumed being in-line with other water intake estimations (Parsons et al., 2013). There were a limited number of properties involved in the study and these were purposively selected to be either high or low risk. The results should therefore not be considered representative but rather show the range of lead consumption likely to be expected for different risk groupings of households. Other factors such as the impact of disinfection strategy should also be further investigated because changing chemical and the amount of residual disinfectant has been observed to have some effect on lead dissolution from pipes (Boyd et al., 2007). Finally, due to the differences in human physiology, uptake of lead from food and drink varies significantly between individuals. The only true way to establish this is to measure body lead. A consumption study such as that carried out here should be used to focus where blood lead surveys should be carried out.

4. Significant New Findings and Conclusions

The results offer a unique insight into the variability of lead concentrations individuals are exposed to when taking water from the home for different risk categories, with respect to the P dosing regime. Lead intake was then weighted based on actual water consumption, rather than estimates which are highly unrepresentative. This has not been presented previously. This produced a number of previously unreported observations:

- Variability in lead concentrations in household tap water was high and did not follow an obvious pattern with respect to stagnation or consumer drinking behaviour. This variability has not been reported and captured in exposure assessments for the same individuals in the same household.
- The effectiveness of P dosing was very different in the two regions studied, with some very high lead concentrations observed.
- Water consumption increased in summer by 24% and lead concentrations were lower in winter. Other methodologies would not, and do not, pick up these differences.
- Variability in lead consumption within a household was clearly demonstrated. For example, two adults in the same home were consuming vastly different levels of lead (100 μg/day compared to 4.3 μg/day), primarily driven by different consumption rates.
- The results provide a methodology for better assessing human exposure to lead at the tap, which in turn, could serve as a basis for improved cost:benefit analysis and policies protecting consumers from water lead risks.

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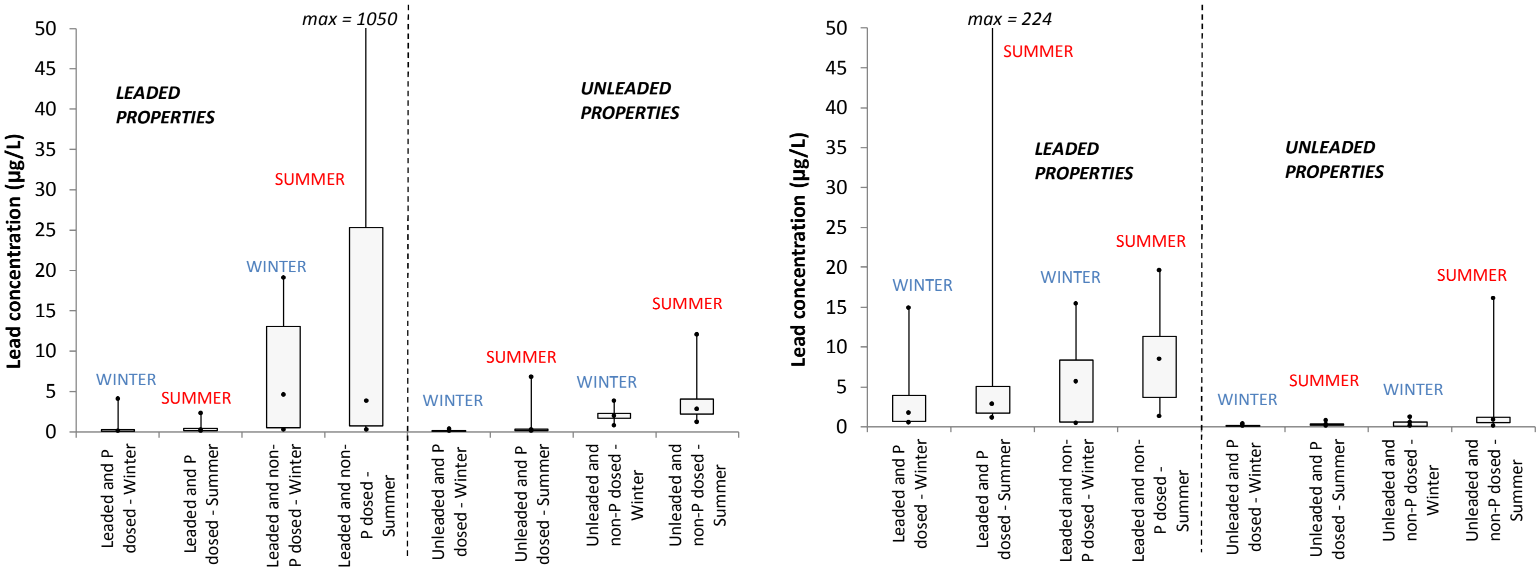


WC1 WC2

Figure 1. Box, tail and whisker plots for lead concentrations in samples taken from WC1 and WC2. The central marker represents the median lead concentration, the upper and lower bounds of the box represent the 75th and 25th percentile lead concentrations respectively and the lines at either end of the box, the 'whiskers', go to the extreme values for the lowest and highest lead levels

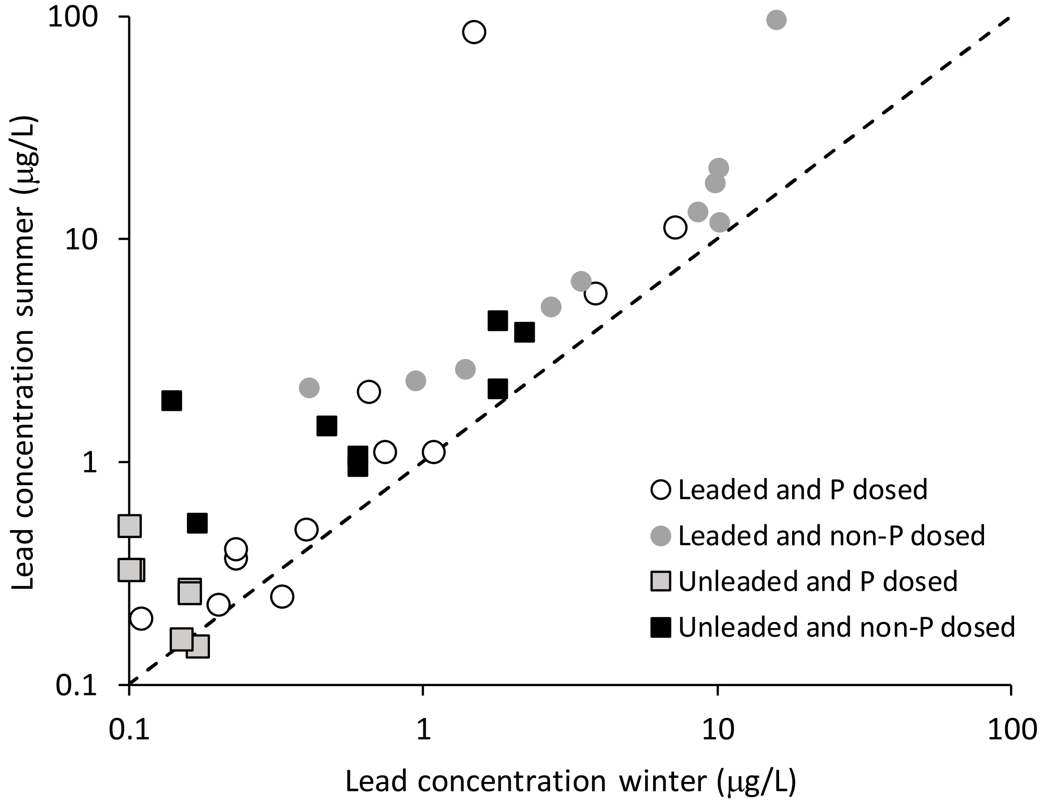
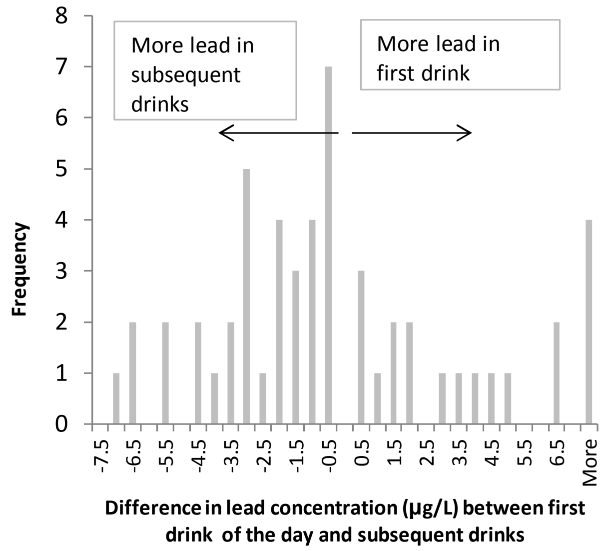
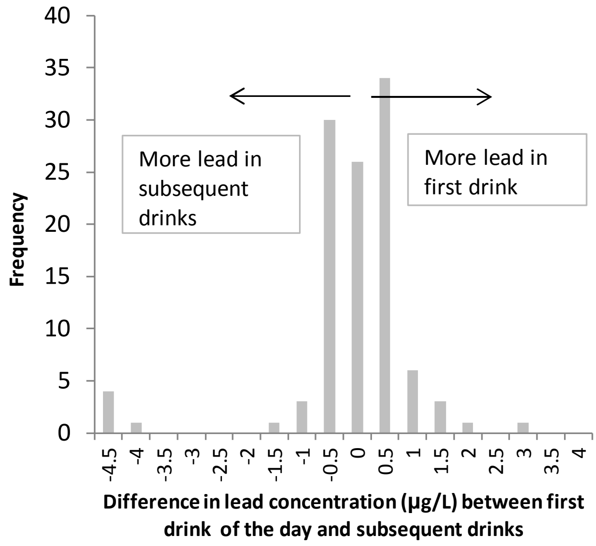


Figure 2. Difference between mean lead concentrations in drinks consumed in winter compared to the summer for each participant in the study.



Leaded and P dosed properties



Leaded and non-P dosed

Figure 3. Difference in leaded properties in lead concentration for first drink of the day and subsequent drinks.

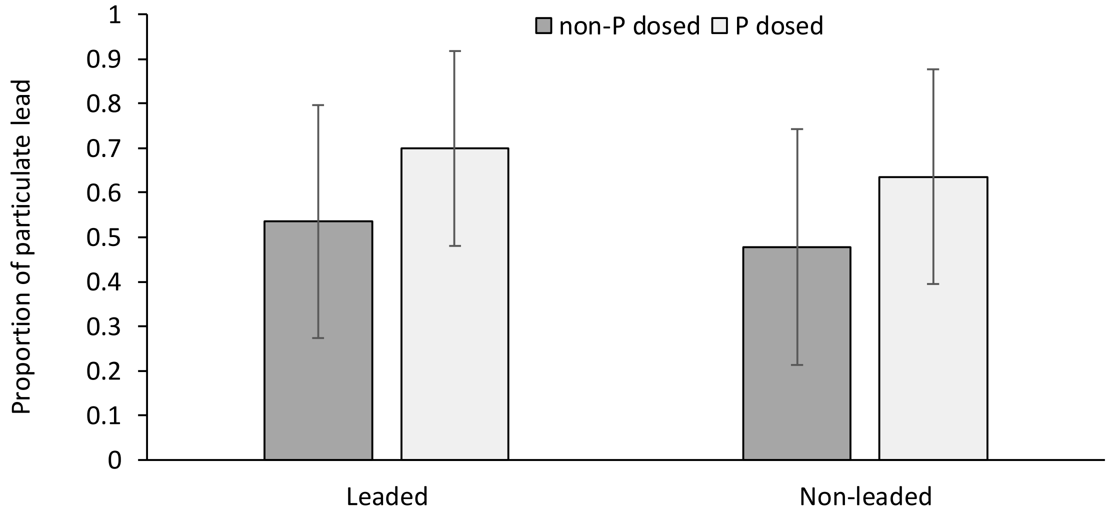
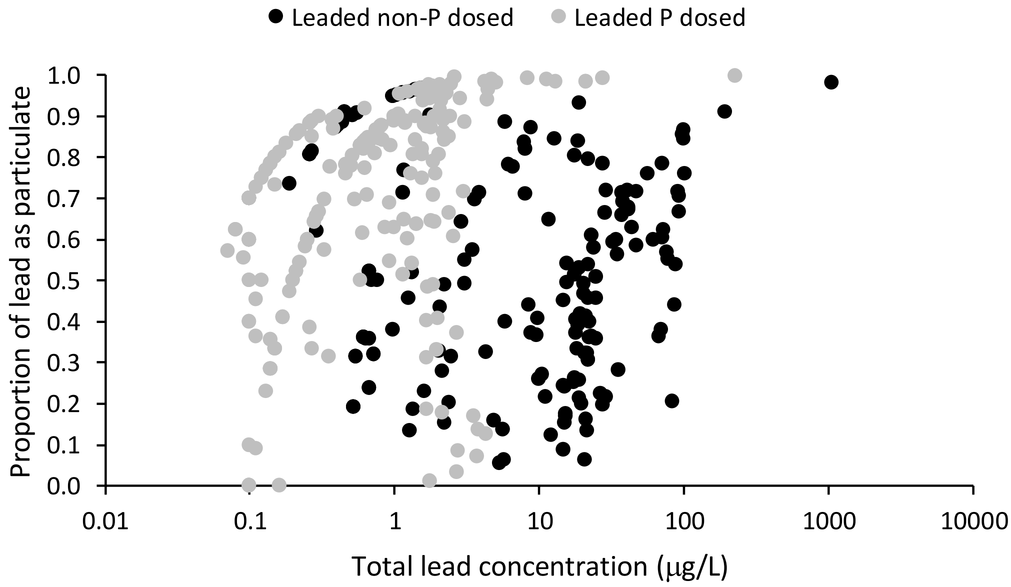
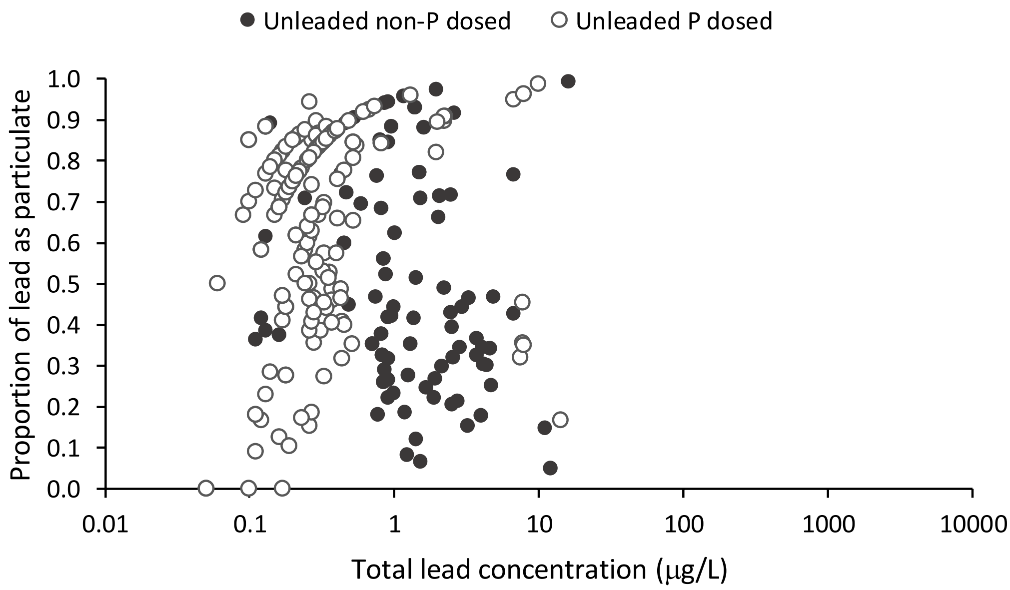


Figure 4. Proportion of particulate lead in all samples for leaded and unleaded samples, with and without P dosing.

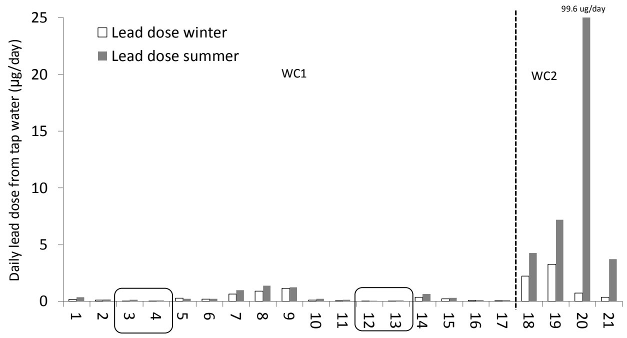


Leaded properties

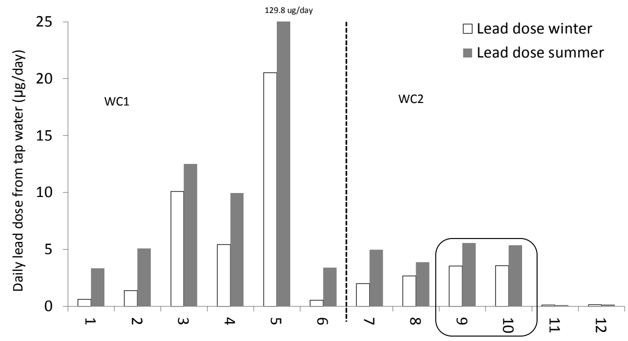


Unleaded properties

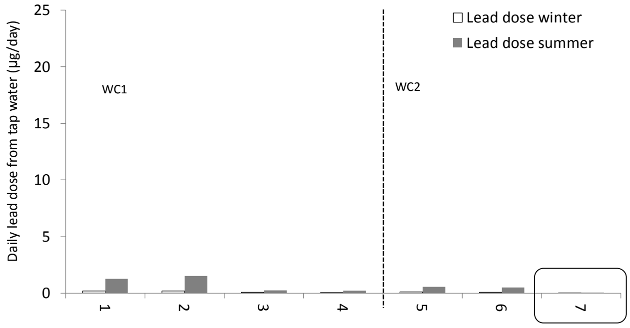
Figure 5. Proportion of particulate lead in samples for unleaded homes, with and without P dosing as a function of the total lead concentration in the sample.



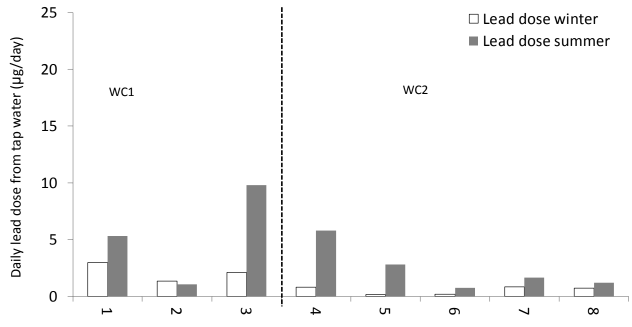
Leaded and P dosed



Leaded and non-P dosed



Unleaded and P dosed



Unleaded and non-P dosed

Figure 6. Daily lead consumption all individuals in the study in the different risk categories of property. Data enclosed by a box indicate child participants in the study.

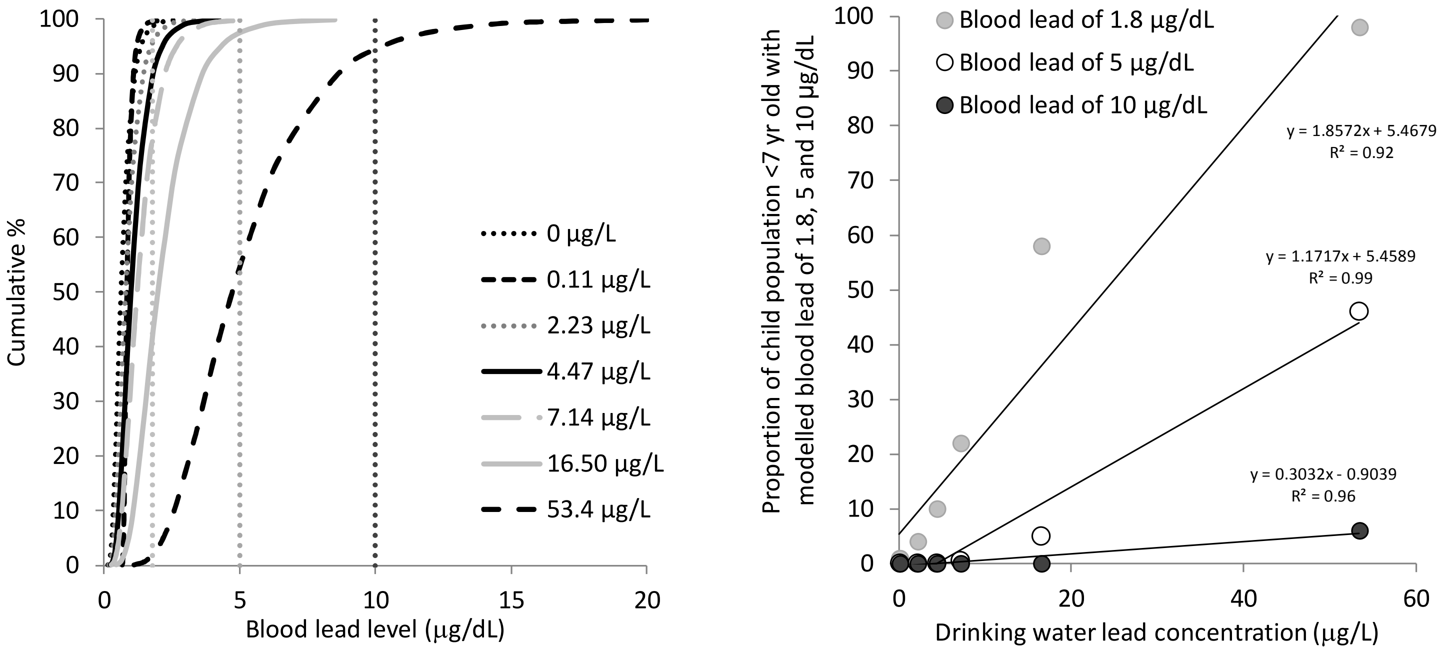


Figure 7. a) Cumulative percentage distribution of estimated BLLs in children <7 years old using the IEUBK model. b) Proportion of population with predicted BLL above 1.8, 5 and 10 μg/dL with increasing tap water concentration. Six tap water lead concentrations have been selected based on data collected during the consumption study.

Table 1. Recruitment statistics for each regional area involved in the study.

Water Supply Region	1: Leaded properties (non-P dosed)	2: Leaded properties (P dosed)	3: Unleaded Properties (non-P dosed)	4: Unleaded properties (P dosed)	TOTALS
WC1					
Properties	5	7	2	2	**16**
Individuals	6	17	3	4	**30**
Total number of drinking events sampled (winter/summer)	85/92	207/197	32/27	58/72	**382/388**
WC2					
Properties	1	2	2	2	**7**
Individuals	4	4	5	5	**18**
Total number of drinking events sampled (winter/summer)	28/27	22/29	69/81	40/49	**159/186**
TOTALS					
Properties	**6**	**9**	**4**	**4**	**23**
Individuals	**10**	**21**	**8**	**9**	**48**
Total number of drinking events sampled (winter/summer)	**113/119**	**229/226**	**101/108**	**98/121**	**541/574**

Table 2. Summary data for lead concentrations in drinking water in the two study areas.

	N	5th Percentile (μg/L)	Median (μg/L)	95th Percentile (μg/L)	Maximum concentration (μg/L)	Proportion of samples >10 μg/L
WC1						
1: Leaded and non-P dosed - Winter	85	0.2	4.5	17.0	19.0	38.8
1: Leaded and non-P dosed - Summer	92	0.3	3.7	74.5	1050.0	43.5
2: Leaded and P dosed - Winter	207	0.1	0.1	1.1	4.0	0.0
2: Leaded and P dosed - Summer	197	0.1	0.2	1.2	2.3	0.0
3: Unleaded and non-P dosed - Winter	32	0.8	1.9	3.2	3.8	0.0
3: Unleaded and non-P dosed - Summer	27	1.6	2.7	9.7	12.0	7.4
4: Unleaded and P dosed - Winter	58	0.1	0.1	0.2	0.3	0.0
4: Unleaded and P dosed - Summer	72	0.1	0.2	2.1	6.7	0.0
WC2						
1: Leaded and non-P dosed - Winter	28	0.5	5.7	13.9	15.4	17.9
1: Leaded and non-P dosed - Summer	27	1.5	8.5	17.4	19.6	33.3
2: Leaded and P dosed - Winter	22	0.5	1.7	10.5	14.9	9.1
2: Leaded and P dosed - Summer	29	1.3	2.9	24.8	224.0	20.7
3: Unleaded and non-P dosed - Winter	69	0.1	0.5	0.8	1.2	0.0
3: Unleaded and non-P dosed - Summer	81	0.1	0.9	2.5	16.1	1.2
4: Unleaded and P dosed - Winter	40	0.1	0.1	0.2	0.4	0.0
4: Unleaded and P dosed - Summer	49	0.1	0.3	0.6	0.7	0.0