Geophysical Research Abstracts,
Vol. 10, EGU2008-A-05621, 2008
SRef-ID: 1607-7962/gra/EGU2008-A-05621
EGU General Assembly 2008

Microbial reduction of iron(III)-rich smectite and uranium(VI)

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To assess the dynamics of microbially mediated U-clay redox reactions, we examined the reduction of iron(III)-rich nontronite NAu-2 and uranium(VI) by *Shewanella oneidensis* MR-1. Bioreduction experiments were conducted with combinations and varied concentrations of nontronite, U(VI) and the electron shuttle anthraquinone-2,6-disulfonate (AQDS). Abiotic experiments were conducted to quantify U(VI) sorption to NAu-2, the reduction of U(VI) by chemically-reduced (CBD) nontronite, and the oxidation of U(IV) by nontronite. Solids were characterized by X-ray diffraction, scanning electron microscopy, and X-ray absorption spectroscopy. When we incubated *S. oneidensis* MR-1 with nontronite and U(VI), little U(VI) reduction occurred compared to nontronite-free incubatons, despite the production of abundant Fe(II). The addition of AQDS to U(VI)- and nontronite-containing incubations enhanced both U(VI) and nontronite-Fe(III) reduction. While U(VI) was completely reduced by *S. oneidensis* MR-1 in the presence of nontronite, increasing concentrations of nontronite led to progressively lower rates of U(VI) reduction. U(VI) enhanced nontronite-Fe(III) reduction and U(IV) was oxidized by nontronite-Fe(III), suggesting that U(VI) served as an effective shuttle electron from *S. putrefaciens* MR-1 to nontronite-Fe(III). The electron-shuttling activity of U may explain the lack of U(VI) reduction observed in the bulk solution. Little U(VI) reduction was observed in incubations that contained chemically reduced nontronite-Fe(II), suggesting that the reduction of U(VI) by structural Fe(II) in nontronite contributes minimally to the overall process of U(VI) reduction. Our results suggest that abundant Fe(III)-rich clays in U(VI)-contaminated

systems may prevent U(VI) bioreduction.