

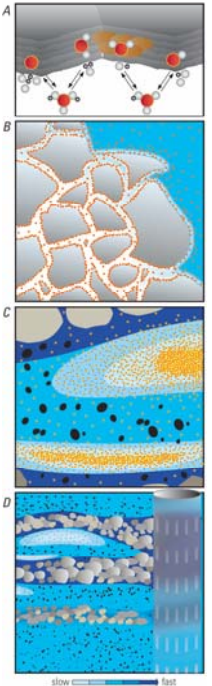
Conceptual Model Development: Site geochemical reaction and mass transfer model for U(VI) transport and field validation for the Hanford IFRC

Roy Haggerty¹, Douglas B. Kent², Chongxuan Liu³, Jun Yin¹, Deborah L. Stoliker², Chunmiao Zheng⁴, Rui Ma⁴,
Janek Greskowiak⁵, Glenn Hammond³, Peter Lichtner⁶, Jonathan Istok¹, and John M. Zachara³

¹ Oregon State University; ² US Geological Survey; ³ Pacific Northwest National Laboratory; ⁴ University of Alabama; ⁵ CSIRO; ⁶ Los Alamos National Laboratory



Multiscale Mass Transfer



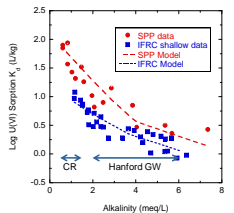
A) Adsorption-desorption. A dynamic equilibrium is established locally between aqueous U(VI) species and surface complexes. Aqueous and surface speciation is controlled primarily by pH and U(VI) (orange), carbonate (white), and calcium (gray). Dominant adsorption sites are on the edges of phyllosilicates and hydrous-iron-oxide weathering products (e.g., goethite).

B) Intragranular diffusion. Most of the contaminant U(VI) is adsorbed within mineral grains or aggregates. Mass transfer of U(VI) occurs via diffusion of dissolved U(VI) species through immobile intragranular water to mobile water outside the grains. Only dissolved U(VI) can diffuse, and that fraction depends on local chemical conditions in the pore space.

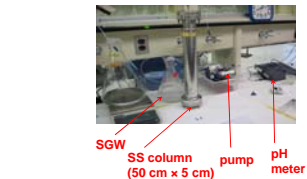
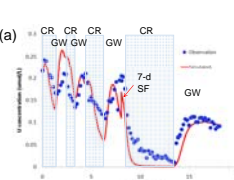
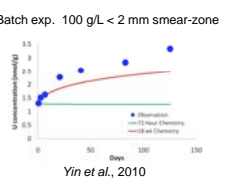
C) Mass transfer across regions with contrasting permeability. Regions (depicted as lenses) enriched in fine-grained material have higher reactive surface areas and, therefore, higher contaminant-U(VI) concentrations than surrounding coarse-grained regions. Lower permeability of these regions results in slower flow rates than surrounding coarse-grained regions.

D) Groundwater flow paths. Groundwater flow rates (different shades of blue) are higher through coarse-grained regions compared to adjacent regions. Wells cut across all flow paths but likely draw water preferentially from high permeability regions.

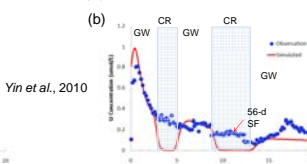
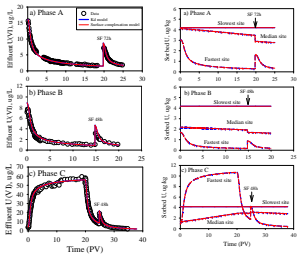
Laboratory Results



Alkalinity dependence of U(VI) adsorption ($\log K_d$) for IFRC smear zone sediments compared to SPP vadose zone sediments compared to SPP vadose zone sediments compared to SPP vadose zone sediments. Dashed curves are represent surface complexation models.

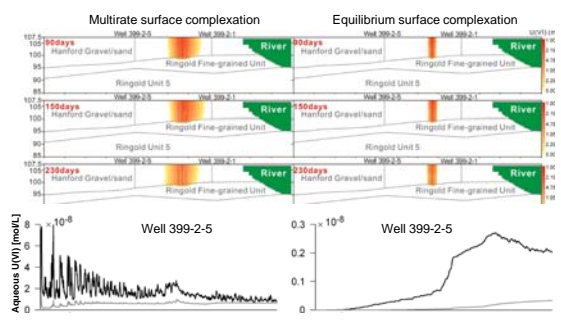


We conducted column experiments examining desorption in the presence of transient water chemistry and desorption from an intact column.

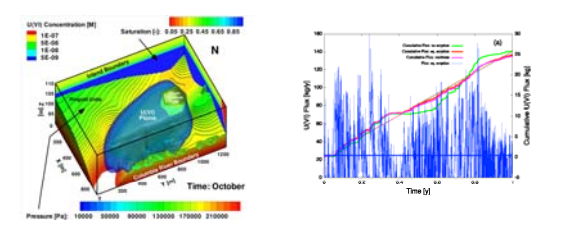


Desorption with transient chemistry from (a) lab-contaminated sed. and (b) field-contaminated sediment. Relatively poor fit in (b) is due to lack of calcite dissolution within model, which results in underestimated Ca during CR injection

Field Modeling



Field model of U(VI) transport in the presence of transient chemistry and transient GW flow [Ma et al., 2010].



Field model of U(VI) transport in the presence of transient chemistry and transient GW flow, showing U(VI) flux to Columbia River [Hammond & Lichtner, 2010].

Field Experiments: Plans

Placeholder – will be replaced with figure and condensed text. (It is also a test to see if you are proof-reading well...)

We will do two field experiments to test the mass transfer and geochemistry model. The experiments will be done under a controlled gradient to minimize river effects. The tests will be long enough to see timescales of many days or even weeks. Both experiments involve a well doublet (dipole, well pair – one injection one extraction).

The first experiment will be smaller scale and shorter in time – a few days. Its objective is to test field-scale mass transfer and geochemistry under controlled hydraulic conditions. The same procedure will be followed, but the wells used are farther apart and each stage of the experiment lasts longer. We will run the experiment until background concentrations are achieved.

The second experiment will be larger scale and longer in time. Its objective is to test field-scale mass transfer and U geochemistry under controlled hydraulic conditions. The same procedure will be followed, but the wells used are farther apart and each stage of the experiment lasts longer. We will run the experiment until background concentrations are achieved.

References

Bond, D. L., J. A. Davis, and J. M. Zachara (2008). Uranium(VI) release from contaminated vadose zone sediments: Estimation of potential contributions from dissolution and desorption. Ch. 14 in M. O. Barnett and D. B. Kent, eds., *Adsorption of Metals by Geomedia II: Variables, Mechanisms, and Model Applications*. Elsevier, Amsterdam, The Netherlands, p. 375-416.

Hammond, G. E., and P. C. Lichtner (in press 2010). Field-scale model for the natural attenuation of uranium at the Hanford 300 Area using high performance computing. *Water Resour. Res.*

Liu, C., J. M. Zachara, N. P. Galaku, and Z. Wang (2008). Scale dependent desorption of uranium from contaminated subsurface sediments. *Water Resour. Res.*, 44, W08413. doi:10.1029/2007WR006476

Ma, R., C. Zheng, H. Prommer, J. Greskowiak, C. Liu, J. Zachara, and M. Rockhold (in press 2010). Field-scale reactive transport model for U(VI) migration influenced by coupled multi-rate mass transfer and surface complexation reactions. *Water Resour. Res.*

Yin, J., R. Haggerty, D. L. Stoliker, D. B. Kent, J. D. Istok, and J. Greskowiak (in review, 2010). Transient groundwater chemistry and groundwater flow near a river: Effects on U(VI) transport near a river. *Water Resour. Res.*