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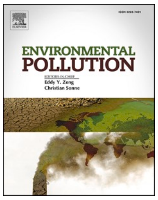

Spatial distribution and composition of mine dispersed trace metals in residential soil and house dust: Implications for exposure assessment and human health☆

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ABSTRACT

Trace metal exposure from environmental sources remains a persistent global problem, particularly in communities residing adjacent to metal extraction and processing industries. This study examines front yard soil and house dust from 62 residences throughout the Australian Ag–Pb–Zn mining city of Broken Hill to better understand spatial variability in metal distributions, compositions and exposures across an industrially polluted urban environment. X-ray fluorescence analysis of paired soil/dust samples indicated that geomean concentrations (mg/kg) of Cu (32/113), Zn (996/1852), As (24/34) and Pb (408/587) were higher in house dust while Ti (4239/3660) and Mn (1895/1101) were higher in outdoor soil. Ore associated metals and metalloids (Mn, Zn, As, Pb) in soil and house dust were positively correlated and declined in concentration away from mining areas, the primary source of metalliferous emissions in Broken Hill. The rate of decline was not equivalent between soil and house dust, with the indoor/outdoor concentration ratio increasing with distance from mining areas for Zn/Pb (geomean = 1.25/1.05 (<1 km); 2.14/1.52 (1–2 km); 2.54/2.04 (>2 km)). House dust and Broken Hill ore Pb isotopic compositions (^{206}Pb/^{207}Pb; ^{208}Pb/^{207}Pb) were more similar in homes nearest to mining areas than those further away (geomean apportioned ore Pb = 88% (<1 km); 76% (1–2 km); 66% (>2 km)), reflecting spatial shifts in the balance of sources contributing to indoor contamination. Incorporation of house dust Pb reduced overestimation of IEUBK modelled blood Pb concentrations compared to when only soil Pb was used. These findings demonstrate that even in contexts where the source and environmental burden of metals are relatively apparent, geochemical relationships and exposures between outdoor and indoor environments are not always predictable, nor easily disaggregated.

1. Introduction

For communities living adjacent to mining and smelting operations, the mobilisation of trace metals and metalloids (hereafter metals) through different environmental mediums has important implications for human exposure and health (Entwistle et al., 2019). Particles dispersed during the extraction and processing of metalliferous ore elevate concentrations of metals in surrounding soil and dust (Csavina et al., 2012). Soils provide a long-term sink for emitted material, leading to the progressive accumulation of metals and forming a reservoir for their potential resuspension and dispersal within the environment (Laidlaw and Filippelli, 2008).

Elevated concentrations of metals in urban soil impacted by mining and smelting operations are often observed to persist in the dust of nearby homes (Argyraki, 2014; Zota et al., 2011). Metals penetrate indoor environments via the entrainment of soil or the direct ingress of airborne particles and amalgamate with other material such as pollen, hair and skin cells to form dust (Lioy et al., 2002). The resulting heterogenous mix of organic and inorganic material is a primary exposure pathway for the ingestion and inhalation of toxic metals by humans (Glorennec et al., 2012; Braun et al., 2020; Kurt-Karakus, 2012). Given that the proportion of time spent indoors is greater than outdoors for the average individual (Brasche and Bischof, 2005; Klepeis et al., 2001), exposure that is attributable to house dust is potentially greater than

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other environmental mediums such as soil (Hogervorst et al., 2007).

Where household level data are not available, indoor exposure risk is often inferred from exogenous geochemical conditions. The US EPA's Integrated Exposure Uptake Biokinetic (IEUBK) Model for Lead in Children, for example, uses a default indoor dust/outdoor soil Pb concentration ratio of 0.7 to estimate the Pb content of house dust from concentrations measured in exterior soil (White et al., 1998). However, given that the contribution of soil to house dust metal concentrations varies considerably (Brattin and Griffin, 2011), reliance on uncontextualized or poorly resolved geochemical relationships may lead to erroneous derivations of exposure risk. A better understanding of the exterior sources and pathways that contribute to indoor contamination is needed to improve exposure risk assessment in different environmental contexts.

With over a century of continuous mining activity and extensive monitoring of contemporary emissions and blood Pb levels in local children (e.g., Dong and Taylor, 2017; Dong et al., 2020; Liu et al., 2021), the Australian Ag–Pb–Zn mining community of Broken Hill provides a relevant context to explore the complexities of metal distribution and exposure in an urban environment impacted by ongoing metalliferous mine emissions. This study investigates the trace metal content and Pb isotopic compositions of deposited dust, residential surface soil and house dust with the specific aims of determining: (1) the contribution of exterior sources and sinks to metal contamination in indoor environments; (2) the transport pathways by which metals are distributed throughout the soil and dust of urban areas; and (3) the influence that these factors have on the assessment of exposure risk in communities impacted by mining emissions.

2. Methods and approach

2.1. Environmental context

The city of Broken Hill, situated in far western New South Wales, Australia, has a population of approximately 17,000. The climate is semi-arid with a mean annual rainfall of 260 mm (Bureau of Meteorology, 2021). Southerly winds prevail annually, with northerlies increasing in frequency during winter (Fig. 1). Vegetation is sparse with minimal groundcover, facilitating dust generation and particle transport.

Mining in Broken Hill is centred on a group of Ag–Pb–Zn rich ore bodies known as the Line of Lode. The Line of Lode intersects the city of Broken Hill along a southwest–northeast axis and is covered by extracted waste rock which divides the city into northern and southern districts (Fig. 1). Metalliferous ore is currently mined by two companies, CBH Resources Limited and Perilya Limited. Mineral extraction now occurs underground, with extracted ore processed and stored aboveground in enclosed areas within the mining leases. Tailings from the operations are stored to the southwest of the city.

2.2. Sample collection and analysis

Indoor vacuum cleaner dust samples were collected from 62 houses in Broken Hill during the Spring of 2018 (Fig. 1). Vacuum dust was collected to provide representative indoor metal concentrations for each house. Participation was voluntary, with residents recruited through Macquarie University's *360 Dust Analysis* citizen science program (360 Dust Analysis, 2020) and via local multimedia and news outlets. Vacuum dust was collected by participants in sealable bags following their regular cleaning routine. One residence provided two vacuum dust samples collected approximately one month apart, which were compared for consistency of metal concentrations. Sample collection was approved via the Macquarie University Human and Research Ethics Committee, number 2446 'New Perspectives on Metals in Indoor Dust in Australian Homes'.

To assess the relationship between metal concentrations in adjacent outdoor and indoor spaces, in situ measurement of front yard surface soil was undertaken at each of the 62 households concurrent with the collection of vacuum dust. Analyses were performed with an Olympus Vanta portable X-ray fluorescence spectrometer (pXRF) fitted with a 50 kV, 4 W tungsten anode X-ray tube. Surface (0–2 cm) soil was measured using the proprietary soil mode for a total of 60 s with 20 s per measurement condition (Beams I–III, 15–50 kV). A composite of five surface soil pXRF measurements was used to capture variability in metal

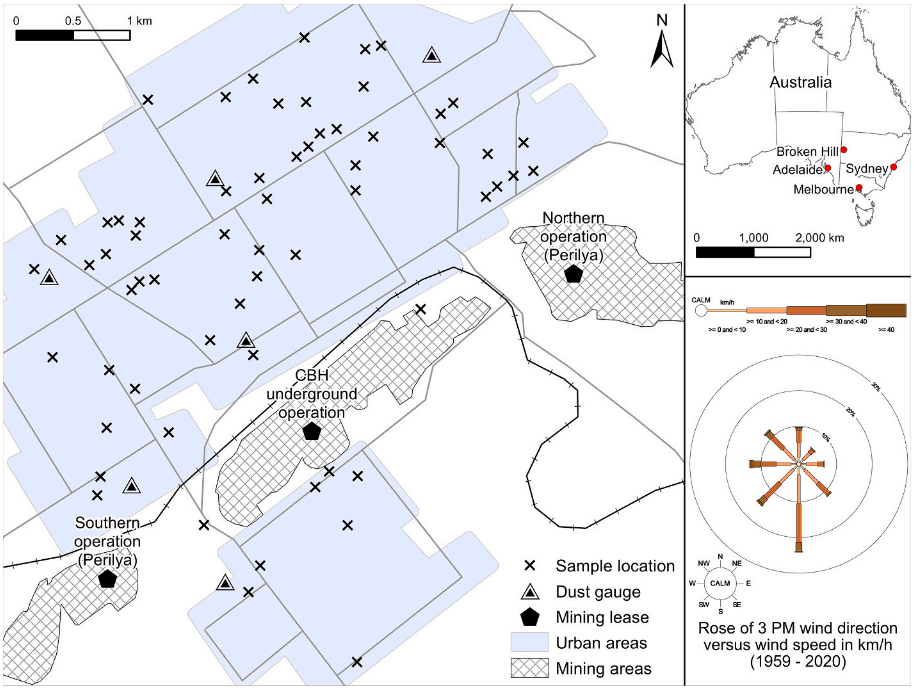

Fig. 1. Map of Broken Hill with sample locations (front yard soil and house dust) and dust deposition gauge sites. Mining areas encompass current mining leases, ore processing and storage facilities, and waste rock deposits. Windrose describes prevailing annual wind direction and speed (Bureau of Meteorology, 2021).

concentrations within front yards. One measurement was taken in the centre of the front yard, and four around its perimeter.

Concentrations are reported in mg/kg for the target metals: Ti, Mn, Cu, Zn, As and Pb. These metals were selected due to their known association with mining in Broken Hill (Mn, Zn, As, Pb) or potential as markers of geogenic soil conditions (Ti) and non-mining related anthropogenic contamination (Cu). The metals Zn and Pb are primary components of the Broken Hill ore body, while Mn and As are secondary contaminants associated with emissions from the mining areas (Taylor et al., 2014; Yang and Cattle, 2018). The abundant geogenic soil metal Ti was included as it was considered unlikely to be strongly influenced by mining emissions or other anthropogenic inputs (Li et al., 2017). In contrast, Cu is an anthropogenic contaminant associated with residential environments (Davis et al., 2001), though is also not strongly enriched within the Broken Hill ore body.

At 10 of the 62 households, approximately 100 g of surface (0–2 cm) soil was collected from immediately below the pXRF analysis point for each of the five front yard analyses, totalling 50 individual soil samples. These samples were collected to estimate the correspondence between in situ pXRF analysis and the analysis of ex situ laboratory prepared samples, as detailed below.

Individual front yard soil ($n = 50$) and house dust ($n = 62$) samples were sieved through a <250 μm stainless steel sieve to remove larger particulate matter and organic debris. Approximately 5 g of sieved material was transferred to XRF sample cups for analysis. Analyses were performed in a test stand using an Olympus Vanta pXRF fitted with a 50 kV, 4 W rhodium (VMR) anode X-ray tube. Samples were measured using the proprietary soil mode for a total of 180 s with 60 s per measurement condition (Beams I-III, 15–40 kV). Duplicate measurements were regularly performed on dust ($n = 6$) and soil ($n = 8$) samples (Supplementary Table S1). Analysis of two house dust samples collected from the same residence approximately one month apart returned consistent metal concentrations, with the relative standard deviation of the two samples ranging from 4% (Pb) to 18% (Zn). Standard pXRF operating procedures for in situ and ex situ analyses involved regular measurements of a silicate (SiO2) blank and National Institute of Standards and Technology (NIST) 2711a (Montana II Soil), 2709a (San Joaquin Soil) and 2583 (Trace Elements in Indoor Dust) Standard Reference Materials (SRM). Mean recoveries were within 15% of certified values for all target metals (86–106%) with the exception of As (98–140%) (Supplementary Table S1).

To enable an equivalent comparison to the <250 μm sieved house dust fraction, the linear equation $y = mx + b$ was applied to adjust metal concentrations returned from individual in situ bulk soil analyses ($n = 310$) relative to the corresponding ex situ analyses of front yard ($n = 50$) soil samples sieved to < 250 μm (Ramsey and Boon, 2012; Rouillon et al., 2017) (Supplementary Figure S2). The mean relative standard deviation of composite ex situ sieved soil samples ranged between 20% (Ti) and 50% (As) compared to composite in situ bulk measurements which ranged between 33% (Ti) and 69% (As). Where concentrations were below pXRF limits of detection, a value equivalent to half the limit of detection was imputed (Supplementary Table S1). The geomean of the five adjusted in situ analyses from each front yard was used to derive a single concentration value for each of the target metals for each residence. Unless otherwise specified, adjusted front yard soil data is referenced for the remainder of this study.

2.3. Inductively coupled plasma mass spectrometry (ICP-MS)

Of the 62 household dust samples, 20 were selected for additional analysis of their Pb concentrations and Pb isotopic compositions. Samples were selected at equal distance intervals from mining areas ($n = 12$) or to identify the source of unusually high Pb concentrations with respect to corresponding front yard soil ($n = 8$). Lead isotopic compositions (^{204}Pb/^{207}Pb, ^{206}Pb/^{207}Pb, ^{208}Pb/^{207}Pb) were measured using quadrapole inductively coupled plasma mass spectrometry (Q-ICP-MS) at the National Measurement Institute (NMI), Sydney, Australia following established methods (National Measurement Institute, 2020; Zhou et al., 2018). Sample digest solutions were diluted as required and analysed with samples bracketed by concentration matched NIST SRM 981 (certified Pb isotopic standard). Raw counts were corrected for blanks and potential Hg isobaric interference, and then corrected for mass discrimination and instrument drift using the average of the bracketing SRM 981 measurements. Method validation data for analytical uncertainties (expressed for NIST SRM 981) were: $^{204}\text{Pb}/^{207}\text{Pb} = 0.0645 \pm 0.0005$; $^{206}\text{Pb}/^{207}\text{Pb} = 1.093 \pm 0.005$; and $^{208}\text{Pb}/^{207}\text{Pb} = 2.370 \pm 0.01$. Instrument performance was assessed relative to NIST SRM 2709a which returned values within the range of published values: $^{204}\text{Pb}/^{207}\text{Pb} = 0.0640$ (±0.0006 measurement uncertainty); $^{206}\text{Pb}/^{207}\text{Pb} = 1.219$ (±0.006); and $^{208}\text{Pb}/^{207}\text{Pb} = 2.485$ (±0.010) (Aung et al., 2004; Reimann et al., 2011; Souto-Oliveira et al., 2019; Takaoka et al., 2006; Unruh et al., 2000). In addition, samples of Broken Hill ore were run on the instrument, which returned mean values ($n = 2$) for: $^{204}\text{Pb}/^{207}\text{Pb} = 0.0648$ (±0.0006); $^{206}\text{Pb}/^{207}\text{Pb} = 1.040$ (±0.006); and $^{208}\text{Pb}/^{207}\text{Pb} = 2.324$ (±0.010), corresponding to published values (Chiaradia et al., 1997; Cooper et al., 1969; Townsend and Hutton, 1998).

Acid digested Pb concentration data were compared to the total Pb concentrations derived from pXRF analysis (Kilbride et al., 2006), and returned a strong positive correlation ($r = 0.91$, $p < 0.01$, $n = 20$). The Pb isotopic composition mixing model developed by Larsen et al. (2012) was used to determine the proportion of Broken Hill ore Pb isotopic compositions (^{206}Pb/^{207}Pb, ^{208}Pb/^{207}Pb) which could be attributed to those in house dust. Background Pb isotopic compositions for Broken Hill soil were derived from the geomean of subsoil samples taken at depths of 30–50 cm ($n = 7$; Kristensen and Taylor, 2016).

2.4. Dust deposition gauges

This study incorporates outdoor Pb loading data from six dust deposition gauges positioned throughout the residential areas of Broken Hill between December 2015 to November 2016 (Fig. 1). Dust gauges were installed and sampled according to Australian Standards (Standards Association of Australia Standards New Zealand, 2003) (Supplementary Text S3A). The mineralogical and morphological properties of the collected dust samples and individual Pb bearing particles were assessed using QEMSCAN® (see generally Pirrie et al., 2009) (Supplementary Text S3B). Rates of external Pb deposition from each dust gauge were compared with the geomean of house dust and front yard soil Pb from the nearest surrounding sampled residences (median distance = 763 m).

2.5. Child blood Pb modelling

The US EPA IEUBK Model for Lead in Children (IEUBKwin2 Build 1.64; US Environmental Protection Agency, 2021) was used to estimate blood Pb concentrations of children aged 1–4 years. To assess the comparative validity of model outputs generated in the absence of site-specific indoor Pb concentrations, two models were run using measured (Model A) and estimated (Model B) house dust Pb values. Model A incorporated site-specific measurements of both front yard soil and house dust Pb concentrations. Model B incorporated measured front yard soil Pb, but with house dust Pb estimated using a conversion factor based on the geomean indoor/outdoor Pb concentration ratio (1.44) measured from residences sampled in this study. Additional model parameters were based on previously published data from Broken Hill for outdoor air Pb (0.28 μg/m^3; Perilya Broken Hill Limited, 2020), drinking water Pb (3 μg/L; Essential Water, 2014), and maternal blood Pb (1.17 μg/dL; Dong et al., 2019), with all other variables set to default. Predicted blood Pb concentrations were summarised using the geomean.

To compare IEUBK model outputs, predicted blood Pb concentrations for each residence were benchmarked against 2018 blood Pb screening results from Broken Hill children aged 1–4 years living within the urban extent of Broken Hill ($n = 604$). Access to blood Pb data was approved following ethics assessment by NSW Greater Western Human Research Ethics Committee (#2020/ETH01783) with concurrence from Macquarie University. Where individual children were tested more than once throughout the year, only the first test result was used. Modelled and measured blood Pb concentrations were compared to the geomean blood Pb of the 10 children living nearest to each sampled residence (median distance = 205 m).

2.6. Statistical and spatial analysis

Data visualisation and statistical and spatial analyses were undertaken using Python 3.9 and ArcGIS 10.6.1. Geochemical data were negatively skewed and non-normally distributed and are therefore described using median and geomean values. Pearson's (r) and Spearman's (r_s) correlations were applied to examine geochemical and spatial relationships and Wilcoxon signed-rank test for differences between exterior and interior metal concentrations. The distance of each residence from mining areas was calculated from the nearest point along the outer boundary of historical and contemporary mining areas (see Fig. 1).

3. Results

3.1. Residential trace metal concentrations

Descriptive statistics for metal concentrations in front yard soil and house dust are reported in Table 1, along with corresponding indoor/outdoor metal concentration ratios. Previous research has identified median background concentrations measured from Broken Hill soil at depths of 30–50 cm for Zn (105 mg/kg) and Pb (100 mg/kg) ($n = 29$; Kristensen and Taylor, 2016). In this study, in situ analysis of front yard surface soil returned median bulk Pb (263 mg/kg) and Zn (602 mg/kg) concentrations approximately 3 and 6 times higher than background levels, respectively. These are primary metals targeted for extraction from the Broken Hill ore body and their enrichment in topsoil is indicative of the deposition of mine dispersed material.

Table 1

Summary statistics for metal concentrations (mg/kg) in front yard soil ($n = 62$) and house dust ($n = 62$). Corresponding indoor/outdoor concentration ratios ($n = 62$) are also provided. Data for individual residences are reported in Supplementary Table S4.

	Ti	Mn	Cu	Zn	As	Pb
Front yard soil (mg/kg)						
Minimum	1417	287	8	32	4	11
10th percentile	3305	978	18	411	10	133
25th percentile	3733	1334	24	654	15	252
Median	4387	1686	31	930	21	382
75th percentile	5146	2551	42	1448	31	672
90th percentile	5868	5655	51	3470	72	1626
Maximum	7406	9267	138	6134	272	3701
Geomean	4329	1895	32	996	24	408
Mean	4482	2396	36	1396	36	635
Standard deviation	1146	1991	21	1315	46	687
House dust (mg/kg)						
Minimum	714	226	35	514	6	118
10th percentile	2376	517	57	872	15	234
25th percentile	3120	734	73	1228	25	334
Median	3942	964	106	1791	34	535
75th percentile	4450	1453	165	2745	53	978
90th percentile	5011	3073	206	3835	82	1635
Maximum	8454	7542	4848	11,692	113	2937
Geomean	3660	1101	113	1852	34	587
Mean	3868	1418	197	2243	41	783
Standard deviation	1225	1269	604	1688	27	657
Concentration ratio (indoor/outdoor)						
Minimum	0.11	0.07	0.65	0.17	0.13	0.07
10th percentile	0.54	0.33	1.58	0.82	0.58	0.65
25th percentile	0.67	0.43	2.24	1.16	1.00	0.93
Median	0.87	0.58	3.52	1.80	1.52	1.33
75th percentile	1.03	0.79	5.37	2.82	2.06	1.88
90th percentile	1.42	1.02	7.51	4.58	4.77	3.51
Maximum	2.74	1.86	35.13	78.78	14.50	32.09
Geomean	0.85	0.58	3.53	1.86	1.43	1.44
Mean	0.93	0.65	4.90	3.38	2.05	2.36
Standard deviation	0.41	0.32	6.05	9.82	2.18	4.39

Distinct geochemical differences were observed between interior and exterior environments in Broken Hill. Geomean Cu, Zn, As and Pb concentrations were higher in house dust than in front yard soil by a factor of 3.53 (113/32 mg/kg), 1.86 (1852/996 mg/kg), 1.43 (34/24 mg/kg) and 1.44 (587/408 mg/kg), while Ti and Mn were 0.85 (3660/4329 mg/kg) and 0.58 (1101/1895 mg/kg) times lower, respectively (Table 1; Fig. 2). A Wilcoxon signed-rank test indicated a significant median difference between front yard soil and house dust metal concentrations ($p < 0.01$; Supplementary Table S5A). Exterior and interior concentrations were positively correlated for the ore associated metals Mn, Zn, As and Pb ($r_s = 0.38$ to 0.62, $p < 0.01$), though not for Ti and Cu ($r_s = 0.06$ to 0.12, $p > 0.05$) (Supplementary Table S5B), which are not strongly enriched within the Broken Hill ore body.

A clear geochemical signature of decreasing concentration with distance away from mining areas was observed indoors and outdoors with respect to the ore associated metals Mn, Zn, As, and Pb (Fig. 3). The trend was strongest for front yard soil ($r_s = -0.45$ to -0.68, $p < 0.01$), though persisted in house dust ($r_s = -0.36$ to -0.58, $p < 0.01$) (Supplementary Table S5C). The decline in the concentration of the primary ore derived metals was greater in soil compared to house dust, with the indoor/outdoor concentration ratio increasing with distance from mining areas for Zn (geomean = 1.25 (<1 km); 2.14 (1–2 km); 2.54 (>2 km); $r_s = 0.30$, $p < 0.05$, $n = 62$) and Pb (geomean = 1.05 (<1 km); 1.52 (1–2 km); 2.04 (>2 km); $r_s = 0.31$, $p < 0.05$, $n = 62$; Fig. 3).

3.2. House dust Pb isotopic compositions

Geomean Pb isotopic compositions of house dust ($^{206}Pb/^{207}Pb = 1.05$; $^{208}Pb/^{207}Pb = 2.33$) were similar to Broken Hill ore ($^{206}Pb/^{207}Pb = 1.04$; $^{208}Pb/^{207}Pb = 2.31$) and distinct from subsoil samples taken at depths of 30–50 cm ($^{206}Pb/^{207}Pb = 1.11$; $^{208}Pb/^{207}Pb = 2.36$) (Kristensen and Taylor, 2016; see also Kristensen et al., 2015) (Fig. 4A). House dust Pb isotopic compositions overlapped with previously published Pb isotopic data from Broken Hill outdoor deposited dust (Dong and Taylor, 2017), surface soil (Kristensen and Taylor, 2016), ceiling dust and house dust samples (Gulson et al., 1994) (Fig. 4A). House dust Pb isotopic compositions were distinct from sources of Broken Hill Pb petrol and displayed considerably lower variation than Pb paint sampled from Broken Hill homes (Gulson et al., 1994).

Using the isotopic compositions of Broken Hill ore Pb and subsoil (30–50 cm) Pb as endmembers, source apportionment modelling (Larsen et al., 2012) indicated that the composition of house dust Pb corresponds closely to the ore body source with a geomean apportioned ore Pb contribution of 76% (Fig. 4B). Using the same endmembers, geomean apportioned contributions of previously analysed samples of outdoor deposited dust (Dong and Taylor, 2017), surface soil (Kristensen and Taylor, 2016), vacuum dust and ceiling dust (Gulson et al., 1994) from Broken Hill were 88%, 83%, 87% and 95%, respectively. Spatial variability in Pb isotopic compositions reported for atmospherically deposited Pb (Dong and Taylor, 2017) was also observed in house dust Pb, which grew increasingly disparate from the Broken Hill ore body with distance from mining areas (geomean apportioned ore Pb = 88% (<1 km); 76% (1–2 km); 66% (>2 km); $r = -0.58$, $p < 0.01$, $n = 20$) (Fig. 4B).

3.3. Dust deposition gauges

Table 2 summarises the geochemical and morphological data collected from the six dust deposition gauges installed in Broken Hill and sampled monthly from December 2015 to May 2016. Deposition rates

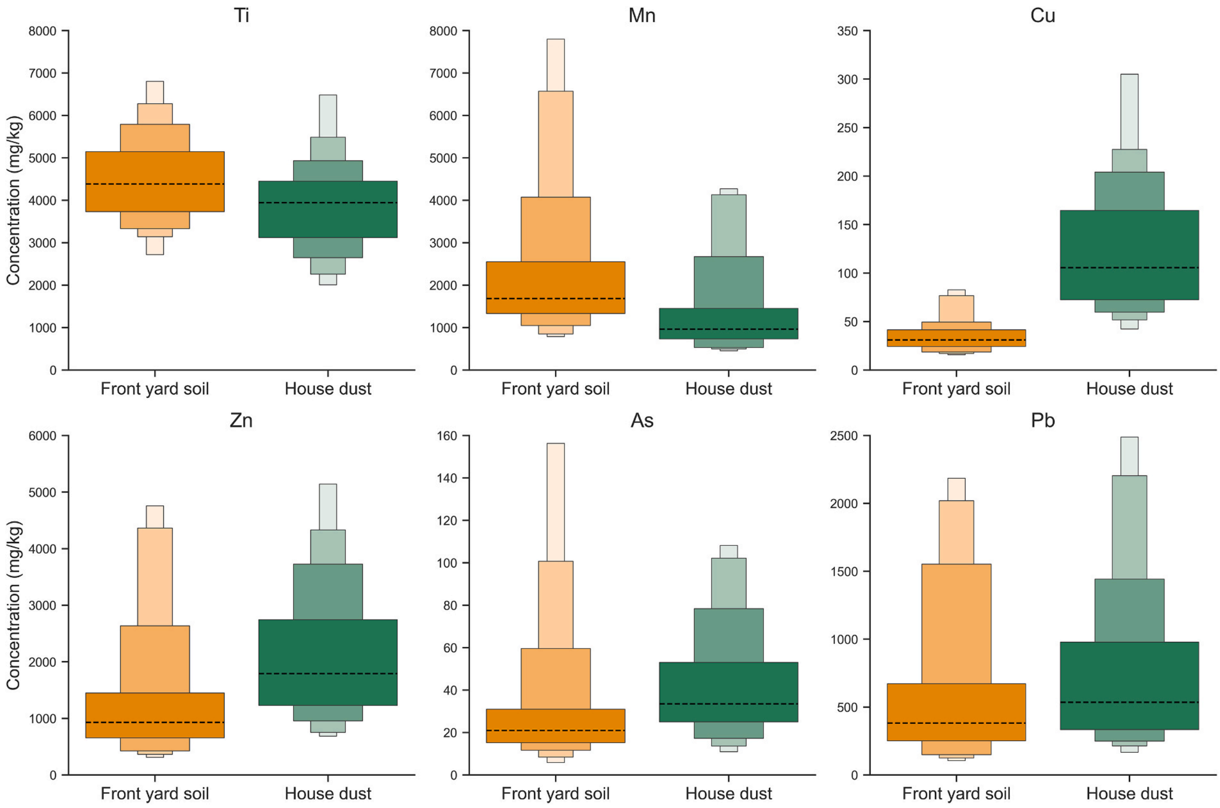

Fig. 2. Percentile distribution of metal concentrations (mg/kg) in front yard soil and house dust. The dashed line designates the median and box ends correspond to the 6.25th (1/16), 12.5th (1/8), 25th (1/4), 75th (3/4), 87.5th (7/8) and 93.75th (15/16) percentiles, respectively. Values beyond the outer percentiles are not shown.

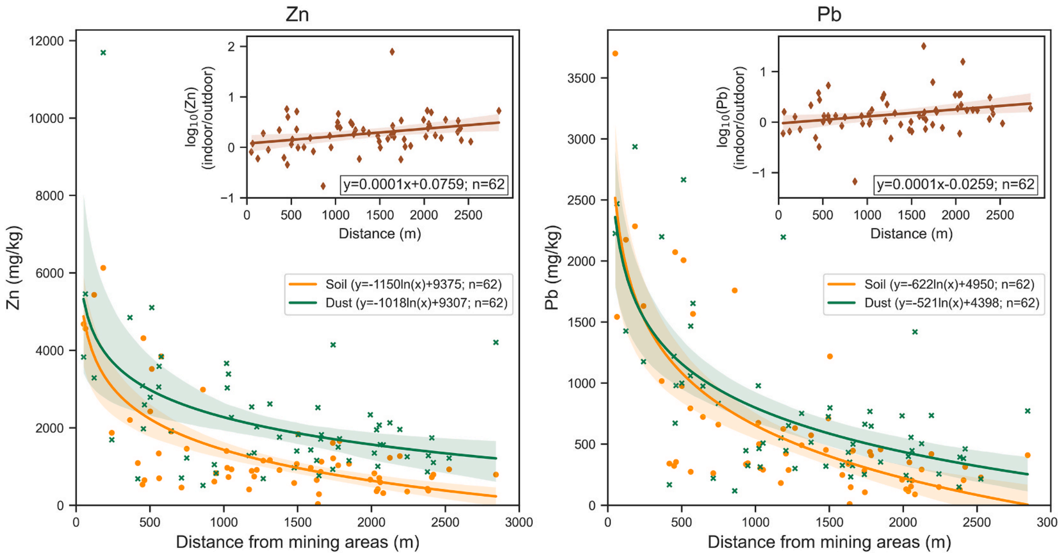

Fig. 3. Front yard soil (●) and house dust (×) Zn and Pb concentrations relative to distance from mining areas (logarithmic regression ± 95% confidence interval). Inset graphs show the corresponding $\log_{10}$ transformed indoor/outdoor Zn and Pb concentration ratios (linear regression ± 95% confidence interval). Statistics reported in text refer to non-transformed data.

for Pb are comparable to those reported at the same sample sites between June and November 2015 (Dong and Taylor, 2017). QEMSCAN® analysis of individual particle morphologies revealed that between 48 to 85% of mapped Pb bearing particles ($\geq$1 μm) were 10 μm or smaller, with gauges D1 and D2 closest to mining areas returning the highest proportion of Pb bearing particles >10 μm (Table 2; Supplementary Figure S6A). Geomean Pb concentrations for residences surrounding each dust deposition gauge showed a significant positive correlation with external Pb deposition rates for house dust ($r = 0.94$, $p < 0.01$, $n = 6$), with a similar, albeit non-significant correlation with front yard soil ($r = 0.80$, $p = 0.06$, $n = 6$) (Table 2).

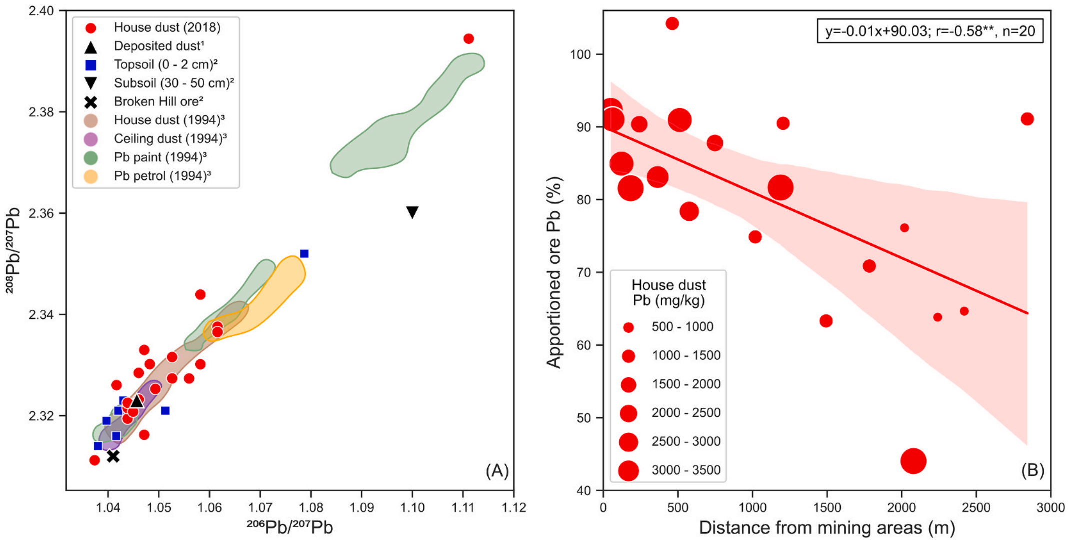

Fig. 4. (A) Pb isotopic compositions ($^{206/207}$Pb and $^{208/207}$Pb) for Broken Hill house dust (n = 20) compared with previous analyses of deposited dust (Dong and Taylor, 2017)[1], topsoil, subsoil, ore (Kristensen and Taylor, 2016)[2], house dust, ceiling dust, paint, and petrol (Gulson et al., 1994)[3] from Broken Hill. (B) Estimated contribution (Larsen et al., 2012) of Broken Hill ore Pb to the Pb isotopic composition of sampled house dust with respect to distance from mining areas (linear regression ± 95% confidence interval; $^{**}p < 0.01$). Point size corresponds to the Pb concentration (mg/kg) returned from ICP-MS analysis.

Table 2
Combined Pb deposition rates at Broken Hill sampling locations (D1 – D6) between the period December 2015 to May 2016. Particle size measurements are derived from QEMSCAN® analysis of individual Pb bearing particles (≥1 μm) (additional information in Supplementary Table S6B). Geomean house dust and front yard soil Pb concentrations are reported for residences nearby each dust gauge site.

Dust gauge site (see Fig. 6)	D1	D2	D3	D4	D5	D6
Collection period (days)	148	148	148	148	148	95[a]
Distance from mining areas (m)	513	532	654	2215	1914	1709
Pb deposition rate (μg/m^2/day)	289	179[b]	99	5	39	12
Solids deposition rate (μg/m^2/day)	54,133	48,475	44,805	7225	23,779	18,046
Solids sample weight (mg)	140	130	120	20	60	30
Pb bearing particle size ≤ 10 μm (%)	59	48	76	85	69	76
Pb bearing particle size range (μm)	1–95	1–68	1–105	1–46	1–44	1–59
Pb bearing particle size count (n)	229[c]	178[c]	203[c]	27	29	25
Nearby house dust Pb (mg/kg)	1464	745	601	424	514	496
Nearby front yard soil Pb (mg/kg)	1268	444	954	220	259	400
Nearby sampled residences (n)	8	5	6	12	15	16

[a] Sample from one month lost due to equipment disturbance.
[b] Does not include dissolved Pb due to inadvertent discarding of supernatant water.
[c] Does not include a 40 mg subsample used for ICP-MS analysis.

3.4. *Child blood Pb modelling*

The central distribution of IEUBK modelled child blood Pb concentrations (US Environmental Protection Agency, 2021) corresponded with concentrations measured from Broken Hill children aged 1–4 years living nearby each sampled residence in 2018 (Fig. 5A). This was consistent between model outputs incorporating measured indoor dust Pb values (Model A) and those estimating indoor dust Pb concentrations from corresponding soil Pb (Model B) using the indoor/outdoor Pb concentration ratio of 1.44 reported herein. Model B, incorporating estimated indoor dust Pb values, was more susceptible to overestimation of blood Pb levels for residences near mining areas with elevated soil Pb, and underestimating blood Pb where soil Pb was low (predicted/measured geomean (μg/dL) = 8.52/5.71 (<1 km); 4.60/4.49 (1–2 km); 3.21/3.61 (>2 km); Fig. 5B). Incorporation of measured indoor dust Pb values in Model A resulted in moderate improvements to model accuracy at the margins of the predicted blood Pb distribution, though continued to overestimate blood Pb in residences near mining areas with high soil and house dust Pb concentrations (predicted/measured geomean (μg/dL) = 7.95/5.71 (<1 km); 4.67/4.49 (1–2 km); 3.78/3.61 (>2 km); Fig. 5B).

4. Discussion

4.1. *Trace metal sources*

The results of this study align with anticipated links between mining activity and environmental contamination in Broken Hill (Yang and Cattle, 2018). Metals associated with the ore body, including Mn, Zn, As and Pb, are enriched in surface soil at concentrations that do not correspond to subsurface soil nor the background geology of the region (Kristensen and Taylor, 2016). Associations between these primary ore associated metals and the decrease in their enrichment in surface soil with distance from mining areas are characteristic of mine and smelter impacted areas globally (Fry et al., 2020; Soto-Jiménez and Flegal, 2011).

Several studies have highlighted the importance of soil as a reservoir for the transfer of metals into indoor environments (Argyraki, 2014; Zota et al., 2011), and up to 78% of indoor dust is estimated to originate from outdoor sources (Calabrese and Stanek, 1992). The strength of relationships observed between Mn, Zn, As and Pb in soil and house dust suggest a common source is contributing to metal contamination of both indoor and outdoor environments in Broken Hill. This is supported by similarities in the apportioned contribution of Broken Hill ore Pb to house dust and previously sampled surface soil Pb (Kristensen and Taylor, 2016). Comparatively lower variability in the apportioned ore Pb contribution observed for ceiling dust (Gulson et al., 1994) and externally deposited Pb (Dong and Taylor, 2017) may reflect the susceptivity of these mediums to inputs of metalliferous mine dust

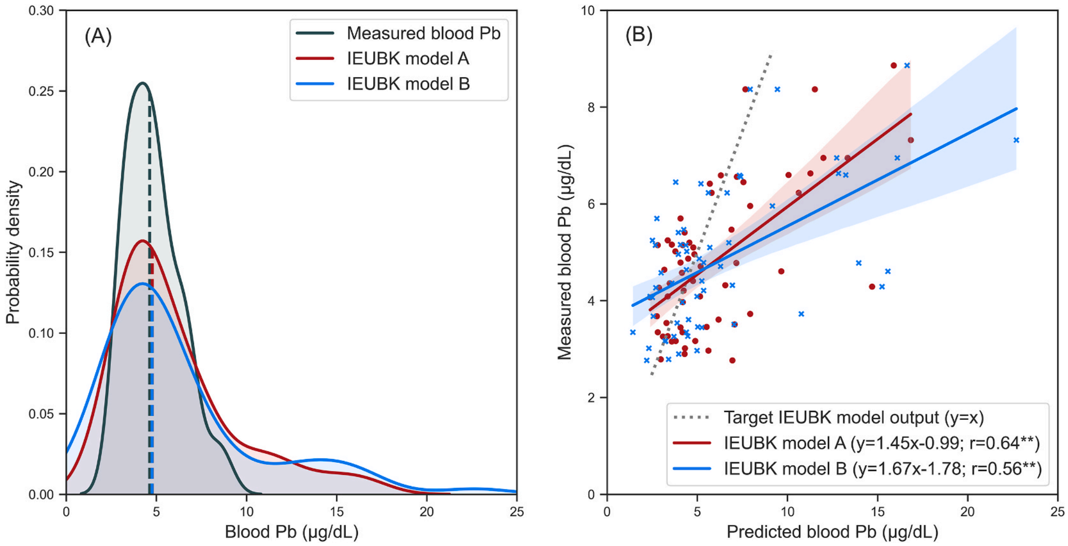

Fig. 5. (A) Kernel density estimate (KDE) showing continuous probability density curve of IEUBK predicted (Model A: measured indoor dust Pb; Model B: estimated indoor dust Pb; n = 62) and measured blood Pb concentrations of Broken Hill children (geomean of a subset of the 10 nearest tested children; n = 62). The dashed line designates the median of each distribution. (B) Relationship between predicted (Model A (●); Model B (×)) and measured blood Pb concentrations (linear regression ± 95% confidence interval; $^{**}p < 0.01$). The dotted line indicates the target linear relationship where predicted blood Pb is equal to measured blood Pb. Summary statistics for IEUBK model outputs are provided in Supplementary Table S7.

dispersed through atmospheric pathways, and additional inputs of secondary Pb sources such as paint, petrol and country rock to soil and house dust. The clear spatial shift in the Pb isotopic compositions of house dust Pb with distance from mining areas nonetheless corresponds with a decline in the deposition and resuspension of metal enriched particles away from the emission source (Fig. 4B). These patterns are contrary to those expected in non-industrial urban environments, where population and traffic density exert a stronger influence on the distribution of metals in soil and dust (Trujillo-González et al., 2016).

4.2. *Trace metal pathways*

Metal enrichment of topsoil surrounding mining areas occurs as particulate matter dispersed during ore extraction and processing settles into the upper layer of the soil profile (Csavina et al., 2011; Mackay et al., 2013). Dust loading and Pb deposition rates from six dust gauges installed across Broken Hill indicate enhanced particle dispersal at sites closest to active mining areas (D1 – D2), where concentrations of Pb in soil and house dust were highest (Table 2; Fig. 6). At these sites, a larger size distribution of deposited Pb bearing particles is indicative of the inverse relationship between the size and density of an airborne particle

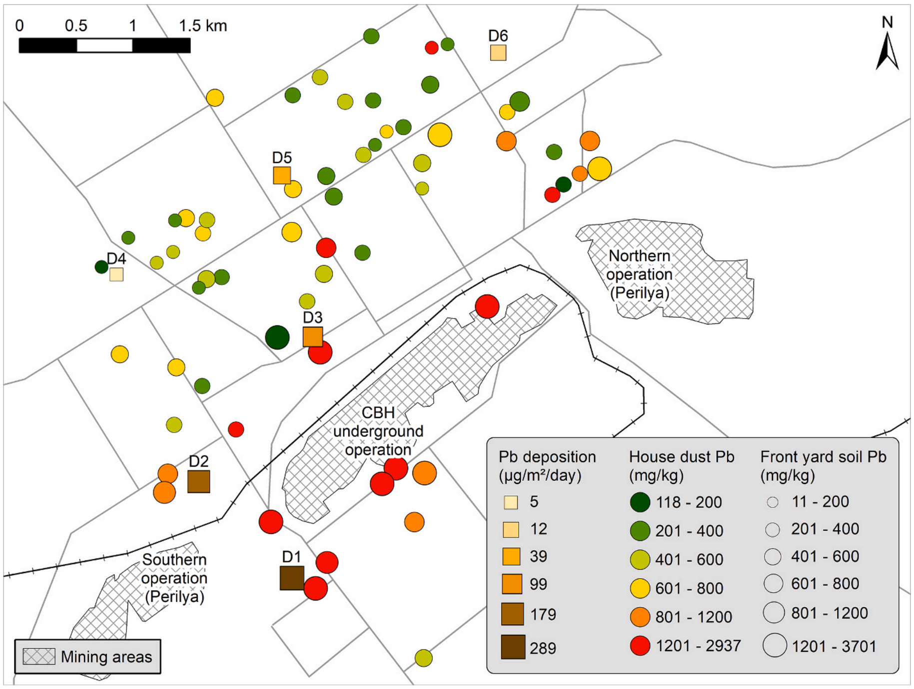

Fig. 6. Front yard soil (● size) and house dust (● colour) Pb concentrations (mg/kg) for all sampled residences in this study (n = 62). External Pb deposition rates (μg/m^2/day) for the six dust deposition gauges positioned throughout Broken Hill are also shown (Table 2).

and its atmospheric residence time (Csavina et al., 2012). Most coarse mine dispersed particles, for example, are reported to settle within 1 km of their source (e.g., Cattle et al., 2012). However, given that the metal enrichment of Broken Hill surface soil persists well outside this 1 km range, a smaller proportion of dispersed particles must either remain suspended for longer periods or be remobilised as windblown dust or through anthropogenic disturbance. The semi-arid climate and sparse vegetative cover of Broken Hill would facilitate the dispersal, resuspension and indoor migration of metalliferous particles (e.g., Csavina et al., 2014), as is evidenced by the correspondence between external Pb deposition rates and house dust Pb concentrations (Fig. 6).

Elevated concentrations of metals in indoor environments have been reported for residential areas with and without major industrial activity (Rasmussen et al., 2001; Zota et al., 2011). In Broken Hill, house dust returned higher Cu, Zn, As and Pb concentrations than was observed for corresponding front yard surface soil (Fig. 2). Elevated indoor concentrations of Cu, which is neither strongly enriched in Broken Hill ore nor correlated with corresponding surface soil concentrations, may be reasonably attributed to indoor sources (cf. Rasmussen et al., 2001). The enrichment of ore associated metals Zn, As and Pb in house dust is more likely due to their disproportionate concentration in the fine particle size fraction following ore extraction and concentrating processes and the corresponding susceptibility with which this fraction is entrained and transported indoors (see Hunt et al., 2006; Laidlaw and Filippelli, 2008). For example, Pb bearing particles captured by dust deposition gauges were concentrated in the finer and more readily airborne $\leq$10 μm particle fraction (Table 2; Supplementary Figure S6A). Particles of an equivalent size ($\leq$10 μm) are reported elsewhere to account for 50% of the weight of indoor dust samples under 80 μm (Rasmussen et al., 2018). This may explain the strength of the relationship between indoor dust Pb concentrations and the Pb deposition rate at nearby dust gauges, as the majority of atmospherically deposited Pb particles are within the size fraction most likely to infiltrate indoor environments.

By comparison, the more uniform distribution of the naturally abundant soil metal Ti throughout the soil matrix may have contributed to its roughly equivalent concentrations in front yard soil and house dust (see Li et al., 2017) (Fig. 2). This, however, does not explain the apparent depletion of Mn in house dust, which like Zn, As and Pb, is enriched in Broken Hill ore and local surface soil (Yang and Cattle, 2018). Comparative depletions of Mn in house dust relative to garden soil have been reported in different environmental and geological contexts (Argyraki, 2014; Rasmussen et al., 2001; Zota et al., 2011). The interaction of Mn and Fe oxides in soil might explain this trend, with the formation of Mn–Fe nodules possibly leading to the adsorption and immobilisation of deposited Mn in soil (see Gasparatos et al., 2005).

4.3. Implications for exposure assessment

The amplification of metal concentrations between soil and house dust in Broken Hill correspond with previous findings that characterise indoor environments as a more constant and potentially more important route of exposure than outdoor areas (Hogervorst et al., 2007; Turner, 2011). Geochemical and morphological factors influencing the transfer and accumulation of metals indoors also influence human exposures at household levels (Beamer et al., 2012) and over larger spatial scales.

Indoor/outdoor concentration ratios for Cu, Zn, As and Pb varied considerably across Broken Hill homes (Table 1). The consistency of house dust Pb isotopic compositions with respect to Broken Hill ore (Fig. 4) suggest that household sources of Pb did not contribute as significantly to this heterogeneity as is reported for non-industrial residential environments (Rasmussen et al., 2001). In a highly industrialised context such as Broken Hill, different household conditions, cleaning regimes, and routes of metal ingress may instead explain the variability between front yard soil and house dust Pb concentrations. For example, transport pathways including the influx of windblow dust, release from legacy reservoirs of ceiling dust, and entrainment on the clothing and footwear of mine workers could bypass existing soil reservoirs within the vicinity of the home, amplifying inconsistencies between exterior and interior metal concentrations (e.g., Davis et al., 2016; Gulson et al., 1996). Such pathways undoubtedly contribute to harmful accumulations of toxic metals inside homes where the assessment of exterior soil would identify no significant human health risk.

The observed increase in indoor/outdoor Pb and Zn concentration ratios with distance from mining areas (Fig. 3) has implications for metal exposure over larger spatial scales. Size selective sorting of metalliferous dust dispersed from mining areas may lead to the increase in indoor/outdoor metal concentration ratios, as the proportion of mobile particles within the size fraction most susceptible to migration indoors would increase with successive episodes of transport away from the emission source. This is reflected by an increase in the proportion of deposited Pb bearing particles $\leq$10 μm in dust gauge sites distal to mining areas (Table 2). While the residential extent of Broken Hill is limited, in larger industrialised urban settings (e.g., Fry et al., 2021; Soto-Jiménez and Flegal, 2011), an increase in indoor/outdoor metal concentration ratios away from emission sources could more significantly shift the balance of exposure risk to indoor environments.

Despite its relevance to human health, exposure and uptake in indoor environments is often determined by way of simple extrapolation of external contaminant concentrations (e.g., Department for Environment, Food and Rural Affairs DEFRA and The Environment Agency EA, 2002; McKone, 1993; White et al., 1998). In this study, significant heterogeneity and spatial asymmetry observed between the metal content of soil and house dust underscores the limitations of estimating exposure risk based on assumed geochemical relationships between different environmental mediums. To this effect, incorporation of measured indoor dust Pb enabled spatial variability in indoor/outdoor Pb concentrations to be captured, improving the overall correspondence of IEUBK model outcomes with respect to local scale heterogeneity in adjoining childhood Pb exposures (cf. Brattin and Griffin, 2011; Gulson et al., 2018). Nonetheless, the continued overestimation of blood Pb in children living near mining areas suggests there is a disconnect between childhood exposures and total soil and dust Pb concentrations observed in the more contaminated residential areas of Broken Hill.

Localised inaccuracies in IEUBK model outputs highlight the limitations of total metal concentrations as a measure of site specific exposure risk. In this study, the default IEUBK model value of 30% absolute Pb bioavailability (the fraction of ingested Pb absorbed into the bloodstream) was retained for soil and house dust (US Environmental Protection Agency, 2021). This value corresponds with the mean absolute Pb bioavailability estimated for Broken Hill surface soil (26%; Yang and Cattle, 2015) yet does not account for increases in Pb bioaccessibility (the fraction of ingested Pb that is solubilised within gastrointestinal fluid) nearer to mining areas where total Pb concentrations are highest in soil and dust (Dong and Taylor, 2017; Yang and Cattle, 2015). This suggests that relative to total Pb concentrations, exposure risk is disproportionately higher in homes surrounding mining areas, which does not appear to be the case given the relatively moderate increase in measured child blood Pb concentrations observed in these areas.

Other factors such as spatial variability in the size distribution of metal enriched particles may also alter rates of uptake through ingestion, inhalation and dermal pathways (see Li et al., 2017; Youn et al., 2016). Coarse, mechanically generated mine particles, for example, may pose a lower exposure risk than is implied by their total Pb concentration as they are less likely to adhere to the hands of children and may contain lower concentrations of bioaccessible metals than finer particles (Li et al., 2021; Yamamoto et al., 2006). Given these uncertainties, incorporation of spatially orientated measures of metal bioaccessibility and particle size distributions in soil and house dust, even at relatively coarse scales, would benefit the assessment of exposure risk across industrialised urban gradients.

4.4. Limitations

As this study relied on the voluntary participation of Broken Hill community members, sampling is potentially skewed towards participants that are most concerned, and therefore most proactive, about addressing metal contamination within their homes. Consequently, geochemical data reported here may underestimate residential metal contamination in Broken Hill.

Previous research has highlighted the limitations of vacuum dust as a measure of indoor metal concentrations compared to other sampling methods such as the collection of deposited dust (e.g., Rasmussen et al., 2018; Rasmussen et al., 2013). In this study, inconsistencies associated with different participant cleaning regimes, vacuum cleaner models, and the possibility of particle fractionation are all recognised to potentially contribute to additional heterogeneity in the metal concentrations of analysed house dust samples. For example, variable efficiency in the removal of fine dust particles during vacuuming may lead to the underreporting of metal concentrations, especially for flooring types with high levels of surface-particle adhesion (Hunt et al., 2008).

5. Conclusions

This study explored indoor and outdoor metal contamination in an urban context impacted by metalliferous mine emissions to determine: (1) the contribution of exterior sources to the metal contamination of indoor environments; (2) the distribution of metals throughout residential areas and living environments; and (3) how these factors may influence exposure assessment in industrialised urban environments.

Spatially variable associations between the Pb isotopic compositions of Broken Hill house dust, exterior dust and soil, and the local orebody are indicative of the source, distribution and environmental fate of Pb mining emissions. Accompanying declines in the metal concentration of surface soil and house dust with distance from mining areas reflect variability in the transfer and accumulation of contaminants between outdoor and indoor environments. The decline is not constant between these environments, with elevated metal concentrations in house dust persisting at greater distances from emission sources than in corresponding surface soil.

Variability in the metal concentration of potential exposure sources has important implications for evaluating risk in communities impacted by polluting industries. In the context of Broken Hill, it is evident that estimating risk based on a single exposure source or static relationship between these sources – in this case soil and house dust – may result in a misleading assessment of blood Pb exposures amongst the community. Given the importance of the home environment to childhood exposures, estimates of risk based solely on exterior mediums should be viewed with caution. This is particularly the case where metal sources and pathways between outdoor and indoor living spaces are dynamic and poorly characterised.

Author credit statement

Max M. Gillings: method and approach, analysis, investigation, writing (original draft, review and editing), visualisation; **Kara L. Fry:** method and approach, investigation, writing (review and editing); **Anthony L. Morrison:** method and approach, analysis, investigation, writing (review and editing); **Mark Patrick Taylor:** obtained funding, conceptualisation, method and approach, writing (review and editing), supervision, project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.118462.

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