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

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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. Urban soil samples contain elevated Pb concentrations (mean 1560 mg/kg), of which 45–85% of the Pb is bioaccessible. The Pb isotopic composition of surface soils (0–2 cm), aerosols and dusts (206Pb/207Pb, 208Pb/207Pb 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 (206Pb/207Pb, 208Pb/207Pb 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 in situ weathered bedrock. Remediation strategies should target legacy and ongoing sources of environmental Pb to mitigate the problem of Pb exposure at Mount Isa.

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1. 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; Bellinger and Needleman, 2003; Canfield et al., 2003; Braun et al., 2006; Bellinger, 2008; Jusko et al., 2008; National Toxicology Program, 2012; Taylor et al., 2012a,b). 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., 2010; Taylor and Schniering, 2010), Port Kembla (Young et al., 2005), Port Pirie (Baghurst et al., 1992, Taylor, 2012; Taylor et al., 2013), 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 (Fig. 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, 2010). 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., 2010) and dust (Queensland EPA, 2008; Taylor et al., 2011), coupled to Mount Isa’s 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
Fig. 1. Location map showing the sample sites for different environmental samples collected and analysed from Mount Isa. Aerosol samples collected as background indicators from the cities of Cairns, Brisbane and Townsville are also shown in the inset along with Mount Isa.
mining towns in Australia, and elsewhere in the world, where mining and industrial activities pose a potential serious risk of harm.

1.1. Study area

Mount Isa is located in north-west Queensland, Australia (Fig. 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, 2013) 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. Pollution 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 smelting and smelting operations (EPA, 2008). Despite upgrades to emission capture technology and ore management technologies 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., 2010, 2011; NPI, 2013).

2. Materials and methods

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

2.1. 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 (Fig. 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) (Fig. 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 ground (SP) (Fig. 1). The selection of the residential lots (P1 to P5); a workplace lot (CP) and; the entrance to a school playground (SP) (Fig. 1) to determine aerosol-Pb isotopic composition.

Total soil samples were collected from the houses on residential lots P1, P2, P3 and P5 (Fig. 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 Wipe™ ~ 240 cm²) 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 HNO₃/HClO₄ at 100 °C for 2 h and analysed for soil-Pb isotope composition using ICP-MS at CDU, Darwin, Australia.

2.2. Aerosols

A total of nine air filters were deployed throughout the Mount Isa urban area at CP and residential lots P3, P4 and P6 (Fig. 1) to determine aerosol-Pb isotopic composition.
The mean concentration of total extractable urban soil-Pb in <180 μm grain size fraction is 1560 mg/kg, of which a mean of 68% is bioaccessible (Table 2; range 45–85%; Supplementary material). Bioaccessible soil-Pb is correlated strongly with total extractable soil-Pb ($R^2 = 0.97$, $p < 0.05$; Supplementary material Fig. 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. The Queensland EPA (2008) also used the value of 430 μg/m² on floors and 2690 μg/m² on interior window sills and are used as a benchmark for the data collected in this study. The Queensland EPA (2008) also used the value of 430 μg/m² in their Mount Isa lead management study.

Benchmarking against the US EPA (2011) 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 (2011) 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/m² of Pb on an air conditioning unit above a child’s bed, kitchen surface and window sill, respectively (Table 2).

### Table 2

<table>
<thead>
<tr>
<th>Mean soil-Pb (mg/kg)</th>
<th>Mean dust-Pb (μg/m²)</th>
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<tr>
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<td>Floor</td>
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<tr>
<td>c&lt;2 mm</td>
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<tr>
<td>&lt;180 μm</td>
<td>1560 (n = 19)</td>
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<tr>
<td>&lt;180 μm bioa</td>
<td>220 (n = 5)</td>
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<tr>
<td>&lt;63 μm</td>
<td>130 (n = 5)</td>
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<td></td>
<td>270 (n = 6)</td>
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<td>77 (n = 1)</td>
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<td>760 (n = 1)</td>
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<td>300 (n = 2)</td>
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<td>NA</td>
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NA = not applicable; n.d. = not determined.

a Sample collected as a grab sample and result in mg/kg.

b Percentage of sample that is bioaccessible; n = number of samples.
3.3. Lead isotopic compositions of dust, aerosol, soil and rock

Aerosol, dust, rock, and surface and subsurface soil-Pb isotope compositions are plotted in Fig. 2a and b. Fig. 2a also shows the Pb isotopic composition of the typical Mount Isa-type ore-derived Pb (206Pb/207Pb ~ 1.043 and 208Pb/207Pb ~ 2.318 – Gulsön, 1985); the present-day average crustal composition (PDAC) (Stacey and Kramers, 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 the primary source. The Pb isotopic composition of soils from the 0–2 cm of vertical soil profiles match closely the aerosol and dust Pb isotopes (Fig. 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 (Fig. 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 that were similar to regional background values. It is worth noting that of the regional aerosol data collected by Bollhöfer and Rosman (2000) (Fig. 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 Cumetal 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. (2010) estimated that >1000 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 (>2 mm), 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 (2010) 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 garden 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 (2011) guideline value of 430 and 2690 μg/m², 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 (2011) threshold value. Floor dust PbB wipe samples from residential lot P2 (child PbB level of 31.5 μg/dL) did not exceed either the Australian (Australian/New Zealand Standard, 1998) or US EPA 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 μg/m² and the child’s bedroom window sill was 2763 μg/m². Further, the air conditioning unit in the main living area, which was not cleaned prior to sampling returned a dust Pb grab value of 3977 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 into the undisturbed long-term Pb dust loadings to the house.

Queensland Environmental Protection Agency (Queensland EPA, 2007) reported that 35% of dust wipes collected during their study exceeded the current US EPA guideline dust floor value of 430 μg/m² (US EPA, 2001). These findings are significant given that the Pb-dust:PbB dose–response relationships identified by Dixon et al. (2009) showed that floor dust Pb loadings as low as 100 μg/m² were not adequate to protect children from adverse blood lead levels.
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 μg/m²/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 μg/m²/day).

Lead-in-dust deposition was also observed to decrease with distance from the XMIM, with average deposition during 2003–07 of 2420 μg/m²/day at 200 m distance, 790 μg/m²/day at 800 m distance and 135 μg/m²/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 μg/m²/day, which is consistent with background Pb-dust deposition rates of 2–20 μg/m²/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 (Mount Isa Mines Limited Agreement Act 1995, (Queensland)) and the Mount Isa Mines Limited Agreement Amendment Act 1997 (Queensland), measured as the quarterly running average concentration of Pb < 1.5 μg/m³ (as PM10). 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 μg/m³ 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 (Cavina et al., 2011, 2012). 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 soils and dusts more elevated with Pb than equivalent coarser fractions, but up to 92% of Pb in the commercial lot (CP) soil is bioaccessible (Supplementary material).

Xstrata Pty Ltd and Queensland Government agencies have disputed the industrial source of environmental Pb and have argued persistently that the elevated Pb of local children and Pb in the Mount Isa urban area are sourced predominantly from natural surface exposures of orebodies. 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; Queensland EPA, 2008, Taylor and Hudson-Edwards, 2008; Munksgaard et al., 2010; Taylor et al., 2010, 2011; 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 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 orebodies 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 and 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., 2010). 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., 2010). 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; and (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 analysed 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 orebody (Fig. 2b), surface soil Pb enrichment from the underlying geological strata (including the Pb–Zn orebodies) 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 underlain predominately with Native Bee Siltstone (Carter et al., 1961) also indicate that these are not likely to be the source of the environmental Pb either. Samples R1 and R2 returned 206Pb/207Pb and 208Pb/207Pb compositions of 1.147, 2.426 and 1.107, 2.346, respectively (Fig. 2a).

4. Conclusion

This study of Pb isotopic composition of aerosols, dusts and soils demonstrates conclusively that atmospheric deposition of mine and smelter-derived Pb are the source and cause of environmental contamination and form a significant pathway for Pb 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.

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Appendix A. Supplementary material

Supplementary material related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2013.05.007.

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