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Geochemical controls on arsenic and antimony in a neutral mixed-waste tailings containment area at the Giant Mine, Northwest Territories, Canada

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The Giant Mine, Yellowknife, Northwest Territories generated 8 Mt of As- and Sb-bearing mill tailings (As: 2890 ± 730 mg kg^{-1}; Sb: 210 ± 71 mg kg^{-1}), deposited in the Northwest Tailings Containment Area (NW-TCA). Mining operations produced a waste mixture consisting of sulfidic flotation tailings (84.8 wt%), Fe-oxide roaster residues (14.4 wt. %), and arsenic trioxide roaster waste (0.8 wt. %). The NW-TCA is utilized as an intermediate in the mine water treatment system, storing mine dewatering effluent high in dissolved As and Sb (mean: 22.2 mg L^{-1} As; mean 0.78 mg L^{-1} Sb). The management of neutral pH, As- and Sb-bearing Au mine waste requires a comprehensive understanding of the geochemical mechanisms controlling contaminant release. An integrated study combining solid-phase characterization (XRD, XRF, SEM-EDS, XANES), porewater geochemistry, and stable isotope analyses ($\delta^{34}S_{SO4}$, $\delta^{18}O_{SO4}$, $\delta^{13}C_{DIC}$, $\delta^{13}C_{DOC}$) identified three sources of dissolved metal(loid)s within the TCA: 1) infiltration of mine dewatering effluent, contributing the largest loading of As, an estimate of 5.3 tonnes a^{-1}; 2) oxidative dissolution of sulfide minerals in the near surface tailings, releasing As, Sb, Zn, Ni, and Co to the porewater, with O_2 ingress extending to 4 mbgs in areas distal to ponded surface water; and 3) reductive dissolution of Fe-oxide roaster residues, resulting in the highest localized concentrations of dissolved As (up to 56.3 mg L^{-1}) observed deeper in the tailings. Isotope analysis and dissolved methane measurements indicate that Fe(III) reduction, sulfate reduction, and methanogenesis are occurring concurrently in the anoxic vadose and saturated zones. Two attenuation processes are likely occurring: 1) precipitation of As-bearing Fe(III)-(oxyhydr)oxide minerals in the vadose zone; and 2) incorporation of metal(loid)s in secondary sulfide minerals produced during sulfate reduction at 10.5 mbgs, the base of the TCA. Assessment of the acid-generation potential of the waste indicated high neutralization capacity and low potential for acidic drainage, but a minimum of 40% S persists as unreacted sulfides even near ground surface. These results indicate that the management strategy for control of contaminant metal(loid)s release under this neutral pH, mixed waste system necessitates consideration of these spatially separated oxidative and reductive dissolution zones.