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Coupled Geochemical and Hydrological Processes Governing the Fate and Transport of Radionuclides and Toxic Metals Beneath the Hanford Tank Farms

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Philip M. Jardine
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831-67038
865-574-8058
jardinepm@ornl.gov

Melanie Mayes
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831-6038
865-574-6398
mayesma@ornl.gov

Scott E. Fendorf
Stanford University
Dept. Geological and Environ. Sci.
Palo Alto, CA 94305
650-723-5238
fendorf@stanford.edu
509-373-6948

Students / post grads: 2 doctoral candidates
2 undergraduate student
1 post-masters
1 post-doctoral

This annual report describes 1.5 y of progress on a 3 y project. The project addresses the goals of the Environmental Management Sciences Program (EMSP) that seeks innovative basic research to benefit cleanup technologies and decision-making strategies for contaminated environments. Our project specifically addresses Hanford research needs in subsurface science by contributing to the objectives of the Tank Farm Vadose Characterization Project and the 200 Area Remedial Action Project which are components of the Hanford Site Groundwater/Vadose Zone Integration Project (Integration Project). The work described within is intended to advance the technological and scientific needs associated with the long-term management of the enormous in-ground inventories of $^{235/238}$U, ^{99}Tc, ^{60}Co, and Cr(VI) present at the Hanford site. We believe that scientifically defensible predictions of contaminant transport and strategies for remediation must be based upon a field-relevant understanding of coupled hydrological and geochemical processes that control subsurface contaminant fate and transport. This research project investigates the migration of $^{235/238}$U, ^{90}Sr, ^{60}Co, and Cr(VI) in undisturbed sediments from the Hanford site using realistic experimental protocols designed to delineate complex hydrological and geochemical processes controlling contaminant movement. The specific research goals of the following proposal are:

- to provide an improved understanding of how preferential vertical and lateral flow, and the formation of immobile water influence the transport of radionuclides and toxic metals in heterogeneous, laminated sediments.

- to quantify the rates and mechanisms of radionuclide and toxic metal interaction with the solid phase under various hydrologic conditions.

- provide new insights into how physical and mineralogical heterogeneities (e.g. stratification, pore regime connectivity, mineral composition along flowpaths) influence contaminant retardation and the degree of geochemical nonequilibrium during transport.

The overall research products will be (1) knowledge and information in previously unexplored areas of vadose zone fate and transport to support EM's performance/risk assessment and decision-making process for Tank Farm restoration; and (2) improved predictive strategies for assessing the transport rate and fate of $^{235/238}$U, ^{90}Sr, ^{60}Co, and Cr(VI) beneath the Hanford Tank Farms. Since this project will unravel fundamental contaminant transport mechanisms in complex porous media, the results will assist in improving our overall conceptual understanding and predictive capability of a variety of vadose zone issues within the DOE system (e.g. INEEL and Sandia).

Technical progress

FY02 technical progress

Significant findings regarding hydrologic processes in sediments beneath the tanks:

(1) The saturated and unsaturated hydraulic conductivity of cores acquired parallel to lithological layering was an order of magnitude more rapid than the conductivity of cores acquired perpendicular to lithological layering. Thus, lateral flow beneath the tank farms is a strong contributor to the spread of contaminants. Also, the conductivity of fine grained cores was an order of magnitude slower than core acquired in sandy media.

(2) The transport of the three nonreactive tracers at different degrees of unsaturation showed essentially no difference in breakthrough patterns when flow remained unrestricted along continuous lithological features as is the case for the horizontal cores. This suggested that physical nonequilibrium conditions were not occurring during lateral flow in these sediments. Thus matrix diffusion processes are not contributing to the attenuation of waste being released from the Hanford tanks allowing for more rapid transport rates beneath the tank farms.

(3) However, when flow is partially restricted by lithologic discontinuities as is the case for the vertical cores (e.g. alternating coarse and fine layers), perched water tables develop resulting in unstable wetting fronts and the prevalence of preferential flow. The early breakthrough and significant separation of tracer in the effluent suggest that large pockets of immobile water develop creating a condition of physical nonequilibrium between flowing domains and immobile domains. From a contaminant transport perspective this process is advantageous since the presence of large pockets of immobile water will significantly impede the vertical migration of waste being released from the Hanford tanks.

Significant findings regarding geochemical processes in sediments beneath the tanks:

(1) Batch and repacked column studies have shown that U, Cs, and Sr are strongly bound to the various sediments beneath the tank farms even under conditions where the solid phase pH is as high as 8.4. Modeled partition coefficients for U were on the order of 10 to 15 cm^3/g corresponding to retardation coefficients of 35 to 50. Modeled partition coefficients for Cs ranged from 30 to 90 cm^3/g, with the Hanford formation exhibiting a significantly larger retention capacity for the Cs versus the Ringold formation. Ternary exchange isotherms and column displacement experiments for the U-Cs-Ca system showed limited competitive sorption effects suggesting independent mechanisms controlled U and Cs sorption. This is consistent with experimental evidence showing that U sorption is often controlled by subsurface Fe-oxides and Cs sorption is often associated with the interlayer spaces of 2:1 phyllosilicates. On the other hand, the Sr-Cs-Ca system shows extensive competition for sorption sites.

(2) X-ray Absorption Spectroscopy (XAS) suggested that inner-sphere ternary carbonate complexes formed on Fe-oxides in sandy Ringold sediments while sorption of sheet silicates was the primary sorption mechanism in silty Ringold sediments. These data underscore the importance of understanding the sediment mineralogy to accurately access the fate and transport of uranium in the environment. The implications of the sorption reactions are that the downward vertical migration of U beneath the tank farms will be significantly slowed by interfacial geochemical sorption reactions.

(3) Reactive transport of Sr in undisturbed Hanford cores and U in undisturbed consolidated material of the Plio-Pleistocene unit have also been conducted. The Plio-Pleistocene unit was not conducive to either preferential vertical finger flow or to lateral flow, as no tracer separation (Br/PFBA/PIPES) was observed under our experimental conditions. The absence of distinct sedimentary bedding in the Plio-Pleistocene probably accounts for this observation. Our results therefore suggest that physical nonequilibrium processes will be minimal in the Plio-Pleistocene unit. Reactive transport of $^{235/238}U$ through the Plio-Pleistocene unit exhibited evidence of nonequilibrium, possibly influenced by kinetics or multi-site/multi-species reactivity. Sorption of $^{235/238}U$ was significantly enhanced under equilibrium conditions when compared to transport or flowing conditions, which suggests kinetic effects on sorption. Transport experiments were conducted in an atmosphere representative of subsurface conditions (1% CO_2), which resulted in the formation of a stable uranyl-carbonate complex. Therefore, it is also likely that the large complex may have different affinities for subsurface sorption sites. Transport through the Plio-Pleistocene unit has demonstrated the presence of geochemical nonequilibrium while physical nonequilibrium processes were minimal, thus resulting in a greater understanding of the relevant mechanisms governing contaminant interactions in the Hanford subsurface.

FY03 technical progress

Collection of additional cores of undisturbed Hanford sediments:

(1) Six large (0.025m x 0.025m) undisturbed cores were collected at the Environmental Restoration Disposal Facility (ERDF) in May for the purpose of conducting additional transport experiments. Three cores in the vertical direction and three cores in the horizontal direction will be used to quantify lateral and vertical flow as a function of water content. Six small (0.008m x 0.006m) cores in both vertical and horizontal directions were collected for the purpose of determining the capillary pressure-saturation curve. Four small (0.008m x 0.006m) cores were collected to

examine the microscopic sedimentary pore structure of the sediments.

(2) Additional samples of Hanford sediments from a variety of boreholes around the Reservation (C3177, C3828, C3391, C4124, and B8810) were obtained from the Hanford Core Library. These sediments will expand our investigation of the Hanford subsurface to depths greater than, and lithologies different from our ERDF sediments, thus increasing the relevance of our work. Disturbed samples of these sediments have been used in batch and kinetic studies of Cr(VI) and U(VI). Intact samples of these sediments will be encapsulated into an apparatus for conducting unsaturated, reactive transport experiments using Cr(VI) and U(VI). This technique will allow us to quantify coupled hydrologic and geochemical processes in deeper cores in which the sedimentary pore structure is reasonably intact.

Significant findings regarding coupled hydrological and geochemical processes in sediments beneath the tanks:

(1) Repacked column experiments were conducted with $SrEDTA^{-2}$ as it has been suggested that organic chelating agents may be responsible for the accelerated transport of strontium. Results indicated that the complex dissociated when contacted with the soil allowing strontium to be transported as Sr^{+2}. It was determined that Fe(III)-bearing oxides striped the EDTA from the Sr thus forming Fe(III)EDTA. This reaction is consistent with the larger association constant for Fe(III)EDTA (log K = 25) relative to SrEDTA (log K= 10). Therefore, Sr^{+2} was used in reactive transport experiments in unsaturated undisturbed Hanford cores. Unsaturated reactive transport of Sr in the undisturbed Hanford flow bedding parallel core shows strong reactivity which is consistent with batch and repacked column studies. The core was conducive to physical nonequilibrium processes (PNE) such as preferential finger flow and the formation of immobile water, as suggested by tracer separation and early breakthrough compared to saturated flow. The Hanford formation is a heterogeneous media of unconsolidated laminated coarse and fine sands that are interbedded with small-scale clay layering. It is this layering that causes PNE processes to develop under unsaturated conditions. Experimental results from batch and repacked columns predicted an increase in sorption of Sr in the undisturbed core due to the increase in sediment mass. However, accelerated transport of Sr was observed in the core possibly due to PNE processes such as preferential flow and media by-pass. Transport through the Hanford formation has determined the presence of physical nonequilibrium processes which influence the geochemical processes controlling reactive transport. These investigations are on-going.

(2) The mobility of Cr(VI) appears to be highly variable in Hanford sediments, with reduction capacity ranging from zero to significant depending on the availability of solid phase Fe(II). Cr(VI) was observed to be stable (i.e., no reduction to Cr(III)) over long time periods in the upper and middle Ringold Formations, and in the fine-grained Hanford from ERDF. These results were confirmed by the introduction of Cr(VI) into a large undisturbed ERDF core, in which complete recovery of Cr(VI) was observed. In contrast, the reduction of Cr(VI) to Cr(III) was robust in sediments from the C3177 borehole and in ERDF coarse-grained sediments, as evidenced by loss of aqueous Cr(VI). The contaminated sediment samples were analyzed using X-ray Absorption Fine Structure (XAFS) at the Advanced Photon Source (APS), which is a technique for determining solid-phase speciation and mineralogy. Reduction to Cr(III) was confirmed, and an association between Fe(II) and Ti suggested that ilmenite and other Fe(II)-bearing minerals acted as the reductants. Within the C3177 sediments, greater reduction to Cr(III) was observed in the more coarse-grained sample taken from 215’ depth; while the lowest amount of reduction was observed in the more fine-grained sample taken from 150’ interval. This coupled with the results from the ERDF samples suggests that there is a correlation between increasing grain size and

reduction of Cr(VI) to Cr(III). The absence of reduced Cr(III) in the Ringold is probably related to its greater age, which resulted in transformation of the original Fe(II) sources to Fe(III). Additional work is ongoing to address the coupled hydrology and geochemistry of Cr(VI) mobility in the Hanford subsurface.

(3) The mobility of U(VI) in intact Ringold and Hanford sediments was tested in three unsaturated, undisturbed cores. Batch experiments suggest that the adsorption of U(VI) is controlled by U-CO_3 complexes at natural sediment pH, and that adsorption was both kinetically-limited and nonlinear. Though the adsorption of U was significant (R ~ 10) under equilibrium conditions, retardation in the large unsaturated cores for equivalent residence times was small (R ~ 2). This is primarily a function of the hydrologic regime in which large proportions of reactive media are bypassed or kinetically-limited under unsaturated conditions, thus reducing available surface sites for U interactions. Subsurface contaminant distributions suggest a correlation between fine-grained beds and U retention, but it is uncertain if this is controlled by the hydrology (water content), geochemistry (mineralogy), or both (based on recently published subsurface inventories beneath the B-BX-BY and S-SX tank farms). The sensitivity of U adsorption and precipitation to changes in flow regime in intact sediments is a topic that needs much more exploration before all relevant mechanisms can be identified.

(4) The rates and mechanisms of Co(II)EDTA interactions with ~20 Hanford area sediments were quantified in batch kinetic experiments, and it was found that capacity of the sediments to produce oxidized Co(III)EDTA ranged from zero to near 100% . This has important implications regarding the enhanced mobility of ^{60}Co due to the high stability constant (log K ~40) of the Co(III)EDTA complex. A good correlation ($r^2 = 0.9$) was found between the sediment Mn content and the production of oxidized Co(III)EDTA, which will improve our predictive capability regarding the mobility of ^{60}CoEDTA in the Hanford subsurface.

(5) The transport of Co(II)EDTA through three large, undisturbed Ringold and Hanford cores was quantified under unsaturated conditions. The oxidation of Co(II)EDTA to Co(III)EDTA was dominant in the cores, which was consistent with the preliminary kinetic experiments. Differences in the amount of oxidized Co(III)EDTA were observed, depending upon the direction of flow relative to the direction of bedding. These results will be used to interpret the influence of coupled hydrological and geochemical processes in Hanford subsurface materials, and this work is on-going.

Implications:

The experimental and numerical results from this research will provide knowledge and information in previously unexplored areas of vadose zone fate and transport to support EM's performance/risk assessment and decision-making process for Tank Farm restoration. By unraveling fundamental contaminant transport mechanisms in complex porous media, we will provide an improved conceptual understanding and predictive capability of a variety of vadose issues within the DOE system. The implications of our work suggest that coupled hydrological and geochemical mechanisms will generally act to retard the migration of contaminants during transport through the vadose zone. The extent to which this occurs will depend upon the sedimentary pore structure, moisture content, the kinetics and the surficial geochemistry of contaminant interactions.

Future accomplishments

FY 2003:

We have communicated these results to the site end-users, Ch2m HILL Hanford Group, and have submitted an informal proposal to CHG at their request. We expect to open a dialogue with the end-users regarding exact needs and directions for the future, which should enable us to strongly tailor our future experiments to suit their needs.

Our future studies will also focus on coupling the mechanisms of geochemical Cr(VI) reduction and U(VI) adsorption with the hydrology of the deep intact cores from the Reservation. Additional U(VI) equilibrium and kinetic studies will be completed on the Core Library sediments, and on clays isolated from the ERDF samples, which will provide additional geochemical parameters. The quantification of geochemical interactions with the isolated clays will provide insights into the connection between decreasing moisture content and increased and/or decreased retardation due to changes in moisture content. The relationship between intact core hydrology and sedimentology will be confirmed by performing dye tracing and core dissection on select Ringold and Hanford cores. Uranium and CoEDTA transport experiments will be conducted on the new Hanford cores from the ERDF, under a variety of hydrologic (% saturation) and geochemical (caustic solutions) conditions. These experiments should provide new information regarding coupled hydrological and geochemical processes that are relevant to the Hanford vadose zone.

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JOHN D
Hertz