SLAC SFA: Coupled cycling of organic matter, uranium, and biogeochemical critical elements in subsurface systems

Program Affiliation: Subsurface Biogeochemical Research
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Natural organic matter (NOM) profoundly affects the behavior of redox-active contaminants and biogeochemical critical elements (*BCEs*, including C, N, S, and Fe) in subsurface systems where NOM is abundant. Many thin, discontinuous bodies of reduced organic-rich sediments have been observed in the uranium-contaminated floodplain at the Rifle, CO DOE research site. These naturally reduced zones have elevated U(IV) concentrations (*ca* 100-fold higher than uranium in surrounding sediments) along with abundant iron sulfides and other redox-active metals. The potential susceptibility of these zones to oxidation raises the concern that U(IV) stored in reduced sediments may be slowly oxidized and released back to groundwater, thus contributing to the ongoing plume-persistence problem at this site. We have recently observed uranium- and sulfide-enriched zones at other uranium-contaminated former ore processing sites, including Grand Junction and Naturita, CO, and Riverton WY, suggesting that organic-rich zones have the potential to mediate uranium mobility at plume persistence sites generally within and peripheral to the upper Colorado River Basin (CRB).

The SLAC SFA is investigating natural organic matter (NOM)-uranium interactions across a range of scales, from molecular to regional, to understand the fundamental biogeochemical properties and processes that govern biogeochemical response of organic-rich reduced zones. Questions being investigated include: (i) What controls the speciation of U(IV) in these zones?; (ii) What/how do microbial interactions control redox transition pathways for uranium, NOM, and BCEs in these biogeochemically complex systems?; (iii) How do organic ligands influence uranium reduction?; (iv) What processes control uranium release from reduced sediments?; (v) What are the physical and biogeochemical characteristics of organic-rich reduced zones?; and (vi) How do organic-rich reduced zones interact with surrounding floodplain aquifers?

We are using an integrated suite of approaches to address these questions, including field-scale investigations at sites across the upper CRB coupled to laboratory investigations of organic-rich sediment properties and reactivity; molecular microbial ecology; stable isotope techniques, spectroscopic and electrochemical studies of the molecular speciation and thermodynamics of uranium, NOM, and BCEs; combined x-ray, electron, and isotope microscopy imaging, and biogeochemical modeling. By collecting and integrating information across a range of scales, we will improve our understanding of these systems at all scales and locations. These activities are leading to new process-level paradigms to quantitatively describe the reactivity of organic-rich zones and their potential to impact uranium mobility, and are helping to advance SRB modeling competencies at floodplain and regional scales.