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Influence and fate of Sb in arsenic trioxide roaster waste from the Giant Mine, NT, Canada

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The former Giant gold mine (Yellowknife, NT, Canada) is among Canada's top five federal contaminated sites and estimated remediation costs currently exceed $4B CAD. Approximately 237 000 tonnes of highly toxic arsenic trioxide roaster waste (ATRW) generated from 1950 to 1999 were stored underground, while an estimated 20 000 tonnes were aerially distributed by stack emissions over the surrounding landscape. The current mitigation approach involves freezing the ATRW in place using thermosyphons; however, a more permanent stabilization method is needed following a full assessment of ATRW composition, reactivity, and updated stabilization technologies.

Bulk X-ray absorption spectroscopy and X-ray diffraction analysis have suggested relative homogeneity of the ATRW, identified as arsenolite [As_2O_3] with small amounts of Sb substitution. In contrast, scanning electron microscopy and electron microprobe analysis have revealed that stoichiometric arsenolite is rare. Rather, individual <1 μm diameter grains of the underground ATRW contain an Sb:As molar ratio up to 1.22, although the majority of the grains average a ratio of 0.01. Experimental work at several pH values, NaCl concentrations, and temperatures indicates that increased Sb substitution inhibits ATRW dissolution. This raises questions regarding the longevity of ATRW in area soils and whether remaining ATRW is progressively more concentrated in Sb, despite years of exposure to precipitation and associated dissolution.

One stabilization strategy used throughout the mining industry locks As in Fe(III) oxide minerals. Consequently, all mine-impacted water at the site is treated with the goal of lowering As concentrations through As(III) oxidation and precipitation of arsenical ferrihydrite. However, the long term fate of As and any associated Sb in these Fe(III) phases is uncertain, especially under storage conditions conducive to reductive transformation of iron oxides. Therefore, experiments were conducted to test the long-term geochemistry of treatment plant solids at equilibrium under different pH values and Fe(II) concentrations, while comparing the results to similar experiments conducted on arsenical ferrihydrite, hydrous ferric arsenate, and scorodite.

We now understand ATRW is a complex and heterogeneous geochemical mixture, where Sb may play a larger role than previously thought. Consequently, many questions remain regarding the importance, prevalence, and influence of Sb in ongoing remediation.