Identification of environmental lead sources and pathways in a mining and smelting town: Mount Isa, Australia

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Abstract

Lead (Pb) concentrations and isotopic compositions from soils, dusts and aerosols from public land and residential lots adjacent to the copper and Pb mine and smelter at Mount Isa, Australia were examined to understand the sources and risks of environmental Pb exposure. Soil samples contain elevated Pb concentrations (mean 1 560 mg/kg), of which 45-85 % of the Pb is bioaccessible. The Pb isotopic composition of surface soils (0-2 cm), aerosols and dusts ($^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ range: 1.049, 2.322-1.069, 2.345) are dominated by Pb derived from the Mount Isa Pb-zinc ore bodies. Underlying soil horizons (10-20 cm) have distinctly different Pb isotopic compositions ($^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ range: 1.093, 2.354-1.212, 2.495). Surface soil-, dust- and aerosol-Pb are derived predominantly from smelter emissions and fugitive mining sources and not from weathered bedrock. Remediation strategies should target legacy and ongoing sources of environmental Pb to mitigate the problem of Pb exposure at Mount Isa.

Capsule

Environmental Pb contamination in the Mount Isa urban area is derived primarily from smelter emissions and local fugitive mining sources and not natural *in situ* surface mineralization.

Keywords: lead (Pb); Blood lead (PbB); contamination; lead isotopes; mining and smelting; Mount Isa.

Introduction

The neurotoxic effect of lead (Pb) has been long established in the scientific literature and children are particularly at risk if exposed (Needleman, 2004). Children presenting with blood lead (PbB) levels well below 10 µg/dL have been linked with decreased Intelligence Quotient (IQ) and academic performance, as well as a range of socio-behavioural problems (Lanphear et al., 2000, 2005; Canfield et al., 2003; Braun et al., 2006; Bellinger, 2008; Jusko et al., 2008; National Toxicology Program, 2012; Taylor, Winder and Lanphear, 2012). Concerns have been raised about the impact of PbB levels in children across a number of Australian mining, smelting and port towns, including Broken Hill (Gulson et al., 1994a, b, c), Lake Macquarie (Gulson et al., 2004; Dalton and Bates, 2005), Mount Isa (Munksgaard et al., 2010; Taylor et al., 2010a; Taylor and Schniering, 2010), Port Kembla (Young et al., 1992), Port Pirie (Baghurst et al., 1992, Taylor, 2012), Rosebery (MCPHR, 1999) and Esperance (Gulson et al., 2009).

A survey by Queensland Health (2008) at the major mining centre of Mount Isa, north-west Queensland (Figure 1), indicated that 11.3 % of 400 local children sampled aged between 1 and 5 years of age had PbB levels above the generally applied risk of harm value of 10 µg/dL, as set by the Australian National Health and Medical Research Council (NHMRC 1993, 2009). In contrast to other Australian mining and smelting towns, Mount Isa has only recently started to receive considerable attention regarding the environmental and human health impacts arising from mining-related activities (e.g. Taylor and Hudson-Edwards, 2008; Noller et al., 2009; Munksgaard et al., 2010; Taylor et al., 2009, 2010a). Research of soil, air and water (including sediments) systems in and around Mount Isa has demonstrated a significant proportion of the urban area and riverine environment is contaminated with Pb and other metals and metalloids (Taylor and Hudson-Edwards, 2008; Taylor et al., 2009; Mackay et al., 2011; Mackay and Taylor, 2013). The combination of elevated Pb in soil (Taylor et al., 2010a) and dust (Queensland EPA, 2008; Taylor et al., 2010b), coupled to the semi-arid climate, provide environmental conditions that enhance the transmission of Pb into residential lots. This makes household dusts and urban soils a likely primary pathway and places Mount Isa children at a potentially significant risk of harm from Pb exposure. This study seeks to identify the sources, pathways and relative risk of environmental Pb at Mount Isa via the analysis of the concentrations and isotopic compositions of Pb in soil, aerosol, rock and dust. The data generated from this research provides valuable information to guide local regulatory authorities in directing environmental remediation in Mount Isa. In addition, the methods and research design used in this study are of direct relevance to other mining towns in Australia, and elsewhere in the world, where mining and industrial activities pose a potential serious risk of harm.

Study area

Mount Isa is located in north-west Queensland, Australia (Figure 1). Settlement occurred in 1923 following the discovery of Pb-silver(Ag)-zinc(Zn) ore, and later copper (Cu) ore, and subsequent mining and smelting commenced in 1931 (Carter et al., 1961). Large-scale open-pit and underground mining and smelting of Pb-Ag-Zn and Cu ore still continues (Mudd, 2007), making Mount Isa one of the largest mining cities in Australia (population 21 237: ABS, 2011). Xstrata Pty Ltd currently own and operate a number of underground and open pit mines and smelters at Xstrata Mount Isa Mines, Mount Isa (hereafter referred to as XMIM).

The National Pollutant Inventory (NPI, 2012) identified the XMIM facility as Australia’s largest emitter of arsenic, cadmium, Cu, Pb, sulphur dioxide and Zn compounds into surrounding air, land

and water systems from the period between July 1999 and June 2008. The modern sources have been identified previously as direct emissions from the Cu and Pb smelter stacks and fugitive emissions from mining, ore transport, ore preparation, waste disposal, and sintering and smelting operations (EPA, 2008). Despite upgrades to emission capture technology and ore management techniques at XMIM, the combined effects of historic and contemporary emissions contribute to significant environmental Pb contamination in and around the Mount Isa urban area (Taylor et al., 2010a, b; NPI, 2012).

Material and methods

A summary of materials collected and analytical methods used during this study are presented in Table 1.

Soils

Sampling of soil in the Mount Isa urban area was conducted between September 2005 and July 2008. The sampling design was aimed at characterising both the lateral spatial pattern and depth profiles of soil-Pb concentrations across the Mount Isa urban area (Figure 1).

Surface soil samples were collected in the Mount Isa urban area between September 2005 and July 2008 (Table 1) from: twenty public use lands (US); five residential lots (P1 to P5); a workplace lot (CP) and; the entrance to a school playground (SP) (Figure 1). The selection of the five residential lots for sampling was based on the PbB levels of the residing child who participated in the Queensland Health (2008) survey. Sampling of three residential lots (P1, P2 and P3) with children who presented with a PbB level exceeding 10 μg/dL was completed in April and May 2008. A further two residential lots (referred to as ‘control’ lots – P4 and P5), housing children with a PbB level below 10 μg/dL, were sampled in July 2008.

Fifteen soil subsamples from eight soil profiles (MIS; Figure 1) at depths of 0-2 cm and 10-20 cm from the ground surface, whose total Pb concentrations are reported in Taylor et al. (2010a), are also used in this study to measure surface (0-2 cm) and proxy background (10-20 cm) soil-Pb isotopic composition (Table 1).

Soil samples were oven dried at 37 ± 2 °C for 24 hours prior to being fractionated into three grain size fractions: < 2 mm, < 180 µm, and the < 63 µm fraction. This study focuses primarily on the < 180 µm fraction because this fraction is readily adhered to hands (hand-to-mouth behaviours), easily inhaled or ingested (Duggan and Inskip, 1985) and is used commonly in human and environmental health risk assessments (Naidu, 2008).

With the expectation of the soil profile subsamples used for determining surface versus subsurface isotopic composition, all soil samples were homogenised and digested in an 1:1 HNO_3/HCl solution to determine total extractable Pb concentrations. Bioaccessibility testing was performed on 36 < 180 µm subsamples using the Relative Bioaccessibility Leaching Procedure (RBALP) (Drexler and Brattin, 2007). This method used 1 g of milled soil digested in a 0.1 M simulated gastric fluid solution (2 g NaCl, 3 g pepsin, HCl and deionised water at pH 1.2) for 1 hour at 37 °C on an end-over-end mixer. This method is considered to be the most similar *in vitro* simulation to the stomach phase of the gastrointestinal tract of a 2-3 years old child (Wragg and Cave, 2002). Digest solutions were filtered through a 0.45 µm cellulose filter. Aliquots were analyzed for total extractable and bioaccessible soil-Pb concentrations using Inductively Coupled Plasma Atomic Emission and Mass Spectrometry (ICP-AES - Varian Vista Pro AX and ICP-MS - Elan 6100DRC Perkin Elmer) at the National Measurements Institute (NMI), Sydney.

Lead isotope compositions were determined in 15 soil subsamples (< 2 mm grain size fraction) from the Taylor et al. (2010a) study. Soils were digested in 1:1 $HNO_3/HClO_4$ at 100 °C for 2 hours and analyzed for soil-Pb isotope composition ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$) using ICP-MS (Agilent 7500ce) at Charles Darwin University (CDU), Darwin, Australia.

Dusts

Dust wipe samples were collected from inside the houses on residential lots P1, P2, P3 and P5 (Figure 1) between April and July 2008 (Table 1). Dust wipe sampling involved marking a 30 by 30 cm area for larger surfaces, such as the tops of refrigerators and bare floor surfaces, or calculating the surface area for confined areas, such as window sills and air vents. Clean nitrile gloves were worn during the collection of each surface wipe sample, which was collected from the selected surface area using a moistened towelette (Ghost WipeTM ~ 240 cm^2) according to the surface dust wipe method E 1728-03 detailed in American Society for Testing and Materials, (2003). Sampled dust wipes were folded and stored in acid-washed 500 mL HDPE bottles. Dust wipe samples were digested in 1:1 $HNO_3/HClO_4$ for 2 hours at 100 ºC. The digest solutions were filtered through a 0.45 µm cellulose filter and an aliquot was analyzed for dust-Pb by ICP-AES at the NMI. The mass on the wipe (µg) was then transformed to $\mu g/m^2$ to standard the values and allow for comparison with the benchmark value provided by the US EPA (2011).

Bulk grab samples of dust were collected from an air conditioning unit inside the house at residential lots P1 and P2 (Table 1). Sample analyse of the dust sample for total extractable dust-Pb was undertaken in the same manner as the soil samples.

In addition, indoor bulk dust samples (DSG Table 1) were collected in acid-washed sealed plastic bags from public buildings in Mount Isa in June 2008 (Figure 1). Dust samples were digested in 10 % HNO_3 (SupraPur grade) in open teflon beakers at 100 °C for 2 hours and analyzed for dust-Pb isotope composition using ICP-MS at CDU.

Aerosols

A total of nine air filters were deployed throughout the Mount Isa urban area at CP and residential lots P3, P4 and P6 (Figure 1) to determine aerosol-Pb isotopic composition (Table 1). Air filters were deployed at each site in early May 2008. These filters were removed at the beginning of July 2008 and replaced with new filters, which were collected in August or September 2008. During this sampling period greater than 70 % of the regional winds originated from south, south-east and east directions (i.e. the prevailing wind blew across the broader urban area, including the sampling sites, towards the direction of XMIM), with no reports of wind speeds exceeding 10 km/hr (BOM, 2010).

Samples were collected using a continuous diaphragm pump, connected to a 0.45 μm polycarbonate air filter capsule (Bollhöfer et al., 1999). The pumping speed ranged between 0.07 and 0.12 m^3/hr at all sample locations. The filters were placed 2-3 m above the ground level, at locations away from main thoroughfares and with the external inlet aperture positioned in a downwards direction. After collection, samples were leached in dilute hydrobromic acid (HBr) for 24 hours at 40 ºC. Pb in the leached solutions was then extracted using HBr/HNO_3 anion exchange. Collected samples were then evaporated to dryness and analyzed for aerosol-Pb isotope compositions using a magnetic sector field, multi-collector VG354 Thermal Ionisation Mass Spectrometer (TIMS, Fisons Instruments) at Curtin University, Perth, Australia.

Rocks

Two local rock outcrops from the Mount Isa urban area were sampled in order to characterise surface bedrock-Pb isotopic composition of the urban area (Table 1; Figure 1). Rock samples were prepared and analyzed in the laboratory in the same manner as the aerosol samples.

Quality control and quality assurance

The reporting and discussion of the quality control and quality assurance protocols are presented in the supplementary data due to the broad environmental media sampled, different analytical programs and techniques (including acids) applied in this study.

Results and discussion

A summary of Pb concentrations in soil, dust and dust wipe samples from the Mount Isa urban area are shown in Table 2. The full dataset is located in the supplementary data.

Soil-Pb

The mean concentration of total extractable soil-Pb in <180 µm grain size fraction is 1 560 mg/kg, of which a mean of 68 % is bioaccessible (range 51 to 92 %). Bioaccessible soil-Pb is correlated strongly with total extractable soil-Pb ($r^2 = 0.97$, $p \leq 0.05$; Supplementary material Figure S1). The health-based investigation levels (HIL) for soil-Pb concentrations (land use category A, NEPC, 1999) of 300 mg/kg has been selected as the comparison guideline in this study as soil samples were collected predominately from areas where young children are active and/or reside. The HIL for soil-Pb is based upon the < 2 mm fraction. The < 180 µm grain size fraction is used here because it is well known that the finer contaminated soil fractions pose a greater risk to young children (Australian/New Zealand Standard, 2000). The mean concentration of soil-Pb (1 560 mg/kg) exceeds significantly the HIL of 300 mg/kg (Table 2). This result is not unexpected given Taylor et al. (2010a) identified that other similar soil samples within 2 km of the Pb stack exceeded the HIL.

The mean soil-Pb concentration from the five residential lots (Table 2) were below the HIL for the < 2 mm and < 180 µm grain size fractions. However, the mean soil Pb (373 mg/kg and 700 mg/kg) in the < 63 µm grain size and < 180 mm fractions of soil from lots P3 and CP, respectively, exceeded the HIL 'A' (Table 2). Overall, the mean soil-Pb concentrations from the residential lots and the school playground were lower than values from the surrounding urban soils located within 2 km of the Pb stack at XMIM (Table 1; Figure 1) and were also lower than in the urban soils measured in a previous study of the Mount Isa urban area (Taylor et al., 2010a).

The soil-Pb concentrations from the residential lots (P1, P2 and P3), control lots (P4 and P5) and the school playground (SP) were not different statistically (ANOVA $p = 0.490$). The bioaccessible soil-Pb in these same samples was also not different statistically (ANOVA $p = 0.610$). However, soil-Pb concentrations at the residential lots and the school playground were different statistically to the total extractable and bioaccessible Pb concentrations in soil samples for all grain size fractions from the workplace (CP) and urban soils (US) (ANOVA $p = < 0.005$).

Dust-Pb

In Australia, there are no statutory surface dust-Pb quantities defined by Federal or State legislation. The closest guidance level for Pb in household dust is the Australian/New Zealand Standard (1998), which was developed for Pb paint abatement work. This document set guideline concentrations of 1 000 µg/m^2 on floors and 5 000 µg/m^2 on window sills. The US EPA (2011) provide surface dust-Pb guideline values of 430 µg/m^2 on floors and 2 690 µg/m^2 on interior window sills and are used as a benchmark for the data collected in this study. The Queensland EPA (2008) used the value of 430 µg/m^2 in their Mount Isa lead management study.

Benchmarking against the US EPA (2001) criteria, all dust samples from residential lots P1 and P3 exceeded the surface dust-Pb threshold value for floors and window sills (Table 2). In contrast, no samples from residential lots P2 and control lot P5 exceeded either the Australian/New Zealand Standard (1998) or the US EPA (2001) threshold values for surface dust-Pb concentrations (Table 2). These lower level floor Pb-dust could be due to increased floor cleaning regimes following the notification of PbB problems in the city more broadly and in the household itself. The highest dust-Pb concentrations were observed in residential lot P3 with concentrations of 15 000 mg/kg and 15 100 and 18 300 µg/m^2 of Pb on an air conditioning unit above a child's bed, kitchen surface and window sill, respectively (Table 2).

Lead isotopic compositions of dust, aerosol, soil and rock

Aerosol, dust, rock and surface and subsurface soil-Pb isotope compositions are plotted in Figure 2a, b. Figure 2a also shows the Pb isotopic composition of the typical Mount Isa-type ore-derived Pb (^{206}Pb/^{207}Pb ~1.043 and ^{208}Pb/^{207}Pb ~ 2.318 - Gulson, 1985); the present-day average crustal composition (PDAC) (Stacey and Kramer, 1975); and representative aerosol values from three other Queensland towns (Brisbane, Cairns and Townsville; Bollhöfer and Rosman, 2000). Samples from Cairns have Pb isotope compositions that are typical of Australian atmospheric background Pb (Bollhöfer and Rosman, 2000).

The Pb isotopic composition of the Mount Isa aerosol samples form a linear trend situated close to a mixing line between the Pb isotopic composition of the Mount Isa ore-derived Pb and PDAC-Pb. The displacement of the aerosol Pb away from PDAC-Pb towards Mount Isa ore-derived Pb indicates the predominance of this as primary source. The Pb isotopic composition of soils from the 0-2 cm of vertical soil profiles coincide closely with aerosol and dust Pb isotopes (Figure 2b). In contrast, soils from the 10-20 cm profile horizons have Pb isotopic compositions much closer to PDAC-Pb than Mount Isa ore-derived Pb isotopic compositions (Figure 2a). The composition of the Mount Isa urban soil profiles is consistent with previous Pb isotope data from broader regional soil profiles outside Mount Isa (Parry, 2000). Parry (2000) showed Pb isotope compositions in surface soils (0-2 cm) approached the isotopic composition of the Mount Isa ore-derived Pb body whereas most soils from deeper horizons (10-20 cm) had Pb isotope compositions were similar to regional background values. It is worth noting that of the regional aerosol data collected by Bollhöfer and Rosman (2000) (Figure 2a), the site of Townsville is the most similar to the Mount Isa data. This is likely to reflect the presence of Pb ore loading operations and Cu metal refining operations in that city, which includes some Pb from Mount Isa.

Surface soil samples from this study reveal that Pb levels are significantly higher in public spaces, such as parks and property verges, compared to residential lots of Mount Isa. Taylor et al. (2010a) estimated that > 1 000 property lots were at risk of having a soil-Pb level greater than 300 mg/kg in the finer (< 180 µm) grain size fraction. Within that envelope of predicted risk the residential lot P3 had mean soil Pb levels of 253 mg/kg (< 2 mm) and 268 (< 180 µm), and workplace lot CP had soil Pb levels of 700 (< 2 mm) and 763 (< 180 µm) (Supplementary material, Table S2). Outside of Taylor et al.'s (2010a) 300 mg/kg soil Pb contour, the remaining sample lots examined in this study returned average soil-Pb concentrations below the HIL value of 300 mg/kg (NEPC, 1999) (Supplementary material, Table S2). The rotovation of garden surface soils with cleaner lower horizons plus the addition of clean soil and organic matter is likely to have contributed to reduced soil Pb levels, compared to the adjacent non-residential areas. Notwithstanding the small sample size of lots examined in this study, the data suggests that it is difficult to ascribe domestic garden soil-Pb reservoirs as the primary cause of PbB levels of the residing child/children, although individual high soil Pb values at the sample lots suggest the risk from soil is not negligible.

In contrast, dust wipe metal results suggest that there is an association between environmental Pb concentrations in property dusts and the PbB level of the residing child. For example, residential lots P1 and P3, where the residing children had PbB levels of 17 and 27 µg/dL, respectively, had floor and window sill dust-Pb levels significantly in excess of the US EPA (2001) guideline value of 430 and 2,690 $\mu g/m^2$, respectively. At the control residential lots P4 and P5, where children had PbB concentrations of 6.5 and 2.1 µg/dL respectively, floor and window sill dust were well below the US EPA (2001) threshold value. Floor dust Pb wipe samples from residential lot P2 (child PbB level of 31.5 µg/dL) do not exceed either the Australian (Australian/New Zealand Standard, 1998) or US EPA (US EPA, 2001) threshold value for Pb in household dusts (Table 1). Increased cleaning of the house floors following the notification of the elevated PbB level resulted was reported by the resident and is likely to have resulted in the low floor dust-Pb values. However, less frequently cleaned areas had markedly higher dust Pb values: the lounge room window sill was 952 $\mu g/m^2$ and the child's bedroom window sill was 2 763 $\mu g/m^2$. Further, the air conditioning unit in the main living area, which was not cleaned prior to sampling returned a dust Pb grab value of 3 977 mg/kg. Although the air conditioning unit measurement is not directly comparable to the standard locations for dust-Pb measures, it provides a reasonable insight of the undisturbed long-term loadings to the house.

Queensland Environmental Protection Agency (Queensland EPA, 2007) reported that 35 % of dust wipes collected during the study exceeded the US EPA guideline dust floor value of 430 $\mu g/m^2$ (US EPA, 2001). These findings are significant given the Pb-dust:PbB dose-response relationships identified by Dixon et al. (2009), who showed that floor dust Pb loadings even as low ~100 $\mu g/m^2$ were not adequate.

The annual average outdoor Pb deposition rate during 2003-07 at 10 of the 11 dust monitoring stations (Queensland EPA 2008) exceeded the German TA Luft (FMENCNS, 2002) air quality standard (Pb < 100 $\mu g/m^2$/day) and at 8 out of 11 monitoring stations the less stringent World Health Organisation (WHO, 2000) air quality standard was also exceeded (Pb < 250 $\mu g/m^2$/day).

Lead-in-dust deposition was observed to decrease with distance from the XMIM, with average deposition during 2003-07 of 2 420 $\mu g/m^2$/day at 200 m distance, 790 $\mu g/m^2$/day at 800 m distance and 135 $\mu g/m^2$/day at 3 km distance (Queensland EPA, 2008). Examining background concentrations provides further context for the effect of the mining and smelting activities on air quality. The Queensland EPA (2008) review estimated that international Pb-deposition for areas without industry or heavy traffic to be in the range 1-40 $\mu g/m^2$/day, which is consistent with background Pb dust deposition rates of 2-20 $\mu g/m^2$/day measured over a 3 year period 600 km NW of Mount Isa (Munksgaard and Parry, 1998). Thus it is clear that the Pb-in-dust deposition rates and Pb concentrations in dust and soil in Mount Isa greatly exceed background levels, demonstrating the impact of industrial emissions on the local environment. The Queensland EPA (2008) concluded that XMIM did not exceed their licence arrangements for Pb in air (see *Mount Isa Mines Limited Agreement Act 1985*, (Queensland) and the *Mount Isa Mines Limited Agreement Amendment Act 1997* (Queensland)), measured as the quarterly running average concentration of Pb < 1.5 $\mu g/m^3$ (as PM_{10}). However, the Queensland EPA (2008) did conclude that air-Pb concentrations in one or more of the years 2005-07 at 4 out of 6 monitoring stations in Mount Isa exceeded the national Australian air quality <0.5 $\mu g/m^3$ goal for Pb (measured as the annual average total suspended particulate Pb concentration; NEPC, 1998).

The source, concentration and pervasiveness of Pb rich dust inside homes suggests that airborne Pb is a significant, if not dominant, pathway for exposures in the Mount Isa community. Particle size influences the dispersal and uptake of Pb dust, and finer sizes are typically more bioavailable (Csavina et al., 2011). The rate and extent of uptake of Pb may be influenced by a range of factors including age, fasting, diet composition (e.g. calcium, fat, iron) and the particle size, mineralogy and solubility of the Pb species absorbed. In terms of source emissions, the only published comprehensive study of Mount Isa smelter emission particle size and speciation showed that very few of the aerosol particulates were > 1 µm (Roberts and Williams, 1979). These Pb exposure risks are a particular concern given that soil and dust Pb derived from the XMIM smelters are partly comprised of Pb sulphate and Pb oxide (Fellows et al. 1980). Elemental Pb, Pb oxide and Pb sulphate are more soluble, and therefore are more likely to be bioavailable, than Pb sulphide (Stopford and Turner, 2000). Data from the present study shows that not only are the finer soil and dust more elevated with Pb than equivalent coarser fractions, but up to 92 % of Pb in the soil is bioaccessible (Table 1).

Xstrata Pty Ltd and Queensland Government agencies have disputed the industrial source of environmental Pb and have argued persistently that the elevated PbB of local children and Pb in the Mount Isa urban area are sourced predominantly from natural surface exposures of ore bodies. This line of argument has been maintained despite the fact that mining operations at Mount Isa have already been connected strongly to environmental contamination (Parry, 2000; Taylor and Hudson-Edwards, 2008; Munksgaard et al., 2010; Taylor et al., 2010a,b; Mackay et al., 2011, Mackay and Taylor, 2013). Further, the surface expression of the main ore body on the lease is no longer present because it has been mined-out and the existing surface is contaminated from ore and concentrate stockpiles and smelting activity (Conaghan et al. 2003).

The evidence for the claim that the source of Pb in the urban environment is derived from natural *in situ* surface minerals is considered implausible for several reasons. Firstly, there is no substantial Pb source from natural surface exposures of minerals in or near Mount Isa’s urban area. The leached and hardened ridges of the Pb-bearing surface rocks, so called ‘gossans’, are now almost entirely covered by current XMIM mining operations, which extract ore from much deeper levels (Conaghan et al., 2003). Therefore, these gossans cannot contribute significantly to contemporary environmental Pb sources. Secondly, there was never any natural surface exposure of the Cu ore body (Carter 1953; Forrestal 1990; Perkins 1990), which was discovered ‘accidentally’ during deep drilling. The Urquhart Shale Formation that hosts the major Pb-Zn-Ag and Cu ore bodies occur to the west of the upper Leichhardt River and do not crop out in the Mount Isa urban area (Carter et al., 1961). Numerous soil profiles in and around Mount Isa show that it is only near-surface layers that are contaminated with a range of metals (Parry 2000; Taylor et al., 2010a). Surface soil (0-2 cm) Pb and Cu concentrations also correlate significantly and are up to 20 times higher at 0-2 cm depth than at 10-20 cm depth (Taylor et al., 2010a). This demonstrates: (1) soil contamination with both Pb and Cu can only have come from particles emitted concurrently into the atmosphere from the smelters because there is no other common unified natural source for both metals; (2) surface soil metal enrichment can only have come from the aerial deposition of contaminated particles, as this study confirms.

The source of Pb in Mount Isa aerosols and surface soils is established by this study of the Pb isotopic composition of aerosols, dusts and surface soils from the urban area. The samples analyzed here (Table 1) had Pb isotope compositions that approximate closely to the Mount Isa ore-derived Pb. Given that subsurface soils (10-20 cm) had Pb isotope compositions distinct from that of the ore body (Figure 2b), surface soil Pb enrichment from the underlying geological strata (including the Pb-Zn ore bodies) is ruled out as a significant contributing source of Pb. Analyses of surface rock samples collected from the urban environment of Mount Isa which is predominately Native Bee Siltstone (Carter et al., 1961) also indicate that these are not likely to the source of the environmental Pb either. Samples R1 and R2 returned ^{206}Pb/^{207}Pb and ^{208}Pb/^{207}Pb compositions of 1.147, 2.426 and 1.107, 2.346, respectively (Figure 2a).

Conclusion

This study of Pb isotopic composition of aerosols, dusts and soils demonstrates conclusively that atmospheric deposition of mine and smelter-derived dusts are the source and cause of environmental contamination and form a significant pathway for exposures in the Mount Isa community. The most appropriate long-term solution to the contaminant problem at Mount Isa is to reduce considerably Pb emissions from the Pb and Cu smelters and the surrounding plant site by the immediate introduction of more effective emissions control technology along with capping of waste, ore piles and unsealed roads. These works need to be undertaken in tandem with the identification and remediation of known contaminant reservoirs held in the urban environment including soils and household dusts (e.g. floors, ceilings, furniture). Such an approach is likely to produce positive outcomes for the community of Mount Isa.

Acknowledgements

The authors thank Macquarie University and Mount Isa Water for financial and logistical support; and Slater and Gordon (Brisbane) for providing financial support for the collection and analysis of selected soil and dust samples. Rhys Hart is thanked for his assistance in the field.

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Figures captions and tables

Table 1. Summary of environmental samples and associated analyses.

Sample Media	N	Date of sampling	Location description^	Summary of analyses
Urban soils (US)*	20	September 2005 & February 2006	Public use lands.	Total extractable and bioaccessible soil-Pb in the < 180 µm grainsize fraction.
Residential lot soils	24	Between April & July 2008	Residential lots (P1 to P5)*; 1 workplace (CP)* and 1 school playground (SP)*.	Total extractable soil-Pb in the < 2 mm, < 180 µm and < 63 µm grainsize fraction; and bioaccessible soil-Pb in the < 180 µm subsamples.
Soils 0-2 and 10-20 cm (MIS)*	15	July 2008	Duplicate soil pit samples from Taylor et al. (2010a).	Pb isotopes.
Dust grab sample	7	June 2008	Air conditioning units (P1 & P2)*; Public buildings in the urban area (DSG)*.	Total extractable dust-Pb for air conditioning unit samples and Pb isotopes for public building samples.
Dust wipes	16	Between April & July 2008	Residential lots (P1, P2, P3 & P5)*.	Total extractable dust wipe-Pb expressed as a loading ($\mu g/m^2$).
Aerosols	9	Between May & July 2008	Residential lots (P3, P4, P6 & CP)*.	Pb isotopes.
Rock (R)*	2	September 2008	Public use lands.	Pb isotopes.

*Code used on Figure 1 is denoted in brackets; ^Locations marked on Figure 1

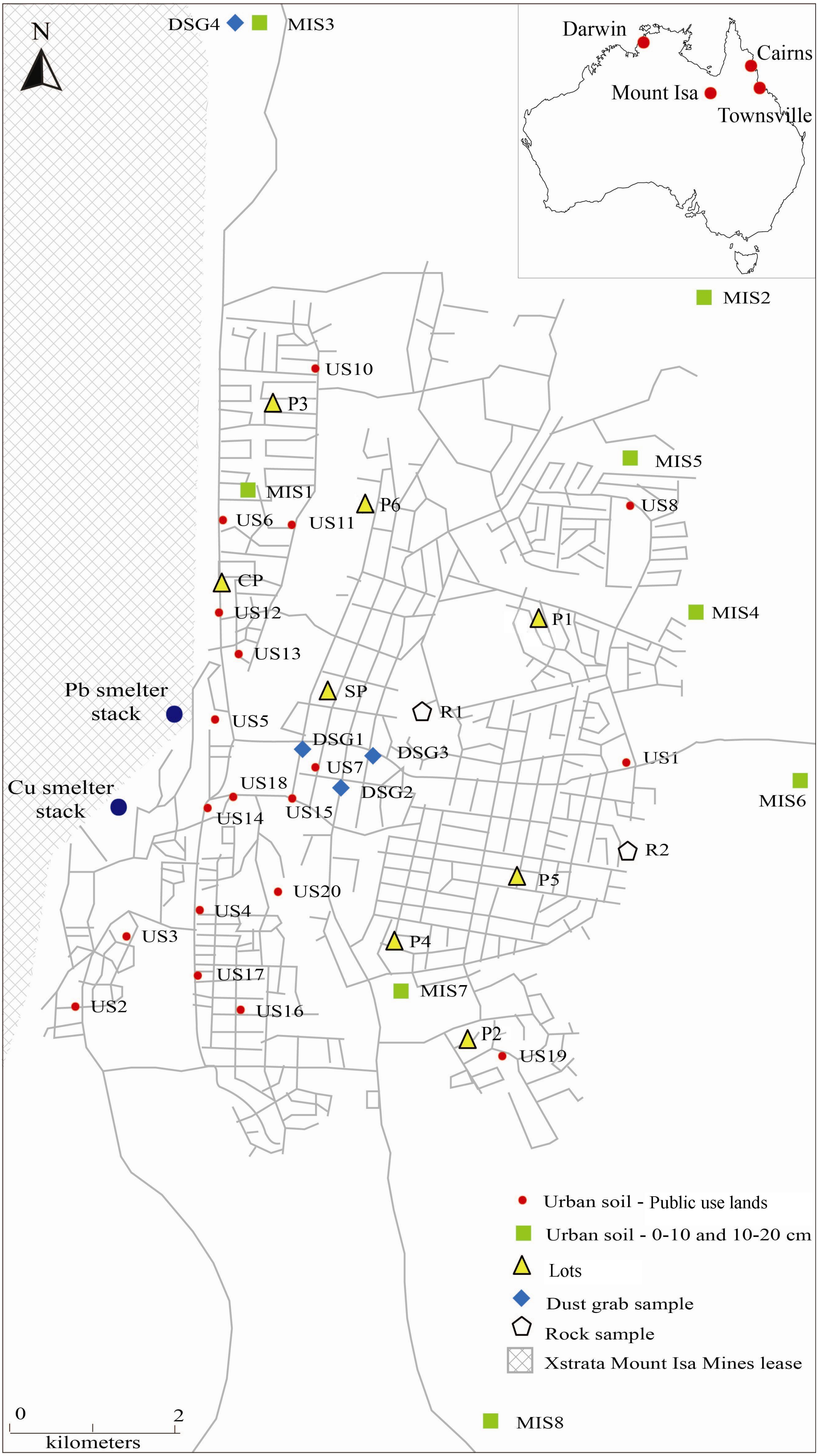
Table 2. Summary of soil- and dust-Pb results.

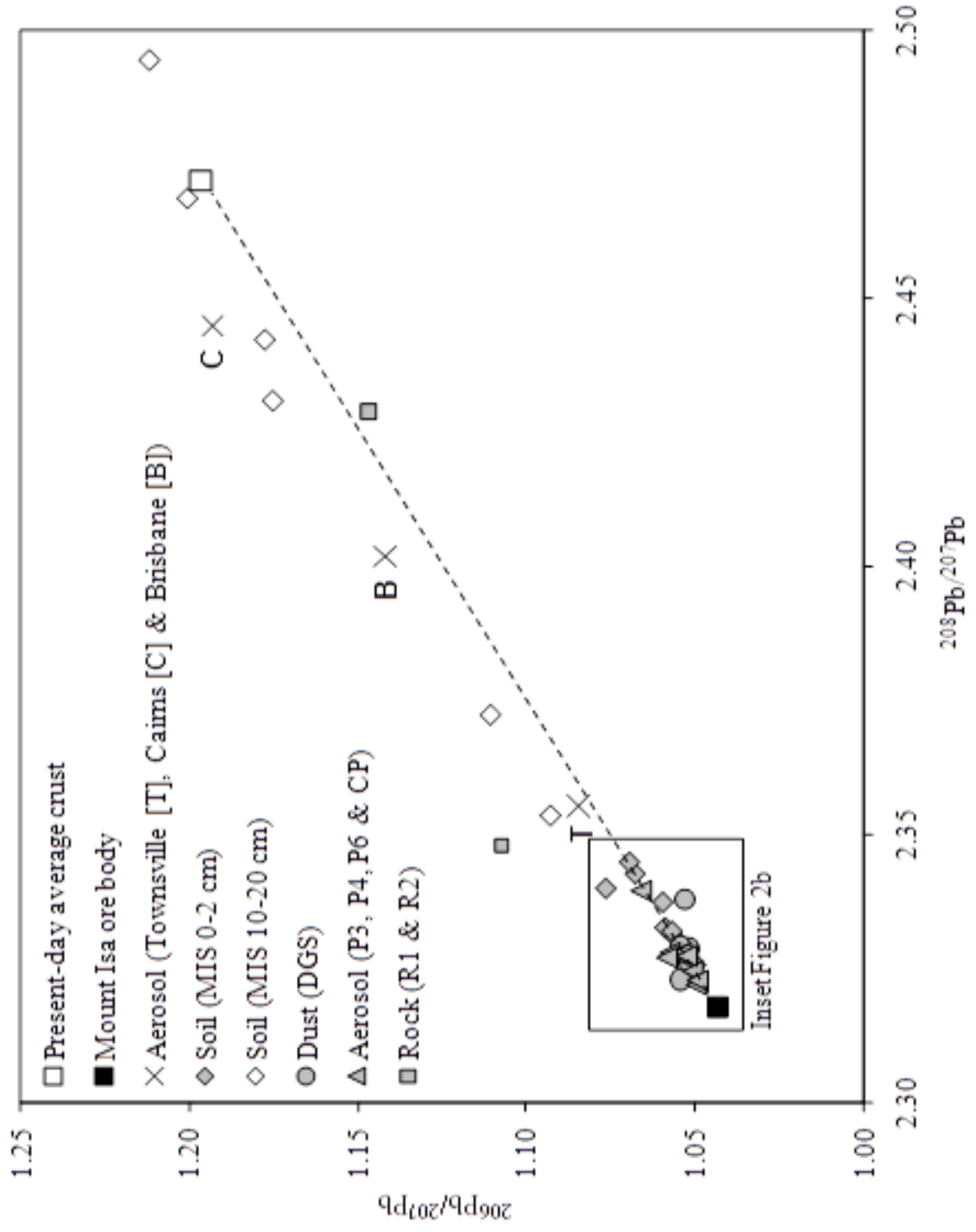
	Mean soil-Pb (mg/kg)				Mean dust-Pb (µg/m^2)			
	< 2 mm	< 180 µm	< 180 µm bio%^	< 63 µm	Floor	Window sill	Kitchen Surface	Air conditioning unit
US	n.d.	1 560 (n=19)	68%	n.d.	NA	NA	NA	NA
P1	160 (n=6)	220 (n=5)	65%	300 (n=5)	940 (n=2)	4 010 (n=2)	1 330 (n=1)	980* (n=1)
P2	90 (n=6)	130 (n=5)	65%	200 (n=5)	80 (n=2)	1 860 (n=2)	n.d.	3 980* (n=1)
P3	250 (n=7)	270 (n=6)	67%	370 (n=6)	830 (n=1)	18 300 (n=1)	15 100 (n=1)	15 000 (n=1)
P4	150 (n=1)	250 (n=2)	88%	n.d.	n.d.	n.d.	n.d.	n.d.
P5	77 (n=1)	77 (n=1)	55%	n.d.	20 (n=2)	180 (n=1)	n.d.	n.d.
CP	700 (n=1)	763 (n=1)	92%	n.d.	n.d.	n.d.	n.d.	n.d.
SP	n.d.	300 (n=2)	63%	n.d.	NA	NA	NA	NA

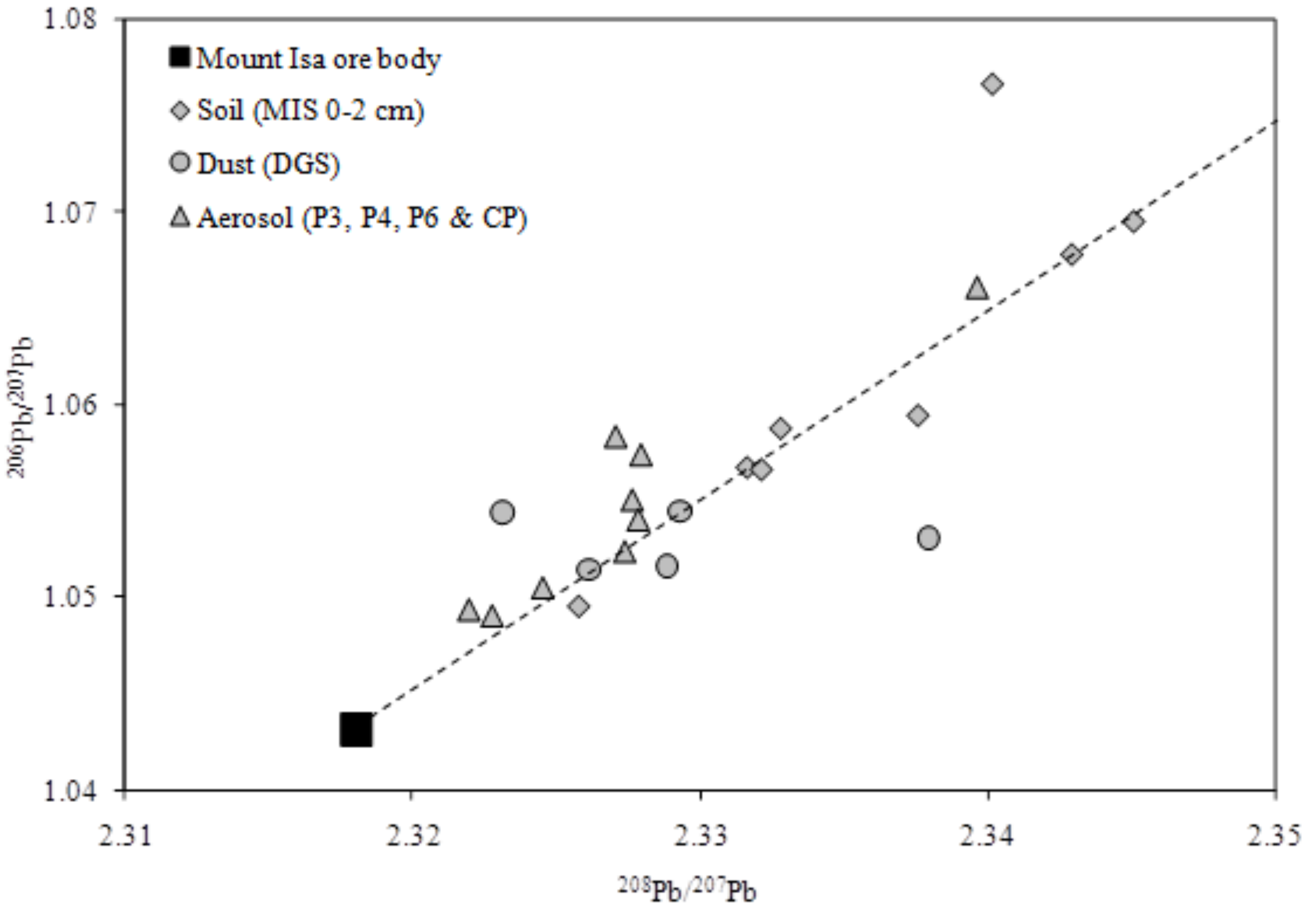
NA – Not applicable; n.d. – not determined; *Sample collected as a grab sample and result in mg/kg; ^percentage of sample that is bioaccessible; n = number of samples

Figure 1. Location map showing the sample sites for different environmental material collected and analyzed from Mount Isa. Dust wipes collected as background indicators from the cities of Cairns, Darwin and Townsville are also shown in the inset along with Mount Isa.

Figure 2a. Pb isotopic composition of the present-day average crust (Stacey and Kramer, 1975); Mount Isa Pb-Zn ore body (Gulson, 1985); aerosols from Townsville, Cairns and Brisbane (Bollhöfer and Rosman, 2000); surface soil (MIS 0-2 cm); subsurface soil (MIS 10-20 cm); aerosol (P3, P4, P6 & CP); dust (DGS) and rock (R1 & R2). The dashed line indicates linear mixing between Mount Isa Pb-Zn ore body and the present-day average crust Pb. **Figure 2b.** Inset from Figure 2a.


N
DSG4
MIS3
Darwin
Cairns
Mount Isa
Townsville
MIS2
US10
P3
MIS5
MIS1
P6
US8
US6
US11
CP
US12
P1
MIS4
US13
Pb smelter stack
SP
R1
US5
DSG1
DSG3
US1
US7
Cu smelter stack
US18
DSG2
MIS6
US14
US15
R2
P5
US20
US4
US3
P4
US17
MIS7
US2
US16
P2
US19
● Urban soil - Public use lands
🟩 Urban soil - 0-10 and 10-20 cm
▲ Lots
◆ Dust grab sample
⬠ Rock sample
▨ Xstrata Mount Isa Mines lease
MIS8
0
2
kilometers


□ Present-day average crust
■ Mount Isa ore body
× Aerosol (Townsville [T], Cairns [C] & Brisbane [B])
◆ Soil (MIS 0-2 cm)
◇ Soil (MIS 10-20 cm)
● Dust (DGS)
▲ Aerosol (P3, P4, P6 & CP)
■ Rock (R1 & R2)
C
B
T
Inset Figure 2b
^{206}Pb/^{207}Pb
1.25
1.20
1.15
1.10
1.05
1.00
^{208}Pb/^{207}Pb
2.30
2.35
2.40
2.45
2.50


Mount Isa ore body
Soil (MIS 0-2 cm)
Dust (DGS)
Aerosol (P3, P4, P6 & CP)
1.08
1.07
1.06
1.05
1.04
^{206}Pb/^{207}Pb
2.31
2.32
2.33
2.34
2.35
^{208}Pb/^{207}Pb

Identification of environmental lead sources and pathways in a mining and smelting town: Mount Isa, Australia
Alana K. Mackay, Mark P. Taylor, Niels C. Munksgaard, Karen A. Hudson-Edwards and Laurie Burn-Nunes
Number of pages = 9
Number of figures = 1
Number of tables = 5

Quality control and quality assurance

The reporting and discussion of the quality control and quality assurance protocols for this study are presented in the supplementary data due to the broad range of environmental media sampled and the different analytical techniques (including acids) applied.

Blanks, replicates and standard certified reference materials were used throughout field and laboratory work for quality control and data assurance purposes. The recovery of certified reference material AGAL-10 (river sediment) was between 99 and 107 % of the listed total extractable soil-Pb value (n = 4). All duplicate and triplicate sample values were within 10 % relative percentage difference. All laboratory and field equipment blanks returned concentrations below instrument limit of detection. The simulated gastric fluid solution used for the RBLAP simulations reported below instrument limit of detection for Pb but revealed trace amounts of Zn with an average of 70 µg/L (n = 7). Analyses of three blank dust wipes revealed minor traces of Pb (0.1 µg per wipe ~240 cm^2). Quality control of Pb isotope ratio analyses of soils were carried out by analyses of the certified reference material NIST-982 Pb as well as 3 in-house (CDU) soil and ore samples for which long-term isotope ratio data exists. The NIST-982 standard was digested in the same manner as the soils, using a 1:1 $HNO_3/HClO_4$ at 100 °C for 2 hours. The average deviation from certified or long-term values for the 4 CDU reference materials was ± 0.25 % for $^{208}Pb/^{206}Pb$ and ± 0.29% for $^{207}Pb/^{206}Pb$. The average internal precision of sample analyses was ± 0.32 % (2. relative standard deviation) for both isotope ratios. Quality control of Pb isotope ratio analyses of the rock and aerosol filter samples was carried out by analysis of blanks and the certified reference material NIST-981 at Curtin University. Analyses of Pb in the filter blanks was less than 0.6 % of the Pb in the prepared aerosol filter sample solutions. The linear mass bias introduced into the isotopic ratio TIMS measurement was assessed from the repeated analysis of the NIST-981 reference material and was 0.05 ± 0.02 and 0.299 ± 0.001 % per atomic mass unit for Faraday cup and Daly detector measurement respectively.

Table S1. Concentrations of total and bioaccessible Pb in urban soils and properties throughout the Mount Isa area. Locations are shown in Figure 1.

Sample ID	Lab ID	Total Pb (mg/kg)	Bioaccessible Pb (mg/kg)	Bioaccessible Pb (%)
US1	UR_1	68	46	68
US2	UR_5	1,130	925	82
US3	UR_6	320	201	63
US4	UR_7	1,440	863	60
US5	UR_8	2,140	1,096	51
US6	UR_9	700	503	72
US7	UR_14	9,780	8,115	83
US8	UR_23	110	66	60
US9	UR_28	110	n.d.	n.d.
US10	MI_9	1,730	785	45
US11	MI_15	240	141	59
US12	MI_16	1,180	682	58
US13	MI_17	1,120	803	72
US14	MI_18	2,270	1,787	79
US15	MI_19	2,480	1,407	57
US16	MI_23	500	361	72
US17	MI_24	1,520	1,266	83
US18	MI_26	1,930	1,647	85
US19	MI_27	150	116	78
US20	SED_7	2,310	n.d.	n.d.
P1	BS_2	120	86	72
P1	BS_3	310	194	63
P1	BS_4	31	20	65
P1	BS_5	140	80	57
P1	BS_6	480	360	75
P2	TR_1	99	60	61
P2	TR_3	140	84	60
P2	TR_4	160	98	62
P2	TR_6	200	150	75
P3	MS_2	470	301	64
P3	MS_3	310	261	84
P3	MS_4	150	96	64
P3	MS_5	340	241	71
P3	MS_6	150	92	62
P3	MS_10	190	108	57
CP	CP_1	763	700	92
P4	SS_2	180	158	88
P5	CS_1	77	42	55
SP	AM3A	330	185	56
SP	AM3B	270	191	71

n.d. not determined

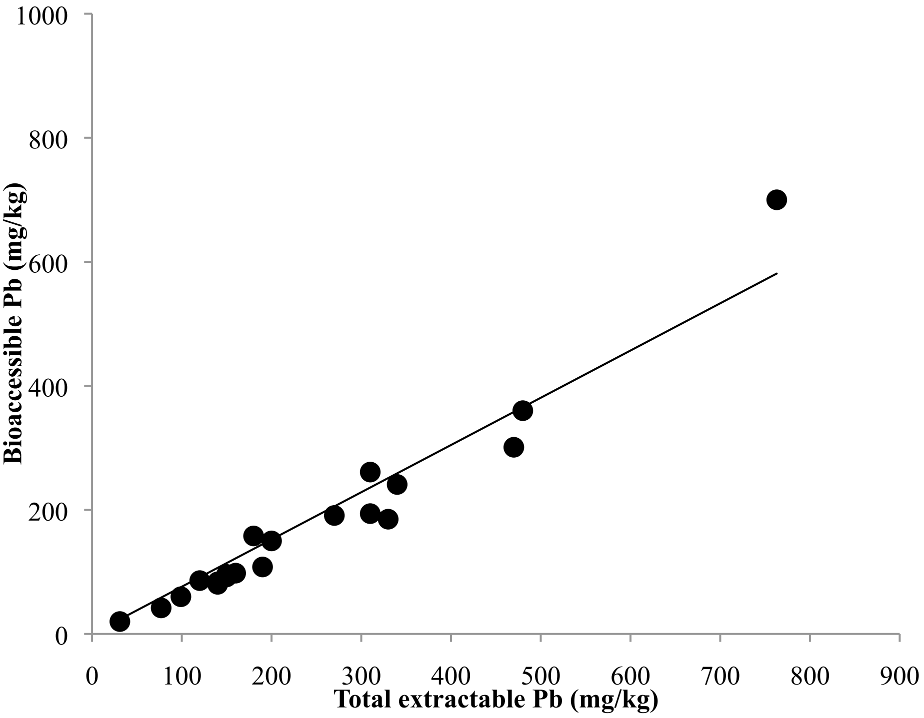


Figure S1. Plot of total extractable Pb (x axis) and bioaccessible Pb (y axis) in urban soils and properties throughout the Mount Isa area ($r^2 = 0.97$, $p \leq 0.05$).

Table S2. Pb concentrations in soil and dust samples from properties throughout the Mount Isa urban area. Sample locations are shown in Figure 1.

Property soil samples					Property dust samples			
Sample ID	Date collected	Pb <2 mm (mg/kg)	Pb <180 µm (mg/kg)	Pb <63 µm (mg/kg)	Sample ID	Date collected	Sample description	Dust-Pb ($\mu g/m^2$) unless noted otherwise
Property 1 (P1)								
AMBS1-B	18/04/08	63	n.d.	n.d.	D1	17/04/08	Floor- lounge room	666
AMBS2-B	18/04/08	75	120	190	D2	17/04/08	Kitchen	1 333
AMBS3-B	18/04/08	250	310	420	D3	17/04/08	Floor- child's bedroom	1 222
AMBS4-B	18/04/08	28	31	69	D4	17/04/08	Window sill- child's bedroom	3 057
AMBS5-B	18/04/08	120	140	180	D5	17/04/08	Window sill- kitchen	4 967
AMBS6-B	18/04/08	400	480	620	D6	17/04/08	Air conditioning unit	980 mg/kg
Property 2 (P2)								
TR1	8/05/08	83	99	160	TRDS_1	2/07/08	Window sill- lounge room	952
TR2	8/05/08	42	54	63	TRDS_2	2/07/08	Floor- lounge room	70
TR3	8/05/08	50	140	230	TRDS_3	2/07/08	Floor- child's bedroom	94
TR4	8/05/08	140	160	250	TRDS_4	2/07/08	Window sill- child's bedroom	2 763
TR6	8/05/08	170	200	320	TRDS_5	2/07/08	Air conditioning unit	3977 mg/kg
TR7	8/05/08	60	n.d.	n.d.				
Property 3 (P3)								
MS_2	8/05/08	340	470	630	MSDS_1	8/05/08	Window sill- lounge room	18 260
MS_3	8/05/08	270	310	380	MSDS_2	8/05/08	Floor- lounge room	833
MS_4	8/05/08	120	150	210	MSDS_3	8/05/08	Air conditioning unit	14 994
MS_5	8/05/08	260	340	480	MSDS_4	8/05/08	Kitchen	15 120

MS_6	8/05/08	130	150	240				
MS_8	8/05/08	230	n.d.	n.d.				
MS_10	8/05/08	420	190	300				
Property 4 (P4)								
SS_2	8/05/08	150	350	n.d.				
SS_2_duplicate		n.d.	180	n.d.				
Property 5 (P5)								
CS_Soil_1	5/07/08	77	n.d.	n.d.	CSDS_1	5/07/08	Kitchen	8
CS_Soil_1_duplicate		n.d.	77	n.d.	CSDS_2	5/07/08	Floor- lounge room	32.2
					CSDS_3	5/07/08	Window sill- child's bedroom	181.8
School Playground (SP)								
AM1	18/04/08	630	n.d.	n.d.				
AM2	18/04/08	140	n.d.	n.d.				
Workplace (CP)								
CP_1	8/05/08	700	n.d.	n.d.				
CP_1_duplicate		n.d.	763	n.d.				

n.d. not determined

Table S3. Pb isotope composition of air filter (aerosols) samples from the Mount Isa urban area, measured at the John de Laeter Centre for Isotope Research, Curtin University, Perth, using a Thermo Finnegan Triton Thermal Ionization Mass Spectrometry (TIMS) instrument.

	Property 6	Property 3	Property 4	CP
Filter Period 1				
Date of deployment	22/06/08	27/05/08	9/05/08	9/05/08
Date of collection	5/07/08	2/07/08	2/07/08	4/07/08
Pumping rate (m^3/hr)	0.08	0.12	0.067	0.076
^{206}Pb/^{207}Pb	1.055	1.049	1.058	1.051
^{208}Pb/^{207}Pb	2.325	2.320	2.324	2.322
Filter period 2				
Date of deployment	5/07/08	2/07/08	2/07/08	4/07/08
Date of collection	3/08/08	25/08/08	6/09/08	7/09/08
Pumping rate (m^3/hr)	0.08	0.12	0.067	0.076
^{206}Pb/^{207}Pb	1.049	1.052	1.057	1.054
^{208}Pb/^{207}Pb	2.319	2.325	2.325	2.325
Filter period 3				
Date of deployment	3/08/08	n.d.	n.d.	n.d.
Date of collection	6/09/08	n.d.	n.d.	n.d.
Pumping rate (m^3/hr)	0.08	n.d.	n.d.	n.d.
^{206}Pb/^{207}Pb	1.066	n.d.	n.d.	n.d.
^{208}Pb/^{207}Pb	2.337	n.d.	n.d.	n.d.

n.d. not determined

Table S4. Pb isotope composition of surface (0-2 cm) and subsurface (10-20 cm) soil samples from the Mount Isa urban area, locations shown in Figure 1. Total metal concentrations for the same sites are provided in Taylor et al. (2010), listed as Pit Sample Sites 1-8. Soils were measured using an Agilent 7500ce ICP-MS at the Environmental Analytical Chemistry Unit, Charles Darwin University, Northern Territory. Rock samples were measured at the John de Laeter Centre for Isotope Research, Curtin University, Perth, using a Thermo Finnegan Triton Thermal Ionization Mass Spectrometry (TIMS) instrument.

Sample ID	Lab ID	Depth	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{207}Pb$
MIS1	SOCC_01	0 - 2	1.050	2.326
		10 - 20	n.d.	n.d.
MIS2	SOCC_02	0 - 2	1.068	2.343
		10 - 20	1.177	2.443
MIS3	SOCC_03	0 - 2	1.077	2.340
		10 - 20	1.093	2.354
MIS4	SOCC_04	0 - 2	1.057	2.332
		10 - 20	1.200	2.469
MIS5	SOCC_05	0 - 2	1.059	2.333
		10 - 20	1.111	2.373
MIS6	SOCC_06	0 - 2	1.069	2.345
		10 - 20	1.279	2.545
MIS7	SOCC_07	0 - 2	1.057	2.332
		10 - 20	1.175	2.431
MIS8	SOCC_08	0 - 2	1.059	2.338
		10 - 20	1.212	2.495
R1	R1	surface	1.147	2.426
R2	R2	surface	1.107	2.346

n.d. not determined

Table S5. Pb concentrations and Pb isotope ratios from indoor settled dust samples from Mount Isa, Darwin and Cairns June 2008, measured an Agilent 7500ce ICP-MS at the Environmental Analytical Chemistry Unit, Charles Darwin University, Northern Territory.

Site	Distance from MIM	Pb (mg/kg)	^{206}Pb/^{207}Pb	^{208}Pb/^{207}Pb
Library (DSG1)	1.1km East	572	1.053	2.338
Motel (DSG2)	1.3km East	1220	1.054	2.323
Motel (DSG2)	1.3km East	757	1.052	2.329
Shopping Centre (DSG3)	1.6km East	485	1.054	2.329
Airport (DSG4)	6.8km North-North-East	1 360	1.051	2.326