

Lead isotope fingerprinting used as a tracer of lead pollution in marine sediments from Botany Bay and Port Hacking estuaries, southern Sydney, Australia

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ABSTRACT

Anthropogenic lead has been released into environmental ecosystems via human activities; essentially from mining, smelting of lead, leaded gasoline and industrial activities. In order to explore the source apportionment and historic record of lead pollution in the marine sediments, concentration of lead was determined from the Botany Bay and Port Hacking estuaries, south of Sydney, Australia. Areas with the highest concentrations of lead in the sediment samples were analysed by inductively coupled plasma-mass spectrometry (ICP-MS) for lead isotopes to effectively identify the metal contamination source. In addition, other sediment samples were collected from cores at 40 cm depth to represent the natural background composition. The study found that the total lead in the tested marine sediments varied from 75.6 mg/kg to 582.2 mg/kg. The $^{206}\text{Pb}/^{204}\text{Pb}$ showed a decline towards the current surface sediment. Assuming that the natural background source of lead remains the same in terms of both isotopic signature and accumulation rate, the decline in $^{206}\text{Pb}/^{204}\text{Pb}$ indicated a rise in the contribution of old lead to the sediment, mainly from gasoline fumes (car and boat exhausts) and paint. This is because the samples came from close to water discharge points which have concentrated the catchment lead via stormwater runoff.

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1. Introduction

Lead is considered a poisonous metal that is harmful to the human body and can cause serious diseases such as central nervous system disorders, kidney brain damage and high blood pressure (Needleman, 2004). Lead pollution above background concentrations in ecosystems is derived from anthropogenic activities such as industry, mining, coal burning, paint and gasoline-fumes. It has increased since the early 1950s from lead-based paints and gasoline-fumes in developed countries, as well as emissions from industrial activities (Needleman, 2004; Gutiérrez-Caminero et al., 2015). It is an important issue in environmental investigation to identify the source of pollution and determine the transport history of pollutants (Cheng and Hu, 2010). A large number of sources of lead pollution occur in ecosystems, including major contributions from urbanization such as gasoline-air emissions, paints

and insecticides, as well as from natural sources such as bedrock (Lu et al., 2011; Zhang et al., 2016). In marine environments, trace metals such as lead can be associated with particulate matter because of their hydrophobic nature and are finally deposited into the underlying sediments (Gerritse et al., 1998). Atmospheric deposition of trace metal pollution is directly documented through several types of environmental ecosystems, such as lake sediments, marine deposits and terrestrial soils (Brännvall et al., 1999; Gobeil et al., 1999; Kelly et al., 2009; Outridge et al., 2011). The distributions of total Pb in sediments can be derived from statistical analysis of a large sample dataset to categorize the source and transport of metal pollution. The chemical composition of trace metals can be an effective method to recognize the source of resources and to compare them by using “fingerprinting” (Qishlaqi and Moore, 2007; Hosono et al., 2016). Several attempts have been made to fingerprint the isotopic composition and ratios of Pb and other metals, which can play an important and powerful tool in tracking sources of pollution (Chiaradia et al., 1997; Franco-Uría et al., 2009; Cheng and Hu, 2010; Popescu et al., 2015; Wen et al., 2015; Hu et al., 2015). Thus, using isotopic ratios of elements is a more sound tracking method than using their concentrations alone to identify sources of contamination (e.g. carbon and chlorine Sueker,

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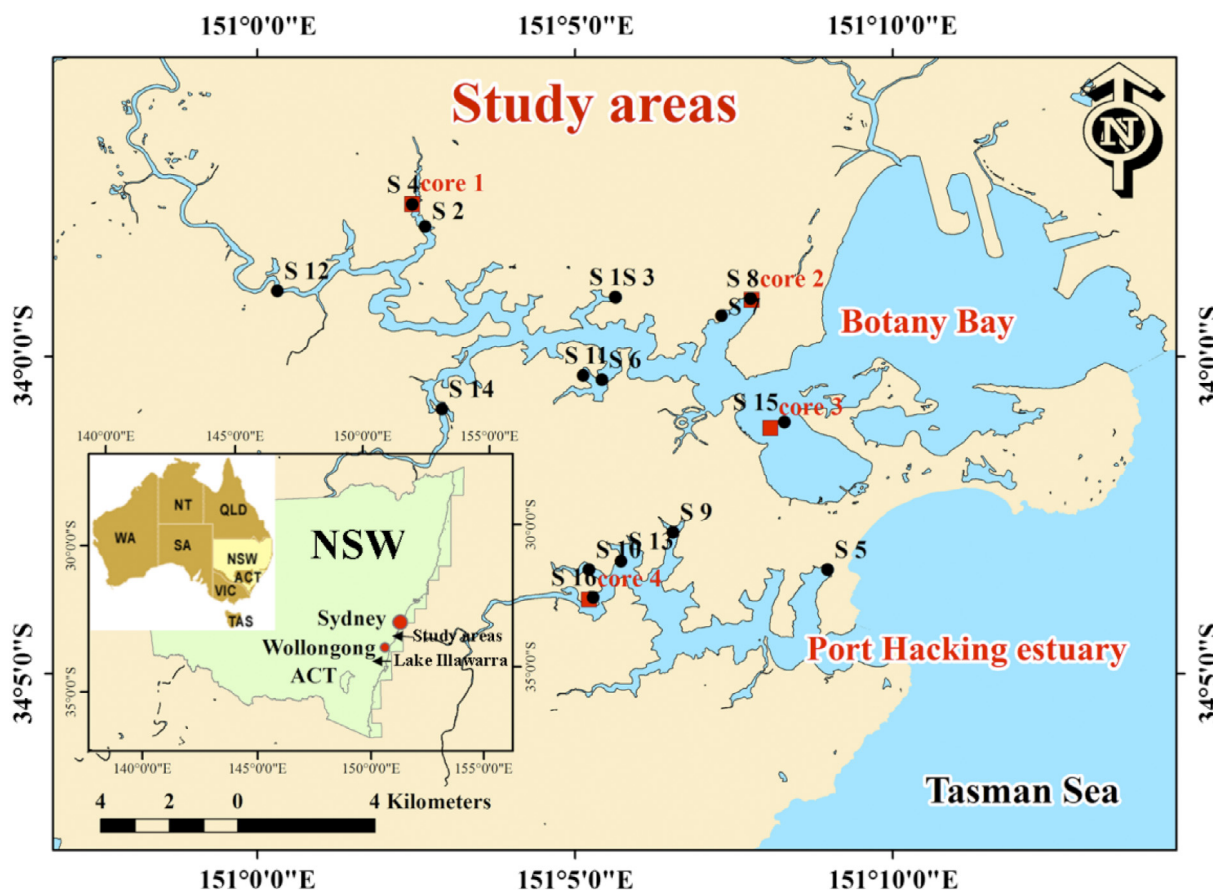


Fig. 1. Surface (dots) and subsurface (red squares) sample locations for lead isotopes in the study areas. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2001; Philp et al., 2002). In the same way, Pb isotopic composition may be used to trace the source and transport history of Pb (Chiara-dia et al., 1997; Cheng and Hu, 2010; Lu et al., 2011; Allan et al., 2015). The main objective of investigating lead isotopes is to support a source apportionment and historic record of lead pollution in the study areas.

2. Methods and materials

2.1. Study areas

This study covers two coastal areas classified as open estuaries, the Port Hacking and Botany Bay areas in New South Wales, Australia (Fig. 1). The Port Hacking catchment consists of approximately 60% natural bushland with the remainder being urbanized areas. It has a population of approximately 226,220 with a population density of 6 persons per hectare. Contamination is emerging from catchment discharge points as well as recreational activities such as watercraft. Port Hacking contains several bays and some shallow rivers and creeks. Consequently, these deeper bays form sediment traps for fine and very fine particles and detritus that are discharged from the rivers and creeks (Smith et al., 1990). In contrast, the catchment areas of Botany Bay are urbanized with population estimated at 264,624 and a population density of 28 persons per hectare. The sources of contamination are discharge points, stormwater, farming, fishing, transportation, light industry (including commercial and agricultural), as well as moored watercraft. It is described as a semi-enclosed estuary with a wide coastal embayment that joins with the open sea. The hydrodynamics of Botany Bay have been changed by anthropogenic construction and dredging, which includes the construction of a shipping container

port as well as the reclamation effects of Sydney's main airport on the northern side of Botany Bay (Gray et al., 2001; Fraser et al., 2006).

2.2. Sample collection

Surface sediment samples were collected using a grab sampler. The surface 5 cm of sediment was reserved for analysis. In addition, subsurface samples were collected using a push core. Fourteen surface sediment samples from the initial 428 samples, which have the highest concentrations of lead, and two samples from a previous study were selected and analysed for lead isotopes. Also, four subsurface sediment samples from different locations were selected to use as background values (Fig. 1 and Table 1). ArcGIS desktop software, version 10.2, was used to create bay boundaries around the collected plot sediment samples of each bay to produce maps for lead, bromine distributions and muddy particles. Spline with Barriers was chosen as the interpolation method for this part of the analysis due to the ability to input a barrier feature when interpolating a raster surface from points using a minimum curvature spline technique (Lin and Chen, 2004; Lark et al., 2006).

2.3. Analytical methods

Powdered samples were analysed ($<4 \mu\text{m}$) at the National Measurement Institute (NMI), NSW, Australia. X-ray fluorescence was used to measure concentration of total lead in sediments. Furthermore, approximately 2 gm of each sample was digested in acid (nitric and hydrochloric) then measured for lead using inductively coupled plasma–mass spectrometry (ICP-MS) using a

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