Geochemical controls of competitive inhibition of microbial uranium reduction by iron oxides

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Uranium bioreduction, the microbial reduction of soluble U(VI) to sparingly soluble U(IV), is a promising bioremediation strategy to remediate uranium contamination in subsurface environments. In the presence of Fe(III) oxides, however, electrons may be diverted from U(VI) to Fe(III) as a result of the similar Gibbs free energy (ΔG_{rxn}) associated with anaerobic respiration of Fe(III) oxides and U(VI). The objective of this work was to identify the geochemical controls of the competition between U(VI) bioreduction and Fe(III) oxide respiration within the same microbial community and propose a new rate law to account for such change in terminal electron acceptor (TEA). Batch incubations with *Shewanella putrefaciens* strain 200 were conducted with either U(VI), 2-line ferrihydrite, or a combination of both as TEAs, and two pH and carbonate concentration conditions were selected to alter the ΔG_{rxn} and promote either U(VI) or ferrihydrite respiration. Uranium bioreduction was inhibited under geochemical conditions that thermodynamically favored ferrihydrite respiration. Under geochemical conditions which favored U(VI) respiration, however, uranium removal and Fe(II) production occurred simultaneously. In the same incubations, enhanced Fe(II) production occurred after total dissolved uranium concentrations were below detection limit. These findings suggest that under the experimental conditions ferrihydrite and uranium serve as TEAs simultaneously while uranium also acts as an electron shuttle between metal-reducing bacteria and Fe(III) oxides. A new uranium bioreduction rate law is proposed that explicitly accounts for competitive inhibition in the precense of ferrihydrite. This rate law is able to reproduce U(VI) bioreduction rates across all experimental conditions. These results advance the current understanding of the coupled biogeochemical cycling of iron and uranium and further the ability to predict the fate of uranium in complex subsurface environments.