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URANIUM DIFFUSION IN SOILS: EVALUATING SORPTION AND SURFACE DIFFUSION INFLUENCES

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Diffusive transport often severely limits subsurface mass transport, and is therefore important in both contamination and in-situ remediation processes. This is especially true for strongly sorbing metal and actinide contaminants such as uranium (U). Uranium contamination of soil and groundwater has occurred at many sites associated with its mining, milling, processing, use, and disposal. Typical U waste solutions are either acidic (e.g., U ore milling) or alkaline (weapons and fuel processing), such that the initial stages of soil contamination involve substantial geochemical disequilibrium. Since U(VI) sorption, solubility, and redox transformations with U(IV) are all strongly pH dependent, its reactive transport can be very difficult to predict under variable pH conditions. Thus, an understanding of U(VI) diffusion rates in soils is needed to better predict its behavior and to evaluate remediation options.

In this study, we measured transient U(VI) diffusion into soils from initially acidic (pH 2) and initially alkaline (pH 11) reservoirs. Two soils, one from Oak Ridge, Tennessee (OR, pH \approx 6.5), and the other from Altamont, California (AL, pH \approx 8.5) were exposed to each of these U(VI) solutions. The diffusion process was monitored through periodic measurements of U concentrations in boundary reservoirs (kinetic phosphorimetry), and through measurements of concentration profiles within soil columns (micro x-ray absorption spectroscopy, conducted at the GSECARS microprobe beamline, Advanced Photon Source, Argonne National Laboratory). These measurements of U time trends and spatial profiles were compared solutions to diffusion equations to obtain apparent diffusivities and in-situ partition (sorption) coefficients (K_d values).

Analyses of U depletion rates in the boundary reservoirs were consistent with measured U(VI) diffusion profiles within the soil columns. In the calcareous AL soil, the initially acidic and alkaline solutions were quickly buffered back to pH \approx 8. The OR soil exposed to acidic U(VI) solution became acidic (pH 4), while the OR soil exposed to the alkaline U(VI) solution became neutral (pH \approx 7). The results from the AL diffusion systems show that batch-based measurements of the U(VI) K_d overestimate sorption and retardation by over an order of magnitude. Our diffusion-based U(VI) K_d values are in the range of 2 to 3 mL/g, whereas batch-based values from other studies are in the range of 20 to 50 in this calcareous soil. In other soils, K_d values at pH = 8 are as high as 300 mL/g. Thus, diffusive transport in calcareous soils is much more efficient than expected from previously reported K_d values. Diffusion of U(VI) in the OR soil was more complex, exhibiting widely varying retardation. Equilibrium calculations also suggest that much of the U(VI) in the alkaline solution precipitated as a schoepite-like phase upon pH neutralization in the OR soil. X-ray absorption spectroscopy confirmed that U remained as U(VI) in these AL and OR soils.