

Mechanism of uranium reduction and immobilization in *Desulfovibrio vulgaris* biofilms

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The prevalent formation of non-crystalline U(IV) species in the subsurface and their enhanced susceptibility to re-oxidation and remobilization as compared to crystalline uraninite raise concerns about the long-term sustainability of the bioremediation of U-contaminated sites. A proposed model showed that abiotic U(VI) reduction with biogenic mackinawite is the major mechanism of U reduction and sequestration following bioremediation at the Old Rifle field site in Colorado [1]. In contrast, a separate study, relying on U isotopic fractionation during acetate amendment at the same site, showed that biological reduction was the principal U immobilization pathway [2]. The focus of the present study was to resolve this discrepancy.

In particular, the main goal of this study was to tackle the remaining uncertainty concerning the formation mechanism of non-crystalline U(IV) in the environment. This question was probed using controlled laboratory biofilm systems (biotic, abiotic and mixed biotic-abiotic) and a combination of U isotope fractionation and X-ray Absorption Spectroscopy (XAS). Regardless of the mechanism of U reduction, the presence of a biofilm resulted in the formation of non-crystalline U(IV). Furthermore, biotic U reduction is the most effective way to immobilize and reduce U. However, the mixed system resembled an abiotic system most closely: (i) the U(IV) solid phase lacked a typical biotic isotope signature [3] and (ii) elemental sulfur was detected which indicates the oxidation of sulfide coupled to U(VI) reduction. The predominance of abiotic U reduction in our systems is due to the lack of available aqueous U(VI) species for direct enzymatic reduction. In contrast, the presence of bicarbonate boosts the aqueous U(VI) species allowing biotic U reduction to outcompete the abiotic processes.

[1] Bargar et al. 2013, *PNAS*, **v110**, p 4506-4511 [2] Bopp et al. 2010, *ES&T*, **v44**, p5927-5933 [3] Stylo et al. 2015, *PNAS*, accepted