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Coupled Cycling of Organic Matter, Uranium, and Biogeochemical Critical Elements in Subsurface Systems. Overview of the SLAC SFA

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Research performed by our group over the past 3 years has shown that organic-enriched sediments are common in shallow sand-pebble-cobble alluvial aquifers in the upper Colorado River Basin (CRB). Organic-enriched bodies are generally sulfidic and referred to as, 'naturally reduced zones' (NRZs), and host large inventories of biogeochemically critical elements and contaminants, including Fe, C, S, N, P, U, Mo, and V. NRZs frequently reside within the capillary fringe where they are exposed to seasonal wet-dry cycling, creating conditions conducive to redox-driven nutrient and contaminant mobilization, with important implications for groundwater quality. Uranium-enriched NRZs are spatially coincident with persistent uranium groundwater plumes, raising the likelihood that sediment-uranium interactions contribute to the longevity of these contaminant hot spots. Seasonal hydrologic variability and organic content are characteristic of floodplains worldwide, and alluvial redox cycling is globally important to water quality. In spite of its importance, there are numerous gaps in our understanding of these hydro-biogeochemically active systems.

We are investigating the fundamental hydro-biogeochemical mechanisms by which NRZs mediate the speciation, behavior, and fluxes of carbon, uranium, and other elements across a range of scales, from molecular to regional. Our goal is to identify, interrogate, and model key processes in shallow alluvial aquifers to improve our understanding of water quality and to support SBR computational capabilities. Questions being investigated include: (i) What are the dominant reactions that occur when NRZs undergo redox transitions?; (ii) What biological, kinetic, and thermodynamic factors control the speciation and behavior of uranium?; (iii) Does microbial N cycling mediate uranium mobilization?; and (iv) How do spatial constraints imposed on metal reducing microorganisms by the insolubility of TEAs influence microbial syntrophy and uranium reduction in NRZs?

We have conducted field-scale investigations at sites across the upper CRB and performed laboratory-based molecular scale investigations using x-ray absorption spectroscopy, x-ray, electron, and isotope imaging, gene sequencing, electrochemistry, and stable isotope techniques. Major scientific contributions over the past year include publication of new process models to describe iron and sulfur cycling in NRZs and their impact on U mobility, a new biogeochemical model for U(IV) behavior in organic-enriched sediments, which is dominated by adsorption to particulate organic carbon, and a new mechanism for organic carbon preservation in anoxic sediments. These findings advance the SBR mission by providing new knowledge about biogeochemical controls over water quality and by providing insights and biogeochemical process representations needed to model contaminant and nutrient behavior.