Impacts of Mineralogy and Competing Microbial Respiration Pathways on the Fate of Uranium in Contaminated Groundwater

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This is a field-oriented project designed to elucidate the microbiological and geochemical factors controlling U(VI) reduction/immobilization in subsurface environments at the Field Research Center (FRC). Since the majority of U(VI) contamination in the FRC subsurface is associated with acidic sediments where the groundwater contains extremely high nitrate concentrations, we have focused our efforts on these acidic sediments. Our objectives are to (1) determine the dominant Fe minerals likely to limit U speciation, (2) directly quantify reaction rates and pathways of terminal-electron-accepting processes that control subsurface sediment chemistry, and (3) characterize the anaerobic microbial consortia—Fe(III)-reducing bacteria (FeRB)—likely to catalyze U reduction.

FeRB were enumerated and characterized from most probable number (MPN) enrichment cultures. Phylogenetic analysis of the 16S rRNA gene sequences extracted from positive MPN enrichments revealed that the only FeRB cultivated at acidic pH were gram-positive organisms not closely related to any previously cultured FeRB. In contrast, members of the family *Geobacteraceae* predominated at neutral pH in enrichment cultures from background sediments. Geochemical parameters likely to limit the activities of FeRB in situ were studied in parallel with freshly collected samples of all of the subsurface sediments studied. Chemical analyses in groundwater included nitrate, ammonium, U(VI), and pH. Solid phase characterization included porosity, density, organic matter content, sorbed nitrate, and sediment pH. Using wet chemical extractions and Mossbauer spectroscopy, aluminosilicates (66%) and Al-substituted goethite were the predominant Fe minerals observed. A small but substantial percentage (<5%) of the Fe minerals present were associated with poorly crystalline minerals such as ferrihydrite. Iron mineralogy did not appear to vary between sites or with sediment pH. Carbon metabolism (as CO_2 accumulation) and available electron acceptors were monitored at regular intervals in sediment microcosms. Sulfate concentrations in microcosms were generally very low, and little or no Fe(III) reduction activity was observed, whereas millimolar amounts of nitrate were depleted over time. Little or no methane accumulated in these incubations, while millimolar amounts of carbon dioxide accumulated in conjunction with the observed nitrate depletion and in response to carbon substrate addition. High denitrification rates, monitored using the acetylene block technique, were observed but only in neutral pH microcosms.

Our results indicate that the contaminated FRC subsurface is a heterogeneous "extreme environment" where the metabolism of FeRB is likely to be controlled by acid tolerance and competition with nitrate as an electron acceptor. Results also point to nitrate removal and neutralization of pH for establishment of conditions conducive to U(VI) reduction/immobilization by FeRB. Currently, the approaches of our multidisciplinary team (mineral characterization, rate measurements, microbial community analysis) are being combined to determine the interactions between Fe mineral transformation and U(VI) solubility during biostimulation in sediment microcosms.