



Arsenic and antimony geochemistry of mine wastes, associated waters and sediments at the Giant Mine, Yellowknife, Northwest Territories, Canada



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ABSTRACT

Elevated levels of arsenic (As) and antimony (Sb) in water and sediments are legacy residues found downstream from gold-mining activities at the Giant Mine in Yellowknife, Northwest Territories (NWT), Canada. To track the transport and fate of As and Sb, samples of mine-waste from the mill, and surface water, sediment, pore-water, and vegetation downstream of the mine were collected. Mine waste, pore-water, and sediment samples were analyzed for bulk chemistry, and aqueous and solid-state speciation. Sediment and vegetation chemistry were evaluated using scanning electron microscope imaging, synchrotron-based element mapping and electron microprobe analysis. The distributions of As and Sb in sediments were similar, yet their distributions in the corresponding pore-waters were mostly dissimilar, and the mobility of As was greater than that of Sb. Competition for sorption sites is the most likely cause of elevated Sb concentrations in relatively oxidized pore-water and surface water. The aqueous and solid-state speciation of As and Sb also differed. In pore-water, As(V) dominated in oxidizing environments and As(III) in reducing environments. In contrast, the Sb(V) species dominated in all but one pore-water sample, even under reducing conditions. Antimony(III) appears to preferentially precipitate or adsorb onto sulfides as evidenced by the prevalence of an Sb(III)-S secondary solid-phase and the lack of Sb(III)(aq) in the deeper zones. The As(V)-O solid phase became depleted with depth below the sediment-water interface, and the Sb(V)-O phase persisted under relatively reducing conditions. In the surficial zone at a site populated by *Equisetum fluviatile* (common horsetail), As and Sb were associated with organic material and appeared mobile in the root zone. In the zone below active plant growth, As and Sb were associated primarily with inorganic phases suggesting a release and reprecipitation of these elements upon plant death. The co-existence of reduced and oxidized As and Sb species, instability of some phases under changing redox conditions, and plant uptake and release pose challenges for remediation efforts at the mine.

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1. Introduction

The mobility, attenuation, and transformation mechanisms of metal(loid)s in the environment is important for understanding and evaluating the potential risks to ecosystems and humans, for predicting their stability over time, and for designing effective remediation strategies of contaminated sites. Arsenic and Sb are in the same group in the periodic table, have similar electronic structures (ns^2p^3), and are strongly associated in some hydrothermal mineral deposits. For these reasons it is often assumed that they exhibit similar geochemical behavior. Research conducted at the Giant Mine, a former gold mine in Yellowknife, NWT, Canada

(Fig. 1) offered an opportunity to compare the geochemical behavior of As and Sb because both elements are elevated in mine waste and in the surrounding environment. Ore was roasted at Giant to extract gold, resulting in a complex range of solid As and Sb-bearing phases in the mine waste. Also, a water treatment plant has been operating at the site for 17 years before sampling, influencing the speciation of metalloids downstream in Baker Pond and Baker Creek. Determining the fate of As and Sb in the environment requires an understanding of the effects of anthropogenic processing on speciation as well as identification of the major controls on As and Sb mobility and speciation under different natural environmental conditions. An understanding of the mobility and possible attenuation of As and Sb is important because Baker Creek is the main conduit for mine waste into Yellowknife Bay (Fig. 2). The aim of this study was to determine the distribution and speciation

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of As and Sb throughout the paths taken by these contaminants, from the roaster at the mine through water bodies, sediments, and vegetation, and to infer the processes that control the mobility and attenuation of these two elements.

1.1. Mobility and attenuation of As and Sb

The oxide minerals of As and Sb are highly soluble (Greenwood and Earnshaw, 1984), so it is only at very high concentrations of these elements that pure As and Sb oxides are expected to control the activities of As and Sb in solution. Concentrations can be attenuated by sorption to Fe and Mn oxides and oxyhydroxides (Ackermann et al., 2008; Kneebone et al., 2002; Leuz et al., 2006a; Manning et al., 1998; Martinez-Llado et al., 2008; Mitsunobu et al., 2006; Morin et al., 1999; Scheinost et al., 2006; Takahashi et al., 2010; Waychunas et al., 1993), although the strength of those bonds differs for the various species. For example, Sb(III) strongly adsorbs to goethite over a much wider pH range (3–12) and forms the stronger bonds, compared to Sb(V) (Leuz et al., 2006a). In contrast, As(V) binds more strongly and over a lower pH range on the surfaces of iron oxide minerals than does As(III) (e.g., Howell, 1994; Dixit and Hering, 2003). In reducing environments, As and Sb can be attenuated by adsorbing onto sulfide surfaces or by precipitating as sulfides (Chen et al., 2003; Farquhar et al., 2002; Gallegos et al., 2007; Kirsch et al., 2008; Wolthers et al., 2007).

Although neither As nor Sb are essential to plants, uptake by vegetation and interactions with root processes are also potential controls on As and Sb mobility. Hydrophytes can act as a biological barrier to contaminant movement into the environment, but may also pass contaminants into the food chain and increase mobility through the exudation of organic acids and acidification due to plant processes (Hozhina et al., 2001). Arsenic and Sb concentrations were found to be elevated in the fluvial horsetail (*Equisetum fluviatile*), the same species found in the aqueous environment near the Giant Mine (Brooks et al., 1981; Hozhina et al., 2001).

1.2. Controls on As and Sb speciation

In water, the redox couples (As(III/V)) and (Sb(III/V)) commonly do not reach equilibrium and these species are often found in concentrations that are in excess of what would be predicted thermodynamically (Filella et al., 2002; Inskeep et al., 2002). Discrepancies between thermodynamic predictions and field observations may be attributed to slow reaction kinetics, biological activity, and photo-oxidation processes (Bertine and Lee, 1983; Filella et al., 2002; Krupka and Serne, 2002; McCleskey et al., 2004). Although they are often closely associated and have similar electronic structures, As and Sb exhibit different redox properties whereby the oxidized species of Sb and reduced species of As can coexist in solution (Mitsunobu et al., 2006; Mok and Wai, 1990).

Oxidation of As and Sb by oxygen alone has been shown to be slow, ranging from months to years (Cherry et al., 1979; Eary and Schramke, 1990; Leuz et al., 2006b), but Fe and Mn oxides and humic acids have been shown to catalyze the oxidation of both As and Sb (Belzile et al., 2001; Buschmann and Sigg, 2004; Kneebone et al., 2002, and references therein; Kuehnelt et al., 1997; Leuz et al., 2006a; Manning et al., 2002; Watkins et al., 2006). Reduction of Sb(V) on the surfaces of nanoparticulate magnetite and mackinawite has also been observed (Kirsch et al., 2008). Both As(III) and Sb(III) can be oxidized by microbial processes of chemolithoautotrophy, respiration, and detoxification (Ehrlich, 2002; Filella et al., 2007; Stolz et al., 2006). Microbially mediated oxidation rates for As(III) are greatly enhanced over inorganic rates (e.g. Nordstrom, 2003). Little is known about

microbially mediated Sb(III) oxidation rates but results from Asta et al. (2012) indicate that such rates also are enhanced over inorganic rates but are substantially slower than microbially mediated As(III) oxidation and this is likely to be the same for rates of reduction.

1.3. Study area

Giant is an underground gold mine located 5 km north of Yellowknife, NWT, Canada, on the northern arm of Great Slave Lake (Fig. 1). The mine was active for fifty years, between 1948 and 1999. The mine area includes a roaster and settling and tailings ponds. Baker Creek flows through the mine property (Fig. 2) and has been impacted by tailings spills and decants, discharge from the water treatment plant and possibly historic stack emissions (Jamieson, 2014). Baker Pond (BP) lies on the stream path immediately downstream of the settling pond and polishing ponds associated with the water treatment plant (Fig. 2). From BP, water empties into Baker Creek, which flows through the mine site and discharges into Yellowknife Bay. Two sampling sites, Baker Creek Vegetated (BCV) and Baker Creek Channel (BCC), are located farther downstream at the outlet to Yellowknife Bay (Fig. 2).

1.3.1. Ore geology and processing

The ore deposits are located in the Archean Yellowknife Bay Supergroup greenstone belt, and hosted in deformation and alteration zones that crosscut the Kam Group mafic volcanics (Canam, 2006). The ore zones contain up to 15%, but more often less than 5% sulfide minerals, which include pyrite, arsenopyrite, sphalerite, chalcopyrite, stibnite, Sb-bearing sulphosalts and pyrrotite (Coleman, 1957). The Au was refractory and hosted primarily in arsenopyrite, and to a lesser extent in pyrite and quartz (Coleman, 1957). To extract gold, processing consisted of four steps: crushing, floatation, roasting, and cyanidation. Roasting was conducted at 500 °C over two stages to oxidize the As and S and convert the arsenopyrite and pyrite into porous iron oxides (maghemite and hematite) amenable to cyanidation. The volatile phase of As was As₂O₃ which was condensed and collected in the baghouse. The Sb gaseous phase was most likely Sb₂O₃ (Marsden and House, 2006; Riveros et al., 2000); the mixed oxidation-state Sb mineral, Sb₂O₄, could also have been thermodynamically stable under the roasting conditions (Asryan et al., 2003), and a vitreous and sparingly soluble (As,Sb)₂O₃ phase also was reported (Riveros et al., 2000). Details of ore processing at Giant is covered by More and Pawson (1978), McQuiston and Shoemaker (1975), and Walker et al. (2005, 2015). The same roaster was used from 1958 to 1999 and changes were reportedly minor and would have ensured a consistent calcine (Marsden and House, 2006).

Arsenic and Sb are associated with maghemite and hematite as As(III) and As(V), and Sb(III) and Sb(V) (Fawcett and Jamieson, 2011; Walker et al., 2005). The association of both oxidation states with the roaster oxides suggests that complex solid–gas phase reactions did occur in the roaster and multiple forms of the metalloids were available to associate with the Fe-oxides.

Processing produced three tailings streams: the floatation tailings which consist dominantly of silicate minerals remaining after sulfide removal, the calcine which consists of As and Sb-rich cyanided roaster-derived material, and the Electrostatic Precipitator (ESP) residue consisting of the cyanided particulate material in the roaster discharge gas. The calcine is composed of fine grains, 90% < 0.045 mm, and the ESP is composed of very fine grains, 90% < 0.014 mm. The floatation tailings comprised the largest volume waste stream but had the lowest As and Sb concentrations, whereas the calcine and ESP dust comprised the smallest volume but contained the greatest concentrations of As and Sb (Fawcett et al., 2006; Walker et al., 2015). The dominant minerals in the

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