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The Legacy of Arsenic Contamination from Mining and Processing Refractory Gold Ore at Giant Mine, Yellowknife, Northwest Territories, Canada

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INTRODUCTION

The case of the Giant mine illustrates how a large, long-lived Au mine has resulted in a complex regional legacy of As contamination and an estimated remediation cost of almost one billion Canadian dollars (AANDC 2012). The mine, located a few km north of the city of Yellowknife on the shore of Great Slave Lake (Figs. 1, 2) produced more than 7 million troy ounces of Au (approximately 220 tonnes) from a largely underground operation. Giant mine was the largest producer in the Yellowknife greenstone belt, which produced more than12 million troy ounces (~370 tonnes) in total (Bullen and Robb 2006). Arsenopyrite-bearing Au ore was roasted from 1949 to 1999 as a pretreatment for cyanidation (Fig. 3a). Poor or nonexistent emission controls in the early years resulted in the release of an estimated 20,000 tonnes of roaster-generated As_2O_3 to the surrounding environment through stack emissions (CPHA 1977; Wrye 2008). Over the lifetime of the mine, however, most of the As_2O_3 (237,000 tonnes) was stored in underground chambers (Fig. 3b) and is a now an ongoing source of As to groundwater and surface water (INAC 2007; Jamieson et al. 2013). Other roaster products include As-bearing maghemite and hematite (calcine) were deposited with tailings and re-mobilized into creek and lake sediments. Under reducing conditions, post-depositional remobilization of As associated with roaster-generated Fe oxides results in release of As to sediment pore water and reprecipitation of some As as a sulfide phase (Fawcett and Jamieson 2011). However, As(III) in maghemite and As_2O_3 persists in the oxidizing conditions of near-surface tailings and soils (Walker et al. 2005; Jamieson et al. 2013).

Ore roasting increases the solubility, toxicity, and bioaccessibility of As by converting sulfide-hosted As to oxide-hosted As. At Giant, understanding the processing history is critical to characterizing mine waste, assessing the risk to human and ecosystem health, and predicting long-term stability and optimal management. Roasting is still used worldwide today for improving recovery of refractory Au ores and for removing As from copper ores, but in modern operations As-bearing roaster products are managed as hazardous waste or sold for use in various industrial applications. Other Au mines in the Yellowknife area also roasted arsenopyrite in the early years of mining and undoubtedly have contributed to As contamination in the region. At the Con mine, the largest other Au mine in the region, managers were able to cease roasting when they encountered less refractory ores, and then converted to pressure oxidation in the late 1980's, resulting in an Fe arsenate waste product considered to be less problematic than As_2O_3 (Walker et al. 2014). Giant mine is considered the largest industrial source of bioavailable arsenic in the region (MVRB 2013).

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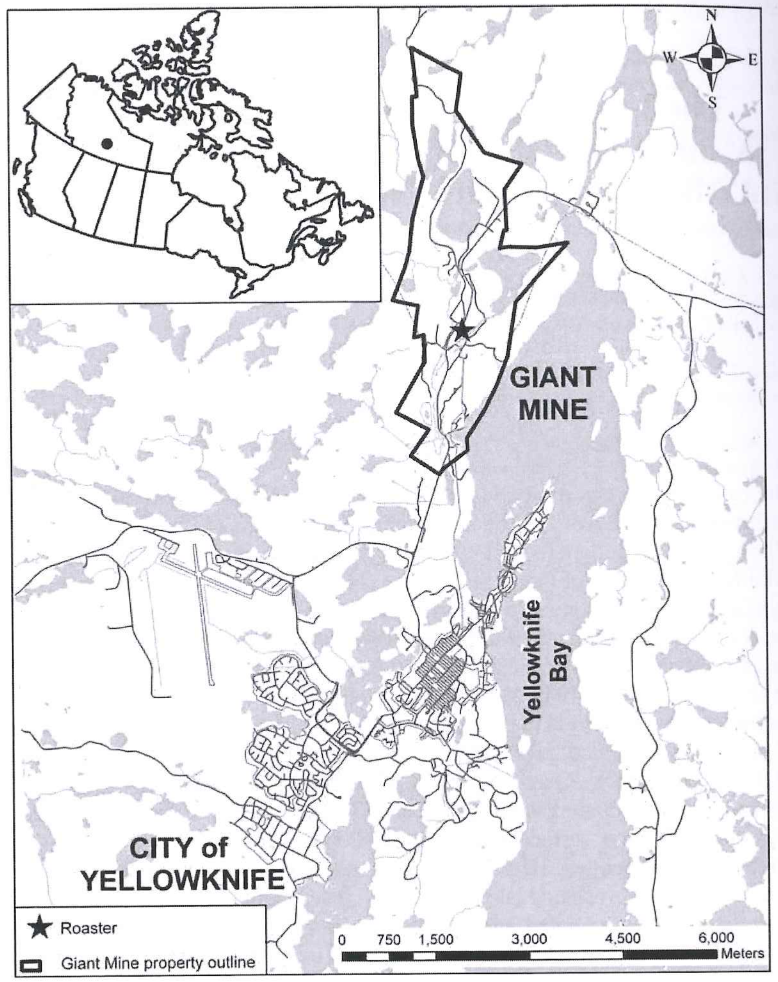

Figure 1. Map showing location of Giant mine and the City of Yellowknife, with inset showing location of Yellowknife, NWT, Canada. Solid square indicates the sample site for the data shown in Figure 4. Map is modified from Bromstad and Jamieson (2012).

The history of Giant mine is also a remarkable example of how relations between mining companies, local population, and government authorities have evolved over the last 75 years. Local Aboriginal communities were not consulted when mines were first established in the 1930's and 1940's yet were affected by removal of access to their traditional land and exposed to arsenic contamination along with other local residents (Sandlos and Keeling 2012). A Dene child died of arsenic poisoning in 1951 in an area near the mine. Pollution controls were put in place following this incident but emissions and spills of As persisted. Giant was operated by a series of private mining companies for fifty years but was placed into receivership in April 1999, and after no purchasers were identified by the receiver, ownership of the mine reverted to the Crown (federal government). Giant mine is now considered an abandoned mine and the environmental liability is public. Modern mining operations in the Northwest Territories require multi-stage permitting and extensive community consultation. The City of Yellowknife, the capital city and home for half the population of the Northwest Territories, developed as a

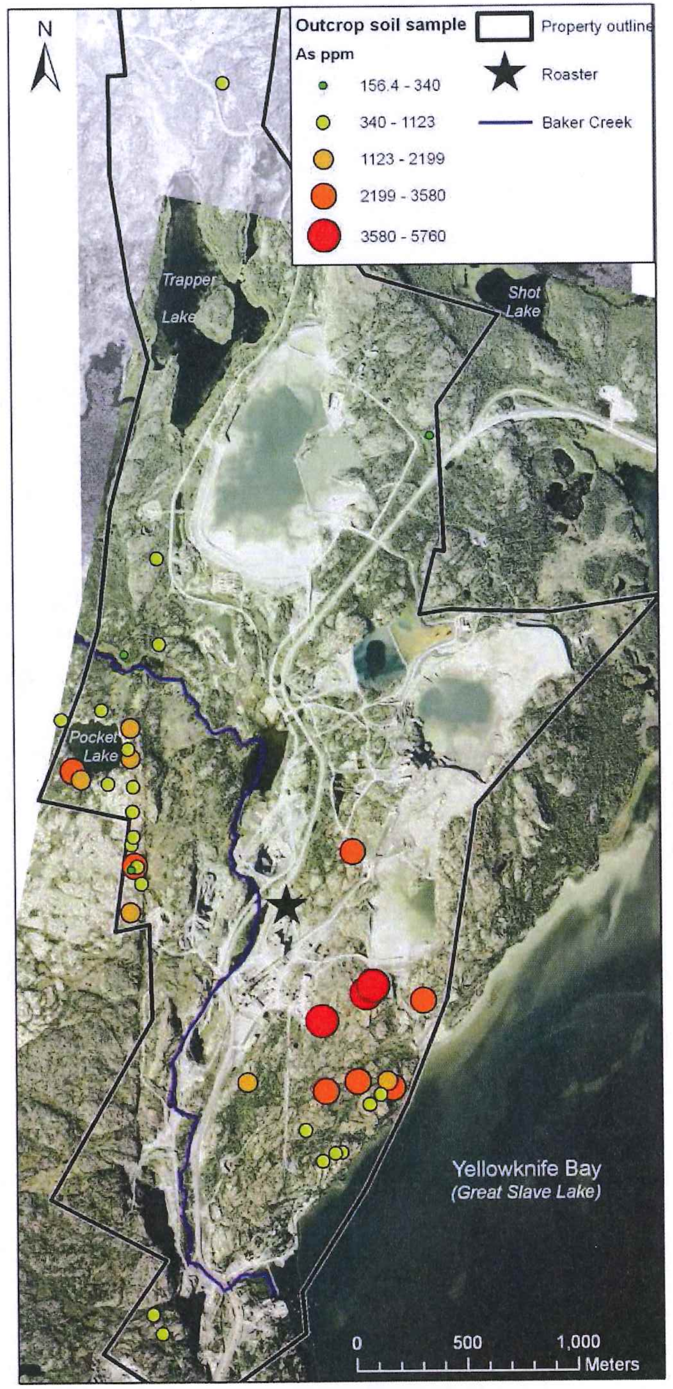

Figure 2. (*for color see Plate 10*) Air photo of the Giant Mine property ca. 2000. Data show total As concentration in outcrop soil samples from Bromstad (2011). The industrial remediation guideline for Giant mine NWT is 340 mg kg^{-1}.

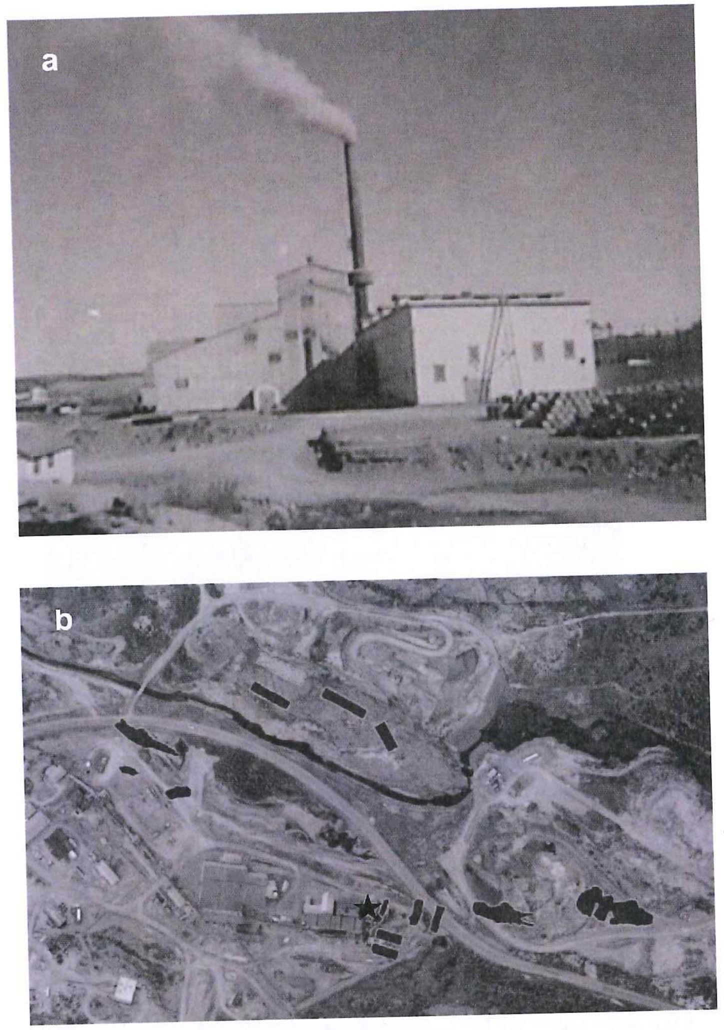

Figure 3. (a) Ore roaster at Giant mine operating in 1950's. Courtesy GNWT Archives (b) Solid black areas show location of underground storage chambers for As_2O_3 dust. Roaster location indicated by star. [Used with permission of the Giant Mine Remediation Team from MVRB (2013).]

center of Au mining. In the last fifteen years, the economy of the region has been fuelled by diamond mining, a sector operating with environmental controls and community participation that would have been unimaginable in the early days of the Giant mine.

The objective of this chapter is to report the lessons learned from research on the history and nature of As contamination at the Giant mine site. The focus is on the geochemical and mineralogical speciation of As in tailings, soils, sediments and pore water, and the long-term stability of these materials. Previous work has documented the concentration and distribution of As in these media (INAC 2007; Bromstad and Jamieson 2012). Factors that resulted in this widespread and complex contamination are discussed in the hope that modern mining will avoid such potentially damaging and costly problems and that risk assessment of other abandoned sites will benefit from this example.

FACTORS INFLUENCING THE LEGACY OF ARSENIC CONTAMINATION AT GIANT MINE

Nature of mineralization

The predominance of As and Sb as elements of environmental concern, the absence of acidic drainage, and the requirement of oxidative pretreatment of the ore are all factors resulting from the nature of the Au ore at Giant, and thus may be applicable to other deposits of this type. Gold mineralization at Giant mine occurs within shear zones as disseminated sulfides in broad, silicified zones or quartz–carbonate veins bounded by muscovite or chlorite schist (Boyle 1961; Siddorn et al. 2006). The deposit lies within the Yellowknife Supergroup, part of the Slave Structural Province of the Canadian Shield. The Yellowknife Supergroup consists of Archaean metavolcanic and metasedimentary rocks intruded by younger granitoids. Several early Proterozoic gabbro and diabase dikes crosscut the area, and several faults divide the volcanic and granitoid rock units (Siddorn et al. 2006). The eastern half of the Giant Mine property lies principally over the Kam Group of the Yellowknife Bay Formation, which consists of variolitic pillowed and massive flows dominated by basalts and metamorphosed to greenschist facies. Ore shoots typically contain less than 10% sulfide and commonly less than 5% (Coleman 1957). The sulfide and sulfosalt mineralogy at the Giant mine includes (in decreasing order of abundance): pyrite (average As content 0.68 wt%), arsenopyrite, sphalerite, chalcopyrite, sulfosalts (jamesonite, berthierite, bournonite and tetrahedrite), pyrrhotite, and galena. However, the actual abundance is variable within individual ore shoots and different parts of the mine (Coleman 1957; Canam 2006; Hubbard et al. 2006; Walker et al. 2014). A study of flotation concentrate in 1990 (originating from stopes being mined at the time) indicated that pyrite and arsenopyrite combined account for 95% of the sulfides and that marcasite, chalcopyrite, sphalerite, acanthite, boulangerite, tetrahedrite, berthierite, gudmundite, stibnite were present in trace amounts (Walker et al. 2014).

Giant falls into the deposit type known as orogenic Au deposits, or lode Au deposits, which are typically associated with quartz-carbonate veins and have relatively low sulfide content (Groves et al. 1998; Seal and Hammarstrom 2003; Siddorn et al. 2006). Most of these ores have associated As and Sb in sufficiently high concentrations that these elements are enriched in tailings and waste rock and are potentially leached into groundwater and surface water. Other elements such as Cu, Pb, and Zn are present in flotation tailings at Giant, as would be predicted from the ore mineralogy described above. As shown in Figure 4, these elements have been remobilized into sediments (Fawcett 2009; Andrade et al. 2010). Although the concentrations of these other elements may, in some cases, exceed guidelines for sediment and soil quality, the focus of the risk assessment and remediation at Giant has been on As (MVRB 2013).

The concentration of As in tailings at Giant (1,000 to 5,000 mg kg^{-1}) is governed by the nature of the mineralization and modifications related to processing methods. Pyrite and arsenopyrite are the dominant sulfide minerals but overall sulfide concentrations in tailings are relatively low (total S typically <1%; INAC 2007). Furthermore, much of the As was diverted from the tailings by ore roasting, and is now present as As_2O_3 in underground storage chambers and as airfall deposits in soils and sediments in the area (Wrye 2008; Bromstad 2011). In comparison, historic tailings from orogenic Au deposits in Nova Scotia contain a wider range of total As concentrations (10 mg kg^{-1} to 31 wt%; median 2,550 mg kg^{-1}; Parsons et al. 2012), reflecting the dominance of arsenopyrite and the highly variable sulfide content in the ore veins (0.1 to 5%, locally 50 to 75%; Kontak and Jackson 1999). The extremely high As content of some of the Nova Scotia tailings is associated with the disposal of arsenopyrite-rich gravity concentrates (Walker et al. 2009). Overall, this type of Au deposit is generally low in total sulfide content with arsenopyrite and/or pyrite as the dominant sulfide minerals (Groves et al. 1998).

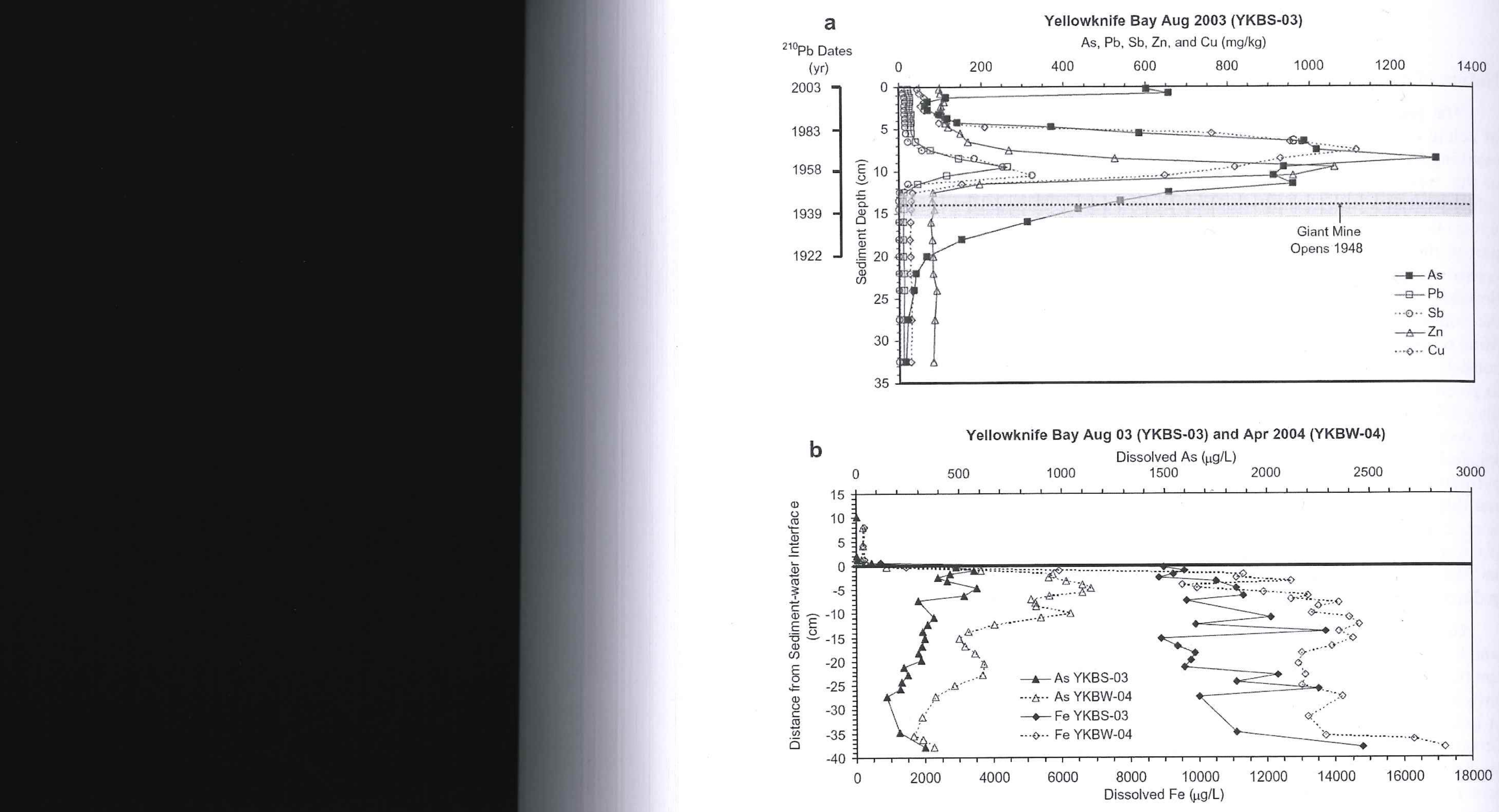

Figure 4. (a) Solid phase metal(loid) concentrations from sediment core with associated timeline. (b) Dissolved As and Fe in sediment pore waters. Location of the sample site in Yellowknife Bay is shown by solid square in Figure 1. Water depth at this location is 13 m. {Reprinted with permission of Elsevier from Andrade et al. (2010), *App Geochem*, Vol 25, Fig, 5 p. 204.]

Critical to the environmental legacy at any mine is the ratio of neutralizing carbonate minerals to acid-generating iron-sulfide minerals because this governs the tendency of tailings and waste rock to produce acid leachate (Bowell et al. 2000; Seal and Hammarstrom 2003). Groves et al. (1998) described orogenic Au deposits as typically having 3-5% sulfide minerals, mainly Fe-sulfides, and 5-15% carbonate minerals. Kontak and Jackson (1999) report generally subequal modal amounts of sulfide and carbonate in the auriferous veins from Nova Scotia. This suggests that acid rock drainage may or may not be associated with mine waste from these deposits. Measured pH values associated with streams in the Nova Scotia Au mining districts are typically greater than 4 and often near-neutral, suggesting effective carbonate buffering of pH (Parsons et al. 2012). However, acidic pore waters are found in the hardpans produced from weathering of arsenopyrite-rich gravity concentrates (DeSisto et al. 2011).

At Giant mine, a significant excess of carbonate, mostly ferroan dolomite, relative to Fe sulfide (pyrite and arsenopyrite) in the tailings precludes the formation of acidic drainage. Acid-base accounting tests demonstrated high neutralization potential (111 to 234 tonnes $CaCO_3$ equivalent per 1000 tonnes of tailings) and low acid generation potential (2.5 to 32.8 tonnes $CaCO_3$ equivalent per 1000 tonnes of tailings) (INAC 2007). Tailings pore waters and streams on the mine site have pH values consistently between 5.5 and 8.5 (INAC 2007; Walker 2006; Fawcett et al. 2014). If the balance between acid-generating and acid-neutralizing minerals in the tailings had been such that acid leachate was produced, it is likely that the drainage would have contained higher concentration of Cu, Zn, Pb, Ni and other metals present in sulfide and sulfosalt minerals vulnerable to acidic dissolution. Given that As and Sb are mobile as oxyanion species at near-neutral pH and tend to adsorb at low pH (Wilson et al. 2010), it is possible that lower pH leachate would have produced lower concentrations of these metalloids. Remnant pyrite and arsenopyrite grains in the Giant mine tailings and waste rock exhibit rims of As-bearing secondary Fe oxyhydroxides, suggesting that sulfide oxidation occurred during weathering. The net neutralizing nature of the mine waste is inherited from the original, carbonate-rich mineralogy and enhanced by sulfide removal during processing. The relatively low ore-roasting temperatures used at Giant (ca. 500 °C) destroyed much of the arsenopyrite but did not affect the ferroan dolomite or calcite (Walker et al. 2014).

The third influence of the nature of the mineralization on the environmental legacy at Giant is that it dictated the choice for ore processing. Some free-milling Au, which is usually defined as ore where >95% Au recovery can be achieved with simple cyanidation or gravity separation (Marsden and House 2006), was present in parts of the ore body exploited early in mining (Tait 1961), but it was soon realized that most of the Au was refractory, incorporated submicroscopically within arsenopyrite, and thus required pre-treatment (Halverson 1990; Stefanski and Halverson 1992; Walker et al. 2014). The presence of Sb in the ore complicated the roasting process because Sb concentrations exceeding 0.75% may cause clinkering (formation of glassy phases not amenable to cyanidation) of the roaster bed (Marsden and House 2006; Fawcett and Jamieson 2011). The remoteness of the site precluded shipping a concentrate to another facility. Ore roasting was likely the obvious choice for Au recovery from the Giant mine ore in 1949. Coupled with the lack of emission controls and regulations, roasting led to the dispersion of large amounts of As_2O_3 through stack emissions and the accumulation of even larger amounts of As_2O_3 in underground storage chambers.

Ore roasting

The influence of ore roasting on the environmental legacy at Giant is a direct consequence of the nature of the As-hosting solid species in the tailings, stored roaster waste, soils, and sediments in the area. Processing converted most of the original arsenopyrite to As-bearing roaster products: Fe oxides (maghemite and hematite) and As_2O_3 (arsenolite). These phases are potentially more soluble and more bioaccessible than the original arsenopyrite, and are vulnerable to reductive dissolution.

The As gases produced during roasting of arsenopyrite condensed to As_2O_3 with a crystallographic structure similar to arsenolite (Wrye 2008; Bromstad 2011; Walker et al. 2014). Because the As_2O_3 at Giant is an industrial product, in contrast to that produced by natural weathering of As-rich sulfides (Nordstrom and Archer 2003; Drahota and Filippi 2009), it is referred to as As_2O_3. The solubility of arsenic trioxide-bearing dust from the Giant mine site has been reported as 11,000 to 15,000 mg L^{-1}, based on laboratory measurements of samples from Giant (Riveros et al. 2000); the As solubility may be limited by Sb content, but this is not well documented. Seepage from underground drillholes and fractures near the storage chambers contain up to 4,000 mg As L^{-1} in pH-neutral water (INAC 2007; Jamieson et al. 2013). Approximately 70% of the As is As(III). Near these seepage points, and on stope walls where low-flow drips from old drill holes and fractures produce As(III)-rich water, biofilms develop (Fig. 5). These contain psychrophilic As(III)-oxidizing bacteria (Osborn et al. 2010). As discussed by Bowell and Craw (2014, this volume), As(III) oxidation to As(V) is an acid-generating reaction. In this case, carbonate in the wallrock and concrete in the bulkheads of the storage chambers neutralizes the water, and yukonite precipitates as a product of the reaction (Fig. 5).

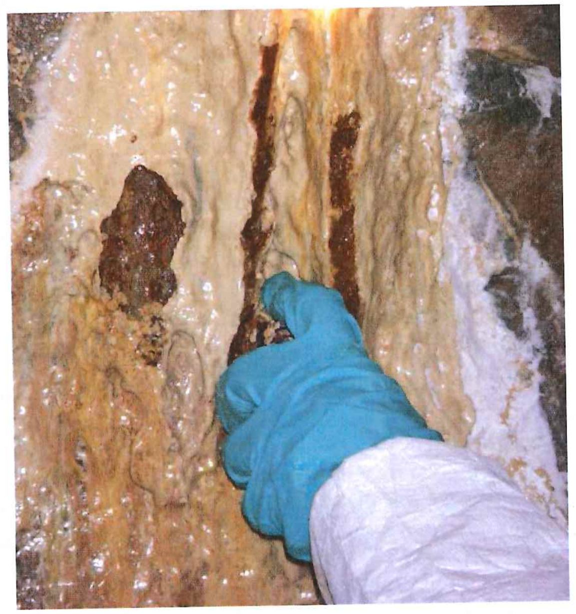

Figure 5. (*for color see Plate 11*) Biofilm developed on underground stope wall where As-rich water drips from an drillhole. Mineral precipitate is yukonite $Ca_7Fe_{12}(AsO_4)_{10}(OH)_{20}\cdot 15H_2O$.

Walker et al. (2005, 2014) have described the As-hosting roaster-generated Fe oxides in detail. They are nanocrystalline composite grains, predominantly maghemite (γ-Fe_2O_3) with a smaller amount of hematite (Fig. 6). Arsenic is incorporated during roasting in a mixed oxidation state with the ratio of As(III)/As(V) determined by the partial pressure of O_2 at the reacting surface during combustion of the sulfide (i.e. lower P_{O2}, higher As(III)). Gas phase As chemisorbs to the maghemite surface. The As concentration of maghemite grains in the tailings range from <0.5 to 7 wt% As. Where further transformation of maghemite to hematite (α-Fe_2O_3) occurs, the transformation is accompanied by rapid crystallite growth, a decrease in surface area, and concurrent decrease in the As content (<2 wt%).

Two types of roasted material were collected and treated with cyanide for Au recovery, electrostatic precipitator (ESP) dust, and calcine (roaster bed product). Fawcett and Jamieson (2011) have shown that in the ESP waste stream, As(III) and Sb(III) are dominant in the bulk sample and in the roaster-generated iron oxides that host these elements, but As(V) and Sb (V) are dominant in the bulk calcine sample and in the roaster Fe-oxide grains therein. As explained in the next section, both ESP and calcine waste streams were combined with flotation tails, resulting in a mixture of oxidation states and mineralogical hosts of As in tailings deposits.

The most important consequence of ore roasting at Giant, in terms of the environmental legacy, is the production of approximately 300,000 tonnes of As_2O_3 as waste during a time of few emission controls, few regulations, and limited options regarding re-use of the As_2O_3. In many cases, mining operations eliminated or reduced the amount of As_2O_3 they need to store

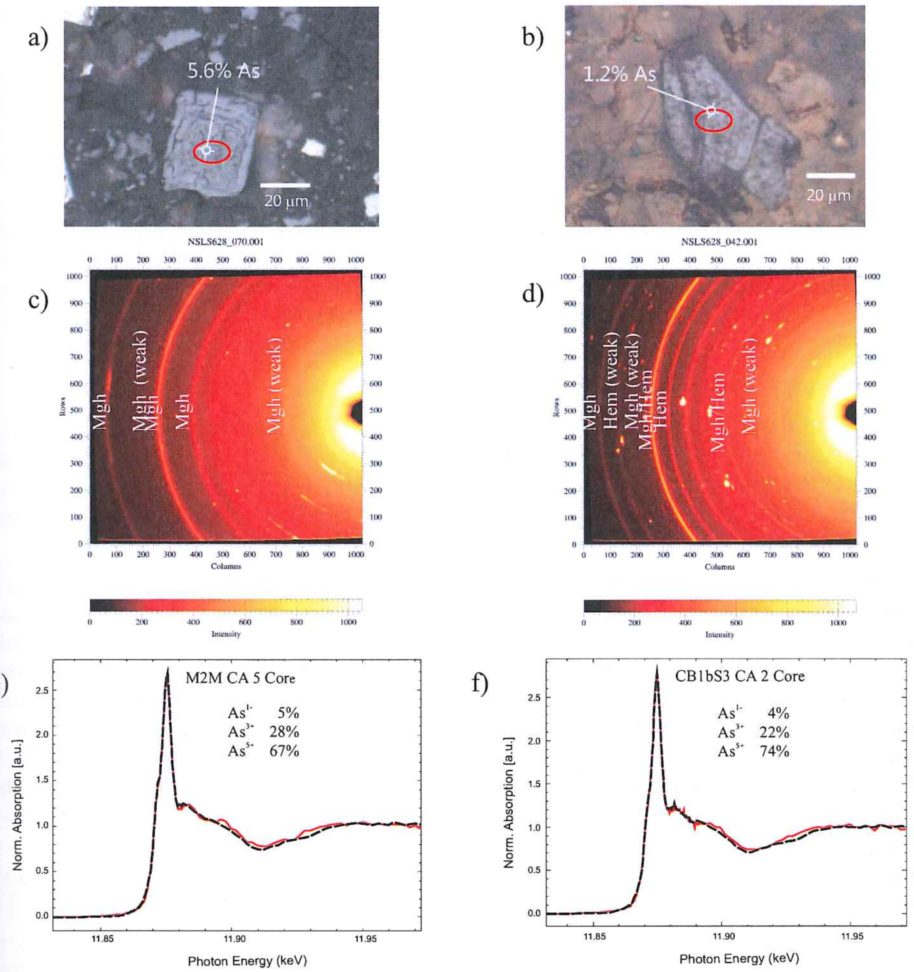

Figure 6. (*for color see Plate 13*) Selected analyses of two grains of roaster iron oxides. a) Reflected-light photomicrograph of square concentric roaster iron oxide from calcine sample (M2M). Total As established by EPMA (designated in white). b) Transmitted- and reflected light photomicrograph of target grain from shoreline tailings sample (CB1bS3). Total As by EPMA as indicated. c) micro-XRD image of target indicated by ellipse in (a). Pattern corresponds to maghemite. Three arcs in lower right-hand corner are chlorite reflections. d) Micro-XRD image of target indicated by ellipse in (b). Pattern is a mixture of maghemite and hematite. e) Micro-XANES analysis of target in (a). Sample spectrum is lighter undashed line, dashed line is best-fit linear combination for result shown. f) Micro-XANES analysis of target in (b). [Reprinted with permission of the Mineralogical Association of Canada, from Walker et al. (2005) *Can Mineral*, Vol. 43, Fig. 8. p. 1218.]

by selling a high-purity version for use in herbicides, insecticides, or the arsenic acid used in the formulation of chromated copper arsenate (CCA) preservatives for the pressure treating of lumber, all products used during most of the years that the Giant mine operated. Other operations, however, also stored arsenic-rich roaster waste. Across town, the Con mine stored their As_2O_3, considerably less in volume, on surface. The Campbell mine in NW Ontario, which, like Giant, has operated continuously since 1949, has 20,000 tonnes of As_2O_3 stored underground.

The remote location of Giant mine precluded effective marketing and sale of As_2O_3. Attempts were made to develop markets for As_2O_3 in the USA and South America although re-mining the storage chambers would have been challenging. As late as 2002, one of the alternative solutions for the As_2O_3 stored underground at Giant was extraction, treatment, and sale of high-purity As_2O_3 to American wood preservative manufacturers. This alternative was subsequently dropped from consideration because changes to U.S. regulations severely restricted the use of arsenic as a wood preservative, and it was considered unlikely that a future market would exist for the Giant mine As_2O_3 (INAC 2002). Thus, a potential resource became an environmental liability.

Modern alternatives for oxidative pre-treatment include hydrometallurgical processes such as low- or high-pressure oxidation, nitric acid treatment, chlorination, and bio-oxidation (Marsden and House 2006). Roasters are still effective in terms of recovery and cost in many cases, and are still used. However appropriate emission controls limit dispersion of As_2O_3 and regulations require disposal in an environmentally acceptable manner.

Waste disposal practices

The distribution of As-bearing waste material is an important factor regarding the environmental impact of Giant mine operations. In the first two years of operation, mill tailings were discharged directly onto the shore of Yellowknife Bay (Fig. 2). These shoreline tailings were stabilized with rip-rap in 2002, but much of the material had already been eroded and dispersed into the bay (Andrade et al. 2010).

Beginning in 1951, tailings were deposited into several former lakes on mine property. Later, tailings dams and several impoundments were constructed (Fig. 2). The processing produced three solid waste streams including flotation tails, cyanided calcine residue, and cyanided ESP dust. Table 1 shows that the lower tonnage streams (calcine and ESP) carry high concentrations of As as fine-grained particles, mostly roaster-generated maghemite and hematite, and possibly some As_2O_3 (Walker et al. 2005, 2014). The total As concentration in tailings ranges from 1,100 to 5,000 mg kg^{-1}, with an average approximately 2,700 mg kg^{-1}. Petrographic examination and electron microprobe examination of the tailings indicate the presence of unroasted arsenopyrite in addition to roaster-generated iron oxide (Walker 2006). Pyrite exhibits oxidation rims of Ca-Fe arsenate (yukonite). The combination of sulfide-hosted and oxide-hosted arsenic requires a carefully-designed cover that will maintain the stability of both. If a thick cover containing organic material is used, there is a risk that oxide-hosted As, particularly roaster-generated Fe oxides, could destabilize and release As, as described from other Au-mining areas (e.g., Martin and Pederson 2002).

Table 1. Arsenic contributions from the main solid effluent streams at Giant.

Tailings Stream	Year†	Approximate Discharge Rate (tpd)	Arsenic Concentration (wt%)	Arsenic Loading (tpd)
Flotation (70-80% <0.075 mm)	1999	1000	0.09	0.9
	1963	794	0.28	2.2*
Calcine Residue (90% <0.045 mm)	1999	170	2.5	4.2
	1963	122	1.2	1.5*
ESP Residue (90% <0.014 mm)	1999	9	4.4	0.4
	1963	9	6.2	0.6

tpd = dry tonnes per day.

† For 1999, discharge rate extrapolated from Royal Oak Mines (1992) and As concentration from Walker et al. (2014). For 1963, data from Grainge (1963).

* Estimated loading. Actual loading to tailings is unknown, since some tailings were cycloned for mine-backfill from 1957 to 1976 removing approximately 50% of the coarsest material and an unknown proportion of As.

Prior to sedimentation control, a significant amount of fine tailings, including ESP dust and calcine, flowed over the ice-covered tailings dams each spring and was deposited at the upstream end of Baker Pond, a natural water body located along Baker Creek (Bérubé et al. 1974). Some fine-grained material travelled down Baker Creek into Yellowknife Bay (Mudroch et al. 1989; Fawcett and Jamieson 2011). A breakwater that extends across most of the mouth of Baker Creek was built in 1964 (Fig. 2). Air photos show that this resulted in the capture of fine sediment behind the breakwater, which is covered by dense aquatic vegetation (*Equisetum fluviatile*), and the establishment of a narrow creek channel (Fawcett and Jamieson 2011). Baker Creek was also impacted by numerous tailings spills originating from faulty tailings pipelines (Fawcett 2009).

Giant mine tailings and fine-grained tailings fractions are now present in both subaerial and subaqueous settings, as both near-surface and buried sediment. Arsenic is mostly in the form of As(III) and As(V) associated with maghemite and lesser amounts of hematite. In some cases, the As appears to be hosted in stable form, but in other cases the As-hosting phases are unstable or soluble, and are likely to release As to groundwater and surface water. Walker et al. (2005) used microXANES to show that the ratio of As(III)/As(V) is similar in maghemite grains collected directly from the roaster in 1999 and grains exposed subaerially in shoreline tailings for 60 years. The lack of oxidation of the As(III) over time in these phases requires an explanation, because surface-sorbed $(As^{III}O_3)^{-3}$ might be expected to oxidize to $(As^{V}O_4)^{-3}$. More work is needed to understand the kinetics of As oxidation in this environment. Walker et al. (2014) suggested that the most plausible reason is that some of the As(III) is incorporated within the maghemite framework associated with structural vacancies. Andrade (2006) sampled shoreline tailings that had been eroded and redeposited at 1 m depth in Yellowknife Bay. The mixed oxidation state of As in the roaster-generated iron oxides was similar to that on the onshore tailings, indicating that these materials are stable in the oxygenated, high-energy, organic-poor, near-shore environment (Andrade 2006).

There is clear evidence of the release of As from buried tailings in sediment in Baker Pond, in the vegetated area behind the Baker Creek breakwater, and at depth in Yellowknife Bay (Andrade 2006; Fawcett et al. 2014). Figure 4 shows post-depositional mobility and upward migration of pore-water As from the mid-core enrichment zone from Yellowknife Bay sediment, which is related to high-As releases in the early years of operation, likely due to spills, tailings pond decanting, and possibly elevated stack emissions. These results are from the deep-water (13 m) site, approximately 1 km from the mouth of Baker Creek (Fig. 2). Andrade et al. (2010) described this release of As from contaminated sediments, which are suboxic, relatively rich in organic material, and contain arsenate- and sulfate-reducing microbes. Some of the upwardly-migrating pore-water As is recaptured at the sediment-water interface by a thin layer of Fe-Mn oxyhydroxides. Post-depositional mobilization of As is also apparent in sediments from Baker Pond and Baker Creek channel. Roaster-generated maghemite exhibits lower ratios of As(III)/As(V) (and Sb(III)/Sb(V)) in sediments at 10 to 20 cm depth compared to near-surface material, suggesting either *in situ* reduction of the metalloid or preferential release of As(V). At depth, approximately 10 to 20% of the arsenic released to pore water in these locations is attenuated via precipitation as sulfide (realgar) or sorption to sulfide surfaces (Fawcett and Jamieson 2011).

Stack emissions of As_2O_3 have left an important environmental legacy at Giant. Table 2 shows the daily emissions rate, which decreased from more than 7 to 0.1 tonnes per day from 1949 to 1990, as control measures such as ESPs were installed. By the time the emission-control process was completely refined in 1963, 86% of the total airborne As emissions at Giant had been released into the surrounding area. Stack emissions consisted of SO_2 and

As-bearing gases, both produced by roasting of arsenopyrite. The As gases condensed into a dust with As-hosting phases consisting mostly of As_2O_3 in the mineral structure of arsenolite, with some As-bearing, roaster-generated Fe oxides in the form of maghemite (Bromstad and Jamieson 2012). Wrye (2008) and Bromstad (2011) have shown that particles of aerially distributed As_2O_3 persist in the near-surface soils over much of the Giant mine property. Figure 7 shows examples of As_2O_3 and the rarer roaster-generated Fe oxide particles, which are clearly visible in samples examined by scanning electron microscopy and identified by synchrotron-based microXRD.

There is little visual evidence of dissolution or instability of the As_2O_3 particles in the soils near Giant mine. Arsenic from stack emissions appears to have concentrated in soils that are found in the small pockets of soil downwind of the roaster on the large outcrops that cover approximately 30% of the mine property (Fig. 2). Outcrop depressions are known sinks for runoff after rainfall and freshet at Giant mine, trapping water and promoting evaporation (Spence and Woo 2002), a feature that may be important at other smelter and roaster sites. Most of the outcrop soil pockets were not included in the delineation of soils destined for removal in the remediation plan, although some of these areas may become publicly accessible. Total As concentrations in soils sampled by Wrye (2008) and Bromstad (2011) range up to 5,760 mg kg^{-1}. The highest values are similar to concentrations in the tailings, demonstrating that the ore roaster effectively diverted As from the tailings to the soil and water in the surrounding environment, particularly in the early days of operation. Based on As-Sb-Au soil concentrations and documented improvements in recovering Au and Sb from roaster emissions,

Table 2. Estimates of Aerial Emissions of As_2O_3 from the Giant Mine Roaster between 1949 and 1999. (tpd = tonnes per day).

Year	**As_2O_3 Emissions † (tpd)**	**As_2O_3 Emissions‡ (tonnes)**
1949-1951*	7.3	7963.6
1952*	5.5	1989.3
1953*	5.5	1989.3
1954	5.5	1990.9
1955	2.9	1061.8
1956	2.7	995.5
1957	3.0	1078.4
1958*	1.5	547.5
1959	0.1	19.1
1960	0.1	27.4
1961-1963*	0.2	163.3
1964	0.3	114.5
1965	0.0	0.0
1966	0.2	88.8
1967	0.1	47.3
1968	0.2	83.0
1969	0.3	109.5
1970	0.2	80.5
1971	0.9	320.2
1972	0.4	145.2
1973	0.4	147.7
1974	0.2	80.5
1975-1992*	0.3	1698.9
1991-1993*	0.1	54.8
1994	0.01	3.6
1995	0.01	3.5
1996	0.01	3.5
1997	0.02	7.6
1998	0.015	5.3
1999	0.01	3.7
Total As_2O_3 Emissions (tonnes)		**20,824**

† As_2O_3 emissions estimates were measured from the top of the roasting stack and were reported by following sources; CPHA 1977, EnviroCan 2007, GNWT 1993, INAC 2007, Tait 1961.

* Emissions for this year or period of years have been estimated based on information from the above sources and known changes to roaster operations.

‡ As_2O_3 emissions for the year or period of years assuming roasting occurred for 365 days. This is an overestimate of emissions, however the dates when the roaster was not in operation are unknown and the majority of As_2O_3 generated was during the first 10 years of roasting.

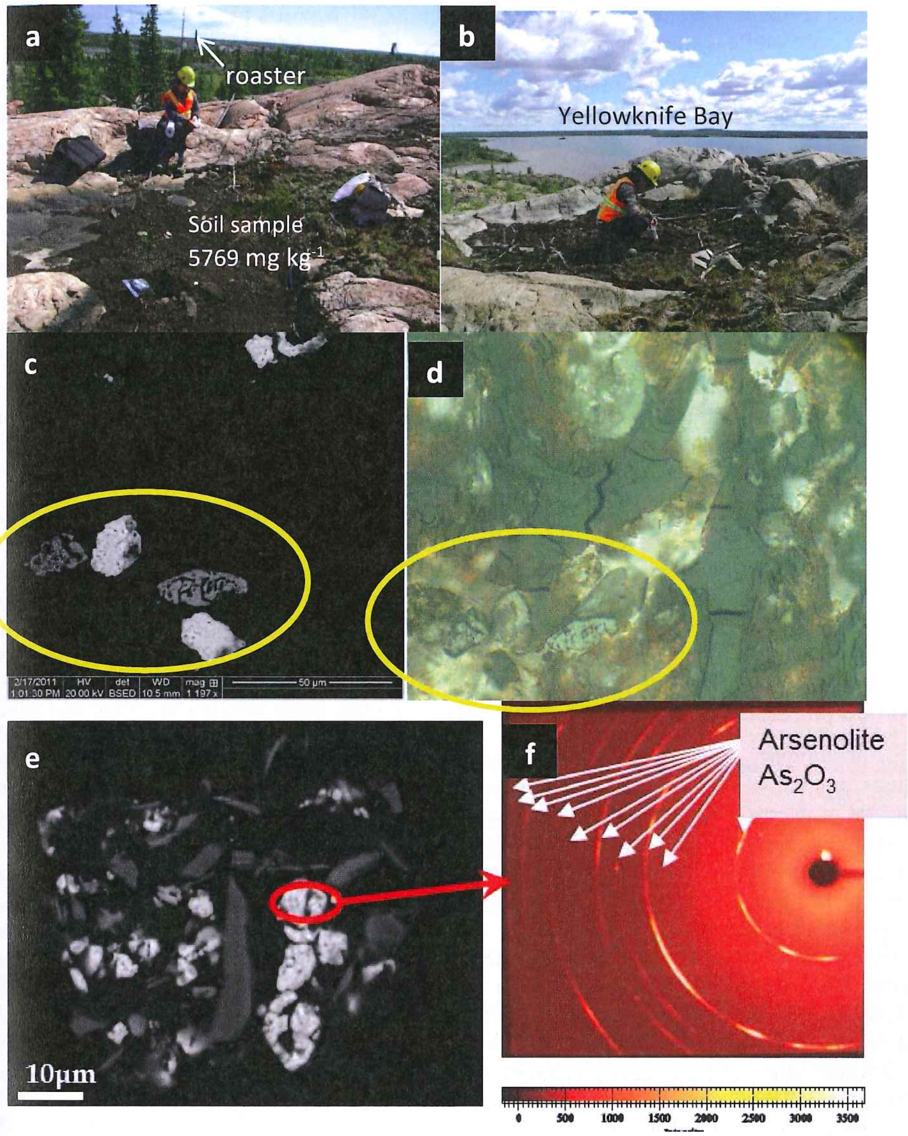

Figure 7. (*for color see Plate 14*) Roaster-derived As-bearing particles in soils near Giant mine. (a) and (b) show typical soil pockets on large outcrops, the sites that contained the highest concentration of roaster-derived soil. (c) back-scattered electron image of a thin section of soil showing light (As_2O_3) and dark (roaster-generated Fe oxide) particles. (d) petrographic image of the same area as (c), combined transmitted and reflected light. (e) As_2O_3 particles in soil (f) microXRD of one of these particles indicating the pattern corresponding to arsenolite. From Wrye (2008) and Bromstad (2011).

Bromstad (2011) concluded that most of the arsenic trioxide in the soils is more than 50 years old, persisting from early stack emissions despite its high solubility and the expectation that arsenic trioxide would have dissolved after years of soil exposure. Haffert and Craw (2008) document reaction rims of scorodite on particles of roaster-generated As_2O_3 probably caused by exposure to acidic, Fe-bearing fluids. In contrast, the As_2O_3 particles in soil near Giant do not have reactions rims, likely because they were never exposed to acid mine drainage, and exhibit only subtle textural evidence of dissolution (Bromstad 2011).

Suction lysimeters were used to sample pore water in soil pockets immediately following summer rainfalls. Soil pore-water concentrations are as high as 2 mg L^{-1} of As, considerably less than the underground seepage waters, but still more than two orders of magnitude higher than the World Health Organization drinking water guideline of 0.01 mg L^{-1}. The pore waters are pH-neutral and the major ions are Ca^{2+}, Na^+, and SO_4^{2-} (Bromstad 2011; Jamieson et al. 2013).

Giant mine has been a managed site during and since mine closure with limited public access except for the area around the outlet of Baker Creek. However, post-remediation plans call for some areas to be unfenced, and human exposure to As_2O_3-contaminated soil may be increased. The risk associated with possible ingestion of soil can be evaluated using bioaccessibility testing designed to evaluate the solubility of As in body fluids (see Basta and Jurasz 2014, this volume). Plumlee and Morman (2011) have ranked gastric-intestinal bioaccessibility of various As compounds, and include arsenic trioxide as one of the most bioaccessible forms and arsenopyrite as one of the least.

Bioaccessible As in three soil samples from a single soil pocket was measured at the US Geological Survey laboratories in Denver using *in vitro* techniques (Bromstad 2011). A simulated gastric fluid was used for the size fraction less than 250 μm and indicated that 29 to 42% of the total As (238 to 4,760 mg kg^{-1}) would be bioaccessible if ingested. A second series of tests using a simulated lung fluid to leach the size fractions less than 20 μm indicated that 15 to 20% would be bioaccessible if inhaled. In both cases, there is no correlation between the percentage of bioaccessible As and the total concentration of As, suggesting homogeneity in As host properties (Bromstad 2011).

In summary, three key factors—the nature of the mineralization, the ore-roasting procedures, and the waste-disposal practices—were influential in creating the complicated environmental problems near the Giant mine. The nature of the mineralization resulted in high concentrations of As in various wastes, abundant neutralizing capacity limiting acid drainage and most importantly, arsenopyrite-hosted refractory Au, which required oxidative pre-treatment for Au recovery. The choice of ore roasting created oxide-hosted arsenic (As_2O_3 and roaster-generated maghemite and hematite) in mill tailings and other waste emissions, materials which are more soluble, more bioaccessible, and vulnerable to reductive dissolution compared with sulfide-hosted As which is present at other mine-waste sites. Finally, the practice of co-depositing fine-grained roaster waste with flotation tailings, and the poorly-controlled release of As_2O_3 from stack emissions resulted in widespread distribution of As in soils, sediment, and water. Much of the roaster-generated As(III) has persisted under oxidizing conditions but shows evidence of post-depositional transformation under reducing conditions.

Modern mining and processing practices can limit As contamination by understanding that this type of Au deposit is typically accompanied by metalloids (As and Sb) that are mobile in neutral-pH water, employing alternative methods of oxidative pre-treatment of refractory ore that do not produce As_2O_3, or using modern ore roasting with efficient emission controls, and employing appropriate tailings and processing waste methods and monitoring. The story of Giant mine also provides some lessons for remediation design of older and abandoned As-contaminated mine sites, namely the importance of understanding processing and disposal history, distinguishing between natural and anthropogenic As, and recognition of post-depositional As mobility.

REMEDIATION PLAN

A comprehensive remediation plan, based on approximately five years of work, was filed for environmental assessment in 2007 (INAC 2007). The "developer" of the site is identified as the Giant Mine Remediation Directorate, which involves both the federal and territorial governments. As a result, the entire submission, including approximately 50 consultants' reports, is available to researchers. The Mackenzie Valley Environmental Impact Review Board, which is the regulatory body in the Northwest Territories, responded in June 2013 to the proposed remediation plan (MVRB 2013). The complexity of the site, the amount of technical information gathered, the requirement to involve other parties, notably the local First Nations communities and NGOs (Non-Governmental Organizations), and the nature of the environmental assessment process have all contributed to the long timeline and large volume of documentation. At the time of writing of this chapter, the Minister of Aboriginal Affairs and Northern Development Canada has provided preliminary response to the regulator's suggested modifications to the remediation plan. Very limited remediation has taken place to date, although some buildings, including the roaster, are being dismantled, and a highway bypass designed to avoid the area overlying the arsenic trioxide storage chambers has been constructed.

Relevant to the consideration of other sites where roaster waste has accumulated are the alternatives considered for the 237,000 tonnes of As_2O_3 stored in underground chambers. More than 50 possible techniques for reducing the risk associated with these chambers were considered, with the top two being *in situ* freezing or removal and encapsulation in cement or bitumen. The selected alternative is the frozen-block method which involves pumping coolant underground and then maintaining frozen ground using thermosyphons, filled with compressed carbon dioxide, as passive heat pumps (INAC 2007). This technology is used commonly in the Canadian north for ice core dams and other applications. Yellowknife is located in an area of discontinuous permafrost, but the large underground opening and subsequent partial flooding of the underground workings has all but eliminated permafrost in the mine area. The remediation plan considered the effects of global warming and concluded that, even after 100 years of sustained temperature increases based on estimates from the ICCP (International Climate Change Partnership), the thermosyphon installation should be adequate. There has been considerable discussion about other aspects of climate change, including the risk of flooding and potential effects on both the frozen chambers and other features on site.

The remediation plan calls for removal or covering of approximately 960,000 m^3 of contaminated soils to industrial site-specific standard of 340 mg kg^{-1} total As which was established by the City of Yellowknife in 2003 (GNWT 2003). A residential guideline of 160 mg kg^{-1} was established at the same time. These criteria are much higher than the Canadian Soil Quality Guideline of 12 mg kg^{-1} (CCME 1997), and were based on the expectation that exposure would be limited by the cold climate and that the "average natural background concentration of arsenic in and around Yellowknife was determined to be 150 ppm." The industrial guideline was also based on the expectation of "little or no public access." Given more recent evidence that the roaster stack emissions have introduced widespread contamination by As_2O_3, and the construction of a highway bypass adjacent to some of the highly contaminated outcrop soil pockets, the evaluation of natural background and future public access may need to be re-examined.

The remediation plan indicates that monitoring, maintenance, and management is designed for perpetuity. This perpetual aspect has significance for funding, the emergence of new technology that might be better than the frozen-block method, climate change (including global warming or glaciation), possible seismic activity, and community anxiety. Given that the funding source for perpetual care would be the Canadian federal government, the source of funding for perpetuity is likely more reliable than at privately-owned sites. Kempton et al. (2010) have described perpetual environmental management at hardrock mines as a global dilemma and noted that, while perpetual treatment is usually less expensive than permanent stabilization, it results in increased social and financial complexity. An analysis of perpetual care at contaminated sites by Kuyek (2011) highlighted the need for "public engagement and activism in order to create the political will for the enormous funding that is required" to maintain funding.

The remediation plan proposes collection and removal of As from surface and groundwater that would reduce the loading of As to Yellowknife Bay, part of Great Slave Lake. The contaminated water would be treated on site and released to Yellowknife Bay via a diffuser system. This would allow year-around release of treated water instead of the current situation which requires winter storage of water in tailings ponds, increasing the possibility of flooding and uncontrolled release of untreated water.

One of the important lessons learned from Giant mine concerns the limitations of remediation. The As released from the site will decrease from the current level of approximately 900 kilograms per year to less than 610 kg per year (MVRB 2013). This may be disappointing to community members in Yellowknife, and in other areas impacted by historic mining activities, who expect that remediation means a complete "clean-up" of contamination. The Giant plan is essentially a control measure because without any remediation, various failures of infrastructure and natural events could results in releases in thousands of kg of As per year.

Although the estimated cost of the proposed remediation is very high ($CA 903,535,080, as reported by AANDC 2012), it is exceeded by the monetary value of the Au extracted over the lifetime of Giant Mine. Bullen and Robb (2006) have determined that the cumulative revenue for Giant and two other nearby mines exceeded $CA 5.5 billion, with the contribution from Giant estimated at $CA 2.75 billion, recalculated in terms of 2006 dollars. In their analysis, Bullen and Robb (2006) described the impact of the revenues on the Yellowknife economy as money flowing to employees and local businesses. Unlike many mining districts, the pattern of revenue flow to the NWT gross domestic product did not follow a boom-and-bust cycle because of the staggered timing of the two large mines (Con and Giant) and the historical change in Au price. Thus the Au mines provided a sustained economic benefit that launched the city of Yellowknife, led to its establishment as the capital of the NWT in 1967, and lasted until the diamond mines provided a new source of mineral wealth.

Local First Nations were largely excluded from these direct economic benefits during the operating years of the Au mines, although a few individuals found employment in the mining industry. Indirect benefits to First Nations included improved access to health care and transportation infrastructure (Bullen and Robb 2006; Sandlos and Keeling 2012). In general, First Nations have opposed Au mining in the Yellowknife area (Bullen and Robb 2006) and have been impacted historically by the environmental degradation of the region and direct exposure to arsenic contamination, partially because of the proximity of their community and the lack of resources to defend their interests in the past (MVRB 2013).

The real cost of Giant mine to the Yellowknife community, in terms of health impacts and environmental damage, cannot be easily evaluated, nor can the ongoing value of the site as a source of future business opportunities and employment. For better or for worse, a significant portion of the economic history and future of Yellowknife is associated with Giant mine.

CONCLUSIONS

Ore and host-rock mineralogy and geochemistry have a direct influence on the environmental legacy at any mine. In the case of Au deposits, where significant processing occurs on the mine site (such as roasting, pressure oxidation, heap leach, mercury amalgamation, and/or cyanidation), solid waste to be managed includes processing waste in addition to waste rock and mill tailings. Orogenic Au deposits such as Giant have refractory ore that requires destruction of the host arsenopyrite to enable cyanidation to recover Au. Ore roasting is an effective pre-treatment for refractory Au but has the unfortunate consequence of producing significant quantities of As_2O_3. In the case of Giant mine, much of the mining and processing took place before containment technology for this hazardous waste was developed, and the remoteness of the location meant that the As_2O_3 could not be sold, as was the case for other roaster operations of the same vintage. The environmental legacy involves both surface and subsurface As-rich material present as sulfide minerals (primary arsenopyrite and secondary realgar) and oxide minerals (roaster-generated Fe oxides and As_2O_3, weathering products of arsenopyrite such as yukonite and As-bearing goethite). Although no acidic drainage is present at Giant mine due to a generous supply of carbonate minerals, the sensitivity of As phases to changes in redox results in post-depositional mobility.

Conversion of arsenopyrite to As_2O_3 has significantly increased As bioaccessibility in potentially ingested dusts and soils at Giant mine. More importantly, this conversion also increased the concentration of dissolved As in groundwater and surface waters, and thus, the general mobility and bioavailability of As. The proposed remediation plan involves a series of measures that would reduce but not eliminate the release of this dissolved As to Great Slave Lake, one of the largest lakes in Canada.

For the current residents of Yellowknife, the mine that helped to launch the city during the gold rush of the 1930's and 1940's is now, for many, an ongoing source of concern as well as an alarming expense, albeit one shared with other Canadian taxpayers. The voice of the original First Nations residents of the area, barely heard in the early years of mine development, is now a critical part in site management decisions. Remediation has replaced mining as the focus of employment associated with Giant mine. Diamond mines and other potential mineral developments in the Canadian North routinely incorporate community consultation.

For researchers outside Yellowknife, the case of Giant mine provides lessons regarding the particular complexities that mining and processing As-bearing Au ores involve. The remoteness of the location and the severity of the climate have influenced the environmental legacy, but more important factors are the nature of ore and host-rock mineralization, and the history of processing and waste management, factors that influence mine waste anywhere on the globe.

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