Mineral Solubility and Free Energy Controls on Microbial Reaction Kinetics: Application to Contaminant Transport in the Subsurface

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Traditional kinetic and thermodynamic models incorporating microbial processes are typically developed using data from batch reactors in nutrient rich media characterized by rapid microbial growth, high respiration rates, and high cell densities. In the subsurface, however, microbial communities survive under energy and nutrient limiting conditions resulting in slow to zero growth conditions that cannot be reproduced by conventional kinetics. In this project, new rate laws able to predict microbial respiration in natural conditions are developed using two approaches. First, microbial growth under bioenergetic limitations is simulated using a novel flow-through bioreactor designed to include a retention filtration unit (retentostat) and accumulate biomass over time. Maintenance energy requirements for dissimilatory reduction of nitrate, a common groundwater contaminant, were determined by running the retentostat under limited conditions with lactate as electron donor. Initial rates of ammonium production by Shewanella oneidensis as model microorganism were similar in the bioreactor and otherwise identical incubations conducted in batch reactors. ATP measurements, however, showed slow yet increasing microbial growth over time in the retentostat while cells in the batch reactors did not grow significantly and died within 2 weeks of inoculation. In a second approach, rates of enzymatic U(VI) reduction were measured under variable, but controlled, geochemical conditions to determine the effect of uranyl speciation on bioreduction rates by S. putrefaciens as model metal-reducing microorganism. High pH, DIC, Ca2+, and Mg2+ suppressed the formation of labile U(VI) complexes and retarded uranium bioreduction. These results indicate that the main reducible fraction of U(VI) consists of non-carbonate complexes despite being the least abundant species in solution. At elevated concentrations of bioavailable uranyl species, however, bioreduction was inhibited by the toxicity of uranium to cells. Viability assays confirmed that the 'free' uranyl ion is responsible for uranium toxicity to S. putrefaciens, in agreement with the biotic ligand model of metal toxicity. A U(VI) bioreduction rate law that accounts for the speciation of U(VI) species is able to reproduce bioreduction rates in all pH, carbonate, calcium, and magnesium conditions. Future retentostat experiments will be designed to examine the thermodynamic controls on the bioreduction of non-carbonate U(VI) species in the presence of alternative electron acceptors. The kinetic and thermodynamic parameters determined in the current study will be directly applicable in reactive transport models to predict the rates of microbial respiration in both natural and contaminated subsurfaces.