Physical, Chemical, and Microbial Processes Controlling Uranium Mobility at the Old Rifle UMTRA Site

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Previous and ongoing laboratory and field studies at the Old Rifle UMTRA site in western Colorado indicate that the biogeochemical transformation of uranium to less mobile forms is a dynamic function of specific microbial populations (i.e., iron reducers and sulfate reducers) responding to biostimulation (amendment with acetate) in the context of site-specific hydrologic and geochemical conditions. Using data that are now available from these experimental studies, we are creating a quantitative model of the mechanistic contribution by individual subsurface processes to observed uranium behavior. Specific issues to be addressed include: (1) the impact of hydrologic conditions on bromide transport and oxygen stratification; (2) the mechanisms controlling the rate of microbially mediated uranium reduction during and after biostimulation; and (3) multicomponent surface complexation controlling the sorption of U(VI) species. Multicomponent, biogeochemical reactive transport simulations based on coupling of these conceptual process models will then be used to interpret and unravel interactions between processes under the dynamic and spatially variable hydrologic and geochemical conditions at the Old Rifle UMTRA field site. The goal is to enhance understanding of the scale-up of fundamental mechanisms, which is critical to the engineering of in situ conditions favorable to bioremediation.

Analysis of the hydrologic conditions for the 2002 and 2003 field experiments, including the transport conditions governing the migration of the bromide tracer and the delivery of the electron donor, acetate, is in progress. The resulting velocity fields will be used to investigate the stratified distribution of dissolved oxygen and U(VI) found at the site. In conjunction with the velocity field identified for the 2002 field study at Old Rifle, we have implemented a biogeochemical reaction network using a dual Monod kinetics formulation that includes response to the acetate injection by iron reducers that also use U(VI) as a terminal acceptor, and sulfate reducers that succeed the iron reducers after depletion of available Fe(III) terminal electron acceptors. Analyses of mechanisms controlling microbially mediated uranium reduction during the biostimulation events in 2002 and 2003 will be the basis for the collaborative analysis of the Princeton (Jaffe et al.) laboratory studies and refinement of the conceptual model for all terminal electron acceptors. This will include the determination of rate laws for the transformation reactions involving oxygen, nitrate, uranium, Fe(III) minerals, and sulfate with potential dependencies on biomass generation and decay. The ongoing development and testing of a preliminary multicomponent, generalized composite, surface complexation model will focus on characterizing uranium sorption as a function of solution chemistry. The final task will be to integrate the updated hydrologic, biogeochemical, and surface complexation process models to investigate site-specific hydrologic and biogeochemical conditions affecting the development of chemically reducing conditions, including the dissolution of Fe(III) minerals and the liberation of U(VI) adsorbed to those minerals; and the identification of mechanistic linkages between biostimulation in the Old Rifle aquifer and subsequent secondary mineral reactions, associated mineral volume changes, and the impact of these changes on accessibility to Fe(III) and U(IV) minerals.