

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

Program Affiliation: Subsurface Biogeochemical Research

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Organic-rich sulfidic sediment lenses hosting relatively high uranium concentrations have been observed at DOE's Rifle, CO site. Subsequently we have observed similar bodies at other uranium-contaminated DOE sites in the upper Colorado River Basin (CRB), including Grand Junction and Naturita, CO; Shiprock, NM; and Riverton, WY. These naturally reduced zones (NRZs) also store large inventories of nutrients and biogeochemical critical elements (BCEs, including C, N, S, and Fe) and they lie within or below the zone of annual groundwater table fluctuations where they are subject to radical hydrologic changes from saturation to highly unsaturated. Thus, NRZs constitute a class of regionally-important biogeochemical hotspots that have the potential to modify the mobility of nutrients, BCEs, uranium, and other contaminants within otherwise oxic nutrient-poor floodplain systems. Large-scale ecosystem perturbations such as extended drought and flooding have the potential to alter NRZ biogeochemistry and to discharge contaminant and nutrient loads to connected ground and surface water systems. There is concern that NRZs are contributing to uranium plume persistence regionally or could do so in the future.

We are investigating the fundamental hydro-biogeochemical mechanisms by which organic matter in NRZs mediates the speciation, behavior, and fluxes of BCEs and uranium across a range of scales, from molecular to regional. Our goal is to identify, interrogate, and model critical processes in these subsurface ecosystems to improve BER's computational subsurface research capabilities and to anticipate impacts to floodplain systems and society under changing climate conditions. Questions being investigated include: (i) What are the physical and hydro-biogeochemical characteristics of NRZs? (ii) What biological, kinetic, and thermodynamic factors control the speciation and behavior of uranium and BCEs? (iii) How do NRZs interact with surrounding floodplain aquifers? (iv) How does microbial N cycling mediate redox conditions? and (v) What are the mechanisms and rates of uranium release to surrounding 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-based molecular scale investigations using x-ray absorption spectroscopy, x-ray, electron, and isotope imaging, molecular microbial ecology, electrochemistry, and stable isotope techniques. Over the past year, these investigations have advanced our knowledge of NRZ biogeochemistry and are leading to new process-level models to understand their function at floodplain to regional scales.