Systematic variations in U isotope ratios of sediments from reactive transport of U

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U isotope ratios are being developed as reliable indicators of redox reactions in the environment with increasing applications in palaeo-redox studies, contaminant transport and remediation monitoring. The environmental redox chemistry controls U mobility, and reactions that immobilize U are closely linked to the distribution of microbial and abiotic reductants in the subsurface. Therefore, understanding and quantifying the reactions using U isotope ratios in relation to the reactive transport of U in groundwater has major implications for the environment and energy.

Here, we present a combined U isotope approach using redox-sensitive $\delta^{238}U$ and and non-redox ($^{234}U/^{238}U$) of mineralized sediments for detection and characterization of hydrological influence on U isotopic distribution. The U mineralized sediments are collected from the Needle's Eye natural analogue site in Scotland, UK. The overall variation in $\delta^{238}U$ of U minerals ranges from -0.35‰ to -1.04‰ in the top 35 cm of the sediment core while ($^{234}U/^{238}U$) varies from 0.96 to 1.01. Our results reveal a systematic inverse relationship between ($^{234}U/^{238}U$) and $\delta^{238}U$ in the sediments; the samples with low $\delta^{238}U$ exhibits high ($^{234}U/^{238}U$), particularly at high U concentration (>500 ppm). The systematic relationship between ($^{234}U/^{238}U$) and $\delta^{238}U$ in sediments from Needle's Eye site closely resembles the isotopic signature (($^{234}U/^{238}U$) and $\delta^{238}U$) of a roll-front U deposit from Smith Ranch Highland, WY, USA, suggesting that the reactive trasnport of U influencing both isotope ratios. The reductive immobilization of U at redox boundaries sets the $\delta^{238}U$ and ($^{234}U/^{238}U$) in sediments while α recoil and preferential leaching of ^{234}U during dissolution tend to decrease the ($^{234}U/^{238}U$) of U minerals. Therefore, the continuous recycling of U across the redox boundary may give rise to the observed patterns. Future work will develop a quantitative model of U isotopic distribution of U minerals across redox boundaries.