

Application of X-ray Absorption Spectroscopy to Assess Arsenic Mobility and the Effectiveness of a Passive Treatment System at an Abandoned Au-Mine

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Management of As-rich groundwater associated with the oxidation of arsenopyrite and arsenian pyrite in gold mine wastes is a global environmental concern. Passive treatment methods provide a low-cost, energy-efficient approach for removing As prior to discharge from mine-waste storage areas. Detailed characterization of biogeochemical processes was conducted at an abandoned mine site where extensive sulfide oxidation in tailings impoundments over the past 100 years has resulted in formation of acid-mine drainage. Measurements of pore-water chemistry, together with mineralogical and microbiological studies, indicate the presence of low pH (2.0-3.9) pore water, elevated concentrations of dissolved As (~400 mg L⁻¹) and metals, depletion of sulfide minerals, and elevated proportions of sulfur- and/or iron-oxidizers (mean of 32.3 % of total reads) in the near surface tailings. High energy resolution fluorescence X-ray absorption near-edge structure (HERFD-XANES) spectra collected for the shallow oxidized tailings are consistent with the presence of scorodite (FeAsO₄·2H₂O), suggesting this phase was the predominant form immobilizing As in the upper tailings. Below the depth of active oxidation, the predominant form of As was arsenopyrite. Secondary As sulfides in the form of realgar/orpiment were observed at the base of the tailings, corresponding to partial removal of As from the pore water. In the absence of an effective remediation strategy oxidation of sulfide minerals and release of dissolved As are expected to continue. Laboratory column studies were conducted to evaluate the potential for enhanced removal of As using different mixtures of organic carbon, zero valent iron and limestone. Extensive removal of As to <10 µg L⁻¹, representing >99.9% removal, and an increase in pH from 3.5 to circumneutral values were observed. Microbial community structure assessments indicate increased activity of sulfate-reducing bacteria in the treatment columns relative to a control column. Synchrotron-based As-µXRF, As µXANES and S µXANES analysis of samples collected at the termination of the experiments indicates As removal occurred through a combined mechanism involving precipitation of a crystalline As-Fe sulfide phase and adsorption as As(V) on ferrihydrite. These results show that enhanced removal of As from acidic, mine-impacted water can be effectively achieved using passive treatment mixtures.