Biotic uranium mobilization of noncrystalline U(IV) and associated U isotope fractionation

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Environmental uranium (U) contaminations may be remediated via microbial reduction of mobile hexavalent [U(VI)] to more immobile tetravalent U [U(IV)] [1]. However, the subsurface stability of U(IV), usually present as solid-phase noncrystalline U, may be affected by reactions with ligands, oxidants or bacteria. Uranium isotopes are proposed as a tool to monitor these (im)mobilization processes. However, U isotope fractionation during potential U(IV) remobilization is not yet fully understood, limiting the applicability of U isotope signatures for prediction of the sustainability of microbiallyreduced U(IV) at U remediation sides.

In this work, the efficacy of *Acidithiobacillus (At.) ferrooxidans* to mobilize non-crystalline U(IV) and the associated U isotope fractionation was investigated in controlled laboratory experiments. The starting material was produced by reduction of a U(VI) isotope standard with *Shewanella oneidensis* MR-1 in a phosphate-containing medium (WLP) [2]. *At. ferrooxidans* mobilized between 74% and 91% U after one week, and interestingly, U mobilization was equally observed for both, living and inactive cells.

The similar mobilization capability of active and inactive At. ferrooxidans cells suggests that the mobilization is based on the reaction with the cell biomass. Contrary to anoxic U mobilization with ligands, where 238 U is enriched in the mobilized phase [3], but similar to abiotic oxidation and mobilization of U(IV) from uranite [4], U mobilization by At. ferrooxidans did not cause significant U isotope fractionation. Potentially, this is because the speed-limiting process during U mobilization, e.g. U adsorption to the biomass in our case, does not generate significant isotope fractionation. Thus, U isotope signatures are unsuitable for tracking bacterial U remobilization and isotopic signatures found in nature are more likely generated by U reduction, or mobilization with ligands. The results of this study, combined with those of [3], challenge the long-term sustainability of in-situ bioremediation measures at Ucontaminated sites.

[1] Basu, A. et al. (2015), Environ. Sci. Technol., 49 (10), 5939–5947.

[2] Stylo, M. et al. (2013), Environ. Sci. Technol., 47 (21), 12351–12358.

[3] Roebbert, Y. et al. (2021), *Environ. Sci. Technol.*, 55 (12), 7959–7969.

[4] Wang, X. et al. (2015), Geochim. Cosmochim. Acta, 150, 160–170.