Integrating Geochemical, Reactive Transport, and Facies-Based Modeling Approaches to Assess U(VI) Contamination at the Savannah River F-Area

LBNL SFA (Laboratory Research Manager: Susan Hubbard)

S.A. Bea, N. Spycher, H. Wainwright, S. Mukhopadyay, S.S. Hubbard,
Carl Steefel and J. Davis (PI) – LBNL

Acidic waste solutions containing low level radioactivity from numerous isotopes were discharged to a series of unlined seepage basins at the F-Area of the Savannah River Site, South Carolina, from 1955 through 1989. A nearly 1 km long acidic plume has developed in the groundwater under this site in relatively permeable, mostly sandy sediments. Although the site has gone through many years of active remediation, the groundwater remains acidic, and the concentrations of U(VI) and other radionuclides are still significant. Monitored Natural Attenuation (MNA) is a desired closure strategy for the site, based on the premise that clean background groundwater will eventually neutralize the groundwater acidity and drive natural immobilization of U(VI) through sorption. Understanding key hydrochemical controls and the impact of chemical and physical heterogeneities on contaminant mobility is of particular interest for assessing s MNA and in-situ treatment over the long term.

The first part of this study involves the development of reactive transport models to explore key controls on the long-term U(VI) plume mobility at this site through considering U(VI) adsorption by sediments and key hydrodynamic processes. Various geochemical, horizontal 1D, and 2D cross-section reactive transport simulations were conducted. These include the saturated/vadose zones and the effects of mineral dissolution and precipitation, as well as H^+ and U(VI) sorption using surface complexation models. The results indicate that H^+ sorption reactions on goethite and kaolinite (the main minerals at the site besides quartz), and the precipitation of Al minerals could delay the pH rebound for decades. Such slow rebound is likely to be exacerbated by residual saturation of the plume below the discharge basins.

The concept of "reactive facies" was explored to spatially distribute linked physical and chemical heterogeneities at local and field scales (see poster by Wainwright et al.). This concept integrates sediment chemical and hydrophysical properties (determined through laboratory and field experiments) with geophysical signatures. Obtained estimates of reactive facies were subsequently used to parameterize our reactive transport models. Because reactive properties at this site relate dominantly to sorption/desorption, we find that heterogeneous distributions of these properties (compared with homogenous distributions) affect the predicted pH and U transport mostly at early times and at plume edges, and over the long term once the concentrations of contaminants have decreased below sorption saturation levels.

The second part of this study involves uncertainty quantification performed in conjunction with the reactive transport simulations in order to: (1) identify the complex physical and geochemical processes that control the migration of the acidic-U(VI) plume in the pH range where it is highly mobile, (2) evaluate those physical and geochemical parameters that are most controlling, and 3) attempt to predict the future plume evolution constrained by historical chemical and hydrological data. The uncertainty quantification analysis shows that model results are most sensitive to the reactive surface area available for sorption, discharge rates, the relative rates of H^+ influx and kaolinite dissolution. The plume behavior also appears to be sensitive to parameters controlling the amount of residual U(VI) in the vadose zone, which acts as a buffering zone in the modeled system.