Arsenic and lead distribution and mobility in lake sediments in the south-central Puget Sound watershed: The long-term impact of a metal smelter in Ruston, Washington, USA

James E. Gawel [a],*, Jessica A. Asplund [a], Sarah Burdick [a], Michelle Miller [a], Shawna M. Peterson [a],
Amanda Tollefson [b], Kara Ziegler [a]

[a] University of Washington Tacoma, Environmental Science and Studies, Interdisciplinary Arts and Sciences Program, 1900 Commerce St., Box 358436, Tacoma, WA 98402, USA
[b] Bellarmine Preparatory School, 2300 S Washington St., Tacoma, WA 98405-1399, USA

HIGHLIGHTS

- We analyzed As and Pb in sediment and bottom water of 26 lakes near ASARCO smelter.
- As and Pb levels in surface sediments downwind are significantly elevated.
- As and Pb levels are correlated in cores, and consistent with smelter operation.
- Maximum dissolved As in bottom water is correlated with As in surface sediments.
- Sediment As and Pb in 83% of downwind lakes exceed *probable effects concentration.*

GRAPHICAL ABSTRACT
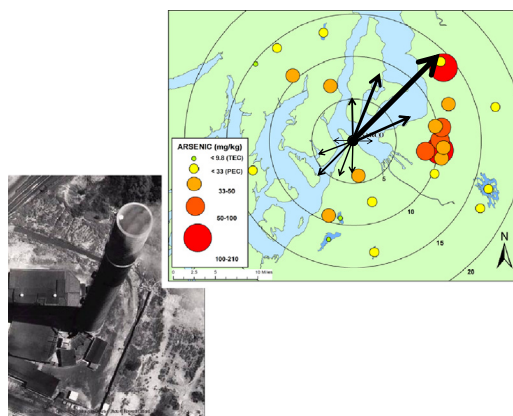
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ABSTRACT

The American Smelting and Refining Company (ASARCO) smelter in Ruston, Washington, contaminated the south-central Puget Sound region with heavy metals, including arsenic and lead. Arsenic and lead distribution in surface sediments of 26 lakes is significantly correlated with atmospheric model predictions of contaminant deposition spatially, with concentrations reaching 208 mg/kg As and 1375 mg/kg Pb. The temporal distribution of these metals in sediment cores is consistent with the years of operation of the ASARCO smelter. In several lakes arsenic and lead levels are highest at the surface, suggesting ongoing inputs or redistribution of contaminants. Moreover, this study finds that arsenic is highly mobile in these urban lakes, with maximum dissolved arsenic concentrations proportional to surface sediment levels and reaching almost 90 µg/L As. With 83% of the lakes in the deposition zone having surface sediments exceeding published "probable effects concentrations" for arsenic and lead, this study provides evidence for possible ongoing environmental health concerns.

* Corresponding author. Tel.: +1 253 692 5815; fax: +1 253 692 4639.
  E-mail addresses: jimgawel@uw.edu (J.E. Gawel), jasplund@uw.edu (J.A. Asplund), burdics8012@gmail.com (S. Burdick), mdmiller2006@msn.com (M. Miller), shawnadonley@gmail.com (S.M. Peterson), astollefson@gmail.com (A. Tollefson), karaziegler@hotmail.com (K. Ziegler).

1. Introduction

Lake sediments act as reservoirs of metal contaminants and long-term sources to overlying waters (Senn et al., 2007; Tanner and Clayton, 1990). Arsenic is of particular concern due to its potential toxicity to humans (Kazi et al., 2008) and other aquatic biota (Knauer et al., 1999; Lee et al., 1991; Wängberg et al., 1991), and its particular susceptibility to remobilization in urban areas where organic matter and nutrient inputs feed bacterial respiration in sediments and bottom waters (Lattanzi et al., 2007). The sediment pool may continue to be a significant source of arsenic to surface waters long after ongoing metal deposition has been abated (Couture et al., 2010; Senn et al., 2007; Tanner and Clayton, 1990).

Arsenic chemistry in aquatic systems is often dominated by the redox status of the system (Ferguson and Gavis, 1972). In many natural waters, the primary inorganic arsenic species of environmental significance are arsenate, $H_2AsO_4^-$:As($+V$), and arsenite, H_3AsO_3:As($+III$). Organic forms of arsenic, such as the monomethyl- and dimethyl-forms, may be significant under certain conditions as well. Arsenate binds to iron oxide solids under oxidizing conditions and precipitates out of the water column (Ferguson and Gavis, 1972). Over time this precipitation results in an accumulation of arsenic in lake sediments. In many urban lakes during summer stratification, bacterial respiration results in anoxic conditions in the hypolimnion as a result of cultural eutrophication. During anoxia, Fe(III)-oxides in the surface sediments are reduced to dissolved Fe^{2+}:Fe($+II$) (Ferguson and Gavis, 1972). In addition, direct reduction of As($+V$) to As($+III$) may occur under anoxic conditions (Harrington et al., 1998). Both mechanisms release arsenic to overlying waters, increasing the potential for downstream transport, biotic exposure and toxicity (Aggett and O'Brien, 1985; Batterson and McNabb, 1983).

The continuing mobilization of arsenic in lakes contaminated by historic sources creates the potential for human exposure and toxicity (Ferguson and Gavis, 1972). Food chain transfer to humans can occur in arsenic-contaminated lakes where fish consumption and irrigation of crops using lake water occur (Arain et al., 2009). In Washington State, there is increasing attention to the potential for contaminant toxicity in particularly susceptible populations consuming large quantities of fish. The Washington State Department of Ecology has stated, "The best current science now indicates that our present fish consumption rates do not accurately reflect how much of our state's fish and shellfish Washingtonians actually eat" (Washington State Department of Ecology, 2013a,b). This review of fish consumption rates is a precursor to review of sediment and water quality standards to protect human health on the basis of realistic fish consumption rates. To inform this review of arsenic standards for freshwater sediments and water resources, it is necessary to examine the spatial extent of arsenic contamination and the magnitude of arsenic bioavailability in fish habitat, including lakes.

The south-central Puget Sound region in Washington State has been heavily impacted by a century of metal emissions from the American Smelting and Refining Company (ASARCO) smelter in Ruston, Washington (Glass, 2003). Long-term emissions from the ASARCO smelter have resulted in the accumulation of toxic metals, including arsenic and lead, in surface soils and sediments of this region (Crecelius and Piper, 1973; Peterson and Carpenter, 1986; Seattle and King County Public Health and Glass, 2000). The smelter, in operation from 1890 to 1986, specialized in the smelting of lead and then copper ores containing high concentrations of arsenic; this facility separated and concentrated arsenic – along with the primary target lead or copper – from these ores for commercial sale. Arsenic and lead contamination is now widespread throughout the region via a combination of atmospheric transport from the emissions stack (at one time the world's tallest) and the use of smelter slag for road ballast (Mariner et al., 1997; Seattle and King County Public Health and Glass, 2000).

While arsenic and lead contamination of marine sediments and soils in the south-central Puget Sound region has been studied quite extensively (Crecelius et al., 1975), the distribution of arsenic and lead in sediments and arsenic mobility in the region's lacustrine systems has received relatively little attention from the research and regulatory communities to date. This region of Washington is home to a myriad of lakes, most of which are popular recreational resources for fishing, swimming and boating, and yet very few of these lakes have been examined for arsenic contamination and mobility, with almost no studies conducted after the closure of the ASARCO smelter.

In this study we describe our investigation of the temporal and spatial distribution of arsenic and lead in the sediments of lakes in the south-central Puget Sound region, as well as the subsequent release of dissolved arsenic to the overlying waters 20 years after the closure of the smelter.

2. Materials and methods

2.1. Site description

Lake sampling took place from 2003 to 2007. In total 26 different lakes (Fig. 1 and Table 1) were sampled for sediment metal concentrations (arsenic and lead) using surface grab samples or sediment cores. A subset of 8 lake sediment cores were dated using ^{210}Pb and a subset of 17 lakes were monitored for water column arsenic concentrations. The choice of lakes sampled was designed to include lakes within the dominant wind direction zones (from East to Northeast and South to Southwest) and outside these zones, but sampling was also influenced by the availability and ease of public boat access and the distribution of lakes in the region.

2.2. Sediment sampling

Surface sediments were collected in 2003 from 9 freshwater lakes (Table 1) within a 20 mile radius of the ASARCO smelter emissions stack. The 20-mile radius was chosen based on data from the Vashon/Maury Island Soil Study 1999–2000 (Seattle and King County Public Health and Glass, 2000) which showed elevated soil arsenic concentrations were limited mostly to distances of 20 miles or less. Duplicate surface sediment samples (approximately the top 10–20 cm of sediment) were collected from separate casts from a boat with a petit Ponar dredge (Wildlife Supply Company) from near the deepest point in each lake (based on available bathymetric maps or local knowledge). Sediments were homogenized with a plastic spoon, and placed in acid-washed Nalgene jars in a cooler for transport to the laboratory.

Sediment cores were collected in 2004, 2005 and 2007 from a subset of 17 lakes (Table 1). Cores were taken from near the deepest point in each lake using a gravity-driven, stainless steel corer with a plastic core catcher for sediment retention during retrieval (K-B corer, Wildlife Supply Company). Separate cellulose acetate butyrate core sleeves were used for each core. The corer was deployed and retrieved by hand from a boat and cores were capped and stored upright on ice for transport to the laboratory. The soft sediments were extruded and separated into 2 cm sections and placed into acid-washed Nalgene jars using a clear-PVC core extruder constructed generally following the design of Kornijow (2013). All surface sediment samples and core sections were dried for 3 days at 80 °C, homogenized in a Wiley mill, and stored in Whirl-pak bags prior to metals analysis.

A subset of eight of the sediment cores (Table 1) collected in 2004 and 2007 were dated in the Nittrouer lab at the University of Washington Seattle using excess ^{210}Pb activity. Excess ^{210}Pb activity was measured in each core section to determine sediment age and section-to-section sediment accumulation rate (Nittrouer et al., 1979). A dried, homogenized sample was spiked with ^{209}Po as a yield indicator. ^{210}Pb activity was determined by alpha spectrometry of granddaughter isotope ^{210}Po