

Poster #21-37

Characterizing Radionuclide Subsurface Transport from Lab to Field Scales Using Multidimensional, Real-Time Imaging Techniques

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BER Program: SBR

Project: University Award

Project Website: <https://www.clemson.edu/centers-institutes/neesrwm/EPSCoR/>

This abstract provides highlights on real-time imaging studies of radionuclide transport performed as part of a DOE Experimental Program to Stimulate Competitive Research (EPSCoR) Implementation grant in South Carolina. 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. Multidimensional tools capable of real time monitoring radionuclide mobility were used to monitor lab to field scale experiments.

Highlights from several ongoing experimental and modeling studies will be discussed including:

1. *Development of novel 1D and 3D photon based measurement techniques:* Ex-situ, real-time 1D gamma ray analysis, 3D x-ray CT and, Single Photon Computed Tomography (SPECT) techniques have been used to examine Tc-99m transport through porous media. Data have demonstrated accumulation of Tc within reducing zones of heterogeneous redox environments and a very slow release of Tc into the aqueous phase.
2. *Quantifying flow in porous media using x-ray CT:* Transfer of mass between fast flow in macropores and slow flow in the soil matrix is an important control on the fate and transport of solutes. We show that CT imaging can be used to investigate film flow along macropores, non-uniform imbibition from macropores into soils, and complex filling processes that will ultimately be critical for controlling the movement of radionuclides through the soil as well as the reagents controlling the chemical environment.
3. *Examination of uranyl phosphate dissolution facilitated by biogenic ligands:* The influence of nutrient availability, plant roots and plant root exudates on preferential water flow have been examined using 3D x-ray computed tomography (CT) and 2D light transmission experiments. Results indicate that root exudates influence the wetting of soil surfaces and has a significant impact on preferential water flow. Microfluidics and flow-through batch reactor experiments verified mass transfer limitations of citrate associate with the mineral surface as the primary control of uranyl phosphate dissolution.
4. *Observations of radionuclide transport in field lysimeters:* Field based lysimeter studies of Tc-99 and Np-237 these ions are highly controlled by oxidation of initially Tc(IV) and Np(IV) sources. The rate and extent of source term oxidation was examined through measurements of Np and Tc in the lysimeter effluents during field deployment as well as leaching, batch sorption, x-ray absorption spectroscopy, and

electron microscopy techniques after the lysimeters were retrieved from the field.

This material is based upon work supported by the U.S. Department of Energy Office of Science, Office of Basic Energy Sciences and Office of Biological and Environmental Research under Award Number DE-SC-00012530.