Identification of lead sources in residential environments: Sydney Australia

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ABSTRACT

Interior and exterior dust, soil and paint were analysed at five brick urban Sydney homes over 15 months to evaluate temporal variations and discriminate sources of lead (Pb) exposure. Exterior dust gauge Pb loading rates ($\mu g/m^2/28$ days), interior vacuum dust Pb concentrations (mg/kg) and interior petri-dish Pb loading rates ($\mu g/m^2/28$ days), were correlated positively with soil Pb concentrations. Exterior dust gauge Pb loading rates and interior vacuum dust Pb concentrations peaked in the summer. Lead isotope and Pb speciation (XAS) were analysed in soil and vacuum dust samples from three of the five houses that had elevated Pb concentrations. Results show that the source of interior dust lead was primarily from soil in two of the three houses and from soil and Pb paint in the third home. IEUBK child blood Pb modelling predicts that children's blood Pb levels could exceed 5 $\mu g/dL$ in two of the five houses.

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1. Introduction

In many urban inner-city areas in the United States, there is an epidemic of childhood PbB poisoning (Gould, 2009). It has been estimated that 24.5%, or 9.6 million US children have a PbB in the 2–10 $\mu g/dL$ range, a level which will cause sub-clinical signs (Gould, 2009). The United States Centers for Disease Control and Prevention (CDC) estimates that in the United States approximately 535,000 children aged 1–5 years had BLLs ≥ 5 $\mu g/dL$ (CDC, 2013). These exposures are quite variably geographical with some locations more significantly affected. For example, in New Orleans children currently have a PbB prevalence (>5 $\mu g/dL$) of 29.6% (Mielke et al., 2013) and Detroit children (aged 0–10 years) have a PbB prevalence of 33% (>5 $\mu g/dL$) (Zahran et al., 2013). In 2012, the United States CDC Advisory Committee on Childhood Lead Poisoning Prevention (ACCLPP, 2012) recommended the adoption of a children's PbB reference level of 5 $\mu g/dL$. While the PbB prevalence has been assessed in the United States, in Australia

childhood PbB surveillance are not collected and reported systematically so spatial and temporal distributions are unknown. The last national PbB testing occurred in 1995, when 1575 children were tested (Donovan, 1996). The arithmetic mean PbB level was 5.72 $\pm$ 3.13 $\mu g/dL$. Currently, PbB testing programs are focussed on Australia's Pb mining and smelting towns: Broken Hill, Mount Isa and Port Pirie (Taylor et al., 2011). Similar to the United States, there is no federal government program for testing or remediation of diffuse non-point source urban soil Pb contamination, although the extent of these sources is increasingly better understood, particularly in urban neighbourhoods (Olszowy et al., 1995; Birch et al., 2011; Laidlaw and Taylor, 2011).

The premise of this study is derived from Laidlaw and Taylor's (2011) review of multiple Australian soil Pb and dust Pb studies that concluded soils and interior dust in many older Sydney suburbs are likely to have been contaminated from industrial and domestic Pb sources. In support of this contention, is the work by Birch et al. (2011) who mapped soil Pb concentrations in the Sydney basin and observed widespread soil Pb contamination with highest concentrations located in the inner parts of eastern, northern and western Sydney.
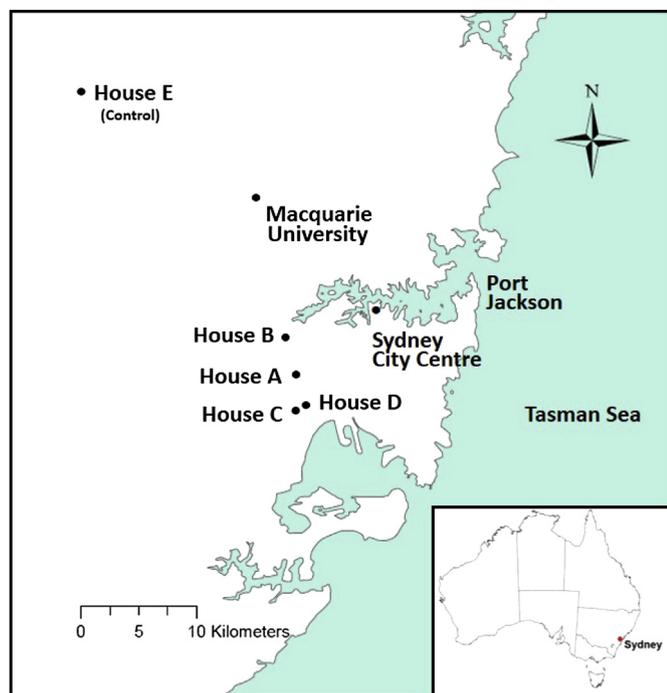
Fig. 1. Site locations of houses sampled in the study. This figure displays the locations of the houses that participated in this study.

This study examines sources of residential Pb and its temporal cycling in 5 brick homes over a 15 month period during 2010—2012 in Australia's largest urban city, Sydney (population 4.61 million; Australian Bureau of Statistics (ABS), 2012). The principal aim of the study was to determine the predominant source(s) of Pb inside typical western Sydney brick homes. Brick homes were selected to avoid the potential confounding effect of exterior lead paint that was used in older homes. Given that the study involved the collection of longitudinal environmental data, it was also possible to evaluate the seasonal variation in exterior and interior Pb quantities.

2. Methods and approach

2.1. House selection

The criteria for the four principal study houses located in the older inner west of Sydney was that each house was greater than 50 years old and was constructed of unpainted bricks. The four houses were built between 1900 and 1918. The houses were selected from three inner west Sydney suburbs: Arnfield (2), Marrickville and Haberfield (Fig. 1). A fifth brick house, approximately 30 years old was also sampled as a reference site. This property was located in an area of low-density bushland in the suburb of Glenhaven, 28 km northwest of Sydney city centre (Fig. 1). The owners of each home gave their consent to environmental sampling.

2.2. Study design

The sampling period was undertaken between November 2010 and January 2012 (15 months). At each house, exterior atmospheric Pb loading rates were measured monthly using passive sampling dust gauges in the rear garden at each home. Interior vacuum Pb in the <75 μm fraction were analysed monthly and attic and interior petri-dish Pb loading rates were measured quarterly. Soil Pb concentrations (<75 μm fraction) were measured in surface soil samples (0—2 cm) collected adjacent to the roadway in front of each house, within 1 m from the front of each house and in the middle of each back yard. In addition, one soil sample was collected at a depth of 50 cm in the middle of each back yard. Using the three inner west homes with the highest total soil Pb concentrations (Houses A, C and D), Pb isotopic composition analysis of selected environmental samples was undertaken using quadrupole Inductively Coupled Mass Spectrometry (ICP-MS). These samples consisted of front yard surface (0—2 cm) and sub-surface (50 cm) soils (<75 μm fraction); a vacuum dust sample (<75 μm fraction); and an indoor paint chip sample from the surface identified previously to have the highest Pb concentration. Finally,

Pb speciation of vacuum dust and surface soil (0—2 cm) samples were determined from the same three houses (Houses A, C and D) using X-Ray Absorption Spectroscopy (XAS) at the Australian Synchrotron facility in Melbourne, Australia.

2.3. Sieving

Soil and vacuum dust samples were sieved using a 75 μm mesh prior to analyses. Sieves were rinsed in a tap water/Alconox™ solution followed by rinsing with type II deionised water (American Society of Testing Materials (ASTM) standard) and then dried at 85° C before and after use. Soil and dust samples selected for XAS analysis were milled to <20 μm with a Retsch MM301 tungsten carbide milling machine in order to mitigate sample thickness effects that could otherwise distort the spectra. The mills were cleaned with fine silica sand followed by Alconox wash and type II deionised water rinse.

2.4. Soil and vacuum samples

Each surface soil sample was a composite of three samples collected approximately 1 m apart using a plastic hand trowel that was cleaned with deionised water and dried between sample locations. Samples were collected and stored in metal-free plastic bags prior to sieving.

Monthly interior vacuum dust samples (1 sample per location per month) were collected from each house over the study period, all of which used High Efficiency Particulate Air (HEPA) vacuum cleaners. Four of the vacuum cleaners (Houses A, C, D and E) were bag-less and one contained a vacuum bag (House B). The entire content of each vacuum sample was placed in a large, resealable metal-free plastic bag. Sieved (<75 μm) soil and vacuum samples were analysed for total extractable Pb concentrations using United States Environmental Protection Agency (USEPA) SW-846-6010 (USEPA, 2013b) method by the ALS Laboratory Group in Sydney, Australia.

2.5. Dust gauge samples

Exterior dust gauges were placed in the rear garden areas of each house in a location where they would not be disturbed. The dust gauge consisted of a 150 mm diameter glass funnel which was inserted into a 2.75 L glass bottle secured in a plastic bucket affixed to a ~2 m high tripod (Australian Standard 3580.10.1-2003; Standards Australia, 2003). Dust gauge bottles were replaced monthly over the study period (1 sample per location per month). Ten ml of copper sulphate solution was inserted into each bottle to prevent algal growth. Dust gauge samples were analysed for total Pb concentrations measured using NexION 300D ICP-MS by the ALS Laboratory Group in Sydney. Total extractable Pb concentration was analysed using USEPA Method SW-846-6020 (USEPA, 2013b).

2.6. Petri-Dish samples

Petri-dish(es) (150 mm or 85 mm diameter) were placed in the attic of each house and on the main living area of each house at approximately 2 m above the floor to collect settled airborne interior dust. The polycarbonate petri-dishes were purchased in an air-tight plastic and were pre-sterilised. Petri-dishes were replaced on a quarterly basis during the study (five quarters over the study period). The main living area petri-dish was placed in the baby's bedroom in house D, and in the living room/family room of houses A, C, B and E. Each petri-dish sample was analysed for the mass of Pb using ICP-MS by the ALS Laboratory Group in Sydney. Samples were digested using a concentration of 7 M nitric acid and 6 M hydrochloric acid according to the method described in Wlodarczyk et al. (1997). Interior and attic petri-dish lead dust loading rate samples were collected quarterly (1 sample per location every three months).

2.7. Paint chip analysis

Paint chip samples were collected from the interior paint inside each house using new disposable razor-blades. Samples were stored in metal free plastic freezer bags prior to analysis for total Pb concentration. Paint chips with the highest concentration in houses A, C and D were analysed for their Pb isotopic composition.

2.8. Pb isotopic composition analysis

Total Pb paint concentrations and Pb isotopes in soil (sieved < 75 μm), vacuum dust (sieved < 75 μm) and Pb paint were analysed using a quadrupole Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) at the ChemCentre in Bentley, Western Australia. In addition, several paint samples used for initial screening were also analysed for total Pb concentrations at ALS Laboratories. Soil and dust particles (sieved < 75 μm) were digested after drying overnight. Analysis was performed using mixed high purity redistilled acid (nitric/hydrochloric) microwave assisted acid digestion (USEPA 3051A modification). Paint samples were digested with strong reflux with nitric acid as per the Association of Official Agricultural Chemists (AOAC) method 974.02 (AOAC, 2013). Sample digests were filtered and volumed in 18 MOhm water before determination of total Pb content by ICP-AES. Samples measured for their Pb isotopic composition were diluted within a concentration range of 10—

Table 1
Descriptive summary statistics of Pb by house across various sampling locations. This table contains the average soil, vacuum dust Pb, exterior dust gauge, attic petri-dish, house petri-dish Pb concentrations for houses A through E.

	House A Marrickville, Parade street	House B Haberfield, Rogers street	House C Arncliffe, Forest road	House D Arncliffe, west Botany street	House E Glenhaven, Bannerman road
Soil (mg/kg)	714 [n = 3] (17)	261 [n = 3] (79)	1203 [n = 3] (482)	451 [n = 3] (287)	47 [n = 3] (10)
Vacuum dust (mg/kg)	575 [n = 13] (240)	265 [n = 12] (177)	1159 [n = 15] (787)	256 [14] (54)	189 [6] (49)
Exterior dust gauge (Pb μg/m^2/28 days)	257 [n = 15] (198)	126 [n = 15] (84)	248 [n = 15] (162)	187 [n = 15] (208)	35 [n = 7] (62)
Petri-dish attic (Pb μg/m^2/28 days)	44 [n = 5] (42)	2 [n = 5] (5)	275 [n = 5] (20)	67 [n = 5] (60)	9 [n = 2] (17)
Petri-dish house (Pb μg/m^2/28 days)	11[n = 5] (13)	<2 [n = 5] (0)	25 [n = 5] (20)	10 [n = 5] (16)	<2 [n = 1] (0)
Paint (mg/kg)	1723 [n = 3] (1260)	7910 [n = 1] (0)	13,748 [n = 3] (13,379)	1750 [n = 2] (717)	NA

Note: Standard deviation in parentheses; n in brackets; *Values represent averages of samples collected from front street, front house side and rear yard. Vacuum Bag Dust and Exterior Dust Gauge − sampled monthly; Petri-Dish Samples − sampled quarterly.

25 μg/L in 0.1% HNO_3 to remain in pulse detector counting mode of the instrument, confirmed for concentration, and then run for Pb isotopic ratios using a pass stable sample introduction (SSI) peltier double cooled spray chamber after optimisation on an Agilent 7500 ICP-MS. Each sample was bracketed before and after by a mass bias correction against the certified reference material National Institutes of Standards and Technology (NIST)-981. In addition, certified reference material NIST-982 was run after every 20 samples as an external quality control. Acquisition was for a total across the isotopes of 24 s for each reading and 5 readings were then averaged for the final result.

2.9. X-ray Absorption Spectroscopy (XAS)

Extended X-Ray Absorption Fine Structure (EXAFS) measurements were carried out at XAS beamline 12ID at the Australian Synchrotron (Melbourne, Australia). Samples were diluted in microgranular cellulose (Sigma−Aldrich) and pressed into pellets of 1 mm thickness using a hydraulic press. The Pb LIII-edge was measured in fluorescence mode with a 100-element solid-state Ge detector at 90° to the incident beam, with 45° sample geometry. Energy was scanned using a liquid nitrogen cooled Si (111) double crystal monochromator, and the energy scale was calibrated on the Pb LIII edge using a Pb standard foil. Samples were measured using energy stepping of 10 eV for the pre-edge baseline, 0.25 eV through the XANES region and steps of 0.035 k in the EXAFS region. Harmonics were suppressed by the use of a collimating mirror upstream of the monochromator, and a sagitally focussing mirror downstream to the monochromator, both of which were Rh coated yielding an energy cut-off of ∼18 keV. During collection the storage ring was operated at 3 GeV with ring current in the range 150−200 mA. At least two spectra for each sample were collected; spectral averaging and dead time correction were carried out using Australian Synchrotron software.

2.10. Quality assurance procedures

Three of soil and three vacuum dust samples were split in the field, each sample homogenized, and sent to the laboratory for analysis.

Table 2
Least squares and least absolute deviation regression coefficients predicting Pb content of exterior dust gauge as a function of soil Pb content. This table presents the statistical results of least squares regressions predicting Pb content of exterior dust gauge as a function of soil Pb content.

	LAD gauge	LAD gauge 3 month moving average	OLS ln gauge	OLS ln gauge 3 month moving average
Soil Pb	0.067* (0.039)	0.076** (0.034)		
ln soil Pb			1.724*** (0.464)	1.530*** (0.452)
Winter (Reference)				
Autumn	−7.228 (48.43)	76.668* (39.23)	−2.316 (1.479)	3.710*** (1.279)
Spring	50.337 (43.26)	47.568 (37.45)	2.655** (1.315)	4.005*** (1.238)
Summer	157.34*** (42.00)	139.12*** (35.88)	5.451*** (1.278)	6.579*** (1.176)
Constant	−22.574 (39.64)	−34.495 (35.50)	−12.42*** (2.933)	−11.06*** (3.004)
Observations	67	57	67	57
F-test			12.01	9.79
Pseudo R-squared	0.274	0.333		
R-squared			0.450	0.430

LAD = least absolute deviation regression; OLS = ordinary least squares regression. Note: Standard errors in parentheses, ***$p < 0.01$, **$p < 0.05$, *$p < 0.1$.

Out of the seven Pb field duplicate Relative Percent Difference (RPD) pairs (3 soil, 3 vacuum dust and 1 paint) reported, all the RPDs were less than 3%, with the exception of paint, which had an RPD of 12.5%. Petri-dish blanks were below instrument detection while dust gauge field blanks returned Pb values of (0.002 mg/L), close to the instrument detection limit (0.001 mg/L). Blank values were subtracted from the dust gauge measurements. The source of the Pb may have originated from the copper sulphate solution that was added to the bottles to prevent biological build-up (ALS Laboratory Group, personal communication). Overall, the results of the RPD calculations between primary and duplicate samples indicate that the analytical data are suitable for interpretive use. Laboratory quality assurance procedures included analysis of matrix spike/matrix spike duplicate (MS/MSD) results data, method blank data, laboratory control spike data, regular sample surrogate data and laboratory duplicate data. The overall results of the laboratory quality assurance are suitable for interpretive use, i.e. between 75% and 120%. The relative standard deviation percentage (RSD%) for Pb isotope analysis was 0.24, 0.22, 0.21 and 0.23 for the lead ratios of $^{207}Pb/^{206}Pb$, $^{208}Pb/^{206}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$, respectively. Overall, the analytical results are considered suitable.

2.11. IEUBK modelling

The USEPA's Integrated Exposure Uptake Biokinetic Model for Lead in Children (IEUBK) software version IEUBKwin1_1 Build11 was used to predict PbB levels in each house (USEPA, 2013a). Average soil Pb and vacuum dust Pb concentrations (Table 1) were used for the soil and indoor dust input variables. The Pb absorption fraction percent used was 23%(46% bioavailable), which is a conservative value based upon Snowdown and Birch's (2004) study of soil bioavailability values in Sydney. An air Pb value of 0.02 μg/m^3 was used in the model, which was based upon the Australian Nuclear Science and Technology Organisation recent air monitoring station data 5 km south of the Sydney CBD (ANSTO, 2013). IEUBK default values were used for the dietary Pb input variables. A value of zero was used for the drinking water contribution given that the most recent data on Sydney's water quality shows values are below detection limits (Sydney Water, 2013).

3. Results

3.1. Lead analysis

Descriptive summary statistics of Pb concentrations in soil, vacuum dust, exterior dust loading, petri-dish (attic and interior) and paint are presented in Table 1. The mean soil Pb concentration at the reference home located in Glenhaven at 47 mg/kg (std. dev. = 10 mg/kg) (House E) is slightly larger than the Sydney soil Pb background level of 16 ± 3.5 mg/kg identified by Birch et al. (2011). The mean of the sample at House E included a roadside soil sample (37 mg/kg), a rear garden sample (56 mg/kg), and a soil sample collected beside the house (49 mg/kg). The soils at this location were present when Pb was still being used as an additive in petrol. House dust Pb concentrations are presented in Supplemental Table S2.

We begin by analysing variation in exterior dust gauge Pb loadings using both *least squares and least absolute deviation regression* procedures (Table 2). Columns 1 and 2 report median regression results. Adjusting for seasonality, Column 1 shows the expected change in the Pb content of exterior dust gauge (at the median of the conditional distribution of exterior dust gauge loadings) per unit (mg/kg) increase in soil Pb. We find that dust gauge Pb loadings increase by 0.066 μg/m^2/28 days for every mg/kg
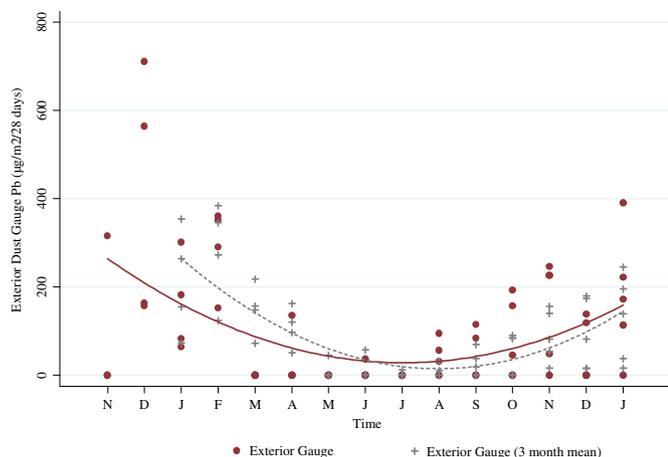
Fig. 2. Exterior Pb dust gauge data at the monthly time-step with fractional polynomial fit. This figure displays monthly variations in atmospheric Pb loading rates (μg/m²/28 days) collected in atmospheric dust gauges that were placed in the rear yards of the study houses between November 2010 and January 2012. The results indicate that atmospheric Pb loading peaks in the summer/autumn and is lowest during the winter. Red dots correspond to the exterior dust gauge Pb loading reading for each dust gauge for the month of observation. Grey crosses corresponding to the 3 month moving average. The red line intersecting the space corresponds to a best fit quadratic line for red dots. The same logic obtains for the grey dotted line. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Least squares and least absolute deviation predicting Pb content of interior vacuum dust as a function of soil and paint Pb content.** This table presents the statistical results of least squares and least absolute deviation regressions predicting the Pb content of interior vacuum dust as a function of soil and paint Pb content.

	LAD vacuum	LAD vacuum 3 month moving average	OLS *ln* vacuum	OLS *ln* vacuum 3 month moving average
Soil Pb	0.803***	1.050***		
	(0.216)	(0.131)		
Paint Pb	0.024	0.024		
	(0.026)	(0.015)		
ln soil Pb			1.185**	1.847***
			(0.367)	(0.260)
ln paint Pb			0.071	0.248**
			(0.146)	(0.103)
Winter (Reference)				
Autumn	−1.00	120.00	0.097	0.496***
	(175.63)	(93.16)	(0.216)	(0.134)
Spring	−24.00	45.00	0.138	0.311**
	(158.70)	(88.96)	(0.196)	(0.128)
Summer	34.00	120.67	0.282	0.549***
	(153.75)	(84.78)	(0.190)	(0.122)
Constant	−172.13	−355.01***	−2.12	−7.86
	(230.86)	(136.36)	(3.31)	(2.35)
Observations	54	48	54	48
F-test			16.41	59.18
Pseudo *R*-squared	0.339	0.613		
R-squared			0.631	0.876

LAD = least absolute deviation regression; OLS = ordinary least squares regression.
Note: Standard errors in parentheses, ***$p < 0.01$, **$p < 0.05$, *$p < 0.1$.

increase in soil Pb. Results, in Column 2, behave near identically if we regress a three-month rolling average of dust gauge Pb loadings on soil Pb ($b = 0.076$, $p < 0.05$). Interestingly, and consistent with a soil re-suspension hypothesis, results show that dust gauge Pb readings are significantly higher in the summer period (Column 1, $b = 157.34$, $p < 0.001$) compared to the reference season of winter gauge readings. The seasonality of exterior Pb loadings is shown in Fig. 2, which reveals dust gauge Pb loadings are lower in the Australian winter months of June to August, and higher in the summer months (December to February). The seasonality of temperature in Sydney follows the same pattern (BOM, 2013). Columns 3 and 4 report least squares regression results. To account for skew, both soil Pb and dust gauge Pb variables are log transformed. By taking the natural log of both sides of the regression equation, allowing us to express the relationship in percentage terms, in Column 3 we find that a 1% increase in soil Pb is associated with 1.72% increase in dust gauge Pb loadings. Results pertaining to the three-month rolling average of dust gauge Pb loadings reported in Column 4 behave similarly.

Given that a substantial fraction of the statistical variation in exterior dust gauge Pb loadings is explained by soil Pb accumulation, the variation in the Pb content of vacuum dust collected from homes over a 15 month period can be determined. Table 3 presents least absolute deviation and least squares regression coefficients predicting the Pb content of vacuum dust as a function of soil Pb, interior paint Pb, and seasonal dummy variables. Table 3 repeats the measurement and analytic logic of Table 2, where monthly, three-month rolling average, and natural log transformations of our response variable are regressed on relevant predictors. In Column 1 of Table 3, we find that each 1 mg/kg increase soil Pb induces a 0.803 mg/kg (95% CI, 0.369–1.238) increase in the median of the distribution of Pb content of vacuum dust. In Column 2, results show that a unit increase soil Pb significantly increases the three-month moving average of the accumulation of Pb in vacuum dust ($b = 1.050$, $p < 0.01$). Interior paint Pb is not statistically associated with Pb vacuum dust in either least absolute deviation regression model. In Columns 3 and 4 results from our log–log least squares

models are reported. Notably, in Column 4, we find evidence of a seasonal effect, with three-month rolling average of Pb in vacuum dust significantly higher in autumn, spring and summer as compared to the reference season of winter.

Figs. 3 and 4, present best-fit linear solutions of the association between soil Pb and exterior dust gauge Pb (Fig. 3), and soil Pb and vacuum dust Pb (Fig. 4) by season. These figures show that the soil Pb effect appears to amplify (slopes rise and steepen) in the summer period as compared to the winter period. Such curve behaviours by season are consistent with a soil dust re-suspension hypothesis.

Next, we analysed the quarterly variation in the Pb content of petri-dishes in sampled houses as a function of soil Pb and exterior dust gauge Pb. In Column 1 and 2 of Table 4, results from least absolute deviation regressions are reported. We find that unit increases in soil Pb ($b = 0.035$, $p < 0.1$) and exterior dust gauge Pb
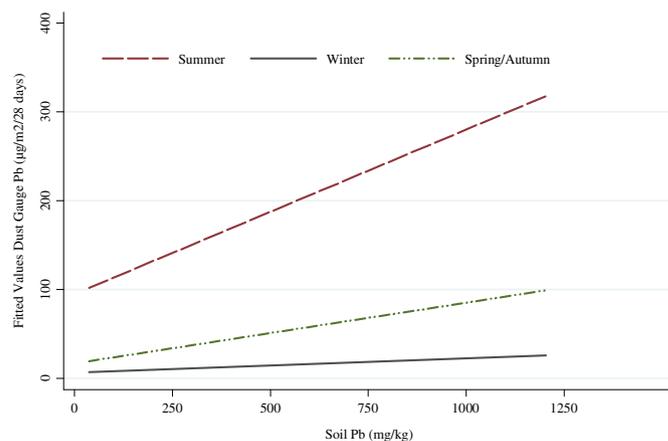

Fig. 3. Soil Pb concentration versus atmospheric Pb loading rates. This figure displays the relationship between soil Pb concentration and atmospheric Pb loading rates (μg/m²/28 days) for the five houses during summer, winter and spring/autumn. Results indicate that the relationship is strongest during the summer.
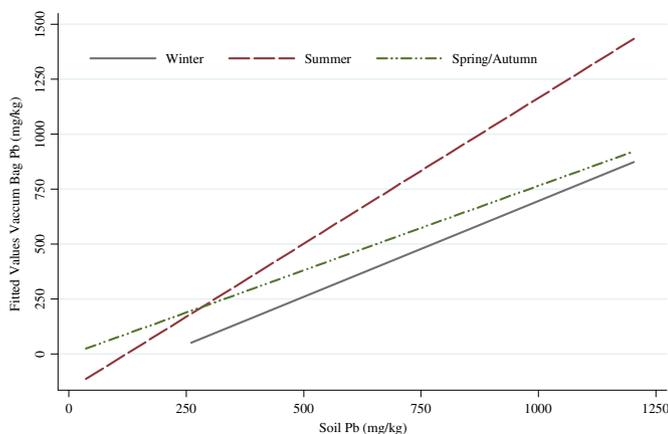
Fig. 4. Soil Pb concentrations versus vacuum Pb concentrations. This figure displays the relationship between soil Pb concentration (mg/kg) and vacuum Pb concentrations (mg/kg) for the five houses during summer, winter and spring/autumn. Results indicate that the relationship is strongest during the summer. The slope between summer is statistically different than the slope for winter and spring/autumn.

($b = 0.163$, $p < 0.05$) are associated with increases in the median of the quantity of Pb accumulated in petri-dishes. In Columns 3 and 4 of Table 4, least squares regression model results are shown. In Column 3, we find that a 1% increase in soil Pb increases the expected quantity of petri-dish Pb by 1.8% (where $p < 0.001$). The results show that a 1% increase in exterior dust gauge induces a more modest increase of 0.166% ($p < 0.05$) in petri-dish Pb (column 4, Table 4). With the exception of model 3, effects of seasonality on petri-dish Pb loadings are not significant.

3.2. X-ray Absorption Spectroscopy analysis of soil and vacuum dust

3.2.1. Soils

The two soil samples, from houses A and C, exhibit simple spectra characterized by a relatively featureless major "beat" pattern, as observed in Fig. 5. Photoelectron backscatter

Table 4
Least squares and least absolute deviation coefficients predicting Pb content of petri-dish as a function of soil Pb and exterior dust gauge Pb. This table presents the statistical results of least squares and least absolute deviation regressions predicting the Pb content of Petri-Dish as a function of soil Pb and exterior dust gauge Pb.

	LAD petri-dish	LAD petri-dish	OLS ln petri-dish	OLS ln petri-dish
Soil Pb	0.035* (0.020)			
ln soil Pb			1.801*** (0.500)	
Dust gauge Pb		0.163** (0.08)		
ln dust gauge Pb				0.166** (0.07)
Interior (Reference)				
Attic	18.32 (15.60)	1.17 (12.55)	2.242** (1.084)	2.25* (1.17)
Quarters 2, 3, 4 (Reference)				
Quarters 1, 5	14.67 (39.51)	−2.61 (16.38)	2.567** (1.126)	1.17 (1.39)
Constant	−15.95 (16.82)	0.00 (10.83)	−13.21*** (3.095)	−1.78* (0.99)
Observations	44	44	44	44
F-test			7.91	4.83
Pseudo R-squared	0.120	0.160		
R-squared			0.160	0.266

LAD = least absolute deviation regression; OLS = ordinary least squares regression. Note: Standard errors in parentheses, ***$p < 0.01$, **$p < 0.05$, *$p < 0.1$.

interference from the closest near-neighbour shell of oxygen atoms yields this simple beat pattern. The pattern indicates a lack of significant electron wave backscatter from second-, third-, or fourth-shell Pb or other heavy-element neighbours. This suggests that Pb is sorbed onto a light-element matrix, e.g., organic matter. In contrast, the repetitive physical structure (spatial arrangement of atoms) in crystalline Pb compounds yields complex spectra due to additional backscatter from Pb near-neighbour shells (see, for examples, the hydrocerussite and Pb oxide spectra). The higher frequency spikes in the samples at about 7k are artefacts due to crystal glitches.

The spectral patterns can be treated as "fingerprints", to be compared to spectra of modelled compounds (Pb humate, Pb particulate matter, hydrocerussite and Pb oxide) of known composition. The similarities of the Pb-humate and Pb-particulate matter are obvious as are the contrasts to hydrocerussite and PbO (Fig. 5). Fig. 5 illustrates a close match between the house C soil sample to Pb-humate and also the house A soil sample to Pb-particulate organic matter. These model compounds are similar: Pb sorbed onto humic acids and Pb sorbed onto organic (plant leaf) particles.

Spectral fits to other Pb species with similar simple beat spectra cannot be ruled out, given that the spectra extend only to 8k. Spectra similar to those of our soils can be produced by Pb sorbed on other forms of organic matter, or on clays that lack stronger backscattering metals, e.g., iron. However, we can say with a high degree of certainty, that the soil Pb is present in a sorbed form, which is most likely the result of reaction and alteration of the form of the Pb after it was introduced or deposited in the yards.

3.2.2. Vacuum dust

The spectra of the three vacuum dust samples, from houses A, C, and D, are similar in their main features, with some secondary differences (Fig. 5). Deviation from the simple beat pattern of the organic sorption model compounds is evident in house D dust in the narrowing or infilling of the 'valley' between the second and third spectral peaks, at about 4-5k. This indicates the presence of a strong backscattering element(s) beyond the first-neighbour shell. A mixture of 62% Pb sorbed on particulate organic matter and 38% hydrocerussite provides a close fit to house dust D (Fig. 6). These compounds were chosen from numerous combinations of up to three model compounds from in-house our spectral library of Pb compounds associated commonly with paint and urban pollution (analysed by the University of Texas at El Paso (UTEP) team at the Stanford Synchrotron Radiation Lightsource). Furthermore, hydrocerussite, or 'white Pb', was the dominant species of Pb in the commercial Pb-based paints of the 20th Century (See et al., 2007).

Fig. 6 indicates that the spectrum of house C vacuum dust closely matches that of Pb sorbed on particulate organic matter, and house A vacuum dust lies between the compositions of C and D (Fig. 6). Thus, the majority of the Pb in the household vacuum dusts has an origin that appears to be external to the home and has probably been either tracked in by foot traffic, or entered the homes as airborne soil and dust. The remainder appears to be old Pb-based paint, which probably originated predominantly inside the house given that the two associated soils did not contain Pb-based paint.

3.3. Pb isotope analysis of vacuum dust

Fig. 7 visually summarizes the Pb isotopic composition ($^{207}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$) data for interior vacuum dust, surface (0–2 cm) and subsurface soil (0.5 m), paint chips, and petrol Pb isotopic composition. To aid interpretation we have also included relevant data on paint and leaded petrol from relevant published sources (Gulson et al., 1983, 1995, 2003, 2006a,b, 2008). In Fig. 7 the vacuum Pb isotope composition for houses A, C, and D
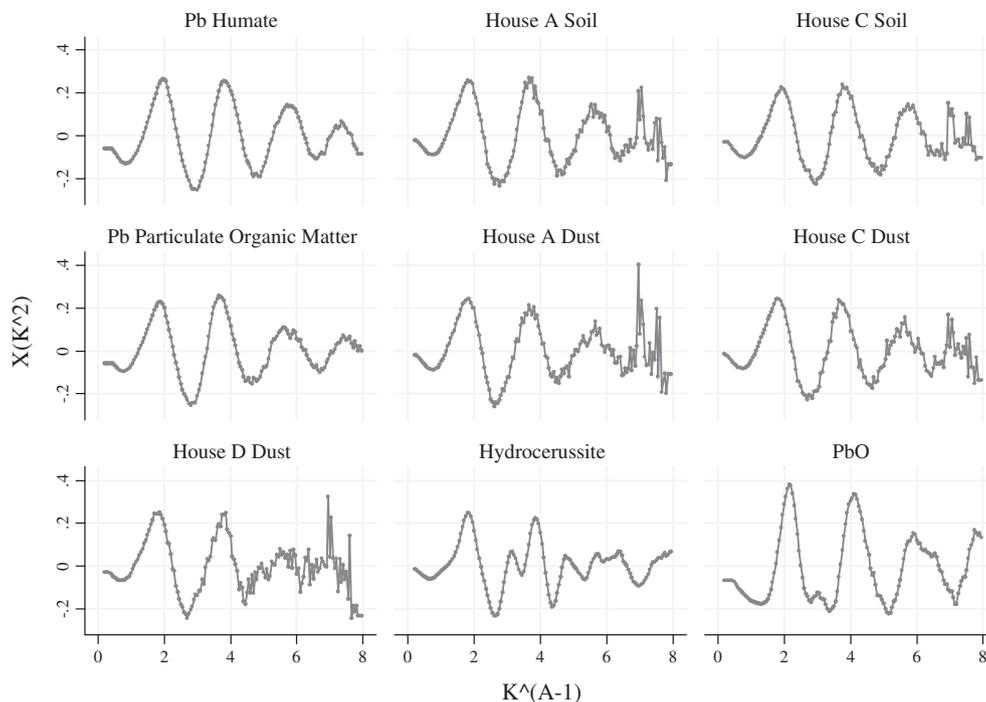
Fig. 5. XAS Pb speciation chart. This figure displays the Pb Spectra for houses A and B soil samples, houses A, C and D vacuum dust samples, Pb humate and Pb particulate organic matter, hydrocerrusite and PbO (lead oxide). The Pb spectra for Pb Humate, House A soil, House B soil, Pb particulate organic matter, House A dust and House C dust are very similar, while being dissimilar to typical paint Pb compounds − hydrocerussite and PbO. The spectra for House D is somewhat similar to the Pb spectras for House A dust and House C dust, while also being dissimilar to typical paint Pb compounds − hydrocerussite and PbO.

correspond more closely to the soil Pb isotope composition values than house-specific paint Pb isotope composition values. To estimate isotope ratio similarity, we calculated the Euclidean distance of each vacuum Pb isotope ratio to corresponding paint, surface soil, soil (0.5 m), and petrol isotope ratio values. Table S1 reports the Euclidean distance between the points where Pb isotopes plot on the Pb isotopes charts. In each house where Pb isotopes were analysed we find that vacuum Pb isotope composition ratios are more similar to surface soils than to subsurface soil (0.5 m) and paint chip samples (Fig. 7).
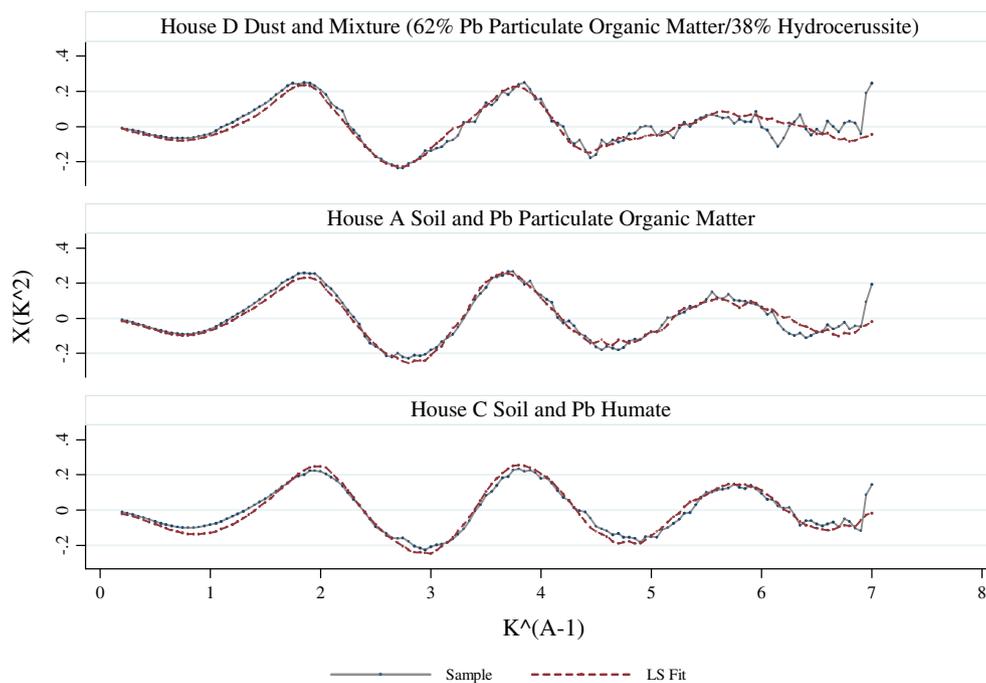

Fig. 6. XAS Pb speciation chart − paired samples. The chart displays the Pb Spectra for House D and the best fit line for Pb particulate organic matter (62%) and hydrocerussite (38%). The chart also displays a good fit between Pb spectra for house A soil and particulate organic matter. The chart also displays a good fit between the Pb spectra for house C soil and Pb humate.
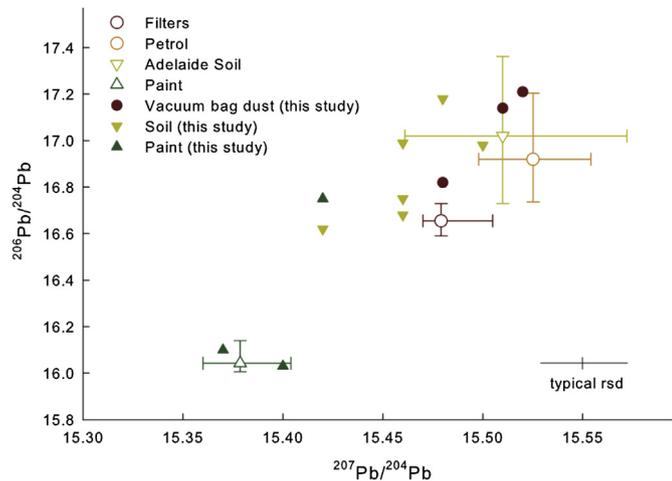
Fig. 7. Scatterplot of $^{207}Pb/^{204}Pb$ by $^{206}Pb/^{204}Pb$. This chart plots the $^{207}Pb/^{204}Pb$ ratio and the $^{206}Pb/^{204}Pb$ ratio of vacuum dust samples, surface soil sample, soil (0.5 m depth) samples and paint samples from houses A, C and D. In addition, Pb isotope ratios from paint samples collected from one house near Sydney (Gulson et al., 2003) and historical petrol Pb samples Pb isotope ratios (Gulson et al., 1983, 1995, 2006a,b) from the Sydney area are plotted as well. The results indicate that the Pb paint samples cluster together in the lower left part of the chart and the Pb isotope ratios for the Sydney petrol and the vacuum dust samples, surface soil sample, soil (0.5 m depth) samples from houses A, C and D cluster to the upper right portion of the chart. This suggests that the origin of the Pb in the houses is from petrol sources.

3.4. Lead deposition rates in house petri-dish, house attic and exterior dust gauge

Fig. 2 details the exterior dust gauge Pb deposition rates in houses A through E between November 2010 and January 2012 (see also Table S3 for additional information). Table 5 displays average quarterly Pb deposition rates for house petri-dishes (HS), attic petri-dishes (AT) and exterior dust gauges (DG) in houses A through E. In general, Pb deposition rates follow the following pattern: exterior dust gauge > attic > house. Interior and exterior Pb loading rates are correlated with exterior soil Pb concentrations and temporal Pb loading rates are highest in the summer and autumn and lowest in the winter and spring (Table 5, Fig. 3), which is consistent with a soil dust re-suspension hypothesis during the drier summer period (Laidlaw et al., 2005, 2012; Laidlaw and Filippelli, 2008; Zahran et al., 2013).

Percentiles for house dust concentration, exterior Pb loading rates, interior petri-dish Pb loading rates and attic petri-dish Pb loading rates are presented in supplementary Table S4.

4. Discussion

Soil Pb concentrations, exterior atmospheric Pb loading rates (dust gauge), interior vacuum dust Pb concentrations and interior petri-dish Pb loading rates correlate ($p < 0.05$) (Tables 2–4). This suggests that re-suspension of exterior soil is a primary source of Pb in the urban homes studied here. Other studies have shown that Australian soil Pb and children's PbB are correlated (Fett et al., 1992; Galvin et al., 1993; Willmore et al., 2006), but the specific pathway of re-suspension and airborne migration into urban homes was not identified, unlike in the USA (Hunt et al., 2006; Layton and Beamer, 2009; Hunt and Johnson, 2012).

In order to evaluate whether interior household Pb is linked to outdoor soil Pb we have examined multiple lines of evidence and have determined the following:

The correlation between exterior soil and interior vacuum dust concentration in this study ($r = 0.659$, $p < 0.001$) was also observed

Table 5
Pb deposition rates – external dust gauge, attic petri-dish and house interior petri-dish. This table presents quarterly average Pb deposition rates collected for the external dust gauge, attic petri-dish and interior petri-dish's collected at 5 houses between November 2010 and January 2012.

Sample location	Season (Quarter)	House A	House B	House C	House D	House E
Interior	Summer1	24	<2	38	<2	NA
Interior	Autumn	22	<2	27	15	NA
Interior	Winter	<2	<2	52	<2	<2
Interior	Spring	<2	<2	<2	<2	<2
Interior	Summer2	NA	NA	14	38	<2
Attic	Summer1	103	<2	917	98	NA
Attic	Autumn	17	12	91	43	NA
Attic	Winter	72	<2	<2	<2	<2
Attic	Spring	<2	<2	123	41	<2
Attic	Summer2	30	<2	244	156	30
Exterior dust gauge	Summer1	354	74	155	265	NA
Exterior dust gauge	Autumn	97	51	162	120	NA
Exterior dust gauge	Winter	<2	<2	12	<2	NA
Exterior dust gauge	Spring	90	85	84	<2	<2
Exterior dust gauge	Summer2	195	38	246	139	16

Units = micrograms/m²/28 days; Quarterly Dust Gauge Samples = average of three monthly dust gauge samples; Quarterly Interior and Attic Samples = 1 quarterly sample each.

in other locations by Bornschein et al. (1986) ($r = 0.75$) and Thornton (1990) ($r = 0.531$, $p = 0.001$, $n = 4512$). The correlation between exterior atmospheric Pb loading and interior vacuum Pb concentration in this study is significantly positive ($r = 0.314$, $p < 0.001$). Adjusting for season of observation, least absolute deviation regression models find that per unit of increase in soil Pb loading increases the median Pb content of vacuum dust by 0.369–1.238 mg/kg. The temporal flux of interior vacuum dust Pb concentrations and exterior dust gauge loadings indicates strongly that exterior soil re-suspension is the driver of the seasonal variations (Figs. 3 and 5). Further, soil Pb concentrations are significantly correlated with the natural log of interior petri-dish Pb loading rates ($p < 0.001$; see Table 4).

Interior vacuum Pb isotope ratios plot closer to the exterior surface soils than interior paint Pb isotope ratios indicating that the source of interior Pb is from exterior surface soils (Fig. 4). In addition, XAS spectra of the exterior surface soil and the interior vacuum dust were nearly identical in the most contaminated houses (Houses A and C) – indicating that they originate from the same source. In House D the data show that the source of the Pb in the house dust was from Pb in soil and house paint (Fig. 3). Finally, in terms of examining the possibility that paint was a primary source of household Pb, the study data show that vacuum dust Pb concentration and paint Pb are not correlated, except when a 3 month moving average is applied (Table 3).

The implications from this study are that in old, densely populated urban areas like Sydney, elevated soil Pb appears to be migrating into homes. The transfer of contaminants from external to internal environments is associated with elevated interior dust Pb concentrations, which in turn have significant potential to raise PbB levels (Layton and Beamer, 2009). If interior dust Pb concentrations similar to the levels found at higher concentrations in this study are widespread in Sydney (and the prevailing soil Pb research indicates that this is the case for the inner west of Sydney – population 2.02 million; NSW Government, 2013), there is a potential for widespread, low-level Pb poisoning, especially in areas with a history of high traffic rates (cf. Laidlaw and Taylor, 2011).

The risk at the lower end of the exposure spectrum is significant because the slope between soil Pb and PbB is steep at low concentrations, resulting in a rapid uptake of Pb in blood as soil Pb levels increase (Mielke et al., 2007). Importantly, the greatest relative decline of IQ due to Pb exposure occurs at the lowest Pb
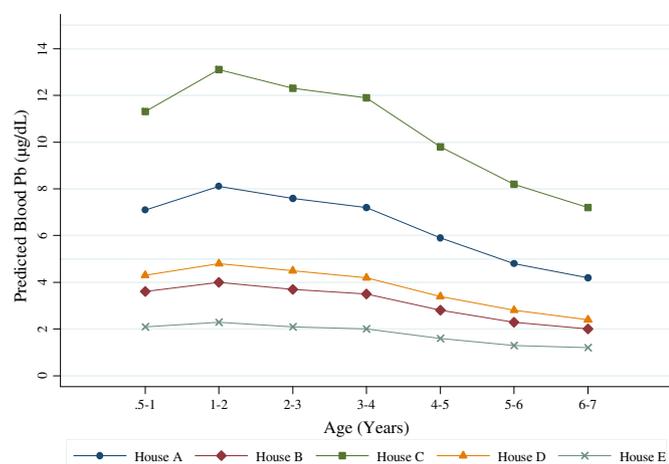
Fig. 8. IEUBK modelled PbB. Plot of predicted childrens's (age 0–7) PbB in study homes A through E which was calculated using the Integrated Exposure Uptake Biokinetic (IEUBK) model (see text for input values).

exposures (Canfield et al., 2003; Rothenberg and Rothenberg, 2005). In terms of population exposures, Taylor et al. (2012) estimated that approximately 100,000 Australian children aged 0–4 years may have PbB levels associated with adverse health outcomes, but because there is no routine or systematic PbB testing of city children in Australia the potential risk of exposure is unconstrained. We estimate exposures in children aged 0–7 in houses A through E using our data via the IEUBK model (USEPA, 2013a) (Fig. 8). This model predicts that children aged 0–7 years in houses A and C have the potential to accrue a PbB >5 μg/dL. Thus, the data from this study supports the argument that both soil lead and blood lead should be evaluated in a systematic manner across our older inner cities to decipher accurately the real risks to young children (Laidlaw and Taylor, 2011). This study supports primary prevention, and precedence for soil intervention as a method of primary prevention has been established by the Norwegian Government whereby soil Pb intervention is undertaken without measuring Pb exposure of children (Ottesen et al., 2008).

4.1. A possible way forward for dealing with potential futures exposures

In New South Wales (NSW), section 149 Planning Certificates are issued in accordance with the *Environmental Planning & Assessment Act 1979* (NSW). The certificates contain information on how a property may be used and the restrictions on development. When land is bought or sold the *Conveyancing Act 1919* (NSW) requires that a Section 149 Planning Certificate be attached to the Contract for Sale. Under sections 149 (5) and (6) of the *Environmental Planning & Assessment Act 1979* (NSW) there is a clear opportunity to identify actual or potential contamination at properties:

(5) A council may, in a planning certificate, include advice on such other relevant matters affecting the land of which it may be aware.
(6) A council shall not incur any liability in respect of any advice provided in good faith pursuant to subsection (5). However, this subsection does not apply to advice provided in relation to contaminated land (including the likelihood of land being contaminated land) or to the nature or extent of contamination of land within the meaning of Part 7A.

Therefore, we contend that following proper environmental assessment of potentially contaminated areas and properties for

dust or soil lead issues, a section 149 certificate should include the assessment information so that prospective purchasers are aware of any potential risk to health.

4.2. Study limitations

There are some contradictions within the data. The atmospheric Pb loading pattern clearly demonstrates that Pb loading rates are highest in the summer and autumn and lowest in the winter. While this is in agreement with temporal Pb concentration patterns identified in the United States (Zahran et al., 2013), it is the opposite signature to that identified in other air and dust lead seasonal studies in Sydney (Chiaradia et al., 1997; Cohen et al., 2005; ANSTO 2013). The ANSTO data reveals that the air Pb (μg/m^3) peaks in May and June, the opposite of the Pb loading rates (μg/m^2/28 days) observed in this study. Chiaradia et al. (1997) suggested that the air Pb concentration peak in the autumn-winter months are probably related to thermal inversion that is typical of Sydney's cooler winter season. Another contradiction is that the quarterly interior petri-dish Pb loading rates did not display a summertime peak that was observed in the monthly exterior Pb loading rates (μg/m^2/28 days) and interior vacuum Pb concentrations (mg/kg). These contradictions further support the need for expanding the study to more Sydney homes to confirm the trends identified within the small group of homes in the study.

5. Conclusion

This study used multiple types of environmental analysis to show the main source of Pb found within interior dust at four inner-city Sydney homes is derived from Pb that has been tracked-in and re-suspended from exterior soil. Lead paint was shown to be a partial source in one of the three homes where Pb isotopes were sampled and Pb speciation was analysed. This study confirms that exterior soil Pb is being transferred to domestic interiors. Given that the greatest relative uptake of Pb into blood occurs at the lowest soil Pb concentrations and the greatest relative decline of IQ due to Pb exposure occurs at the lowest Pb exposures, then Pb contaminated urban roadside and garden soils may require remediation or isolation to provide a margin of safety for children.

Appendix A. Supplementary data

Supplementary data related to this article can be found at doi:10.1016/j.envpol.2013.09.003.

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