Goldschmidt2020 Abstract

Aqueous Chemistry Influences Uranium Isotope Fractionation

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The uranium isotope ratio, $^{238}U/^{235}U$, can be used to better interpret environmental redox conditions. Recently, researchers have applied the $^{238}U/^{235}U$ ratio to study the formation of ore deposits, evolution of O_2 levels over Earth's history, and remediation of contaminated groundwater [1]. However, to reliably apply $^{238}U/^{235}U$ to track and quantify these processes, one must recognize how variable aqueous conditions and reduction mechanics influence the uranium isotope fractionation. U(VI) reduction is the main geochemical reaction that fractionates $^{238}U/^{235}U$. Both microbial and abiotic U(VI) reduction can lead to preferential removal of ^{238}U, leaving less ^{238}U in solution [2,3,4]. However, aqueous chemistry can strongly influence the magnitude and even direction of isotope fractionation during abiotic and microbial U(VI) reduction [2,3,4]. Through numerous experiments varying U aqueous chemistry, we attempt to interpret several of the mechanisms controlling U isotope fractionation.

We conducted batch experiments investigating how aqueous chemistry influenced U isotope fractionation during abiotic and microbial U(VI) reduction. Fe(II) sulfide and Shewanella (with lactate as an electron donor) were used for abiotic and microbial U(VI) reduction experiments, respectively. By varying several chemical parameters, such as pH, bicarbonate, and Ca concentrations, we detected the influence of aqueous chemistry on isotope fractionation. With higher pH and bicarbonate and Ca concentrations, greater isotope fractionation was observed in both abiotic and microbial experiments. With lower levels of these constituents, muted or even reversed (preferential removal of ^{235}U) isotope fractionation occurred. These experiments can be explained by a two-step process of either diffusion or adsorption of U(VI) to a solid surface followed by U(VI) reduction.

[1] Andersen *et al.* (2017) *Rev. Mineral. Geochem.* **82**, 799-850. [2] Stylo *et al.* (2015) *PNAS* **112**, 5619-5624. [3] Brown *et al.* (2018) *PNAS* **115**, 8688-8693. [4] Basu *et al.* (2020) *Envir. Sci. Technol.* **54**, 2295-2303.