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Redox Constraints on Shallow Alluvial Sediments: Implications for U Mobility

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Redox processes are important mediators of nutrient and contaminant storage and release in alluvial aquifers. Our previous studies show that organic-enriched zones are common in sand-pebble-cobble aquifers in the upper Colorado River Basin (CRB) and develop reducing conditions and sulfidic mineralogy. We refer to these as “Naturally reduced zones” (NRZs). Large inventories of uranium accumulate in NRZs located within persistent uranium groundwater plumes, raising the likelihood that sediment-uranium interactions help to sustain these contaminant hot-spots. The susceptibility of accumulated U(IV) to oxidation and remobilization creates the need to better understand and predict U speciation, the biogeochemical controls on redox conditions in NRZs, and their impact on U storage and remobilization.

In this study, we tracked Fe and S speciation in NRZs as a function of depth, organic carbon content, and grain size in order to better define the biogeochemical controls over the distribution and reactivity of redox process in the upper CRB floodplain sediments. Redox reactivity was then compared with the distribution of U species in order to elucidate factors controlling their stability. We hypothesized that U(IV) in NRZs would be bound to particulate organic matter. In this case, the ability of sediment to stabilize U(IV) would depend on the ability of sediments to physically and chemically protect U from oxidation. Here we show that organic carbon content and moisture are critical factors in the stability of U(IV) in NRZs. Coarse-grained textures allow greater exposure of NRZs to oxidants and conversion of U(IV) to U(VI). We also show that fine-grained NRZs can retain U(VI) produced by oxidation of U(IV) by adsorption on surfaces of solid phases. Finally, our data suggest that, even in the absence of oxidative perturbations, U can be mobilized. Knowledge produced by this work will help to develop conceptual and numerical models of the biogeochemical controls over U transport within alluvial aquifers and provides insights into the mobility of nutrients and other contaminants.