Fate and Transport of Radionuclides Beneath the Hanford Tank Farms: Unraveling Coupled Geochemical and Hydrological Processes in the Vadose Zone

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Goals

To provide an improved understanding and predictive capability of coupled hydrologic and geochemical mechanisms that are responsible for the accelerated migration of radionuclides in the vadose zone beneath the Hanford Tank Farms.

Objectives

- To provide an improved understanding of how lithological discontinuities within the sediments influence the propensity for preferential flow and matrix diffusion at different water contents.

- To quantify the significance of downward vertical advection, lateral spreading, and physical nonequilibrium processes on radionuclide transport under variable hydrologic conditions.

- To quantify the rates and mechanisms of ^{137}Cs, $^{235/238}U$, and ^{99}Tc interactions with the solid phase under various hydrodynamic conditions and to determine how physical heterogeneities (i.e. stratification, pore regime connectivity) influence the retardation and degree of geochemical nonequilibrium during contaminant transport.

Rationale

- The accelerated transport of ^{137}Cs, $^{235/238}U$, and ^{99}Tc within the vadose zone beneath the 200-West Area tanks has been recognized, although the mechanisms responsible for the vertical migration of the radionuclides is unclear.

- There is limited experimental data on the hydrological and geochemical processes controlling the fate and transport of radionuclides in the far-field vadose zone beneath the Hanford tanks.

- Fundamental experimental research on the rates and mechanisms of radionuclide migration in the vadose zone will enhance our conceptual understanding and predictive capability of contaminant transport beneath the tank farms.

Approach

- Field relevant, long-term unsaturated flow and transport experiments in undisturbed Hanford sediments.

- Multiple tracer strategies for quantifying preferential flow and nonequilibrium mass transfer at various water contents.

- Use of a variety of novel surface analyses techniques (x-ray computed tomography, x-ray absorption spectroscopy, hyperquenching fluorescence) to quantify the distribution and chemical environment of contaminants as a function of sediment lithology and water content.

Large undisturbed sediment columns (0.3 m diam. X 0.3 m length) were acquired from a road excavation within the Upper Ringold formation and the Plio-Pleistocene unit. Each material had nearly identical physical and geochemical characteristics as sediments beneath the 200-West Area of the Hanford tank farms. The bluffs from which the cores were extracted was a few kilometers east of the Hanford site and about 300 m above the Columbia river.

A rotary coring apparatus, that used a diamond bit core barrel, was fabricated and mounted onto a backhoe bucket so that sediment cores could be obtained at any angle within the various formations.

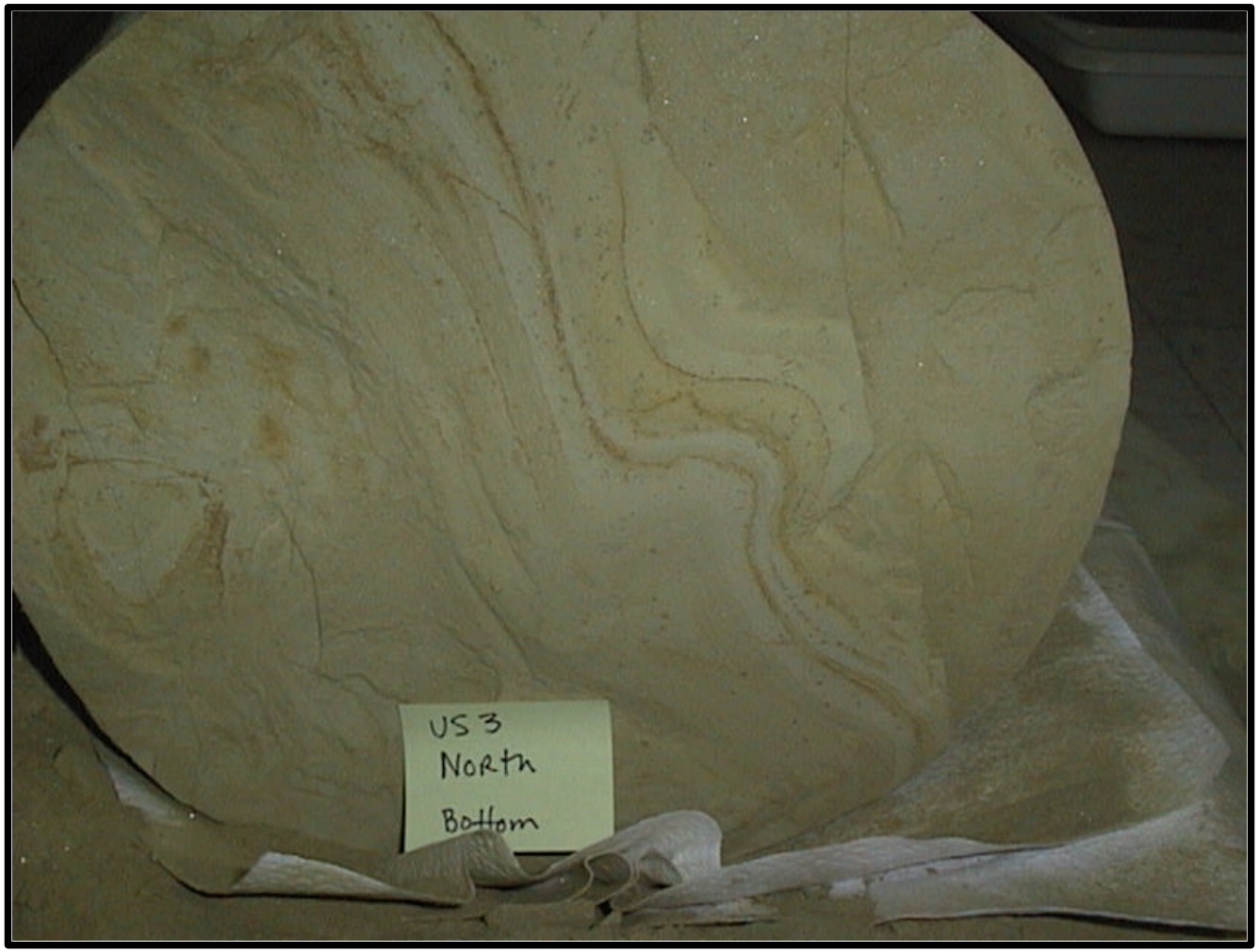
Within the Upper Ringold Formation, undisturbed cores were drilled parallel to lithological layering (horizontal into the formation as shown above) and perpendicular to lithology (vertically downward). The different core types will serve to quantify the significance of downward vertical advection versus lateral movement of contaminants due to tension-dependent anisotropy.

In order to extract the horizontal and vertically drilled cores (vertical cores are shown above) from the formation, the annulus separating the formation from the core was filled with expandable foam and allowed to harden.

The cores were carefully removed from the formation by hand excavation. The rigid foam material not only maintained the integrity of the cores, it also contained an impression of the various sediment layers from the surrounding porous media, including Fe-oxide staining patterns. Thus, it was possible to infer the distribution of flow paths and mineralogical heterogeneities within the cores.

Shown is a representative core that was drilled parallel to lithological layering within the Upper Ringold formation with the harden foam showing fine scale sediment layering with Fe-oxide staining. The sediment face was coated with paraffin wax in order to maintain core integrity during transport to Oak Ridge National Laboratory (ORNL).

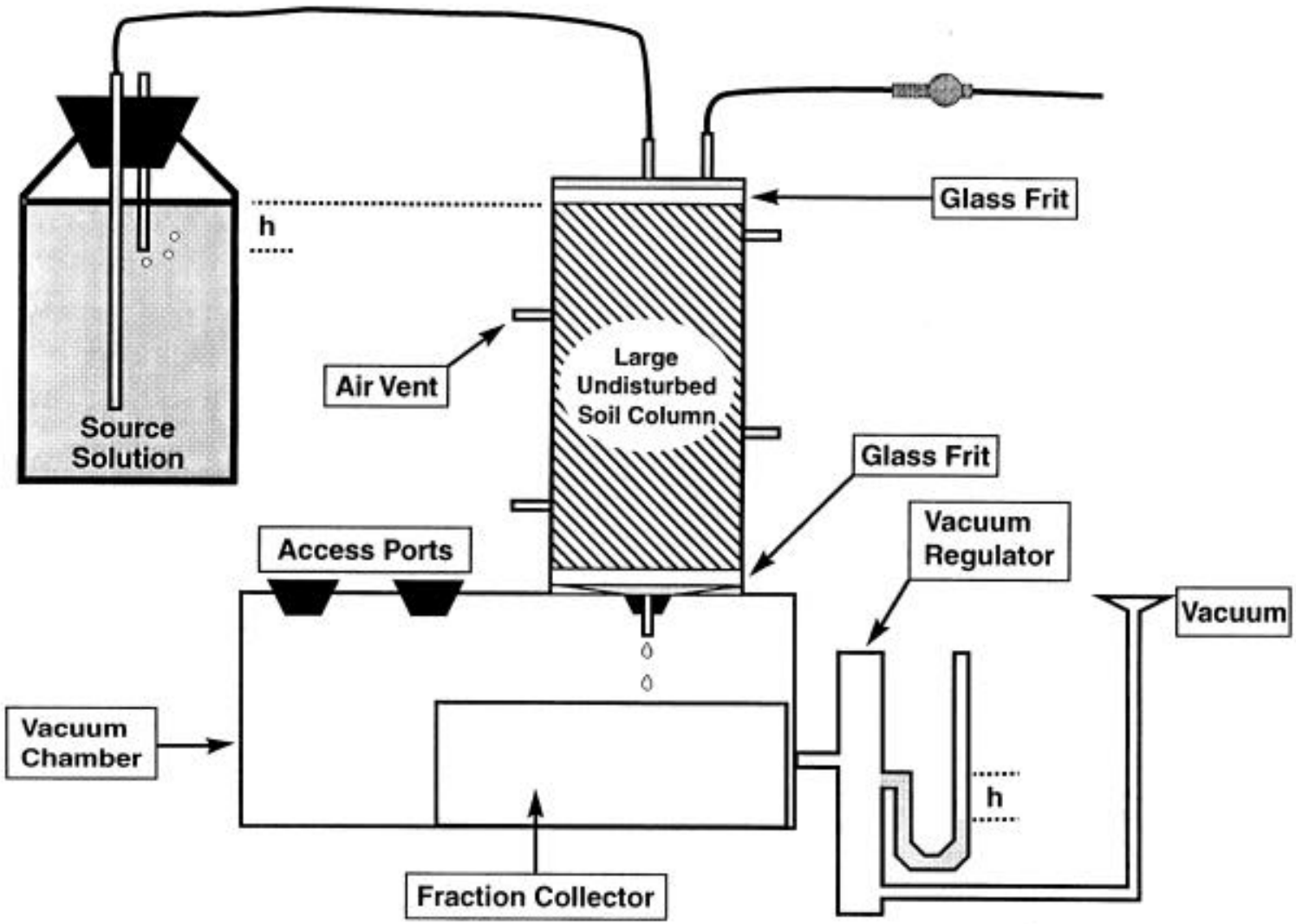
This is the face of a representative core that was drilled parallel to lithological layering showing fine scale laminated silts and sands with Fe-oxide coatings. These mineralogic features are known to strongly influence the migration of radionuclides in the subsurface. Discrete vertical fractures, perpendicular to bedding planes, were also noted in many of the columns.



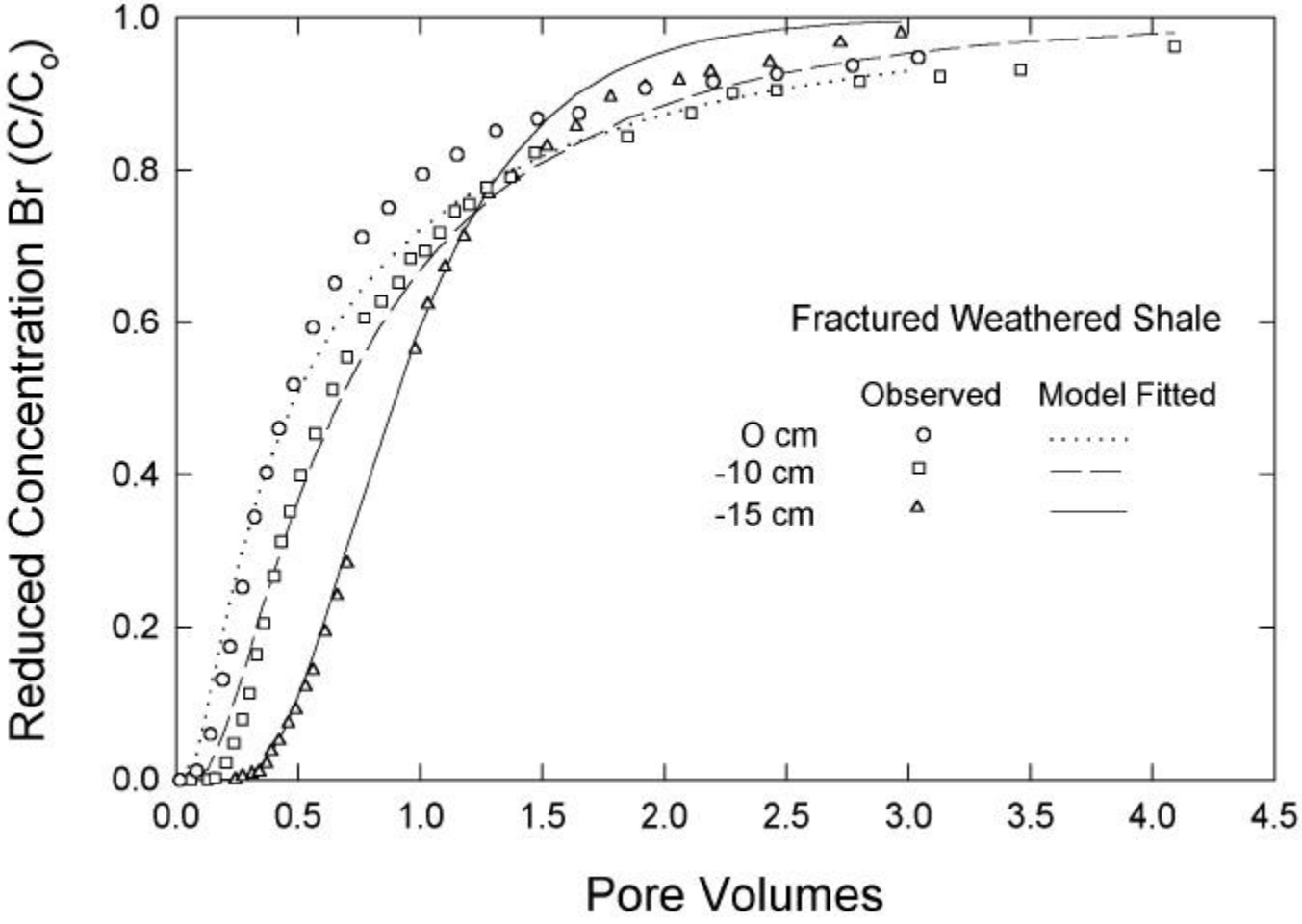
This is the face of a representative core drilled perpendicular to lithology (vertically downward) showing the fluvial depositional environment of the Upper Ringold formation. All cores were carefully transported to ORNL and prepared for unsaturated flow and transport experiments as described in Jardine et al. (1993 a,b Soil Sci. Soc. Am. J.) and Reedy et al. (1996, Soil Sci. Soc. Am. J.).

The rotary coring apparatus was successfully used to extract undisturbed cores from the Plio-Pleistocene unit which is essentially a cemented carbonate. This particular layer beneath the tank farms is of considerable interest with regard to U transport. The interaction of U with this material results in the formation of stable U-CO_3 species which may accelerate the migration of this contaminant through the underlying Ringold formation with subsequent groundwater interception.

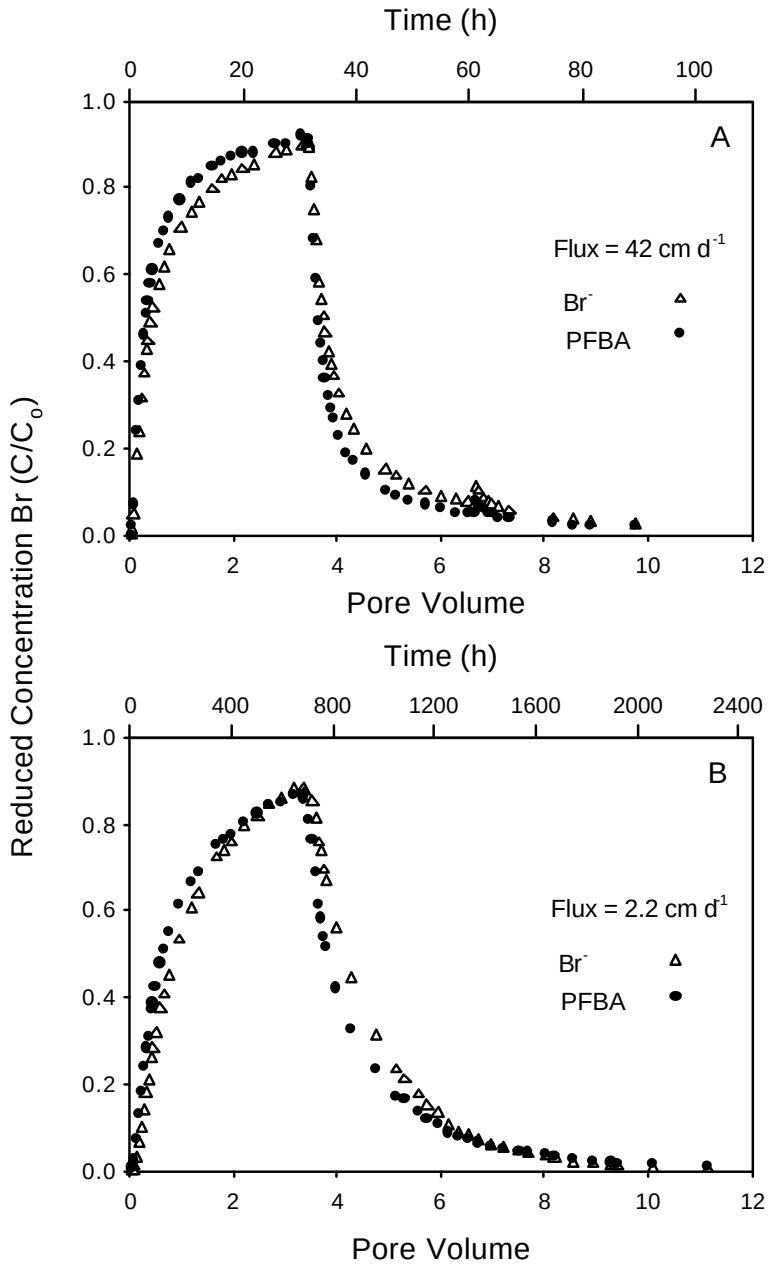
The rotary coring apparatus was not used in the Hanford formation since this material was essentially devoid of small scale structural integrity. Bulk samples were acquired for constructing repacked columns which will be used for assessing radionuclide fate and transport mechanisms in this material. Note the fine photography skills used by this investigator.



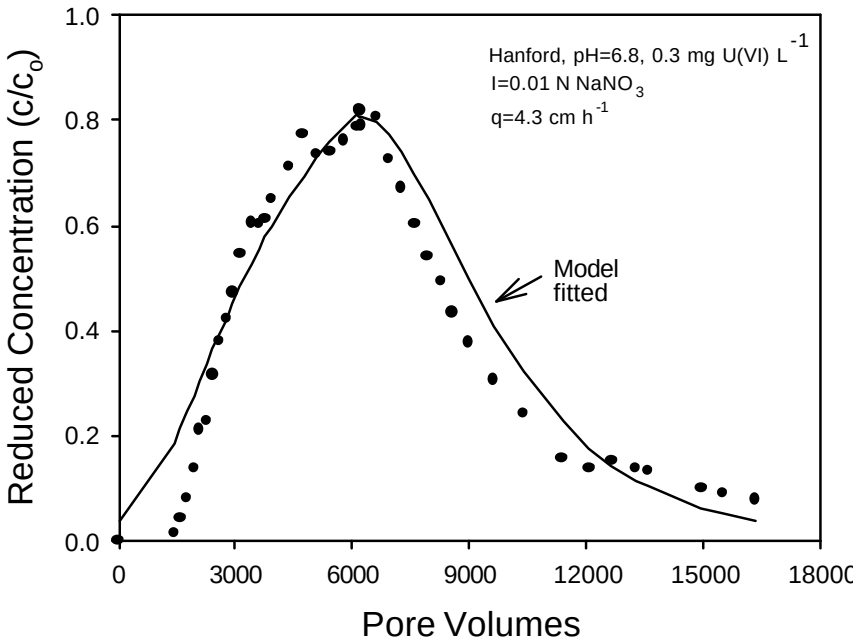
Schematic diagram of the unsaturated flow apparatus illustrating the constant pressure head (h) design. The experimental set-up is designed for investigations of transient or steady-state reactive or nonreactive solute flow and transport in vadose zone soils and sediments. Porous membranes will replace glass frits in the current study.

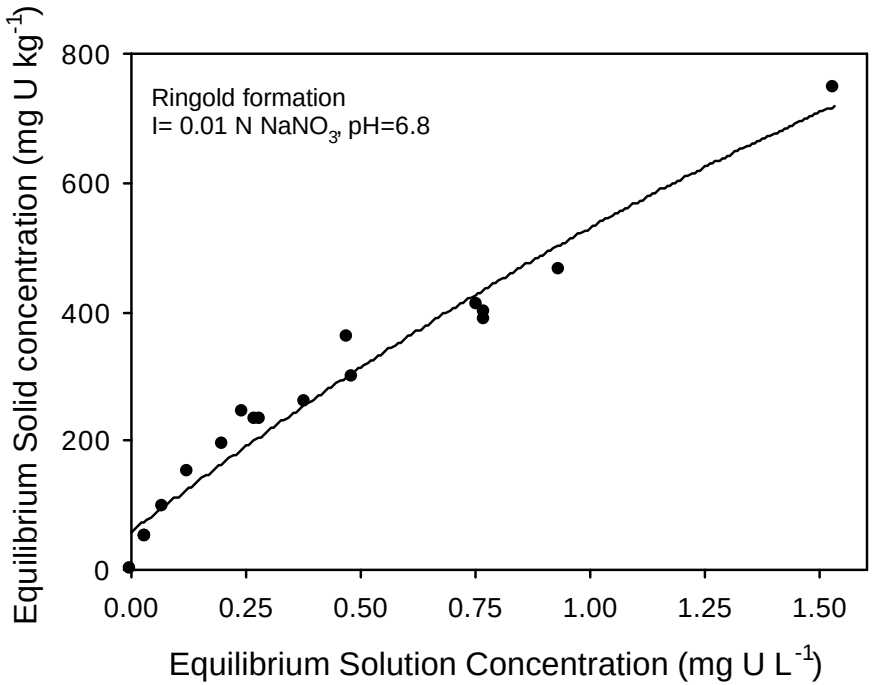


An example of nonreactive Br transport at three different pressure heads (i.e. water contents) for an undisturbed column of structured soil. These experiments showed the dependence of water content on preferential flow and time-dependent mass-transfer between various pore regimes of different size. Similar experiments are underway for the undisturbed cores acquired in this study.

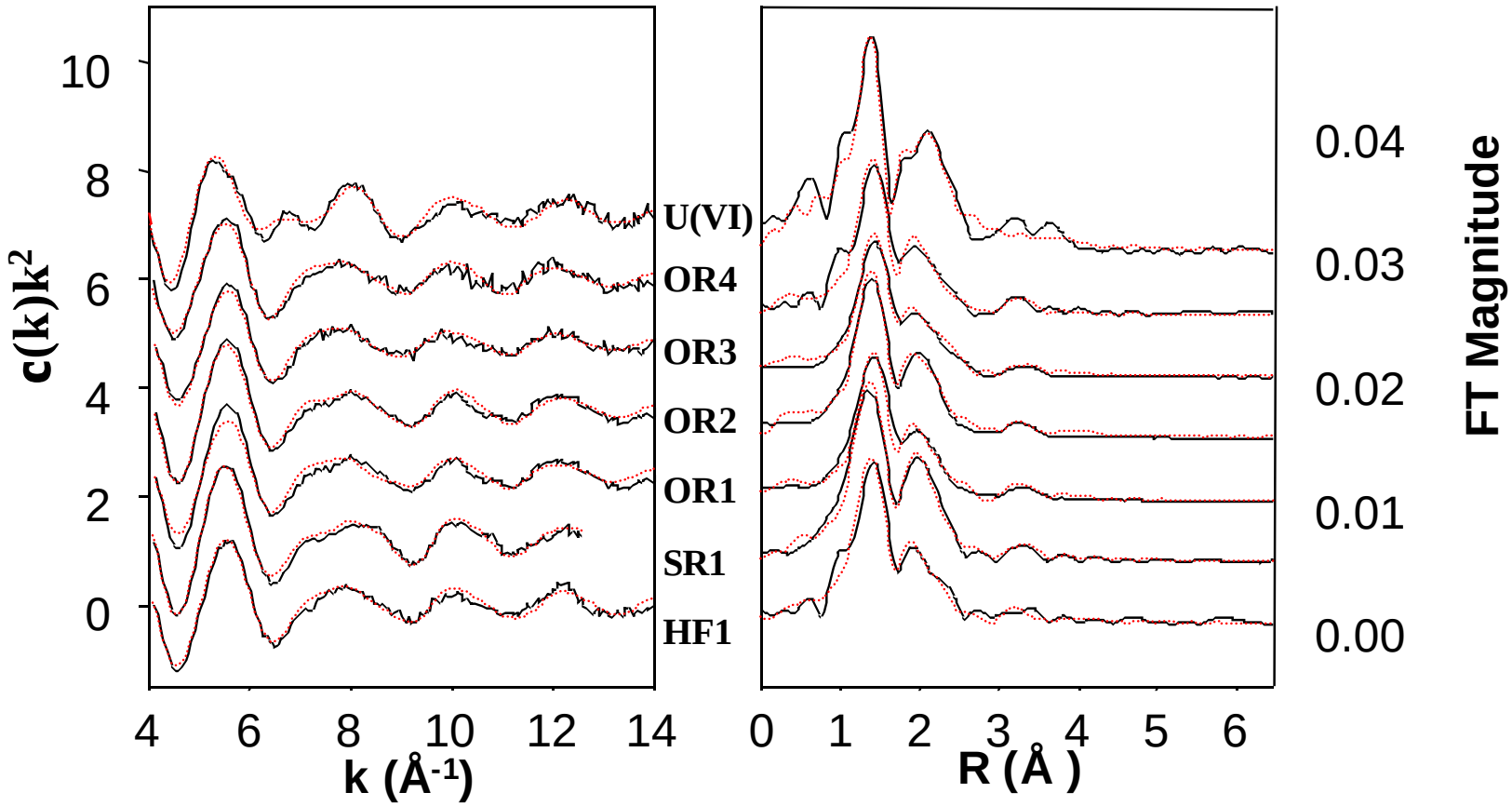


An example of breakthrough curves for the simultaneous injection of two non-reactive tracers, Br and PFBA, at two different fluxes in an undisturbed column of structured soil. The 40% larger molecular diffusion coefficients (D_e) of PFBA relative to Br, results in accelerated breakthrough of the former, thus confirming nonequilibrium mass transfer between different sized pore classes. In the current study, three nonreactive tracers, Br, PFBA, and PIPES will be used to assess nonequilibrium mass transfer as a function of water content for the various undisturbed cores. The D_e for PIPES > PFBA > Br.

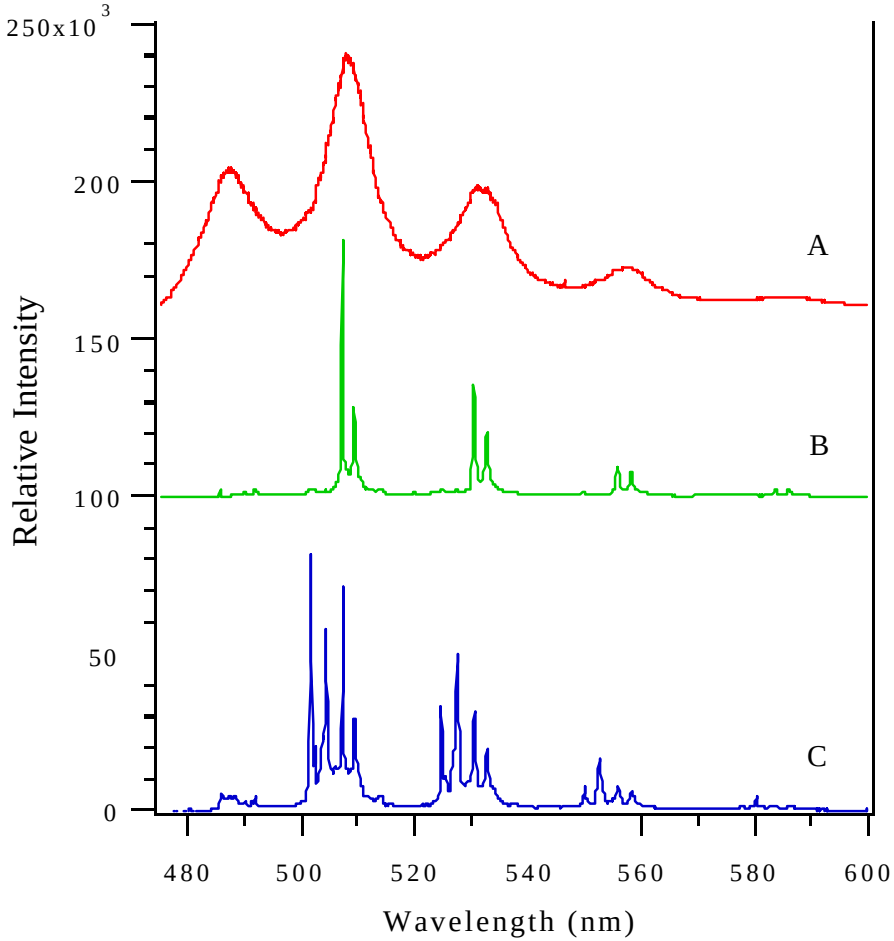




An example of U(VI) transport and reactivity in Upper Ringold sediments. The breakthrough of U(VI) was significantly delayed relative to nonreactive Br (essentially coincided with the Y-axis), with k_d values as high as 1000 cm^3 g^{-1}. Geochemical retardation mechanisms can significantly impede U(VI) transport in subsurface environments.



An example of how X-ray Absorption Spectroscopy (XAS), using synchrotron based radiation, is used to decipher the sorption mechanisms of U on Oak Ridge, Savannah River, and Hanford sediments. Ternary complexes of uranyl carbonate were the dominant surface complexes in the Upper Ringold sediments, the lesser amounts of bidentate inner-sphere complexes on Fe-oxides. However, much of the U is present as disordered inner-sphere and outer-sphere surface complexes suggesting that the surface bound U is labile and that sorption is reversible. This technique will be applied to bulk and core samples used in the flow and transport studies to unravel the mechanisms of Cs and U sorption to the various Hanford sediments.



Shown are spectra of uranyl nitrate crystals at room temperature (A) and 4 K (B, C) using laser induced line-narrowed hyperquenching fluorescence. This exploratory technique is designed to detect and speciate U at very low surface coverages. It will be applied to bulk and core samples used in the flow and transport studies in order to identify U speciation at the mineral-water interface.

Research Products

- A new conceptual understanding of radionuclide migration beneath the tank farms which will enhance our ability to manage the enormous in-ground inventories of ^{137}Cs, $^{235/238}U$, and ^{99}Tc present at the Hanford site.

- 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.

- An improved predictive tool for assessing the transport rate and fate of ^{137}Cs, $^{235/238}U$, and ^{99}Tc beneath the Hanford tank farms.

- Rapid dissemination of results to scientific and user community through peer-reviewed publications and technology transfer.