Zinc Release and Reimmobilization during Fe(II)-induced Ferrihydrite Transformation

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Iron oxide minerals are ubiquitous in sediments and soils and often have great influence on the fate and availability of trace metals. Zn is an essential trace metal but can be toxic at higher concentration. Zn cycling and availability can be affected by its adsorption on, incorporation in, and release from iron oxides. Ferrihydrite is a metastable nanocrystalline iron oxide and transforms to more crystalline phases when exposed to dissolved Fe(II). While such reactions have been widely studied, the effects of ferrihydrite phase transformations on solid-associated trace metal speciation, how metals impact the resulting mineral products, and whether such processes fractionate metal stable isotopes are poorly studied.

We have investigated the phase transformations of Zn-substituted ferrihydrite at pH 7 for 12 days under different Fe(II) concentrations. X-ray diffraction patterns of the reaction products reveal that low Fe(II) concentration (0.2 mM) favors the formation of hematite, whereas high Fe(II) concentration (1 mM) promotes the production of magnetite. Both transformation processes are associated with the transient release of Zn to solution, which during aging is taken up by the mineral products. Reitveld refinement of the XRD data shows a progressive transformation from ferrihydrite to other iron oxides, with new phase nucleating within a few days but changes continuing over the entire experiment. Control experiments with pure ferrihydrite observe less hematite formation, indicating Zn alters ferrihydrite transformation pathways. EXAFS spectroscopy shows changes in Zn coordination state, indicating Zn incorporates into newly formed minerals. Despite the coordination change, Zn isotopes preserve their original fractionations in the solid phase.