Quantifying physicochemical heterogeneity and critical transitions that influence plume behavior using geophysical approaches and reactive transport modeling

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Predictions of subsurface contaminant plume mobility and remediation often fail due to the inability to tractably characterize heterogeneous flow-and-transport properties and monitor critical geochemical transitions over plume-relevant scales. This study presents two different, recently developed strategies to quantify and predict states and processes across scales that govern plume behavior. The first approach, called reactive facies, exploits coupled physiochemical heterogeneity to characterize subsurface flow and transport properties that impact plume sorption and thus mobility. The second approach focuses on identifying diagnostic signatures of critical system transitions. Development of both strategies have involved the use of reactive transport models, geophysical methods, and stochastic integration approaches and take advantage of multi-scale and disparate datasets.

The reactive facies concept is based on the hypothesis that packages of sediments can be defined that have unique and linked distributions of properties that influence reactive transport and that geophysical methods can be used to identify and spatially distribute reactive facies and their associated parameters. We develop and test the reactive facies concept within uranium contaminated Atlantic Coastal Plain sediments that underlie the U.S. Department of Energy Savannah River Site, F-Area, South Carolina. Through analysis of field data (core samples, geophysical well logs, and cross-hole ground penetrating radar and seismic datasets) coupled to laboratory sorption studies, we have identified two reactive facies that have unique distributions of mineralogy, texture, porosity, hydraulic conductivity and geophysical attributes. We develop and use facies-based correlations with geophysical data in a Bayesian framework to provide high resolution (0.25m by 0.25m) estimates of reactive facies and their associated transport properties and uncertainties along local and plume-scale geophysical transects. To illustrate the value of reactive facies, we used the geophysically-obtained reactive facies properties to parameterize reactive transport models that were then used to simulate the migration of an acidic-U(VI) plume through the 2D domains. Modeling results suggest that each identified reactive facies exert control on plume evolution, highlighting the usefulness of the reactive facies concept and approach for spatially distributing properties that control flow and transport over field-relevant

scales as is needed for improved prediction of plume behavior.

The second approach focuses on quantifying and predicting critical redox transitions that control uranium immobilization during a bioremediation experiment performed at the DOE Integrated Field Research Center at Rifle, CO. We first develop and implement a mechanistic reactive transport approach to simulate reaction networks and associated system responses to the bioremediation experiment at the field scale. To complement this mechanistic approach, we also develop a data-driven statistical (or top down) approach that considers measured geochemical concentrations (which are indicative of redox state) as hidden random processes and that consider time-lapse spectral induced polarization data as observations at each location. We use a Bayesian approach and Hidden Markov models to estimate the probability of being in each redox stage over time and space following biostimulation. We view the mechanistic (bottom up) and data-driven (top down) modeling as complementary approaches that can constrain and validate each other, and as a step toward development of autonomous monitoring and assimilation approaches needed to monitor long-term plume behavior.