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Geochemical controls on schwertmannite transformation to goethite under acid rock drainage conditions

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Schwertmannite (ideal composition $Fe_8O_8(OH)_6SO_4$) is a ferric oxyhydroxysulfate mineral commonly found in acid rock drainage from the weathering of hard-rock sulfide deposits. Schwertmannite is poorly crystalline, resulting in a large surface area and high sorptive capacity for metal(loids). Although schwertmannite is predicted to transform to other more stable phases, such as goethite (FeOOH) or jarosite ($KFe_3(OH)_6(SO_4)_2$), the transformation rate varies widely depending upon the aqueous conditions and the ions and metals adsorbed onto the surface. While some conditions have been documented in the literature, the fundamental geochemical controls on the rate of schwertmannite transformation and the fate of adsorbed metals are not well constrained. Using laboratory-synthesized schwertmannite and field samples (greater than 99% schwertmannite) from legacy mines in California and Colorado, the geochemical controls on schwertmannite stability have been evaluated by studying the rate of transformation over a range of conditions relevant to natural acid rock drainage including aqueous sulfate, potassium, ferrous iron, pH, and ionic strength. In addition, we tested whether air-drying and re-wetting of synthetic and natural schwertmannites affected the rate of transformation. Each variable was systematically evaluated in laboratory incubations at 35^{o}C. Aqueous composition, pH, conductivity, and mineralogy were monitored for 2-6 months, depending on the rate of transformation. Mineralogy transformation was determined using a low-mass method for quantitation of poorly crystalline materials by X-ray diffraction, as well as pH and sulfate concentrations. In all cases, the transformation of laboratory-synthesized schwertmannite to goethite was faster than what has been observed in the field. For all materials, low sulfate, low ionic strength, high Fe(II), and high pH all individually contribute to elevated transformation rates. However, natural schwertmannite had a substantially lower transformation rate than synthetic materials under identical laboratory conditions. We hypothesize that trace elements found in natural schwertmannite decrease the rate of transformation. This information is important for the development of robust models of metal mobility in acid rock drainage systems. In addition, the accumulation of metals in iron oxides may make these phases potential targets for metal recovery efforts which will benefit from a fundamental understanding of iron oxide stability.