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Fe and S speciation to track redox constraints on shallow alluvial sediments: Implications for U storage and stability

VINCENT NOËL^{1*}, KRISTIN BOYE¹, RAVI KUKKADAPU², JUAN S. LEZAMA PACHECO³, SCOTT FENDORF³, SCOTT ROYCROFT³, JOHN R. BARGAR¹

¹Stanford Synchrotron Radiation Laboratory, SLAC, USA
(*correspondence: noel@slac.stanford.edu)

²Environmental Molecular Sciences Laboratory, PNNL, USA

³Earth System Science, Stanford University, USA

Redox processes are important mediators of nutrient and contaminant storage in alluvial aquifers. Organic-enriched zones are common in sand-pebble-cobble aquifers in the Upper Colorado River Basin (UCRB) and develop Fe(III) and sulfate reducing conditions. We refer to these as “Naturally reduced zones” (NRZs). Large inventories of U accumulate in NRZs located within persistent U groundwater plumes, raising the likelihood that sediment-U interactions help to sustain these contaminant hot-spots. The susceptibility of U to redox processes creates the need to better predict the biogeochemical controls on redox conditions in NRZs, and their impact on U storage and remobilization. The chemical forms of Fe and S provide precise tracers to better define the distribution and reactivity of redox constraints. We hypothesized that Fe and S combined speciation could help to develop conceptual and numerical models of the biogeochemical controls over redox-active contaminants mobility, as U, within alluvial aquifers.

Organic carbon content, moisture, and particle size of floodplain sediments were found to govern Fe and S mineral transformations and the abundance of reactive FeS and S(0) species in NRZs. Comparison of the distributions of these reactive species with that of U species suggests that: (1) iron sulfide occurrence maps on top of U(IV)-accumulating zones; (2) reactive Fe(II) species relative abundance is a functional proxy to estimate susceptibility of NRZ U(IV) stocks to oxidative remobilization; (3) Fe colloids are likely present and drive U(IV) remobilization under persistent reducing conditions. Finally, our data suggest that NRZ oxidation processes in response to seasonal hydrological variation increase affinity of U for Fe minerals, controlling remobilization and release of U.