Large $^{238}U/^{235}U$ Fractionation in an alkaline, euxinic lake: implications for the marine sedimentary record

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Variations in the ratio of $^{238}U/^{235}U$ in sedimentary rocks have been used to quantify the oxygenation of the Precambrian oceans and changes in the extent of ocean anoxia at the P-T boundary. Despite research demonstrating that U(VI) reduction reactions are the primary driver of fractionation ($\varepsilon^{238}U=\sim1‰$), interpretations of $^{238}U/^{235}U$ remain ambiguous. A recent review of modern marine sediments suggests that the expression of this fractionating U reduction in all anoxic and hypoxic basins is limited by diffusion of aqueous uranium into the sediment column, limiting observed shifts to $\varepsilon^{238}U\leq0.6‰$. In contrast, ancient sediments from the Black Sea suggest $\varepsilon^{238}U$ greater than this diffusive limit [1].

To address this disparity, we characterized the U cycle in Mono Lake (eastern California, USA) in order to better understand the physical and chemical controls on $^{238}U/^{235}U$ fractionation in anoxic settings. Mono Lake is a closed basin alkaline lake (pH 9.8) with characteristically long residence times for actinides. Evaporation has resulted in highly concentrated uranium (500 µg/L) and carbonate (320 mM) within the lake. Anoxic, sulfidic conditions exist at depth in the water column, and support U(VI) reducing conditions.

We characterized the $^{238}U/^{235}U$, $^{234}U/^{238}U$, and U concentrations of lake water as a function of depth through the water column, as well as inputs (streams and springs) and outputs (carbonate tufas and lake botom sediments). The lake water $^{238}U/^{235}U$ is -1.1‰±0.1‰ and is invarient with depth. In contrast, authigenic U in the lake bottom sediments average 0.0‰±0.2‰ and have constant concentrations over the 60 cm depth interval we measured. Freshwater streams and springs deliver U to the lake with a $^{238}U/^{235}U$=-0.2‰±0.2‰. A numerical model of the system suggests that (1) the lake is in steady-state for U and U isotope compositions, (2) U reduction is not limited by diffusion into the sediment and (3) U reduction likely occurs in the water column. These results suggest that inferences of $\varepsilon^{238}U\geq0.6‰$ in the sedimentary record may indicate the onset of U reduction in the water column

[1] Andersen, *et al.* (2017) *RiMG* **82**, 799-850.