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ORIGINAL PAPER

Estimates of potential childhood lead exposure from contaminated soil using the USEPA IEUBK model in Melbourne, Australia

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Abstract Soils in inner city areas internationally and in Australia have been contaminated with lead (Pb) primarily from past emissions of Pb in petrol, deteriorating exterior Pb-based paints and from industry. Children can be exposed to Pb in soil dust through ingestion and inhalation leading to elevated blood lead levels (BLLs). Currently, the contribution of soil Pb to the spatial distribution of children's BLLs is unknown in the Melbourne metropolitan area. In this study, children's potential BLLs were estimated from surface soil (0–2 cm) samples collected at 250 locations across the Melbourne metropolitan area using the United States Environmental Protection Agency (USEPA) Integrated Exposure Uptake Biokinetic (IEUBK) model. A dataset of 250 surface soil Pb concentrations indicate that soil Pb concentrations are highly variable but are generally elevated in the central and western portions of the Melbourne metropolitan area. The mean, median and geometric soil Pb concentrations were 193, 110 and 108 mg/kg, respectively. Approximately 20 and 4% of the soil samples exceeded the Australian HIL-A residential and HIL-C recreational soil Pb guidelines of 300 and 600 mg/kg, respectively. The IEUBK model predicted a geometric mean BLL of 2.5 ± 2.1 µg/dL (range: 1.3–22.5 µg/dL) in a hypothetical 24-month-old child with BLLs exceeding 5 and 10 µg/dL at 11.6 and 0.8% of the sampling locations, respectively. This study suggests children's exposure to Pb contaminated surface soil could potentially be associated with low-level BLLs in some locations in the Melbourne metropolitan area.

Keywords Blood · Model · Prediction · Contamination · Urban

Background

Lead (Pb) is a well-known neurotoxin that is associated with a myriad of health effects in humans (Bellinger 2011). The United States National Toxicology programme has reviewed the effects of low-level exposure to Pb (NTP 2012). The Australian National Health and Medical Research Council (NHMRC) (NHMRC 2015) advises that "…If a person has a blood lead level greater than 5 micrograms per decilitre, it is recommended that the source

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of exposure should be investigated and reduced, particularly if the person is a child or pregnant woman." The United States Centres for Disease Control (CDC) (CDC 2017) and the World Health Organisation (WHO 2017) state that there is no "safe" level of Pb exposure. Thus, understanding the sources of Pb exposure is important in order to prevent potential detrimental health effects caused by Pb.

The main sources of Pb exposure in children are lead-based paint (Rabinowitz et al. 1985), drinking water (Edwards et al. 2009) and soil dust (Mielke and Reagan 1998; Zahran et al. 2013). The exposure pathway of Pb-contaminated soil is primarily through ingestion (Manton et al. 2000) and inhalation (Zahran et al. 2013). Lead-contaminated soil dust can enter homes by adhering to pets fur and paws (Brunekreef et al. 1983), being tracked in on shoes (Hunt et al. 2006; Hunt and Johnson 2012; Johnson 2008), and through re-suspension of soil during the dry months of the year (Laidlaw and Filippelli 2008; Laidlaw et al. 2014). Children are also exposed to Pb by directly ingesting soil when outdoors (Wilson et al. 2013).

Regulators in Australia and internationally have established soil Pb guidelines to account for exposure to Pb in soil. The Australian residential soil Pb guideline is 300 mg/kg for residential areas with gardens or accessible soil and 600 mg/kg for public access recreation areas such as parks and playgrounds (NEPC 2013). Soil Pb guidelines are lower than the Australian guidelines in some countries. The California residential soil Pb guideline is 80 mg/kg (CEPA 2009), the Canadian soil Pb guideline is 70 mg/kg for agricultural land and 140 mg/kg for residential or parkland (CCME 2017), and the Norwegian soil Pb guideline for soil in day care centres, playgrounds and schools is 100 mg/kg (NPCA 2009).

Children's Pb exposure is typically measured with a blood lead level (BLL). In many American states, children's BLLs are screened routinely. For example, in the Detroit metropolitan area, between 2001 and 2009, BLLs for 367,839 children were screened (Zahran et al. 2013). Unlike the USA, no Australian Government body currently screens children's BLLs, with the exception of BLL monitoring that is conducted in mining towns. Thus, the burden of Pb exposure in Australia's major cities is poorly understood. Laidlaw et al. (2017c) estimated children's BLLs in the Sydney metropolitan area from soil Pb data using the United States Environmental Protection Agency (USEPA) Integrated Exposure Uptake Biokinetic (IEUBK) model. Laidlaw et al. (2017c) predicted that the geometric mean BLL of a 24-month-old child was 2.0 ± 2.1 μg/dL with children at approximately 5.6 and 2.1% of the sampling locations potentially exhibiting BLLs exceeding 5 and 10 μg/dL, respectively. The estimated BLL in Sydney by Laidlaw et al. (2017c) could possibly under-predict BLLs because it does not include additional contributions to children's BLLs from exposure to interior Pb paint or Pb exposure from drinking water (Harvey et al. 2016). There has been no attempt to screen BLLs in urban areas since 1995 (Donovan 1996).

Two studies have partially described the spatial distribution of soil Pb concentrations in the Melbourne metropolitan area. Laidlaw et al. (2017b) collected 114 surface soil samples and observed that soil Pb concentrations were elevated in the central and western portions of the Melbourne metropolitan area. Laidlaw et al. (2017b) also observed that soil Pb concentrations decreased with distance away from the roadway and was most elevated within 15 m of the roadway. Laidlaw et al. (2018) analysed soil Pb concentrations in 136 vegetable gardens in the Melbourne metropolitan area and reported that the mean soil Pb concentrations exceeded the Australian HIL-A residential guideline of 300 mg/kg in 8% of 13 community garden beds and 21% of the residential vegetable gardens. Furthermore, Laidlaw et al. (2018) reported that soil Pb concentrations generally increased with age of the home ($p < 0.000$), were higher in homes with painted exteriors ($p = 0.004$), and were higher beneath the household dripline than in vegetable garden beds ($p = 0.040$).

The hypothesis of this study is that soil Pb is potentially contributing to elevated BLLs in children in portions of the Melbourne metropolitan area. The objectives of this study were to (1) map surface soil Pb concentrations in Melbourne using available soil Pb datasets and (2) predict children's BLLs in the Melbourne metropolitan area using the USEPA IEUBK model.

Methods

VegeSafe soil sampling and analysis method

Two-hundred and fifty surface soil (0–2 cm depth) samples were included in this study. One-hundred and thirty six of the soil samples were collected by residents from their home vegetable garden and sent to Macquarie University VegeSafe programme for soil Pb analysis using a portable X-Ray Fluorescence instrument. The site location of the samples was dependent upon the residents' decision to participate in the VegeSafe programme (Rouillon et al. 2017a).

The VegeSafe programme offers a *pro bono* soil metal service to residential and community garden participants across Australia. The programme's website (https://research.science.mq.edu.au/vegesafe/) provides instructions about sampling and requests that participants provide a maximum of five labelled samples from their home yard, from a depth of 0–2 cm. Samples are typically received in the post (mail) and results are emailed back to participants on completion of analysis. Participants submit a consent form with their samples containing some basic metadata including sample location (dripline, vegetable garden), age of house, construction of house exterior (e.g., brick, wood, weatherboard) and whether it is painted or not. Soil processed as part of Macquarie University's VegeSafe programme is processed as received, i.e., there is no further sample preparation. Analysis of received soils is undertaken using an Olympus Delta X-Ray Fluorescence (XRF) 50 kV or a 40 kV instrument. Operational procedures included daily measurements of an energy calibration check, measurements of a silicate (SiO_2) blank and National Institute of Standards and Technology certified reference materials (CRMs: NIST 2710a, 2711a) throughout the measurement process. For the element of concern here, Pb, duplicate values rarely report $> 10\%$ RPD and typically $< 5\%$ RPD from the certified reference materials. Recent evaluation of in situ soil metal(loid) analysis versus laboratory-standard data showed that Pb was within 30% of laboratory data (Rouillon et al. 2017b). While the posted VegeSafe samples are not in situ per se, they are reasonably equivalent because they are raw, unprocessed samples, similar to field samples (Rouillon et al. 2017b).

Laboratory analysed soil samples method

The remaining 114 of the 250 surface soil samples were collected by Laidlaw et al. (2017b) and analysed at the National Association of Testing Authorities (NATA) accredited laboratory at ALS Laboratories in Brisbane, Queensland. The preparation of the soil samples at ALS Laboratory consisted of Hot Block Acid Digestion. An aliquot (1.0 g) of sample was heated with nitric and hydrochloric acids and then cooled. Peroxide was added and samples heated and cooled again before being filtered and bulked to volume for analysis. Lead was determined following an appropriate acid digestion of the soil using an inductively coupled atomic emission spectroscopy (ICPAES) instrument. This method was compliant with the National Environmental Protection Council (NEPC) Schedule B(3) (NEPC 2013).

Laboratory analysed soil samples: quality assurance/quality control

Two certified reference materials (CRM's) with different soil Pb concentrations were submitted for laboratory analysis. The CRM from ERA in Golden Colorado (USA) (catalog number 540) had a certified soil Pb concentration of 254 mg/kg, while the CRM from Sigma-Aldrich (trace metals—sandy loam; CRM020-D020-50 g; Lot D020) had a certified soil Pb concentration of 5110 ± 50.8 mg/kg. Seven duplicate and six triplicate soil samples were analysed for Pb concentrations. The primary samples, duplicates and CRM's were analysed at ALS laboratories, three triplicate samples were analysed at SGS laboratories, and three triplicate samples were analysed at Eurofins MGT laboratory.

The relative per cent difference (RPD) for the two CRM's was 2 and 10%. The RPDs between the original and duplicate samples were 0, 4, 4, 8, 11, 12, and 23%. The RPDs for the triplicate samples analysed at SGS laboratories were 5, 11 and 12%. The RPDs for the triplicate samples analysed at Eurofins MGT laboratory were 37, 48 and 80%. It is suspected that the high RPDs for the SGS laboratory triplicates may be due to either sample heterogeneity or laboratory error at SGS laboratory. ALS laboratory internally analysed duplicate samples for 8 soil samples which exhibited RPDs ranging between 0 and 2.54%. Matrix spikes for lead were conducted on three soil samples with recovery rates of 105, 110 and 57.4%. It is concluded that the quality of the data is acceptable.

Comparability of community and residential garden soil metal datasets

Rouillon and Taylor (2016) observed a very strong correlation between portable XRF (pXRF) and ICPAES Pb data (r^2 0.999) which indicates that pXRF is a robust alternative to ICPAES for the measurement of Pb in soils. The same model pXRF instrument as used by Rouillon et al. (2017b) was used to analyse Melbourne residential vegetable gardens (n = 136) for their Pb concentrations. Soil samples (n = 114) from Laidlaw et al. (2017b), which were mostly sampled near parks and roadside nature strips were analysed for Pb using ICPAES following acid digestion.

IEUBK blood lead model

The IEUBK model was used to calculated the BLL of a 24-month-old child at each soil sampling location (USEPA 2018). The median bioavailability (34%; range = 25–34%) measured for soils in Sydney by Laidlaw et al. (2017c) was used in the IEUBK BLL calculations for Melbourne. A description of the method and default assumptions used for the application of the IEUBK model is presented in (Laidlaw et al. 2017c). The IEUBK model has been used to predict children's BLLs using surface soil Pb concentrations in urban areas such as Toledo, Ohio (USA) (Stewart et al. 2014), Broken Hill, NSW (Australia) (Yang and Cattle 2015) and Sydney, New South Wales (Australia) (Laidlaw et al. 2017c).

Results

Two-hundred and fifty soil samples were collected in this study and analysed for Pb concentrations. The mean, median and geometric soil Pb concentrations were 193, 110 and 108 mg/kg, respectively (Table 1). Soil Pb concentrations ranged from 8 to 3341 mg/kg (Table 1). Approximately 20 and 4% of the soil samples exceeded soil Pb concentrations of 300 and 600 mg/kg, respectively. Figure 1 presents a histogram of sample counts versus soil Pb concentration. Figure 2 shows the spatial distribution of the soil Pb concentrations across the Melbourne metropolitan area. The figure indicates that soil Pb concentrations are highly variable spatially and are generally higher in the central and western portions of the Melbourne metropolitan area.

Table 1 Soil lead summary statistics (n = 250)

	mg/kg
Minimum	8
25th percentile	48
Geometric mean	108
Median	110
Mean	193
Standard deviation	282
75th percentile	247
95th percentile	566
Maximum	3341

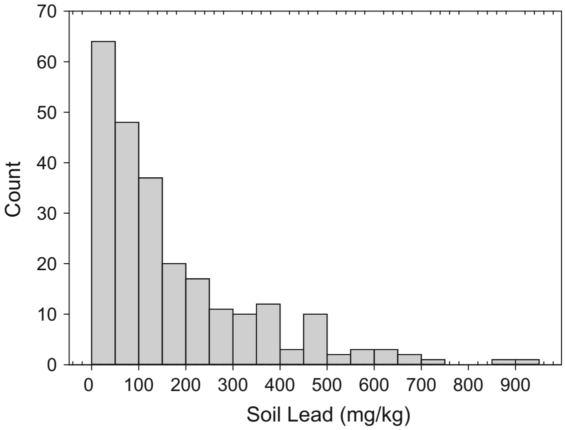

Fig. 1 Histogram of soil lead concentrations in Melbourne (n = 250) (the soil Pb concentrations less than 1000 mg/kg are displayed)

Blood lead levels in Melbourne were estimated for a hypothetical 24-month-old child at each of the 250 soil sampling locations using the median soil Pb bioavailability from Sydney. The estimated mean, median, and geometric mean BLLs using a soil Pb bioavailability of 34% were 2.9, 2.3 and 2.5 µg/dL, respectively (Table 2). The estimated BLLs ranged from 1.3 to 22.5 µg/dL (Table 1). A histogram of the estimated BLL distribution indicated that the BLLs were skewed to the left (Fig. 3). Approximately 11.6 and 0.8% of the sampling locations were estimated to exceed BLLs of 5 and 10 µg/dL, respectively. Figure 4 shows the spatial distribution of the estimated BLLs in the metropolitan area. This map generally indicates that BLLs are estimated to be elevated in the central and western portions of the metropolitan area and exhibit lower concentrations to the north and

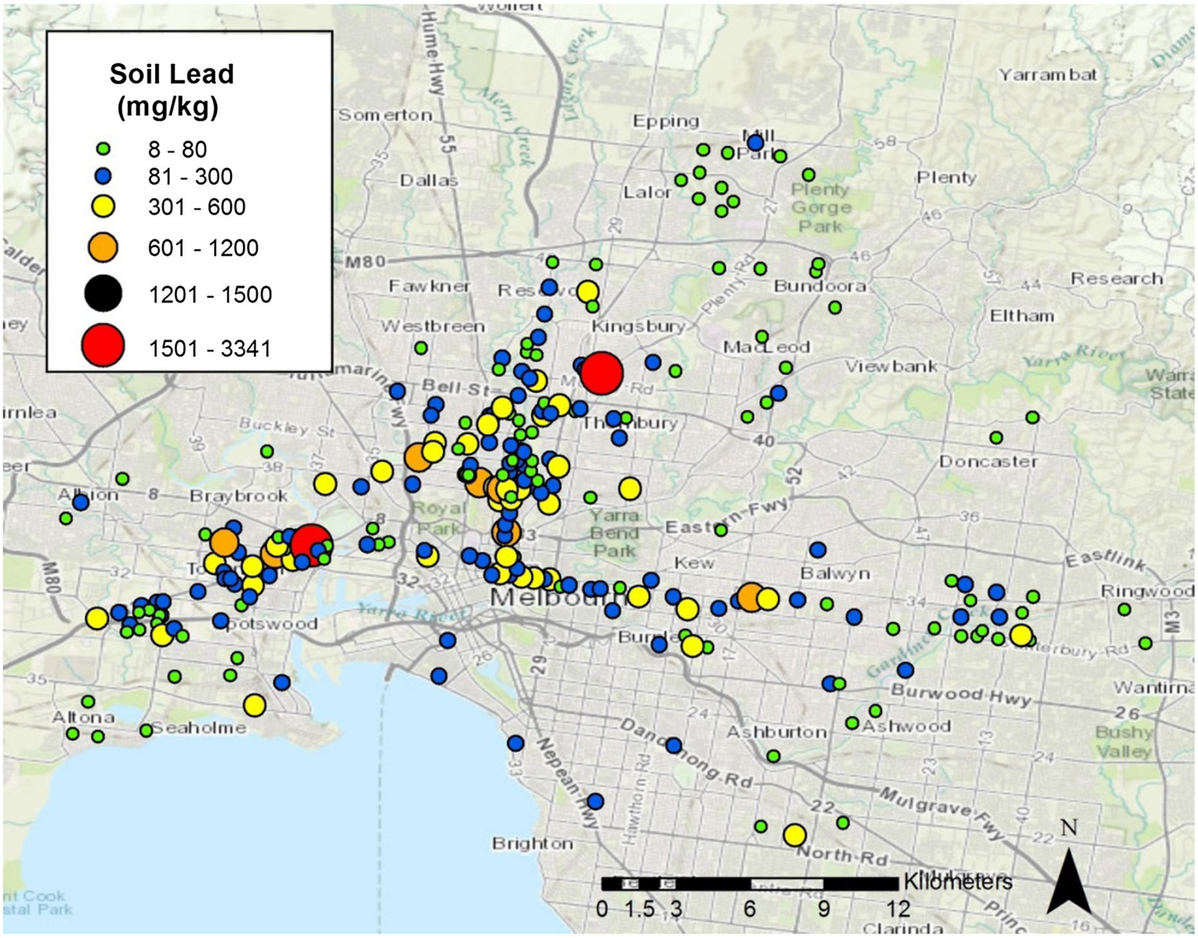

Fig. 2 Map of soil lead concentrations in Melbourne

Table 2 Summary statistics of children's (24-month-old child) BLLs predicted using the IEUBK model (*n* = 250)

	μg/dL
Minimum	1.3
25th percentile	1.7
Geometric mean	2.5
Median	2.3
Mean	2.9
SD	2.1
75th percentile	3.2
95th percentile	6.2
Maximum	22.5
0–2.5 μg/dL	57.6%
> 2.5 μg/dL	42.4%
> 5 μg/dL	11.6%
> 10 μg/dL	0.8%

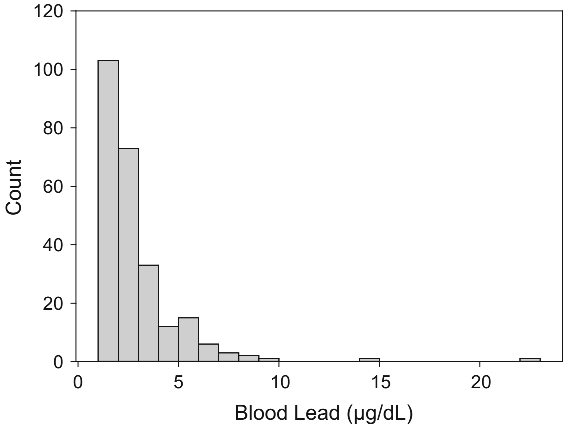

Fig. 3 Histogram of children's (24-month-old child) blood lead levels predicted using the USEPA IEUBK model

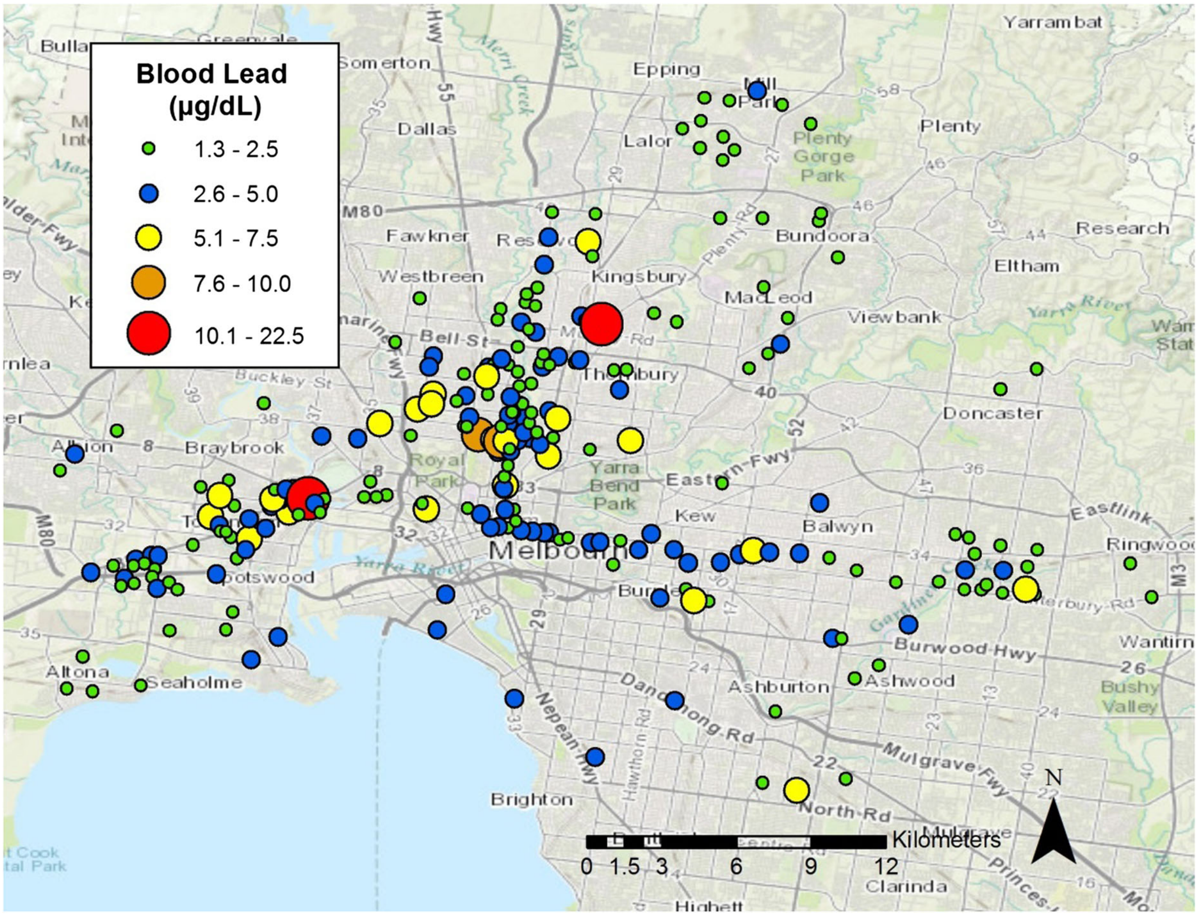

Fig. 4 Map of children's (24-month-old child) blood lead levels estimated using the USEPA IEUBK model

northeast. To assess the sensitivity of the model to the ranges in soil Pb bioavailability, estimated BLLs were calculated using the range of soil Pb bioavailability values observed in Sydney's soil. The estimated geometric mean BLLs using bioavailablity values of 25, 34 and 43% were 2.2 ± 1.7, 2.5 ± 2.1 and 2.8 ± 2.6 µg/dL, respectively.

Discussion

The results of the soil sampling performed in metropolitan Melbourne in this study indicates that surface soil Pb concentrations exceeded the Australian HIL-A residential soil Pb guideline (300 mg/kg) in 20% of sampling locations. It is difficult to discern a spatial pattern of the soil Pb concentrations due to the low density of samples in some locations. Laidlaw et al. (2018) observed that soil lead concentrations in Melbourne's vegetable gardens increased with the age of the homes ($p < 0.000$). Based upon this observation, it would be expected that the spatial variation in soil Pb concentrations and children's BLLs in unsampled areas of Melbourne mirror the age of homes. Mielke et al. (1997) concluded that the amount of soil Pb in New Orleans, Louisiana (USA), followed the age of homes primarily because the ages of homes follow the development of urban traffic patterns and the use of leaded petrol.

The results of this study suggest that there is potential for low-level Pb exposure in children in portions of the Melbourne metropolitan area due to exposure to Pb in soil. The estimates of potentially elevated BLLs in Melbourne in this study are similar to the estimates made by Laidlaw et al. (2017c) in Sydney. In Melbourne (this study), it was estimated that BLLs exceed 5 and 10 µg/dL in 11.6 and 0.8% of the sampling locations, respectively, while in Sydney, Laidlaw et al. (2017c) estimated that BLLs exceed 5 and 10 µg/dL in 5.6 and 2.1% of the sampling locations. It is an important distinction that these estimates were made for each sampling location and should not be construed as representing the prevalence of elevated BLLs in the wider population. Furthermore, the spatial pattern of sampling locations in Melbourne is substantially different than the better spatially distributed sampling locations in Sydney. In Sydney, the sample locations were sampled on a grid with a density of 1 sample/km^2. By contrast, Melbourne not sampled on a grid and many of the sampling locations were located in the central portion of the Melbourne metropolitan area where soil Pb levels are typically elevated. It is likely that the different sampling schemes in Melbourne compared to Sydney account for the higher rates of estimated elevated BLLs in Melbourne.

The availability of data regarding BLLs in children in Australian inner city urban areas is sparse. The only survey of BLLs in children in Australia was conducted on 1575 children in 1995 (Donovan 1996). Donavan found that the geometric mean Pb concentration in 1995 in 1–4-year-olds was 5.1 µg/dL, with 7.3% exceeding 10 µg/dL and 1.7% exceeding 15 µg/dL. These BLLs were likely elevated due to the use of Pb in petrol at that time in Australia (Kristensen 2015). Following the elimination of Pb in petrol in 2002, Gulson et al. (2014) measured BLLs in 108 children (aged 2–3 years) from the Sydney Metropolitan area between 2002 and 2006 and observed a geometric mean BLL of 2.4 ± 2.1 µg/dL. Measurements of BLLs specifically in the state of Victoria historically indicate that children and adults have been exposed to Pb. Taylor et al. (1995) reported that in 1993 the mean BLL of 252 children in Victoria (123 age < 5 years) was 5.7 ± 3.0 µg/dL with 8.1 and 1.6% exhibiting BLLs $>$ 10 and 15 µg/dL, respectively. Kelsall et al. (2013) surveyed the BLLs of Victorian adults ($n = 3622$) between 2009 and 2010 and observed that 68.8% had BLLs < 2 µg/dL, 28.8% had BLLs between 2 µg/dL and ≤ 5 µg/dL, 1.8% had BLLs between 5 µg/dL and ≤ 10 µg/dL, and 0.7% had BLLs ≥ 10 µg/dL. No recent studies documenting children's BLLs in Melbourne have been located. In the absence of children's BLL screening data, it is difficult to confirm whether the IEUBK blood Pb model predictions in this study are accurate. Soil lead concentrations are not the only environmental predictor of urban children's BLLs. In Sydney, Gulson et al. (2018) demonstrated that Pb in petri dish dust and dust sweepings predicted children's BLLs consistent with observed values.

Limitations and further research

One limitation of this study is that there is a lack of screening of children's BLL in Australian cities. Hospitals could potentially screen children's BLLs regularly when they come in for routine testing. If properly planned, this could provide an indication of current lead exposures. Clinical BLL testing would confirm the veracity of the framework of using soil lead as a surrogate for BLL. It is recommended that in order to better define the spatial extent of the soil Pb hazard in the Melbourne metropolitan area, soil samples should be collected on a grid at a high density (4 samples/km^2) using methods similar to those used by the British Geological Survey (British Geological Survey 2017). Potential BLLs could be calculated using the IEUBK model. The soil Pb and estimated BLL maps could be used to inform residents of high-risk areas where residents could conduct further testing of soils at their homes to determine whether their soils are contaminated with Pb. Should elevated soil Pb concentrations be observed, residents could take measures to prevent exposure to soil Pb such as covering bare soils with mulch or clean soil, building raised vegetable garden beds with a permeable geotextile liner placed at the bottom of the bed and importing clean soil, building sand boxes for children to play in, placing stepping stones through frequently travelled pathways, removing outdoor footwear at the door and wearing gloves when handling soil. To prevent further contamination of the soil, flaking exterior lead-based paints could also be carefully encapsulated or removed by professionals. Laidlaw et al. (2017a) reviewed the literature regarding soil Pb remediation in urban areas.

Conclusions

In this study, children's BLLs were estimated from surface soil (0–2 cm) samples collected at 250 locations across the Melbourne metropolitan area using the United States Environmental Protection Agency (USEPA) Integrated Exposure Uptake Biokinetic (IEUBK) model. A dataset of 250 surface soil Pb concentrations indicate that soil Pb concentrations are highly variable and generally elevated in the central and western portions of the Melbourne metropolitan area. The mean, median and geometric soil Pb concentrations were 193, 110 and 108 mg/kg, respectively, with a range of 8–3341 mg/kg. Approximately 20 and 4% of the soil samples exceeded soil Pb concentrations of 300 and 600 mg/kg, respectively. The IEUBK model predicted a geometric mean BLL of 2.5 µg/dL (range: 1.3–22.5 µg/dL) with BLLs exceeding 5 and 10 µg/dL at 11.6 and 0.8% of the sampling locations. This study suggests children's exposure to Pb-contaminated surface soil could potentially be associated with low-level Pb exposure in some locations in the Melbourne metropolitan area. It is recommended that residents in the central and western portions of the Melbourne metropolitan area have their surface soils analysed for Pb concentrations and take remedial measures should elevated soil Pb concentrations be observed. Furthermore, it would be beneficial if soil Pb concentrations were systematically sampled at a high density (4 samples/km^2) across the metropolitan area to inform residents of areas with high risks of exposure to elevated soil Pb concentrations.

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