Reactive transport modeling of Hanford 100H lab- and field-scale experiments

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The Unraveling Biogeochemical Pathways Challenge of the LBNL SFA focuses on identifying and quantifying the critical and interrelated microbial metabolic and geochemical mechanisms associated with chromium *in situ* reductive immobilization and reoxidation from the molecular to the local field scale at the Hanford 100 site. Here we summarize modeling efforts to simulate the reactive transport processes observed in batch, laboratory flow-through column, and field experiments. A range of capabilities available in the in-house general-purpose reactive transport codes CrunchFlow and ToughReact are brought to bear in this effort. A major thrust of this task has been to develop and incorporate new methods to better constrain the rates of biogeochemical reactions. In particular, rates of respiration and fermentation are calculated with a biomass-explicit approach, with the fraction of carbon going to biomass growth (anabolic pathways) estimated from literature values and/or from the increase in reaction rate over time. In addition, isotope systematics are incorporated that account for fractionation processes during respiration and fermentation.

Modeling of flow-through column experiments focused on the two separate experimental conditions that showed the most removal of Cr(VI) from solution: fermentation and denitrification. In each case, parameters for reaction rate constants were derived from cell suspension experiments carried out with bacterial strains isolated from the site and column experiments. Under denitrifying conditions, Cr(VI) reduction was modeled as co-metabolic with nitrate reduction based on experimental observations and previous studies on a denitrifying bacterium derived from the Hanford 100H aquifer. Under fermentative conditions, the model assumed that Cr(VI) reduction was carried out directly by fermentative bacteria that converted lactate into acetate and propionate.

A 3D reactive transport model of the Hanford 100H site was developed for the field-scale experiment. Thermodynamic, kinetic, and Monod parameters were compiled and calculated based primarily on a wide variety of empirical laboratory data, conceptual models for bacterial growth, and field data. The model successfully captured many aspects of bacterial growth accompanying lactate degradation, propionate and acetate formation and transport, ^{13}C pathways to bicarbonate, and sulfate reduction. Carbon ($^{13}C/^{12}C$) and Sr ($^{87}Sr/^{86}Sr$) isotopes were used to independently constrain rates of microbially mediated processes, mixing with groundwater, and calcite dissolution rate. Cr isotopes were included in order to assess the extent and sustainability of Cr remediation by immobilization. Further, to evaluate the contributions on the overall kinetic Cr isotope fractionation factor, α_{kin}, we modeled Cr(VI) reduction along a 1D flow path using a novel multiple continuum approach. Simulations suggest that for a given reduction mechanism α_{kin} can vary over a large range. The low end of the range (=larger fractionation) is defined by the theoretical equilibrium fractionation factor, while high reduction rates and/or transport limitations induced by Cr(VI) transport to reactive sites can shift α_{kin} to values close to 1 (=no fractionation).