Séminaire Seminar

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Examination of actinide subsurface transport from lab to field scales: influence of redox reactions and organic ligand complexation

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Visio via Zoom

The migration of actinides in the environment is dependent on the chemical species which dominate under given geochemical conditions. The mobility can be enhanced or retarded by altering the oxidation state or forming soluble organic ligand-metal ion complexes. This work examines three case studies to evaluate the impacts of these changes in chemical speciation on the transport of trace elements through soil. Our approach seeks to characterize the time and length scales over which non-equilibrium states are maintained by rate-limiting (or rate-enhancing) reactions between radionuclides and co-reactants due to interactions between physical mass-transfer processes (i.e., flow, advection, diffusion) and (biogeo)chemical reactions.

A range of Np and Pu sources with varying chemical forms were deployed in multi-year lysimeter experiments. All plutonium sources are transforming to disordered PuO_{2+x} type phases regardless of the initial oxidation state and the extent of transformation to disordered PuO_{2+x} phases appears to be accelerated in the field lysimeter sources relative to the archived sources. Comparison of the downward migration of each source indicates some notable differences between lysimeters with different initial Pu and Np sources. The current working hypothesis to explain these differences is that colloidal PuO_{2+x} and NpO_2 phases are forming during the chemical/physical transformations of the source materials and downward migration is enhanced by these colloidal phases. Thus, the extent of transport appears to be somewhat dependent on the initial chemical/physical state of the source.

A second mechanism by which the mobility of ions can be altered is through complexation with organic ligands. This work has examined the influence of nutrient availability, plant roots and plant root exudates on preferential water flow through field lysimeters and dissolution of uranyl phosphate minerals through formation of soluble U(VI)-ligand complexes. Results indicate that less soluble phosphate sources lead to enhanced plant exudate production which in turn enhances uranium solubility and mobility. These results were verified using flow-through batch reactor experiments examining the dissolution of uranyl phosphate by citric acid (a common phytosiderophore). Comparable experiments examining the influence of organic matter on Pu sorption indicate similar processes control Pu migration.

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