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Arsenic and mercury in lake whitefish and burbot near the abandoned Giant Mine on Great Slave Lake

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ABSTRACT

Contaminant levels in fish are of public concern in northern Canada where they are an important food source. In this study, we investigated the concentration of total arsenic, four arsenic species (arsenite (AsIII), arsenate (AsV), dimethylarsinate (DMA), and monomethylarsonate (MMA)), and total mercury (Hg) in the muscle and liver of lake whitefish (*Coregonus clupeaformis*) and burbot (*Lota lota*) collected at two sites near the abandoned Giant Mine site (Baker Pond and Yellowknife Bay) and two reference sites more than 25 km away (Chitty Lake and southern Great Slave Lake). Total arsenic concentrations were typically higher in fish tissues collected near the mine site, and higher in burbot than lake whitefish. We found lower concentrations of arsenic in the muscle tissue of adult lake whitefish than juveniles. All four arsenic species were only detected in the liver tissues of adult lake whitefish collected from Baker Pond on the mine site, and juvenile lake whitefish from the adjacent Yellowknife Bay. Mercury levels were highest in fish from Chitty Lake, and higher for burbot than lake whitefish, similar with other research reporting elevated mercury in small northern lakes relative to larger waterbodies. However, mercury levels in fish were not elevated beyond consumption guidelines. Elevated arsenic concentrations in the fish tissues collected near the mine site suggest that the area continues to be a source of arsenic to the aquatic food web; therefore, continued monitoring is warranted, particularly with a large portion of the local population harvesting wild food sources.

Introduction

In Canada's Northwest Territories (NWT), fish are an important and culturally valued resource, with approximately 40% of the population hunting and fishing to supplement their diet (GNWT, 2014), and 25% of the total population partaking in recreational angling, which is, per capita, more than most other jurisdictions in Canada (Fisheries and Oceans Canada, 2012). Yellowknife Bay, on the north shore of Great Slave Lake, supports many species of large bodied fish that are used as food by people in the City of Yellowknife and the nearby aboriginal communities of Dettah and N'dilo. Large bodied fish are known to accumulate contaminants and often have elevated concentrations relative to biota lower in the food chain (e.g., Evans et al., 2005; Kidd et al., 2012). Elevated concentrations of certain metals in fish, such as mercury and arsenic, are of public concern because of the well documented health risks associated with consuming fish with high metal burdens (Canadian Council of Ministers of the Environment (CCME), 2000; Canadian Food Inspection Agency (CFIA), 2014; Health Canada, 2012).

Mercury is of particular concern because of its documented toxicity, persistence in the environment, high potential for bioaccumulation in the aquatic food web and ability to biomagnify with increasing trophic levels (CCME, 2000; Kidd et al., 2012; Wiener et al., 2003). Toxic effects on fish include disrupted neurological function and reduced growth, oxygen uptake, reproductive development, sensory abilities, osmoregulation, and digestion (Kidd et al., 2012; Scheuhammer et al., 2015; Wiener et al., 2003). In the aquatic environment, mercury can be converted through biogeochemical interactions to the more toxic organic methylmercury (MeHg) (CCME, 2000; Chetelat et al., 2015; Jensen and Jernelöv, 1969; Winfrey and Rudd, 1990). Fish accumulate MeHg through their diet (Rodgers, 1994). Fish tissues are typically analyzed for total mercury, since it has been demonstrated that the majority of mercury in fish is present as MeHg (Bloom, 1992; Forsyth et al., 2004). Mercury is naturally occurring in the environment (Lockhart et al., 2005), but levels can be exacerbated by the cumulative impacts of natural disturbances like fire (Garcia and Carignan, 1999; Kelly et al., 2006) and anthropogenic activities such as logging (Garcia and Carignan, 1999), mining (Lockhart et al., 2005), flooding for hydroelectric development (Bodaly et al., 1984), or atmospheric inputs from waste incineration and fossil fuel emissions (Kidd et al., 2012).

Point-source contamination from industrial activities into the aquatic environment is a prime public concern. Arsenic contamination is often associated with historic gold mining activity since arsenic commonly occurs in the ore of gold bearing metal sulfide deposits (Cohen

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and Bowell, 2014). The toxicity of arsenic is well known (Bowell and Craw, 2014), being a carcinogen in humans (Kapaj et al., 2006) and causing toxic and biological effects on fish (Pedlar et al., 2002b). It bioaccumulates in the aquatic environment, but concentrations normally decrease with trophic position (Chen and Folt, 2000; Dutton and Fisher, 2011; McIntyre and Linton, 2012). Adverse impacts to fish include effects to growth and reproduction through diminished appetite, altered feeding behavior, increased abnormalities, reductions in gonadal development, spawning success in adults, hatching success of eggs, and overall health and survival during the early life stages (McIntyre and Linton, 2012; Wiener et al., 2003).

Like mercury, arsenic has different species (or forms), and these species have varying toxicities due to different physical and chemical properties. The inorganic forms of arsenic [arsenite (AsIII) and arsenate (AsV)] are generally considered more toxic than organic species [dimethylarsinate (DMA) and monomethylarsonate (MMA)], with the trivalent form generally considered the more toxic of the two inorganic species (McIntyre and Linton, 2012). Arsenic can be absorbed directly through the gills of fish as well as through the gut, where it can be methylated from an inorganic to organic form (McIntyre and Linton, 2012).

Giant Mine on the shore of Yellowknife Bay in Great Slave Lake operated between 1948 and 2004 and was one of the most productive gold mines in Canadian history (Government of Canada, 2011). Gold production at Giant Mine also resulted in the production and subsequent storage of large amounts of arsenic waste onsite, rendering the mine site one of the most contaminated sites in Canada (Federal Contaminated Sites Action Plan (FCSAP), 2014; Office of the Auditor General of Canada (OAG), 2012; Government of Canada, 2011). Close to 260,000 tonnes of arsenic trioxide (As_2O_3) was generated as a byproduct of gold ore processing at Giant Mine. Two hundred thirty seven thousand tonnes of As_2O_3 was captured by emission control technologies over the operating life of the mine and is currently stored underground; however, 20,000 tonnes was not captured and was released to the surrounding landscape via emissions from the roaster stacks employed at the mine (Jamieson, 2014; Wrye, 2008). Over many decades, in addition to the atmospheric fallout from historic roaster stack emissions, Yellowknife Bay received indirect anthropogenic inputs of arsenic through the discharge of mine wastewater via Baker Creek, and the historical deposition and erosion of tailings along the northeast shoreline of Yellowknife Bay (Andrade et al., 2010). In the early years of mine operations (1948–1951), tailings were deposited directly into Yellowknife Bay in a small embayment on the north shore. These tailings have subsequently redistributed within Yellowknife Bay over time (Golder Associates Limited, 2005).

Arsenic loading to Yellowknife Bay has long been a concern with several earlier studies identifying elevated levels of arsenic in sediment and surface waters of Yellowknife Bay (Jackson et al., 1996; Mace, 1998; Moore et al., 1978; Mudroch et al., 1989). However, relatively little information is available regarding arsenic in fish in Yellowknife Bay (Jackson et al., 1996; de Rosemond et al., 2008), and none from sites directly on the mine site.

The primary objective of this work was to evaluate arsenic and mercury burdens in fish close to Giant Mine and compare these data with that from fish collected from reference lakes beyond the influence of historic mining activity at Giant. We hypothesize that arsenic concentrations will be highest in the fish tissues collected nearest the mine site and higher in fish with lower trophic status, whereas mercury concentrations will be higher in the tissues of fish collected from small lakes and with higher trophic positioning. We analyzed metal concentrations in the liver and muscle tissues from juvenile and adult lake whitefish (*Coregonus clupeaformis*) and adult burbot (*Lota lota*), two large bodied fish species that occur in Yellowknife Bay. These species were selected as they are commonly harvested fish for human consumption (flesh of both species and the liver of burbot are eaten) and represent different trophic positions. Lake whitefish feed mainly on plankton as juveniles, and benthic invertebrates as adults (Scott and Crossman, 1973). Therefore depending on size, lake whitefish can be classified into different trophic levels. Burbot are top-level predators having an almost exclusively fish-based diet as adults (Amundsen et al., 2003) and occupy a higher trophic position than lake whitefish (Cott et al., 2011).

Methods

Sample sites and fish collections

Lake whitefish and burbot were collected at four locations within 200 km of Giant Mine, Yellowknife, NWT. Lake whitefish collected from southern Great Slave Lake were obtained from local commercial fisherman, and lake whitefish from all other sampling locations were captured using multi-mesh gillnets. Burbot were targeted using longlines baited with cisco (*Coregonus artedi*). All fish were immediately killed upon capture, placed on ice, and frozen until dissection. Fork length and total length (±1.0 mm) were recorded for lake whitefish and burbot, respectively, and total body mass (±1.0 g wet) was measured for both fish species. Tissue samples of skinless white dorsal muscle and liver (±10 g) were collected from each fish, placed in individual small plastic bags and frozen at −20 °C for subsequent analyses.

In December 2010, adult lake whitefish (n = 8) were collected from Baker Pond, a reach of Baker Creek (62° 30′ 28 N 114° 21′ 32 W), which flows through the Giant Mine site into Yellowknife Bay (Fig. 1). Historically, Baker Pond was the receiving environment for Giant Mine's tailings and treated waste water (Fawcett et al., 2015).

Concentrations of arsenic in surface waters and sediments in Baker Pond vary seasonally in association with changes in redox conditions and changing inputs from the catchment. Reported values for arsenic in surface waters and sediments range from 200 to 4000 μg/L and from 2000 to 14,000 mg/kg, respectively (Nash, 2015; Fawcett et al., 2015; Walker et al., 2015). At the time of fish collection there was less than 1 m of water below the ice.

In March 2012, adult (n = 8) and juvenile (n = 8) lake whitefish, and adult burbot (n = 8) were collected from Yellowknife Bay, Great Slave Lake (62° 24′ 40 N 114° 20′ 13 W), approximately 1 km from Giant Mine (Fig. 1). Previous research has shown that this area of Yellowknife Bay (Back Bay) has been impacted by historical mining activities, either through the discharge and redistribution of tailings and wastewater or via roaster emissions (Jackson et al., 1996; Andrade et al., 2010). Concentrations of arsenic in surface waters of Yellowknife Bay vary seasonally and recently reported concentrations range between 0.5 and 10 μg/L (Jackson et al., 1996; Andrade et al., 2010). The lacustrine sediments of Yellowknife Bay act as both a source and a sink of arsenic to overlying waters, dependent on redox conditions and other biogeochemical factors (Andrade et al., 2010). Values of reported concentrations of arsenic in sediments from the main basin of Back Bay range between 53 and 1000 mg/kg (Jackson et al., 1996; Andrade et al., 2010).

In June 2012, adult lake whitefish (n = 9) and burbot (n = 8) were collected from Chitty Lake (62° 42′ 48 N 114° 7′ 54 W), approximately 25 km northeast of Giant Mine in an area expected to be beyond the influence of historic roaster arsenic emissions on water, sediment and aquatic biota (Wagemann et al., 1978) (Fig. 1). Wagemann et al. (1978) report arsenic concentrations in water and sediment as <10 μg/L and 28 mg/kg, respectively. Chitty Lake is primarily surrounded by Archean metasedimentary rocks of the Yellowknife Supergroup. Arsenic concentrations are generally lower in this unit compared to bedrock of the Yellowknife Greenstone Belt (Boyle, 1960; Galloway et al., 2015); therefore, geogenic inputs of arsenic to lake sediments and water are expected to be low.

In July 2012, adult lake whitefish (n = 8) were collected from the south side of Great Slave Lake, approximately 10 km north of Hay River, NWT (60° 59′ 32 N 115° 41′ 10 W). The southern and eastern shores of Great Slave Lake are part of the Western Canadian Sedimentary Basin.