Redox, hydrological, and molecular controls over uranium mobility in redox-variable aquifers

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Uranium contamination stubbornly persists as a challenging and costly water quality concern at former uranium mining, ore processing and nuclear complex sites across the U.S. The longevity of uranium plumes implies interactions between sediment and dissolved uranium species. Improved understanding of these interactions is important for risk assessment and contaminated site management. Uranium mobility in natural sediments is controlled not only by its molecular form and solubility, but also by variability in surrounding redox conditions as well as by hydrologic processes, which transport uranium and mediate system-level biogeochemical behavior. Advancing our knowledge of uranium behavior requires that we understand hydrologic-biogeochemical-uranium couplings.

Our team is investigating the impacts of biogeochemical-hydrological cycles on uranium mobility in contaminated aquifers across the upper Colorado River Basin. Spectroscopic, microscopic, and geochemical studies of sediments and model systems show that U(IV) adsorbs to the surfaces of particulate organic mattter and exhibits a molecular structure consistent with 'noncrystalline' U(IV). We have found that organic-enriched sediments are common even in cobble-dominated alluvium and that U(IV) strongly accumulates in organic-enriched sediments at all sites examined, with non-crystalline U(IV) being predominant. The stability of U(IV) and the underlying biogeochemistry of sediments are strongly affected by variability of sediment saturation state, and secondarily by sediment grain size. These factors have major implications for uranium behavior, plume persistence, and water quality under the variable hydrologic conditions that typify the region.