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Bioavailability and uptake of smelter emissions in freshwater zooplankton in northeastern Washington, USA lakes using Pb isotope analysis and trace metal concentrations[☆]

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ABSTRACT

The upper Columbia River and associated valley systems are highly contaminated with metal wastes from nearby smelting operations in Trail, British Columbia, Canada (Teck smelter), and to a lesser extent, Northport, Washington, USA (Le Roi smelter). Previous studies have investigated depositional patterns of airborne emissions from these smelters, and documented the Teck smelter as the primary metal contamination source. However, there is limited research directed at whether these contaminants are bioavailable to aquatic organisms. This study investigates whether smelter derived contaminants are bioavailable to freshwater zooplankton. Trace metal (Zn, Cd, As, Sb, Pb and Hg) concentrations and Pb isotope compositions of zooplankton and sediment were measured in lakes ranging from 17 to 144 km downwind of the Teck smelter. Pb isotopic compositions of historic ores used by both smelters are uniquely less radiogenic than local geologic formations, so when zooplankton assimilate substantial amounts of smelter derived metals their compositions deviate from local baseline compositions toward ore compositions. Sediment metal concentrations and Pb isotope compositions in sediment follow significant ($p < 0.001$) negative exponential and sigmoidal patterns, respectively, as distance from the Teck smelting operation increases. Zooplankton As, Cd, and Sb contents were related to distance from the Teck smelter ($p < 0.05$), and zooplankton Pb isotope compositions suggest As, Cd, Sb and Pb from historic and current smelter emissions are biologically available to zooplankton. Zooplankton from lakes within 86 km of the Teck facility display isotopic evidence that legacy ore pollution is biologically available for assimilation. However, without water column data our study is unable to determine if legacy contaminants are remobilized from lake sediments, or erosional pathways from the watershed.

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1. Introduction

Metallurgic facilities are potential sources of trace metal pollutants that can harm organisms and degrade aquatic and terrestrial ecosystems. Metal pollutants from smelting facilities may be released as direct discharge of mining slag and as diffuse atmospheric emissions; studies have addressed biological responses to pollution arising from both pathways (Il'yashuk and Il'yashuk,

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2001; Il'yashuk et al., 2003; Thienpont et al., 2016). In rural and remote aquatic environments, most trace metal inputs originate from atmospheric deposition (Nriagu, 1990). For example, smelting operations in Trail, British Columbia, Canada, and Northport, Washington, USA, have contaminated the upper Columbia River by direct discharge of metal processing wastes, while airborne metal pollutants have contaminated higher altitude watersheds, lakes, and streams in the upper Columbia River watershed (Child et al., 2018; Johnson et al., 2011, 2013; USEPA, 2006, 2014; Vlassopoulos, 2010, 2014; WADOE, 1998).

The Le Roi smelter in Northport, Washington operated sporadically between 1898 and 1921 (Stevens, 2011; Tai, 2010). Copper and gold ores were processed during the initial 12 years of operation, and in 1909, the smelter temporarily ceased operating, and remained idle for seven years. Lead smelting operations

commenced in 1916, largely driven by Pb demands created during World War I. Lead ores were only refined during the later operational period (1916–1921), and were primarily from mines (i.e., Hercules, Tamarack and Custer) located in the Silver Valley region (i.e., Tiger Peak) of north Idaho (ROT, 1922).

The Teck facility in Trail, British Columbia is one of the largest non-ferrous Zn/Pb smelters in the world, and has been discharging heavy metal laden emissions since 1896. By comparison, smelting operations at the Teck smelter have been much larger and more sustained than the intermittent Le Roi operations (Queneau, 2014; Stevens, 2011; RIAA, 2006). The predominantly north/south orientation of the upper Columbia River allows prevailing winds to funnel emissions into northeastern Washington, where particulate metals are deposited into lakes and watersheds (Goodarzi et al., 2001; Johnson et al., 2013; USEPA, 2006; Vlassopoulos, 2014). Several studies have addressed aerial transport and deposition of smelter emissions to the surrounding area (Goodarzi et al., 2001, 2002a, b, 2003) and the upper Columbia River valley (Child et al., 2018; Johnson et al., 2011, 2013; USEPA, 2006, 2014; Vlassopoulos, 2010, 2014; WADOE, 1998). Emissions from the Teck smelter are the dominant metal pollution source to upland lakes in the region (Vlassopoulos, 2014), and have been detected in lake sediment up to 144 km away from the Teck smelter (Child et al., 2018). Previous studies have not addressed biologic availability of airborne trace metal pollutants. The main objective of this study is to determine whether airborne pollution, either deposited presently or remobilized from legacy pollution in sediments or the watershed, is biologically available for assimilation by zooplankton in northeastern Washington lakes using Pb isotope analysis and trace metal concentrations.

Researchers have successfully utilized Pb isotope analysis to track sources of atmospheric pollution (e.g., petrol additives, smelting emissions) in sediment cores and lichens (Bindler et al., 2001; Bollhöfer and Rosman, 2001; Child et al., 2018; Dolgoplova et al., 2006; Farmer et al., 1996; Goodarzi et al., 2001). Pb isotope analysis has also been used in marine, estuarine, and terrestrial food webs to trace pollutants (Ip et al., 2005; Scheuhammer and Templeton, 1998; Smith et al., 1990; Søndergaard et al., 2010). Fewer studies have employed Pb isotopes in freshwater food webs, especially to track smelter pollution (Gélinas and Schmit, 1997). Although Pb isotope compositions can be altered by mixing with additional Pb sources (e.g., watershed derived Pb), isotopic compositions are unaltered by smelting procedures (Shiel et al., 2010), atmospheric transport and deposition (Farmer et al., 1996), and trophic transfer within freshwater ecosystems (Gélinas and Schmit, 1997).

Major ores processed at the Teck facilities and their isotopic composition have been previously documented (Sangster et al., 2000; Shiel et al., 2010; Vlassopoulos, 2010, 2014; Queneau, 2014). Lead ores from the Sullivan deposit (Kimberly, British Columbia) were first delivered to the smelter in 1910, and were the primary feed ores processed until the deposit was exhausted in 2001 (Queneau, 2014; TECK, 2002; Vlassopoulos, 2014). The Teck facility also processed ores from Pine Point (1964–1988) and Red Dog (1989 to present) deposits located in Northwest Territories and Alaska, respectively (Queneau, 2014). Following the exhaustion of the Sullivan ores, the smelter continued processing Red Dog ores, and added ores from Pend Oreille (Washington, USA) and Bolivia mines (Shiel et al., 2010). Pb isotope analysis is uniquely suited for this study because the isotopic compositions of ores historically processed by the Teck and Le Roi smelters are less radiogenic than natural geologic formations in southeastern British Columbia and northeastern Washington (Cannon et al., 1962; Child et al., 2018; Church, 2010; Höy and Dunne, 2001; Nelson, 2011; Sangster et al., 2000; Shiel et al., 2010; Small, 1973; Vlassopoulos, 2010, 2014;

Zartman and Stacey, 1971).

Metal body burden and Pb isotope analysis were utilized to determine if smelter emissions are biologically available to zooplankton within lakes located in the upper Columbia River watershed. Based on previous surveys, lakes in closest proximity to the smelters and the river valley were suspected to contain more smelter derived contamination (Johnson et al., 2013). Metal cycling in lakes can be significantly influenced by productivity, stability, and redox environment adjacent to the sediment-water interface (Hamilton-Taylor and Davison, 1995). Lakes with the highest level of bioavailable metals should result in increased zooplankton body burdens. Consequently, if legacy or current smelter derived pollution represents a significant fraction of the biologically available metals, zooplankton Pb isotope compositions would be similar in composition to ores processed by the smelters rather than geogenic baseline compositions.

2. Materials and methods

2.1. Site locations and sample collection

A distribution of lakes, ranging from 17 to 144 km from the Teck smelter, was selected based on likelihood of airborne Pb contamination from previous reports (Child et al., 2018; Johnson et al., 2013; Vlassopoulos, 2014), proximity to the smelter, and likely airborne distribution plumes in the Columbia River valley (Fig. 1; ICF, 2011). Lakes varied in maximum depth, surface area and watershed dimensions, but were comparable in elevation and trophic state (Table 1). All the study lakes, except Phillips, typically exhibit stratification periods with subsequent hypolimnetic anoxia.

Sediment samples were collected near the deepest area of the lake in triplicate with a stainless-steel Ekman grab (15.25 × 15.25 × 15.25 cm), and were homogenized in stainless-steel buckets (Johnson et al., 2013). Homogenized samples were placed in polyethylene bags, labeled, and iced for transport to the laboratory, where they were frozen and stored until processed. Zooplankton were collected in triplicate from all lakes, except Phillips, using a 73- μm -mesh net connected to a 250- μm -mesh collection cup (Reed, 2011). 250- μm -mesh collection cup was added to preferentially select larger bodied zooplankton which can make diel vertical migrations (e.g., Cladocera, Copepoda, *Chaoborus* sp.). Vertical net tows commenced 1 m above the sediment-water interface and pulled to the surface. Composite zooplankton samples were collected to ensure adequate biomass for analyses, and contained 20 to 50 tows, depending on zooplankton densities and lake depth. Zooplankton were concentrated in acid washed bottles, triple rinsed with deionized water, filtered on quartz fiber filters, and frozen prior to analysis. All samples were subsequently freeze-dried, homogenized, and processed in the Radiogenic Isotope and Geochronology Laboratory at Washington State University (USA).

Water quality parameters were collected simultaneously during sediment and zooplankton sampling. Oxidation-reduction potential (ORP) and pH were measured at 1-m intervals with a Hydrolab MiniSonde 5 (OTT Hydromet, Loveland, Colorado). Surface water TP and TN concentrations were analyzed using the semi-automated colorimetric method following persulfate digestion (APHA, 2005). Carlson trophic state index (TSI) was calculated using surface water TP concentrations (Carlson, 1977). Sediment organic matter composition was determined by loss-on-ignition at 550 °C (Heiri et al., 2001).

2.2. Laboratory analysis

Total Hg in sediment and zooplankton on a dry weight (dw) basis was analyzed using a Direct Mercury Analyzer 80 (Milestone

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