



Use of lead isotopes to identify sources of metal and metalloid contaminants in atmospheric aerosol from mining operations



Omar I. Félix^a, Janae Csavina^a, Jason Field^b, Kyle P. Rine^a, A. Eduardo Sáez^{a,*}, Eric A. Betterton^b

^a Department of Chemical and Environmental Engineering, University of Arizona, Tucson, AZ, USA

^b Department of Atmospheric Sciences, University of Arizona, Tucson, AZ, USA

HIGHLIGHTS

- Lead isotopes used for source apportionment of lead contamination at mining sites.
- Atmospheric particulate and soils exhibit lead and arsenic contamination.
- Contaminant soil deposition patterns can be determined from lead isotopic analysis.

ARTICLE INFO

Article history:

Received 10 June 2014

Received in revised form 7 November 2014

Accepted 18 November 2014

Available online 12 December 2014

Handling Editor: X. Cao

Keywords:

Lead isotopes

Dust and aerosol

MOUDI

Smelting

Metal and metalloid contamination

ABSTRACT

Mining operations are a potential source of metal and metalloid contamination by atmospheric particulate generated from smelting activities, as well as from erosion of mine tailings. In this work, we show how lead isotopes can be used for source apportionment of metal and metalloid contaminants from the site of an active copper mine. Analysis of atmospheric aerosol shows two distinct isotopic signatures: one prevalent in fine particles (<1 μm aerodynamic diameter) while the other corresponds to coarse particles as well as particles in all size ranges from a nearby urban environment. The lead isotopic ratios found in the fine particles are equal to those of the mine that provides the ore to the smelter. Topsoil samples at the mining site show concentrations of Pb and As decreasing with distance from the smelter. Isotopic ratios for the sample closest to the smelter (650 m) and from topsoil at all sample locations, extending to more than 1 km from the smelter, were similar to those found in fine particles in atmospheric dust. The results validate the use of lead isotope signatures for source apportionment of metal and metalloid contaminants transported by atmospheric particulate.

© 2014 Elsevier Ltd. All rights reserved.

1. Introduction

Metal- and metalloid-laden dust and aerosol particles are produced during mining operations in a wide range of sizes. Coarse particles (>1 μm aerodynamic diameter) are produced mainly by mechanical action like grinding and wind erosion (Csavina et al., 2011) while fine particles (<1 μm) are the result of molten ore processing and smelting, being produced by condensation and coagulation of vapors (Jacob, 1999; Banic et al., 2006). The impact of airborne particles on human health depends on particle diameter and composition. Particles larger than 10 μm are generally filtered out by the upper respiratory tract while particles smaller than 10 μm (PM₁₀) are inhaled and may be transported to the lungs where they are phagocytized by alveolar macrophages and transported to the blood stream (Zheng et al., 2004). The

composition of the particles also plays a key role in health effects. Of particular concern is the presence of lead and other contaminants (e.g., arsenic) in atmospheric dust and aerosol that result from mining activities (Csavina et al., 2011, 2012; Mackay et al., 2013).

The atmosphere is the major initial recipient of lead among all environmental compartments. Global anthropogenic sources of lead are at least one order of magnitude higher than natural sources (Komárek et al., 2008). The determination of potential sources of atmospheric lead is important since its production can be controlled in advance, minimizing unwanted exposures to this toxic metal. However, it is difficult to discriminate between sources of lead and other metal and metalloid contaminants by only knowing their total concentration (Hopper et al., 1991). For this reason, lead isotopic analysis has been introduced as a “fingerprinting” technique for lead contamination. Each source of lead may have a specific isotopic composition and the differences in this composition may be used to discriminate between possible sources (Komárek et al., 2008).

* Corresponding author. Tel.: +1 (520) 6215369.

E-mail address: esaez@email.arizona.edu (A.E. Sáez).

The use of lead isotopes to determine possible sources has been widely reported (e.g., Grousset et al., 1994; Munksgaard and Parry, 1998; Veyseyre et al., 2001; Kurkjian et al., 2002; Zheng et al., 2004; Chen et al., 2005). Lead has four stable isotopes (average molar abundances in parenthesis): ^{204}Pb (1.4%), ^{206}Pb (24.1%), ^{207}Pb (22.1%) and ^{208}Pb (52.4%). ^{204}Pb is the only non-radiogenic isotope and its abundance has remained constant over time (Russell and Farquhar, 1960). The abundances of ^{206}Pb , ^{207}Pb and ^{208}Pb in minerals vary because they are derived from the decay of ^{238}U , ^{235}U and ^{232}Th , respectively (Adgate et al., 1998). Isotopic composition of the radiogenic isotopes varies in different ores, depending on the age of their geological formation (Faure, 1986). An advantage of the use of lead isotopes for source apportionment is the fact that isotopic fractionation does not happen in industrial or environmental processes (Ault et al., 1970), owing mainly to the high atomic mass of lead. Lead isotopic composition is often expressed in terms of the concentration ratios $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ (Zheng et al., 2004; Chen et al., 2008).

Cumming and Richards (1975) observed that a plot of the concentration ratios $^{208}\text{Pb}/^{206}\text{Pb}$ vs. $^{207}\text{Pb}/^{206}\text{Pb}$ led to approximately a single monotonically increasing curve for all mineral samples analyzed. The radioactive decay of ^{238}U , ^{235}U and ^{232}Th in terms of their relative abundances and half-lives has led to an increase in the concentration of ^{206}Pb over time. As a consequence, the ratios $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ have decreased over time (Mukai et al., 2001). Fig. 1 shows the plot devised by Cumming and Richards, often called “growth curve”, and the distribution along the curve of three different ores: The Iron King mine (Arizona, USA) is the oldest ore of those shown with an age of 1800 million years (Anderson and Guilbert, 1979), the Mount Isa ore (Queensland, Australia) has an age of approximately 1600 million years (Cumming and Richards, 1975) and the Ray Mine ore (Arizona, USA) has an age of 70 million years (Bouse et al., 1999).

In this work, we sampled atmospheric dust and aerosol at two different sites in Arizona: The first site is located in the towns of Hayden and Winkelman, where a copper mine and smelter currently operate; while the second site is located in Tucson, representing a local urban environment. The main objectives of this work were:

1. To assess if lead isotopic analysis can be used for source apportionment of airborne particulate matter associated with mining operations.

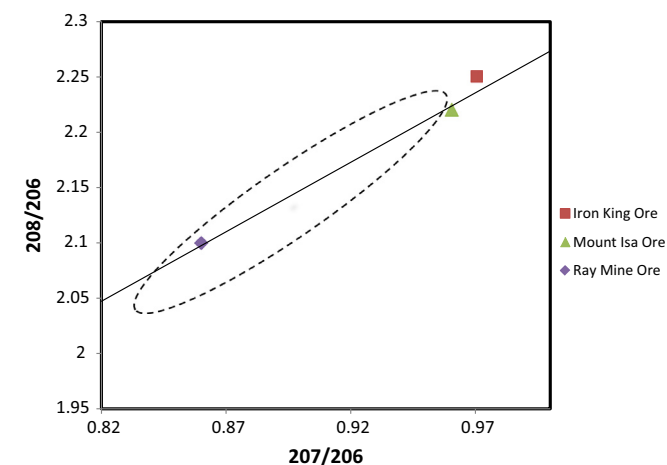


Fig. 1. The Cumming and Richards growth curve (solid line) for lead isotope concentration ratios (adapted from Chen et al., 2008), showing three mineral ores. The dashed region includes data from extensive sources (86 different sites in 35 countries) reported by Bollhöfer and Rosman (2001).

2. To investigate the extent of soil contamination in the vicinity of the smelting complex.

2. Materials and methods

2.1. Study site

The Hayden site is located approximately 80 km northeast of Tucson, Arizona. It is comprised of two towns: Hayden and Winkelman, with a combined population of approximately 1200. Currently, the site includes a concentrator, a copper smelter and tailings facilities. It is located at the confluence of the Gila and San Pedro Rivers. The processed copper ore originates from various mines in the vicinity, including the Ray mine. The Ray mine consist mainly of a porphyry copper deposit hosted in Pinal schist and Pioneer shale (w.mindat.org/loc-3377.html). In 2005, soil analysis showed that arsenic, lead and copper levels exceeded their respective residential soil remediation levels (EPA, 2012). The Environmental protection agency has reported elevated concentrations of arsenic, lead, copper, chromium and cadmium in atmospheric air samples in Hayden and Winkelman (EPA, 2012).

2.2. Sampling

The sampling site was located on the roof of a single-story High School building, approximately 2 km E of the mine tailings impoundment and 1 km SSE of the smelter (Fig. 2). A weather station and data logger (CR800, Campbell Scientific) providing information about temperature, relative humidity, wind speed and wind direction, was also present at the site. Wind speed and wind direction were analyzed with WindRose Pro (Enviroware) software. In Tucson, AZ, the sampling site was located on the roof of the five-story Physics and Atmospheric Sciences building at the University of Arizona.

A ten-stage micro-orifice uniform deposit impactor (MOUDI; M110-R, MSP Corporation) (Marple et al., 1991) was used to collect atmospheric dust and aerosol. The MOUDI was operated at a flow rate of 30 L min^{-1} for 96-h sampling periods. Sampling was performed in two modes: regular sampling was done by operating the MOUDI continuously for 96 h, while programmed sampling was performed by operating the MOUDI only for specified wind speeds and directions until total operating time reached 96 h. The calibrated cut points (d_{50} -values) for the MOUDI sampler are 18, 9.9, 6.2, 3.1, 1.8, 1.0, 0.55, 0.32, 0.18, 0.10 and $0.054\text{ }\mu\text{m}$ equivalent aerodynamic diameters. Teflon substrates (PTFE membrane, 2- μm pore size, 46.2 mm diameter, Whatman) were used for sample collection. The substrates were cleaned sequentially by DI water and methanol before use. They were transported to and from the field site in enclosed impactor holders (MSP Corporation). Substrates were weighed before and after sampling using EPA class I equivalent methods on an ultra-microbalance (Mettler Toledo XP2U). Aluminum substrates (47 mm, MSP Corporation) were occasionally used for electron microscope analysis samples.

Soil samples were taken at Hayden at four different distances from the smelting complex in a straight line transect NE of the smelter, a prevailing wind direction (Fig. 3). Samples were collected at different depths to obtain a vertical profile of the contamination: 0–3 mm, 3–6 mm, 6–9 mm, at 50 mm, and finally at 100 mm below the surface. Final samples at each site and depth were obtained by mixing three different samples. The composite samples were dried for 10 h at a temperature of $110\text{ }^\circ\text{C}$ and then sieved (ASTM, 2010) through a 0.84 mm sieve to discard coarser fractions.

Download English Version:

<https://daneshyari.com/en/article/4408396>

Download Persian Version:

<https://daneshyari.com/article/4408396>

[Daneshyari.com](https://daneshyari.com)