

## Isotopic Mapping of a Redox Boundary: Heterogeneities Across a Roll-front U Deposit

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Interaction between groundwater flow and abiotic and/or microbially induced redox regimes is important for metal transport in the environment. Understanding the relative contributions of groundwater hydrology and pore-scale chemical reactions is crucial for understanding formation mechanisms of redox-interface ore deposits as well as detecting natural reactions using redox-active isotope tracers. The redox-interface responsible for ore formation extends along the hydraulic gradient and gives rise to isotopic heterogeneity.

Here we report U isotopic characterization ( $(^{234}\text{U}/^{238}\text{U})$  and  $\delta^{238}\text{U}$ ) of a roll-front U deposit from Smith Ranch Highland, WY, USA, using sediments from 3 drill cores located along the hydraulic gradient. Our results show substantial variations in  $\delta^{238}\text{U}$  (up to  $\sim 2\text{‰}$ ) and  $(^{234}\text{U}/^{238}\text{U})$  (ranging from 0.77 to 2.26) in U minerals with depth in each core and amongst the different cores. In each core, U concentration is correlated with  $\delta^{238}\text{U}$ ; samples with highest U at the center of the mineralized zone exhibit high  $\delta^{238}\text{U}$  ( $> 0.0\text{‰}$ ) which decreases ( $\delta^{238}\text{U} < 0.0\text{‰}$ ) both upward and downward. The  $(^{234}\text{U}/^{238}\text{U})$  in each core is correlated with  $\delta^{238}\text{U}$ , with the lowest  $(^{234}\text{U}/^{238}\text{U})$  ( $\ll 1.0$ ) at the center of the mineralized zone.

Decreasing  $\delta^{238}\text{U}$  along the hydraulic gradient at the center of the mineralized zone suggests a significant hydrological influence on U isotopic distribution. During ore formation reduction of dissolved U(VI) leads to the formation of U minerals enriched in  $^{238}\text{U}$ . The remaining aqueous U(VI) is progressively enriched in  $^{235}\text{U}$  as it flows downgradient. The resulting U minerals also become  $^{235}\text{U}$  enriched relative to those forming upgradient. Dissolution of these  $^{235}\text{U}$  enriched U minerals would produce isotopically lighter U(VI) in groundwater. To use  $\delta^{238}\text{U}$  of groundwater for detection of U(VI) reduction, the abovementioned dissolution effects must be disentangled from the isotopic fractionation in the same direction generated by reduction. Future work will determine  $\delta^{238}\text{U}$  and  $(^{234}\text{U}/^{238}\text{U})$  in the porewater and adsorbed U(VI) for each sample.