



Uranium Bio-accumulation and Cycling as revealed by Uranium Isotopes in Naturally Reduced Sediments from the Upper Colorado River Basin

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Uranium (U) groundwater contamination following oxidized U(VI) releases from weathering of mine tailings is a major concern at numerous sites across the Upper Colorado River Basin (CRB), USA. Uranium(IV)-bearing solids accumulated within naturally reduced zones (NRZs) characterized by elevated organic carbon and iron sulfide compounds. Subsequent re-oxidation of U(IV)_{solid} to U(VI)_{aqueous} then controls the release to groundwater and surface water, resulting in plume persistence and raising public health concerns. Thus, understanding the extent of uranium oxidation and reduction within NRZs is critical for assessing the persistence of the groundwater contamination. In this study, we measured solid-phase uranium isotope fractionation ($\delta^{238/235}\text{U}$) of sedimentary core samples from four study sites (Shiprock, NM, Grand Junction, Rifle and Naturita, CO) using a multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS). We observe a strong correlation between U accumulation and the extent of isotopic fractionation, with $\Delta^{238}\text{U}$ up to +1.8 ‰ between uranium-enriched and low concentration zones. The enrichment in the heavy isotopes within the NRZs appears to be especially important in the vadose zone, which is subject to variations in water table depth. According to previous studies, this isotopic signature is consistent with biotic reduction processes associated with metal-reducing bacteria. Positive correlations between the amount of iron sulfides and the accumulation of reduced uranium underline the importance of sulfate-reducing conditions for U(IV) retention. Furthermore, the positive fractionation associated with U reduction observed across all sites despite some variations in magnitude due to site characteristics, shows a regional trend across the Colorado River Basin. The maximum extent of ^{238}U enrichment observed in the NRZ proximal to the water table further suggests that the redox cycling of uranium, with net release of U(VI) to the groundwater by non-fractionating oxidation, is occurring within this zone. Thus, release of uranium from the NRZs may play a critical role in the persistence of groundwater contamination at these sites.