U.S. Department of Energy — Office of Science

Integrated Field-Scale Subsurface Research Challenge

Oak Ridge Field Research Center

Oak Ridge IFRC: Multi-Process and Multi-Scale Modeling and Data Analysis at the IFRC site, Oak Ridge, TN

Fan Zhang, Guoping Tang, Jack Parker, Jian Luo, Wei-Min Wu, Gengxin Zhang, Shelly Kelly, Tonia Mehlhorn, Jack Carley, Kenneth Kemner, Craig Criddle, Christopher Schadt, Wensui Luo, Baohua Gu, Brian Spalding, Scott Brooks, David Watson, and Philip Jardine

OAK RIDGE National Laboratory

Abstract

Multi-process and multi-scale modeling and data analysis of the former S-3 Ponds waste disposal site on the Oak Ridge Reservation have been performed to develop a practical model to simulate the coupled biogeochemical processes that control contaminant uranium mobility and ultimately to predict remediation of the geochemically complex site.

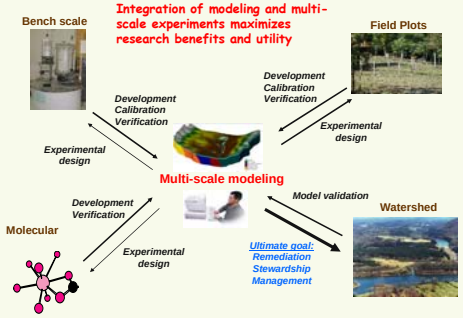
One such investigation was to test the feasibility of U(VI) reduction with slowly degrading electron donor, microcosm tests using uranium (VI) contaminated sediments and groundwater amended with calcium oleate were performed. A kinetic bioreaction model was developed to simulate the rapid removal of U(VI) from the aqueous phase associated with oleate degradation, acetate production and sulfate reduction.

Another investigation was designed to predict a major source of uranium-containing precipitates that have been observed in dolomitic gravel fill as a result of exposure to acidic groundwater contaminated with U, Al, SO_4^{2-} and NO^{3-}. An equilibrium precipitation/dissolution reaction model was developed to simulate the groundwater geochemical composition and U removal resulting from exposure to high pH carbonate gravel.

Local-scale models were developed for Area 2 and Area 3 to facilitate interpretation of flow and tracer studies and to investigate the impacts of aquifer heterogeneities and non-equilibrium mass transfer on degradation rates of contaminants. Reaction models developed through simulation of batch, column, and field experiments will be incorporated into the local-scale models to help interpret future on site investigation.

Long computational times and high spatial and temporal variability have been constraining our ability to predict site-wide contaminant transport in the field. We have made significant progress in reducing the computational time using parallel computers and are addressing observation variability in calibrating the site-wide transport model for ORIFRC site. We are currently moving toward development of a multi-scale, multi-process numerical model for the prediction of contaminant migration in complex watershed-scale environments.

Objectives

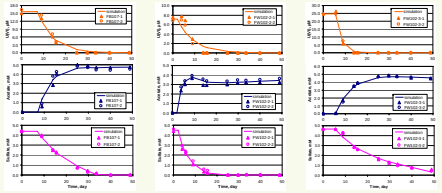


Batch Simulation of Oleate Stimulated U(VI) Reduction

A kinetic bioreaction model was developed to simulate the rapid removal of U(VI) from the aqueous phase associated with oleate degradation, acetate production and sulfate reduction, including

- *oleate degradation;*
- *sulfate reduction associated with oleate;*
- *sulfate reduction by acetate;*
- *sulfate reduction by hydrogen;*
- *U(VI) reduction associated with oleate; and*
- *U(VI) reduction by acetate.*

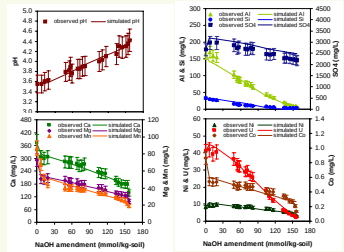
The significance of this investigation is our enhanced ability to predict biologically mediated U(VI) reduction



Small pH Adjustment Column Experiment Simulation

An equilibrium aqueous complexation model was used to compute speciation and saturation indices for considered minerals. Gibbsite, jurbanite, basaluminite and kaolinite were found to be over saturated above pH 3.5 to 4.5 during the experiment. Therefore, dissolution and precipitation kinetics of these minerals were included in simulations.

To model soil buffer capacity and pH-dependent ion exchange capacity, aquifer solids were treated as a hypothetical quadraprotic acid H_4X and a polyprotic base $Y(OH)_2$. The total ion exchange capacity was computed as CEC = H_3X^- + $2H_2X^{2-}$ + $3HX^{3-}$ + $4X^{4-}$ and AEC = YOH^+ + $2Y^{2+}$. A fraction of Al and Fe precipitates generated during titration were also assumed to produce surfaces with neutral or positive charge. Cation exchange reactions involving cationic K, Na, Al, Ca, Mg, Mn, Ni, Co, Sr and U aqueous species and anion exchange reactions involving SO_4^{2-} and anionic uranyl-carbonate species were considered in the model.

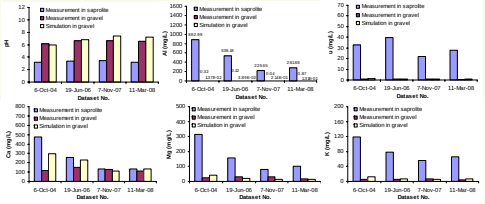


The significance of this investigation is our improved ability to predict geochemical induced U and Tc sequestration and the enhancement for up scaling processes to the field

Field Gravel Pathway U Precipitation Simulation

Precipitation/dissolution reactions of minerals determined using XANES and EXAFS were used in simulations to predict ground water composition in the gravel.

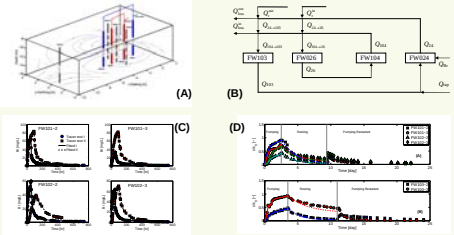
Mineral	Precipitation Reaction	log K
Dolomite	$CaMg(CO_3)_{2(s)} = Ca^{2+} + Mg^{2+} + 2CO_3^{2-}$	-16.54
Calcite	$CaCO_{3(s)} = Ca^{2+} + CO_3^{2-}$	-8.480
$K_4UO_2(CO_3)_3H_2O$	$K_4UO_2(CO_3)_3H_2O_{(s)} = 4K^+ + UO_2^{2+} + 3CO_3^{2-} + H_2O$	-43.9 ± 1.4
Basaluminite	$Al_4SO_4(OH)_{10}{\cdot}5H_2O_{(s)} + 10H^+ = 4Al^{3+} + SO_4^{2-} + 15H_2O$	22.5



The significance of this investigation is our improved ability to predict the precipitation and dissolution processes that control U(VI) migration to and from a very large sink/source of the contaminant. This source contributes to U concentrations well above drinking water standards 3 miles down the watershed.

Field scale simulation

Field pilot-scale U(VI) biostimulation experiments were conducted at Area 3 in Oak Ridge. Model simulations based on tracer test data include analyses of hydraulic control (A) and (B), biodegradation rate of injected electron donors (C), and nonequilibrium mass transfer between matrices and fractures (D). Applied approaches include linear-systems approach, inverse modeling, method of temporal moments, and reactor-system design *thus providing an improved predictive capability of plot scale biologically induced U(VI) reduction.*



Site wide modeling

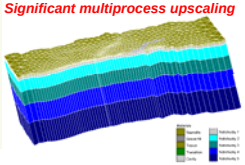
Coupled flow and transport model

- for nitrate migration in Nolichucky shale with density fact
- 3-d model with 10,323 triangular prism elements and 6,320 nodes
- domain 1,500 ft by 5,000 ft by 900 ft with duration from 1951 to 1985 to 2008
- full hydraulic conductivity tensor to account for 45 degree dipping
- exponential depth reduction factor for the hydraulic conductivity and porosity

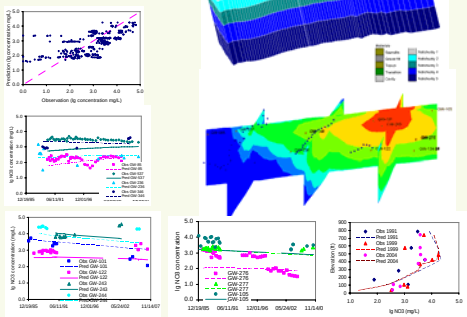
Model calibration

- 687 monitor wells
- Parallelized HGC 5.0 with OpenMP
- Parallel PEST
- 10 computer nodes x 8 CPU

Significant multiprocess upscaling



Calibration results



Publications

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