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Lead isotopic compositions of ash sourced from Australian bushfires

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ABSTRACT

This study identifies natural and industrial lead remobilized in ash deposits from three bushfires in relatively pristine areas of Australia in 2011 using lead isotopic compositions ($^{208}Pb/^{207}Pb$; $^{206}Pb/^{207}Pb$). Lead concentrations in the ash ranged from 1 to 36 mg/kg, bracketing the range of lead (4–23 mg/kg) in surface soils (0–2 cm), subsurface (40–50 cm) soils and rocks. The lead isotopic compositions of ash and surface soil samples were compared to subsurface soils and local bedrock samples. The data show that many of the ash and surface soil lead isotopic compositions were a mixture of natural lead and legacy industrial lead depositions (such as leaded petrol combustion). However, some of the ash samples at each of the sites had lead isotopic compositions that did not fit a simple two end-member mixing model, indicating other, unidentified sources.

1. Introduction

Lead, which has been used extensively for industrial applications for thousands of years (Nriagu, 1983), is the most pervasive and ubiquitous elemental pollutant in the world – including Australia (Bollhöfer and Rosman, 2000, 2001). Australia has a long history of lead mining, dating back to 1841 in Glen Osmond, South Australia, and now ranks second in the world in lead mining, processing and production (Guberman, 2012). In addition to the impact of mining and smelting activities, significant amounts of lead were released in Australia from the combustion of leaded petrol. Between 1932, when the first documented sale of leaded petrol occurred in Australia, and 2002, when leaded petrol was completely phased out (Cook and Gale, 2005; NEPC, 2001), it is estimated that leaded petrol emissions contributed up to 90% of the atmospheric lead in urban areas in the country (NEPC, 2001). In two national assessments of petrol lead emissions, it was determined that 3842 tonnes of lead were emitted in 1976 in Australian capital cities (Farrington and Boyd, 1981) and 2388 tonnes of lead were emitted in 1985 (Farrington, 1988), despite mandated reductions of allowable lead in petrol (NEPC, 2001). Therefore, over the 70-year period of leaded petrol use in Australia tens, if not hundreds, of thousands of tonnes of lead was released into the ambient environment.

These industrial lead emissions have adversely impacted environmental and human health in Australia (Laidlaw and Taylor, 2011). Although industrial lead exposures have declined over the past three decades with the elimination of leaded petrol, as they have elsewhere (Annest et al., 1983), the environmental legacy from those emissions remain a source of potential exposure (Harris and Davidson, 2005; Laidlaw et al., 2012). Moreover, recent studies have determined that relatively low blood lead levels that were previously considered innocuous have been associated with adverse human health problems (ACCLPP, 2012; Lanphear et al., 2005). The most commonly reported health effects of lead exposure in children are reduced intelligence, increased behaviour problems, reduced attention, hyperactivity and cognitive impairment (NTP, 2012).

Consequently, events that remobilize historic lead depositions are an environmental and human health concern. These include fires, which pose potential health risks due to smoke and particulate emissions (Weinhold, 2011) along with their role in redistributing elements such as mercury (Finley et al., 2009) and lead (Nriagu, 1989). Given that fluctuations in atmospheric lead emissions have been linked to changes in blood lead levels (Annest et al., 1983) and that there is still no established safe level for lead (NHMRC, 2009; WHO, 2010), the likelihood that bushfires mobilise historic lead depositions (Odigie and Flegal, 2011) warrants investigation. Previous studies have shown that heavy metals can become enriched in the residues formed from the burning of biomass (Nzihou and Stanmore, 2013). Up to 11.7% of the lead stored in the original biomass may remain as bottom (*in situ*) ash,

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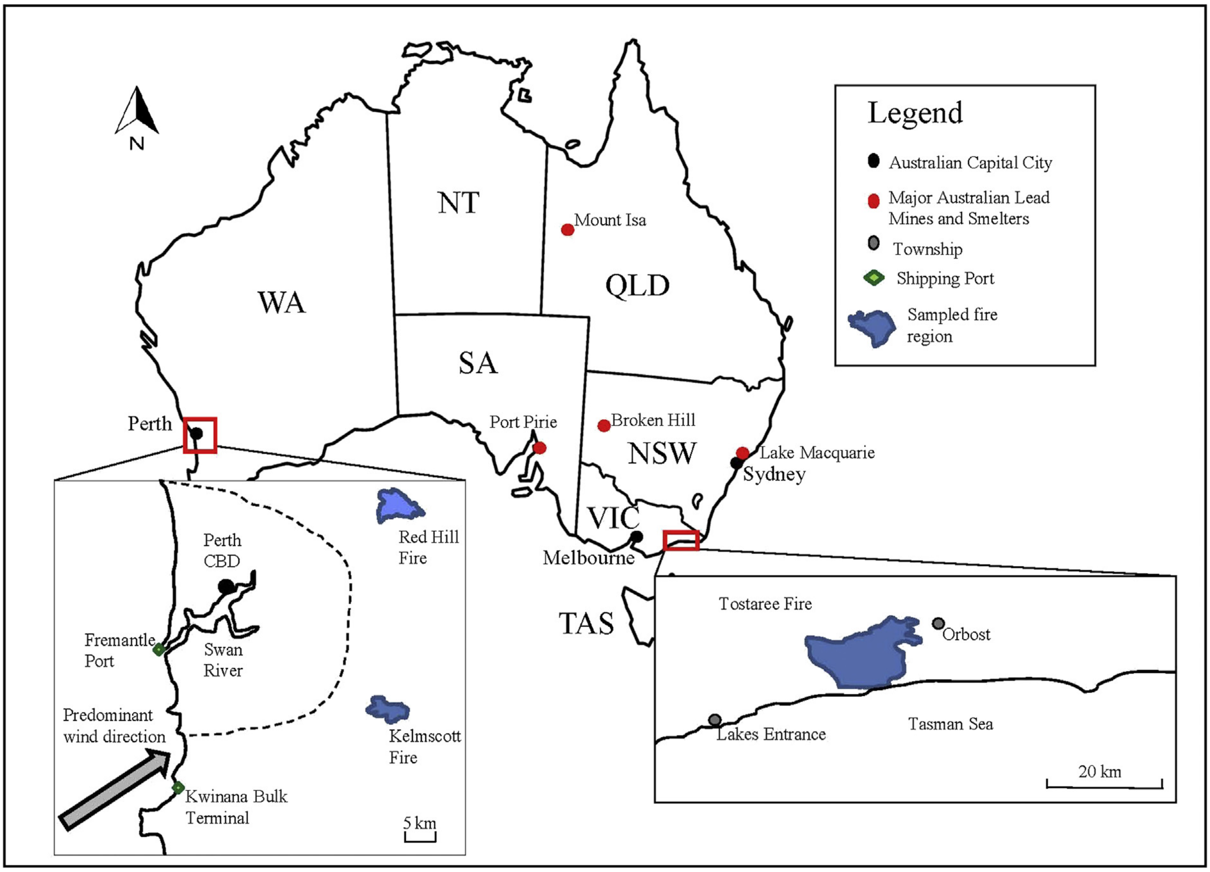

Fig. 1. The location of the three bushfire sample sites and Australia's two major lead mines of Broken Hill (New South Wales) and Mount Isa (Queensland).

with the remainder forming airborne component (fly-ashes and dust) that can be widely dispersed (Narodoslawsky and Obernberger, 1996).

The potential for the re-release of historic anthropogenic lead emissions is a particular concern in Australia because hazard reduction burning (controlled burning) of urban bushland is a standard fire risk-reduction strategy. Furthermore, the Australian continent is prone to frequent and uncontrolled wildfires, and their incidence and severity are predicted to increase due to climate change (Hennessy et al., 2006). Therefore, the primary goal of this study was to ascertain if historic depositions of industrial lead were being released back into the environment during bushfires.

Fortunately, tracing lead contamination in Australia is often rather simple because almost all of its industrial lead came from a few large, geologically old (~1700 million years) ore deposits that have relatively unique isotopic fingerprints (Gulson, 2003). The principal Australian lead ore deposits are in Broken Hill (New South Wales) and Mount Isa (Queensland), which are among the largest in the world and have distinctive lead isotopic compositions (e.g., $^{206}Pb/^{207}Pb$: 1.0390–1.0432; $^{208}Pb/^{207}Pb$: 2.3102–2.3197) (Chiaradia et al., 1997; Cooper et al., 1969; Cumming and Richards, 1975; Gulson, 1985; Gulson and Mizon, 1979; Townsend et al., 1998; Table S2, Supporting Information). Relatively small amounts of leaded petrol, with more radiogenic isotopic compositions, were imported from Singapore and the US prior to 1998, but sales of those imports were restricted to northern Australia, thousands of kilometres from the field sites analysed in this study. Consequently, we hypothesized that lead isotopic compositions of soil and ash samples collected for this study would be composed primarily of natural lead and Australian industrial lead.

2. Methods

2.1. Sample collection sites

Ash and soil samples were collected at three sites across Australia shortly after three large, uncontrolled and intense bushfire events in relatively pristine areas in the first week of February 2011 (Fig. 1). These were in the Waygara State Forest (Tostaree Fire) in Victoria, ~340 km east of Melbourne; the Darling Range Regional Park (Red Hill Fire), ~20 km northeast of Perth; and the Banyowla Regional Park (Kelmscott Fire), ~20 km east of Perth. In all cases, ash samples were collected from the burnt remains at the base of large old eucalypt trees that had been destroyed during the fire events. This targeted sampling approach differs from that used in Odigie and Flegal (2011) who relied on the collection of ash samples accumulated at the surface of soils in the burnt area. Older trees were the focus of the sampling because we hypothesized that these trees would be more likely to contain signatures of historic lead depositions compared to smaller, younger trees that would have grown in period following the introduction and domination of Australian unleaded petrol for all new vehicles from 1986 (Australian Government, Department of Environment, 2001). Sampling of the ashed trees was undertaken using a semi-systematic sampling pattern spread out across the burnt area. While the collected samples were spread widely across each of the locations (Tostaree Fire 110 km^2; Red Hill Fire 11.7 km^2; Kelmscott Fire 10 km^2), the occurrence of ashed older trees dictated individual sample locations (Table S1, Supporting information). The ash was collected in plastic bags, using established trace metal clean procedures (Odigie and Flegal, 2011).

Soil samples were collected from depths of 0–2 cm (surface) and 40–50 cm (deepest), using comparable trace metal clean procedures (Taylor et al., 2010). Additional soil and rock samples were collected from Kelmscott and Red Hill in 2012. The deeper soil samples were used to determine local natural background soil lead concentrations and isotopic composition as no natural lead isotopic compositions had been reported in the literature for the study sites. This assumption was based on previous studies using a similar approach to determine successfully both local natural background lead concentrations and isotopic compositions. Subsurface sampling of soils at depths > 30 cm has been demonstrated previously to be a robust technique for characterizing the natural lead signature of a location (Gulson and Mizon, 1979; Gulson et al., 1981; Mackay et al., 2013; Taylor et al., 2010). Soil and rock samples were collected close to, but outside of, the bushfire areas with the footprint of the samples enveloping the ash sample areas.