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

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Goals

To provide an improved understanding and predictive capability of coupled hydrological 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 the formation of immobile water regimes 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, $^{235/238}U$, ^{90}Sr, 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 both the near-field and 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.

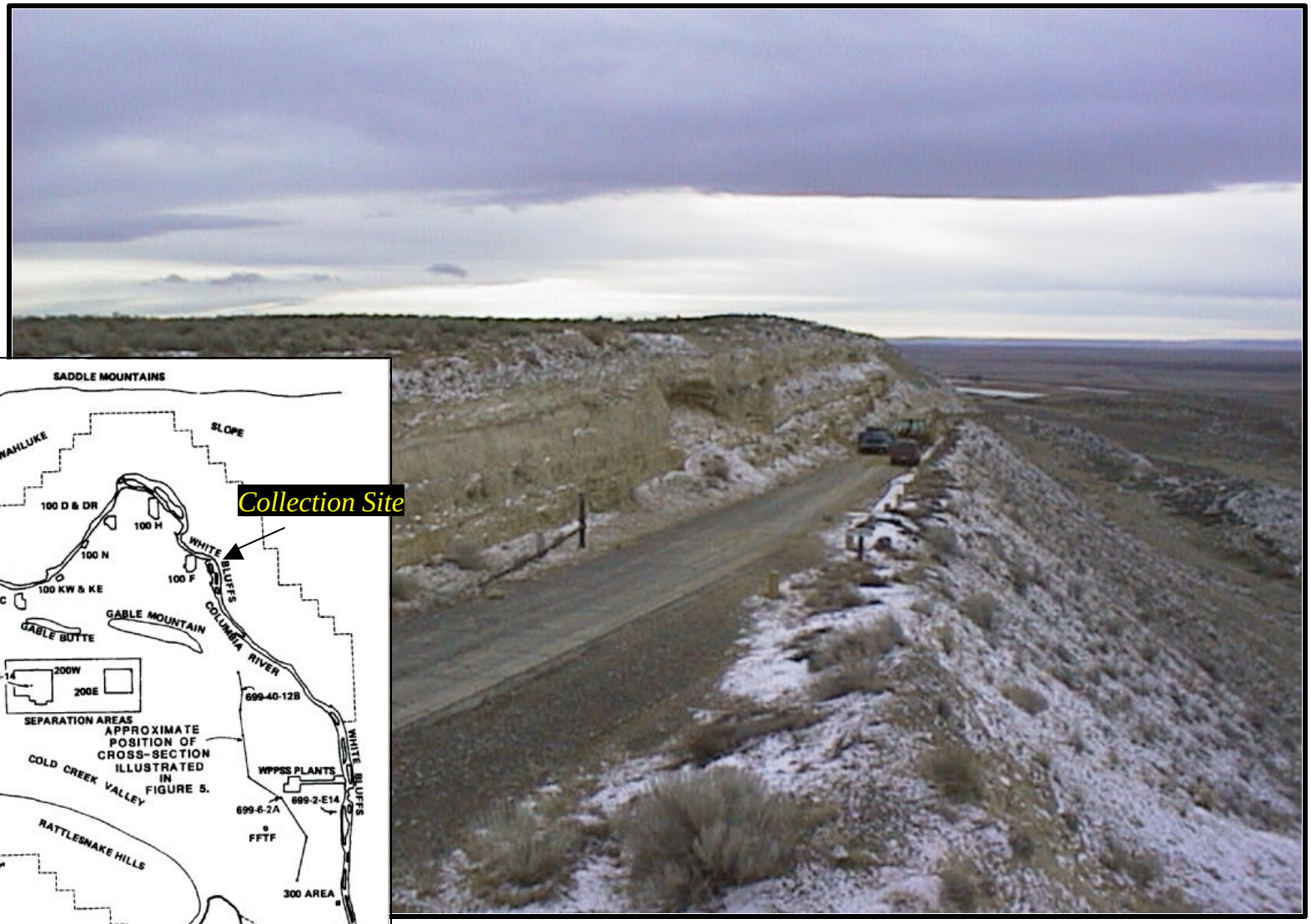
Approach

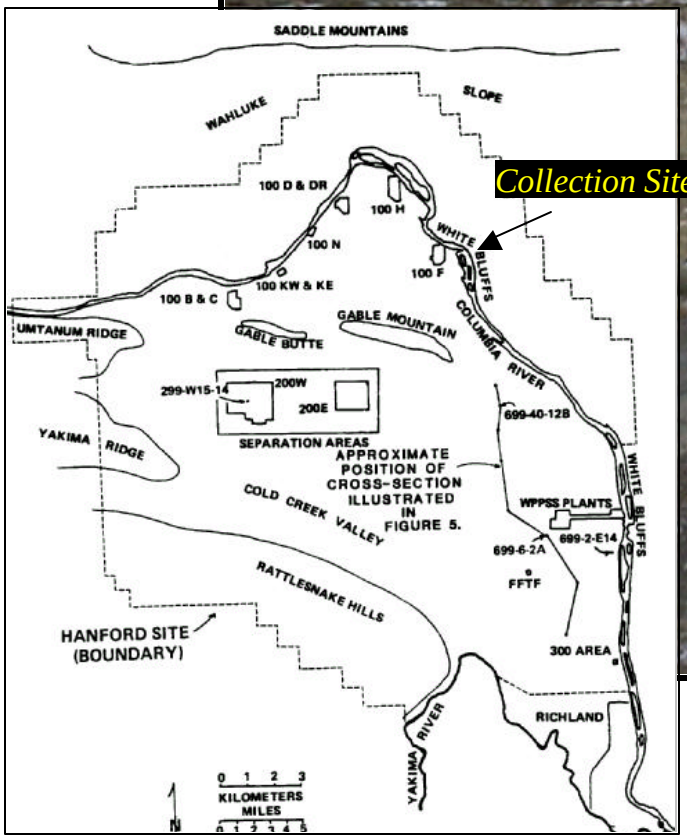
- Field relevant, long-term unsaturated flow and transport experiments in undisturbed sediments within the Hanford and Upper Ringold formations involving ^{137}Cs, $^{235/238}U$, ^{90}Sr, and ^{99}Tc.

- Multiple tracer strategies (Br^-, PFBA, PIPES) for quantifying preferential flow and nonequilibrium mass transfer at various water contents.

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

Upper Ringold Formation and the Plio-Pleistocene unit at the White Bluffs

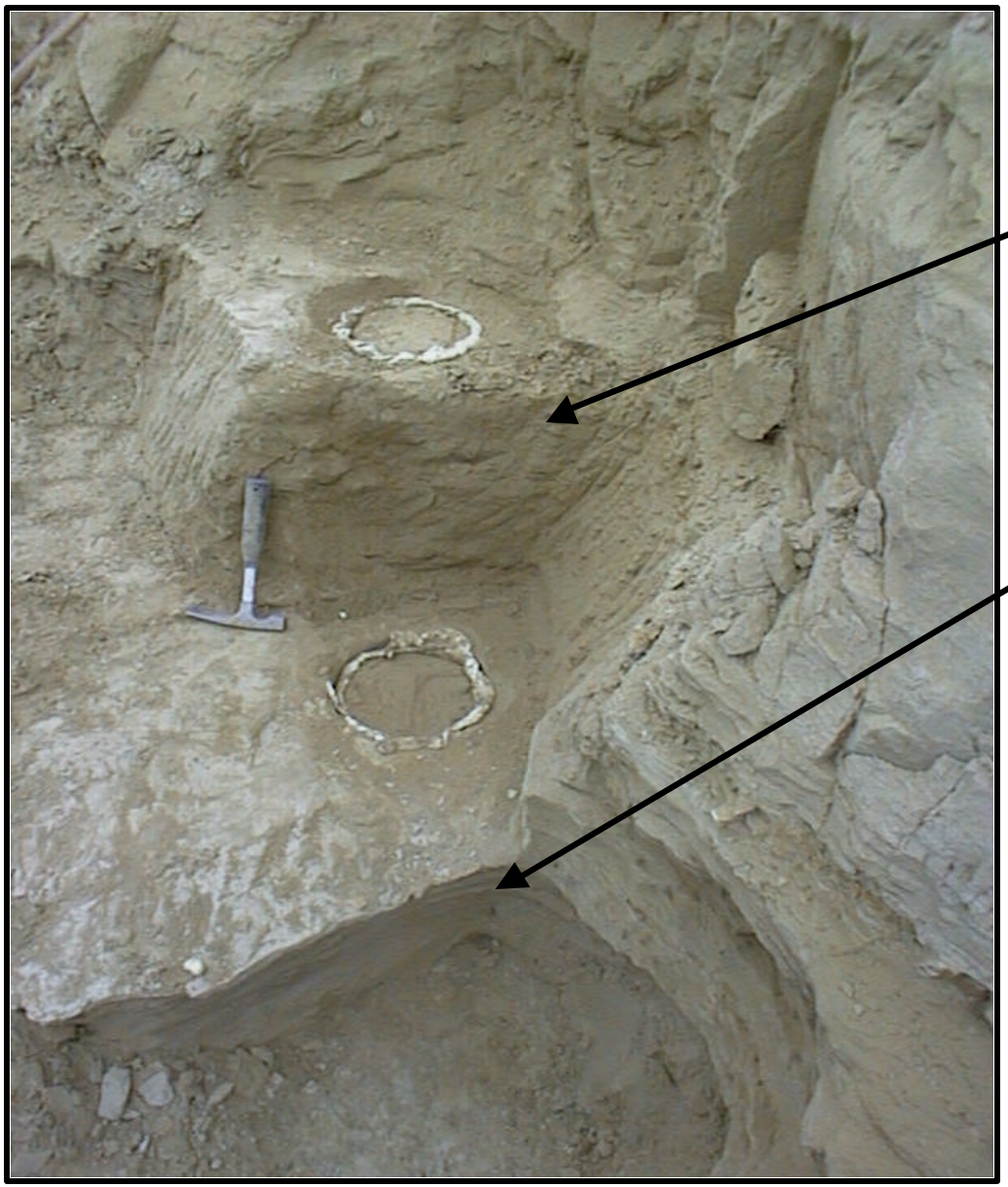




Upper Ringold formation consisting of laminated sands and silts

Rotary coring apparatus for obtaining undisturbed cores at any angle within the formation

Extraction of vertical cores from the Upper Ringold



Upper Silts/Sands

42 % sand, 56 % silt, 2 % clay

Lower Silts/Sands

57 % sand, 42 % silt, 1 % clay

Core extraction involved filling the annulus separating the formation from the core with expandable foam and allowing it to harden.

Vertical Core
Flow Cross-Cuts Beds (FXB)

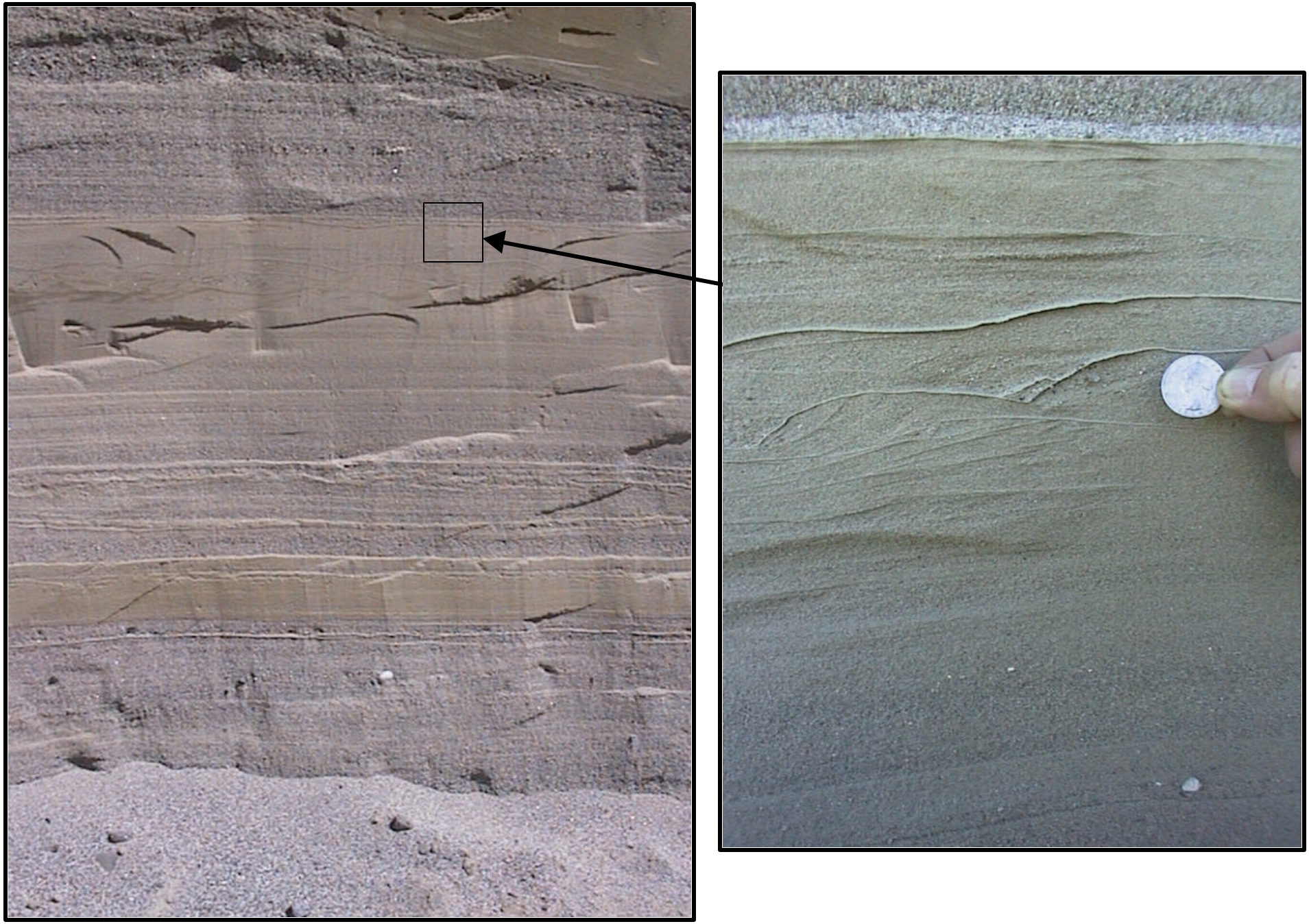
Horizontal Core,
Flow Parallel to Beds (FBP)

Different core types will serve to quantify the significance of vertical versus lateral transport

Plio-Pleistocene unit (caliche)

Hanford Formation

Hanford formation consisting of layered coarse and fine sands with some silt/clay



Extracting a horizontal core from the Hanford formation

Experimental Setup

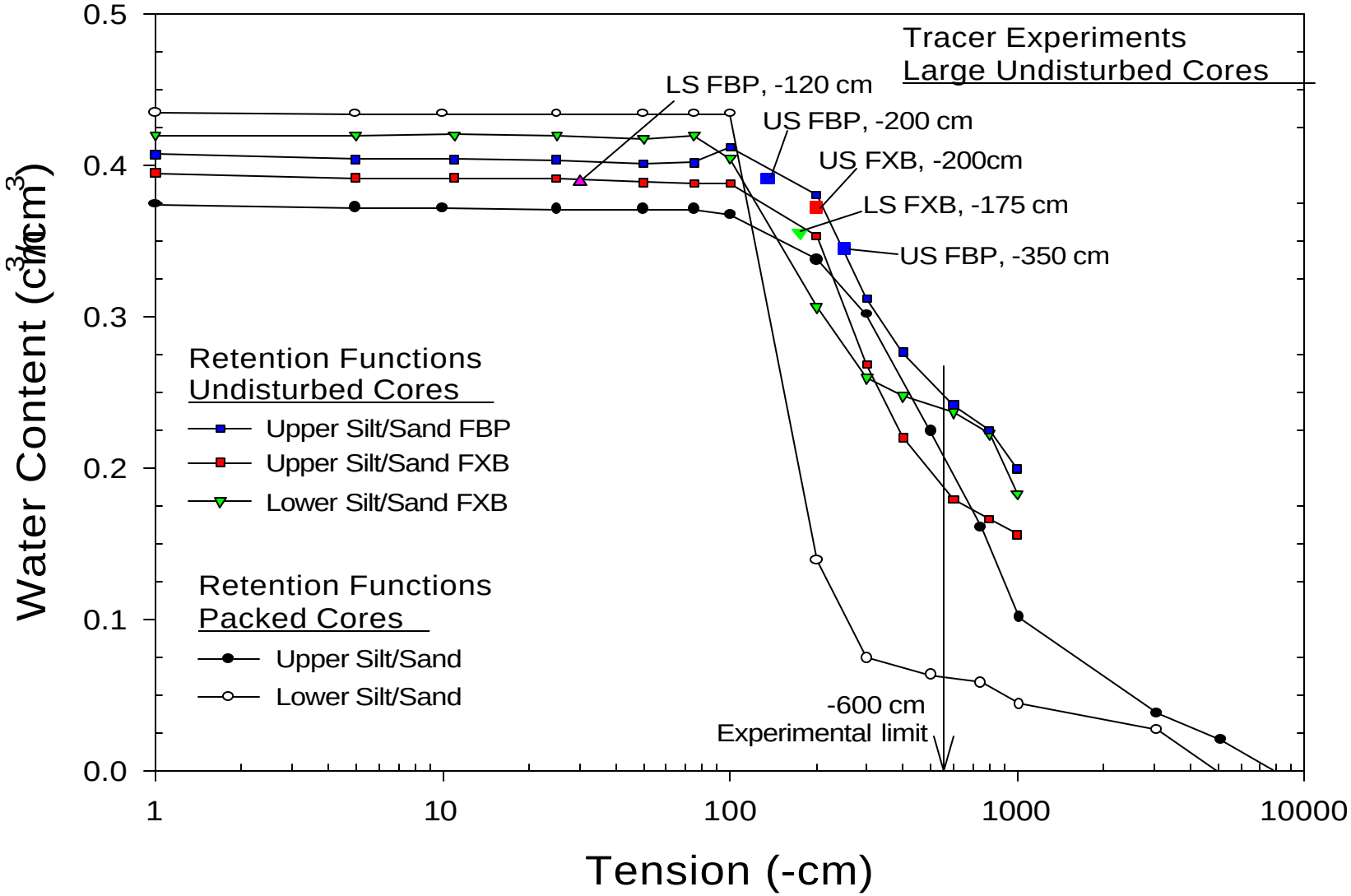
Multi-Channel Influent Delivery Pump

Tensiometers (1 bar)4 per Column

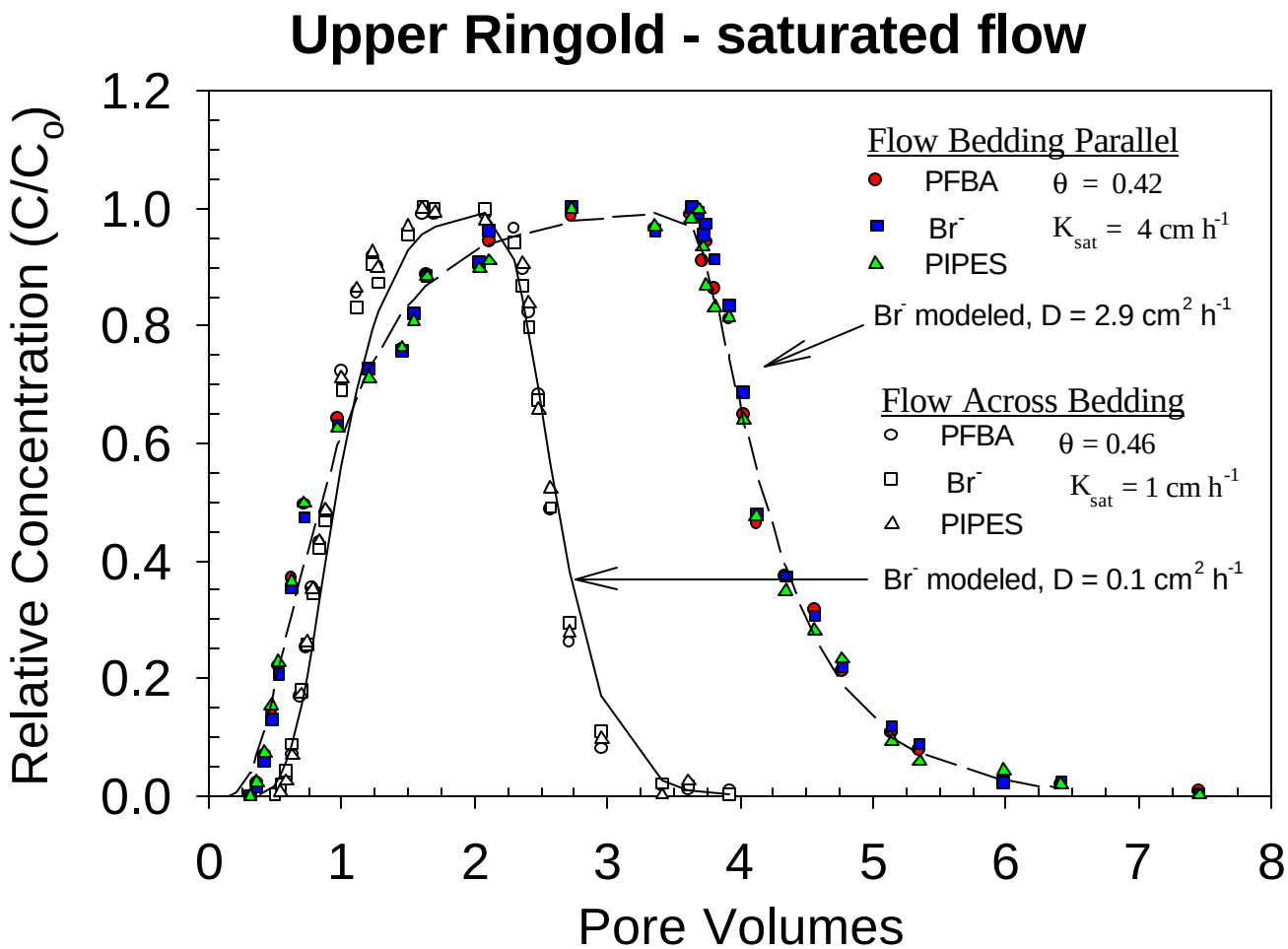
600 mbar membrane in Acrylic/PVC endcap

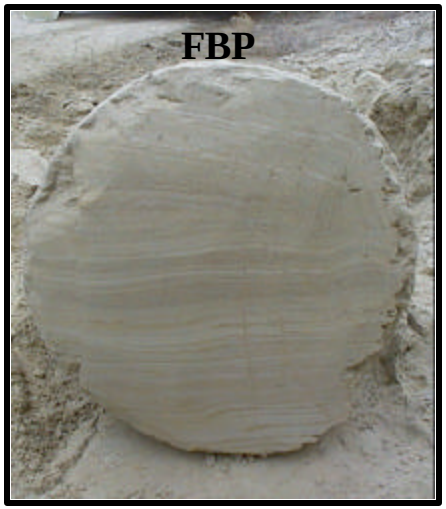
Vacuum Chamber with internal Fraction Collector

Predicting flux at various water contents in undisturbed cores from water retention functions



Effects of Horizontal Stratification

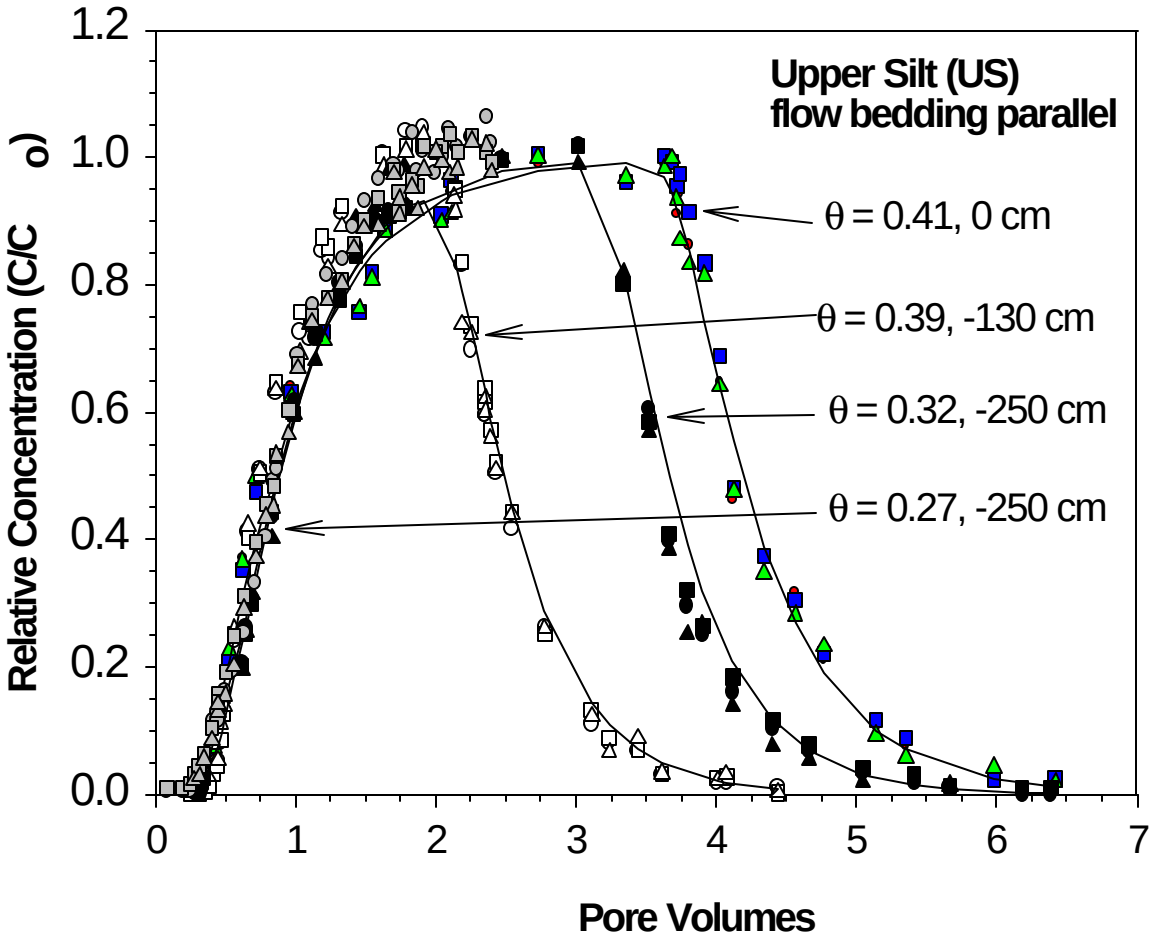




- **Bed parallel preferential flowpaths.**
- **Lateral flow strongly preferred over vertical flow.**
- **Lack of tracer separation suggests no immobile water present.**

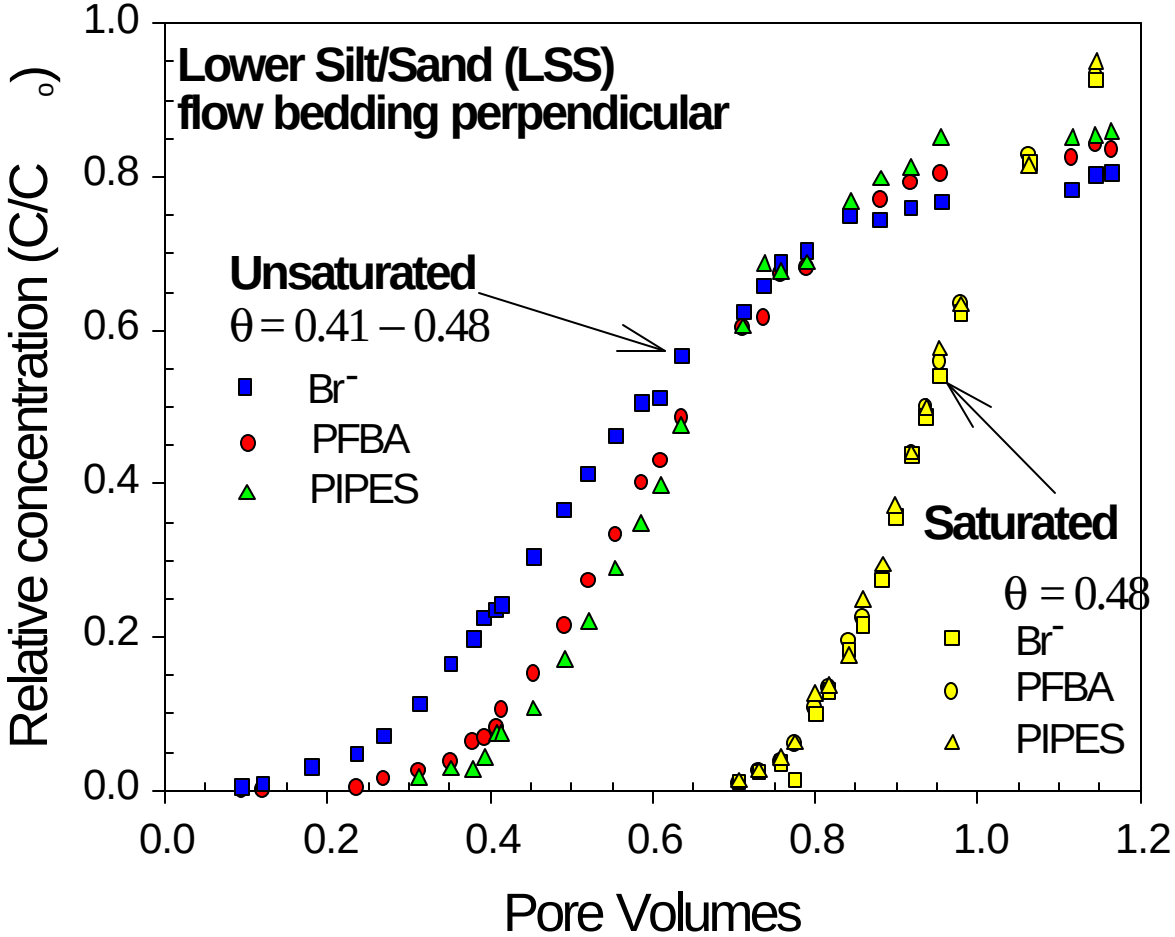
Hydrologic Transport Mechanisms: Lateral Flow

Variably saturated conditions



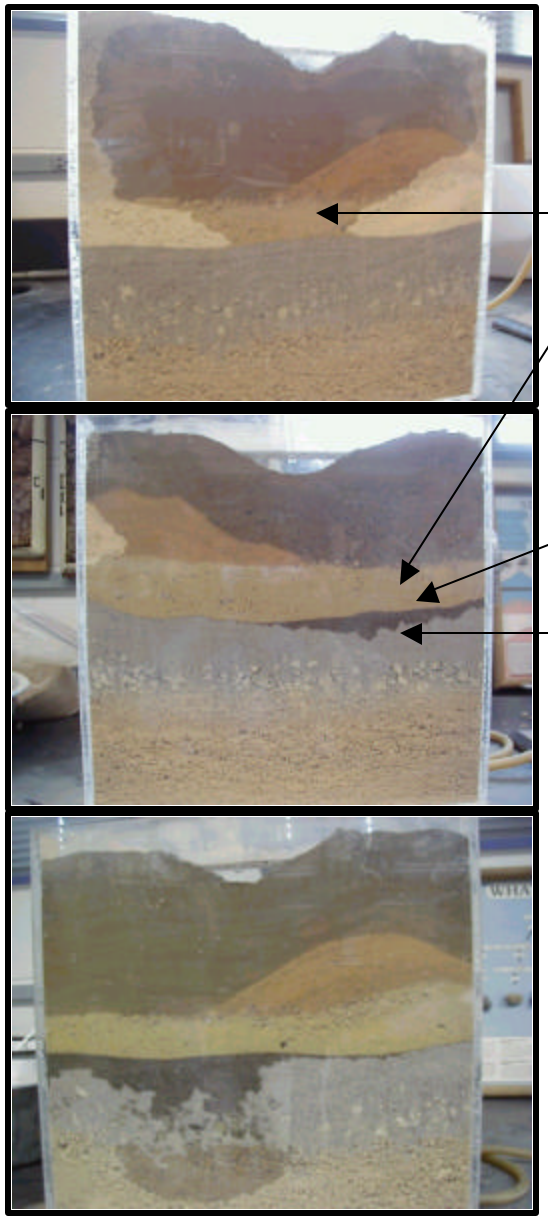
- **Lack of tracer separation suggests no immobile water present regardless of water content.**
- **Continuous layers result in unrestricted lateral flow with no lithologic effects.**

Hydrologic Transport Mechanisms: Vertical Flow



- **Early, asymmetric breakthrough coupled with tracer separation suggests the presence of media bypass coupled with significant immobile water during unsaturated vertical flow.**
- **Alternating layers of coarse and fine grained material results in restricted vertical flow.**
- **Unstable wetting fronts develop as a result of local scale perched water.**

Flow in layered sediments of varying texture



Perching in fines

Unstable wetting front

Fingers along leading edge

- This scenario is most likely present below tanks where preferential vertical flow and significant lateral spreading influence contaminant migration.
- mm scale of influence.

Source: http://www.agronomy.psu.edu/Courses/Soils101/Labs/drngeboxes.html

Semi-Arid Region Vadose Zone Transport Through Fine-grained Media

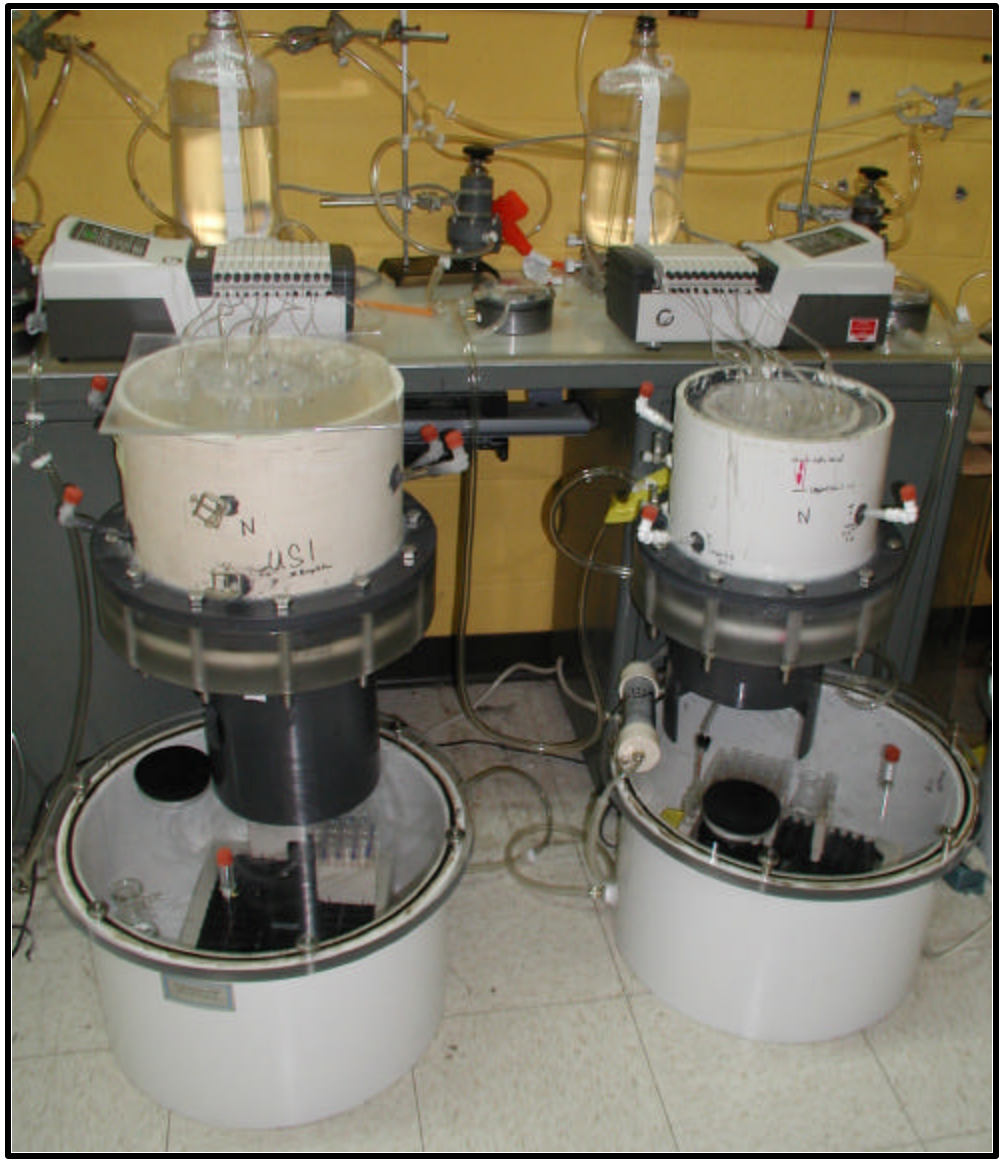
FLOW ALONG CONTINUOUS LAYERS

- Lateral flow
- No immobile water regardless of saturation
- No diffusional component

FLOW ACROSS DISCONTINUOUS LAYERS

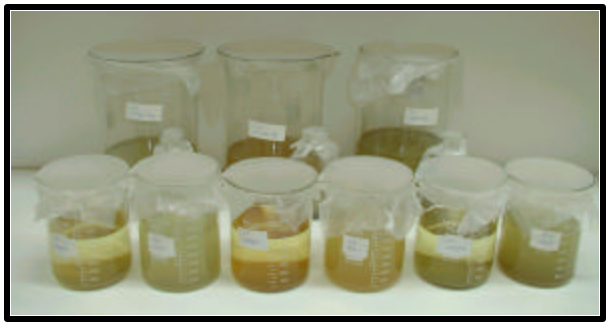
- Perched water
- Unstable wetting fronts
- Preferential finger flow
- Significant immobile water
- Effects are most dramatic in Ringold vs. Hanford formation due to intensive mm scale textural effects in the former.

Initiation of reactive transport experiments involving ^{137}Cs, $^{235/238}U$, ^{90}Sr, and ^{99}Tc.



Sediment Characterization

- Sand/silt/clay separation
- XRD
- SEM with EDX analysis

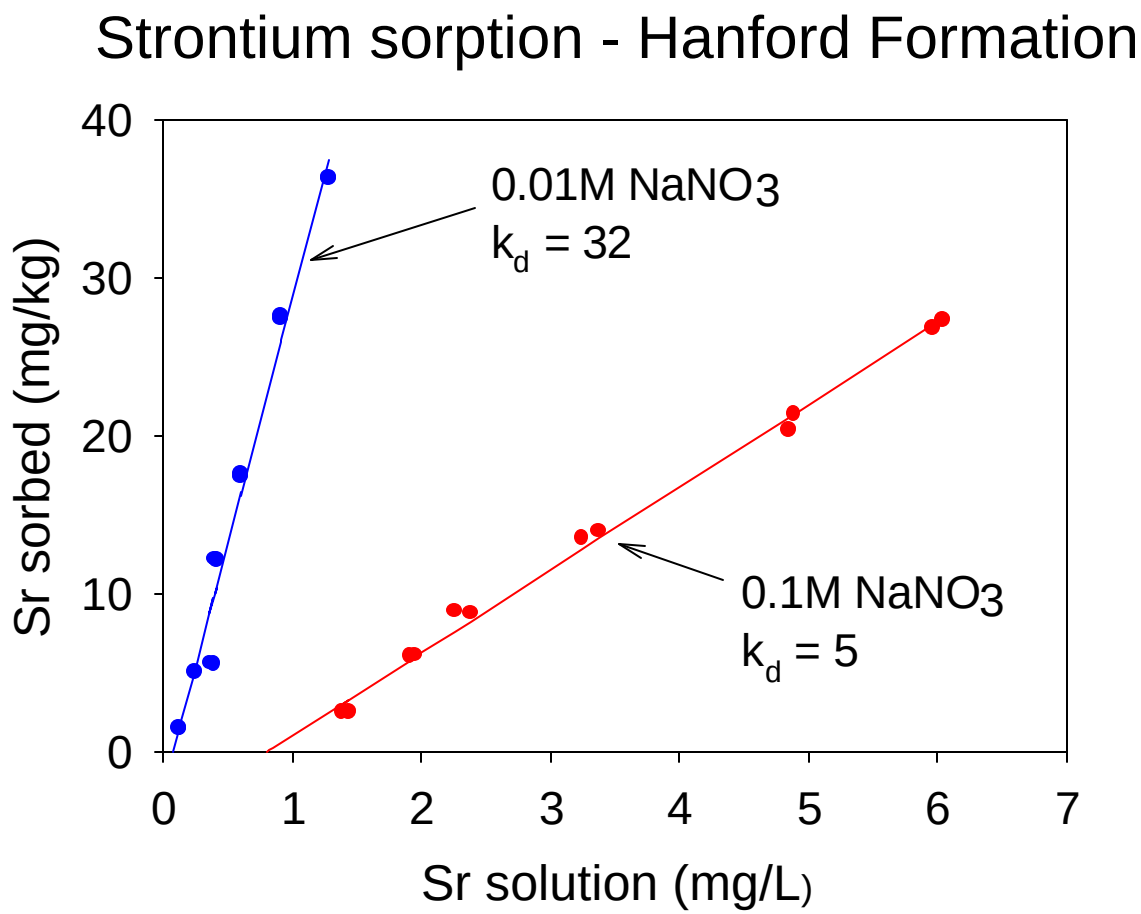


Fe-oxides
Mn-oxides
TOC/TIC

Sample	Sand (>50 micron)	Silt (2-50 micron)	Clay (<2 micron)	Fine Clay (<0.2 micron)
US	42%	56%	2%	
US	Quartz > Mica > Kaolinite > Chlorite > Plagioclase	Quartz > Plagioclase > Mica > Kaolinite > Chlorite	Illite > Kaolinite > Smectite > Chlorite/HIV* > Quartz	Illite > Kaolinite > Smectite > Chlorite/HIV* > Quartz
LS	57%	42%	1%	
LS	Quartz > Mica > Plagioclase > Kaolinite > Chlorite	Quartz > Plagioclase > Kaolinite > Mica > Chlorite	Illite > Kaolinite > Smectite > Chlorite/HIV > Quartz > Plagioclase	Illite > Kaolinite > Smectite > Chlorite/HIV > Quartz > Plagioclase

* Hydroxy-interlayered vermiculite

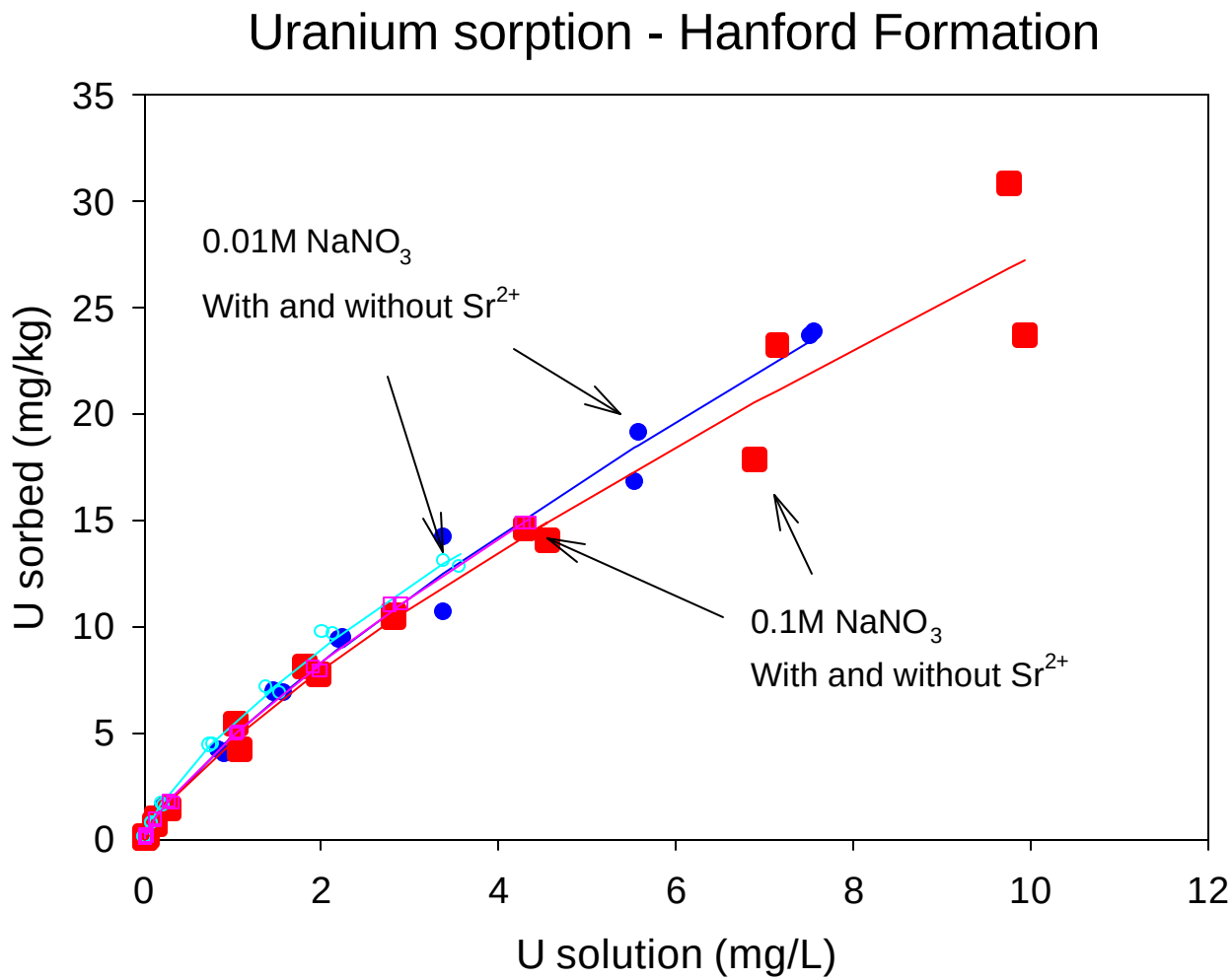
Radionuclide Sorption on Ringold and Hanford Sediments



Large ionic strength effect suggests that Sr^{2+} adsorption is controlled by weak electrostatic interactions with the solid phase.

In the absence of competing cations, Sr^{2+} retardation is appreciable.

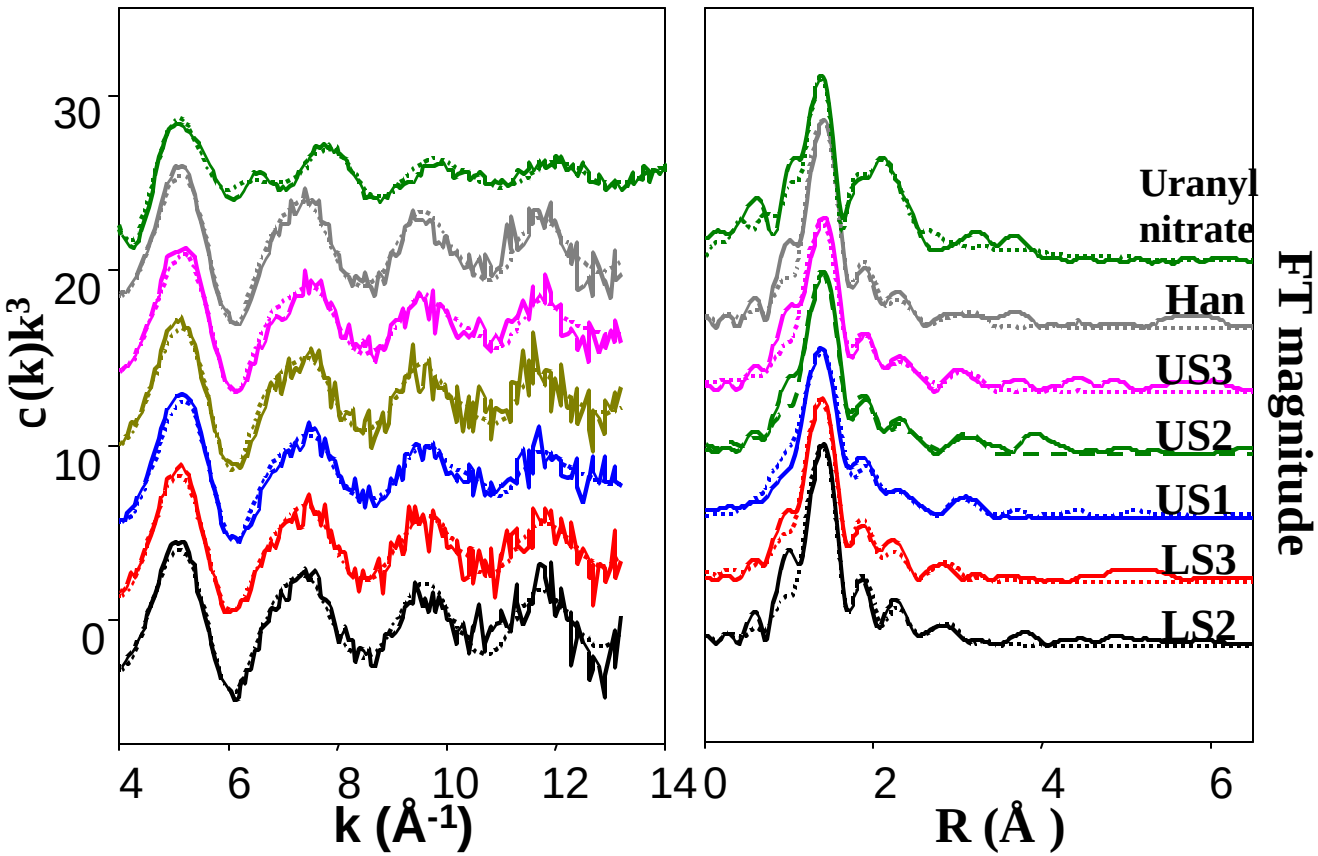
Radionuclide Sorption on Ringold and Hanford Sediments



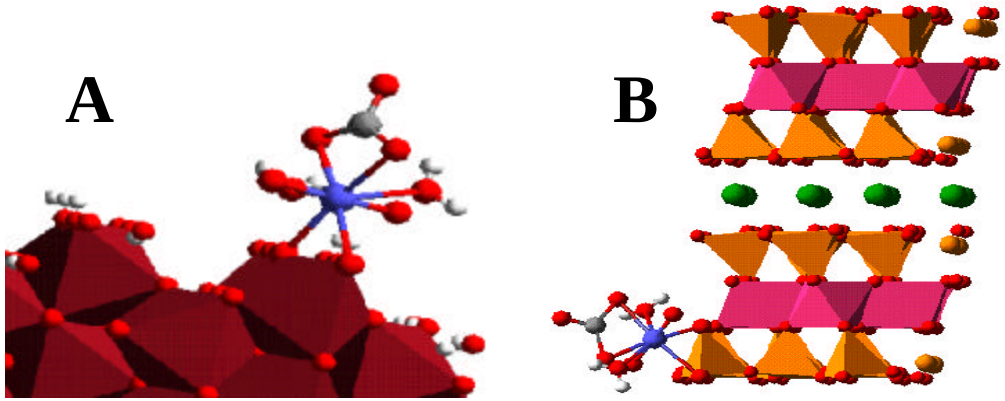
Lack of an ionic strength effect suggests that U adsorption is controlled by strong inner-sphere complexes the solid phase. Results consistent with XAS analysis.

U retardation is not influenced by the presence of Sr^{2+}.

Radionuclide Solid Phase Speciation Using X-ray Absorption Spectroscopy

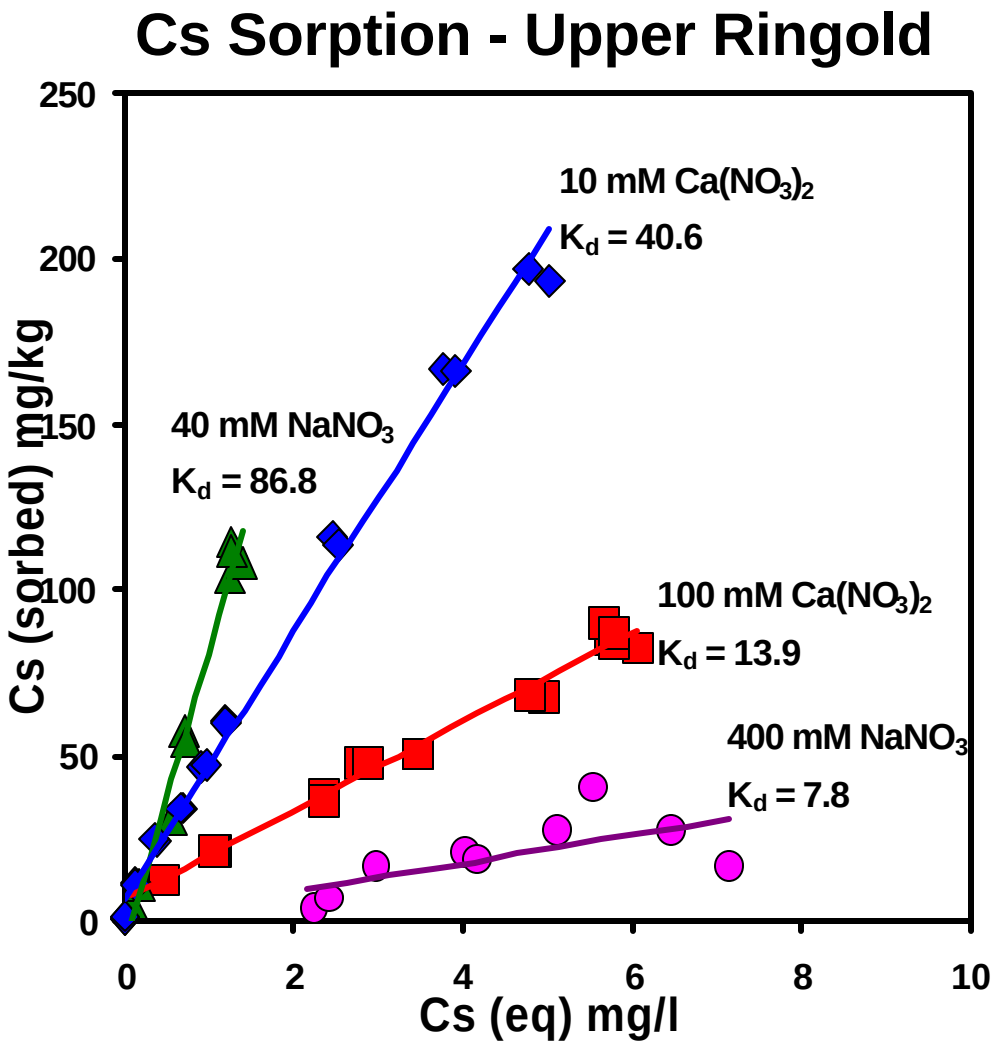


- XAS used to quantify contaminant valance state and sorption mechanisms.
- Inner-sphere ternary carbonate complexes formed on Fe-oxides for sandy Ringold sediments while sheet silicates dominated in the silty Ringold sediments.



U-CO_3 complexes on (A) Fe-oxides and (B) 2:1 layered silicates.

Radionuclide Sorption on Ringold and Hanford Sediments



Large ionic strength and cation effects suggest that a significant proportion of sorbed Cs^+ is not being fixed within the interlayer spaces of 2:1 phyllosilicates. Nonequilibrium conditions may be a factor with enhanced fixation occurring with longer Cs^+ residence times.

Year 1-2 Research Findings

- Lateral flow strongly preferred over vertical flow.
- Continuous layers result in unrestricted lateral flow with no immobile water regardless of saturation.
- Alternating layers of coarse and fine grained material results in restricted vertical flow and the development of unstable wetting fronts as a result of local scale perched water.
- Similar Ringold lithologies, flux differed by an order of magnitude.
- Sediments have a large sorption capacity for impeding contaminant transport.

Implication of Results to Cleanup Issues

Improved Conceptual Understanding of Vadose Zone Processes:

- Lateral flow is a strong contributor to contaminant spreading beneath the tanks.
- Media is conducive to large flow and transport anisotropy.
- Unsaturated vertical flow results from unstable wetting fronts that promote media bypass.
- Significant immobile water regimes can form during vertical flow which contributes to the attenuation of waste being released from the tanks.
- Small-scale mineralogic heterogeneities, such as Fe-oxide and clay lenses, will serve to impede the lateral and vertical migration of contaminants.

Improved Numerical Modeling of Vadose Zone Processes:

- Flow and transport data, coupled with water retention functions, using undisturbed media is extremely valuable for parameterizing simulation models.

Future Plans

- Finalize multiple nonreactive transport experiments at lower water contents within the Ringold and Hanford formations and the Plio-Pleistocene unit, to investigate the significance of downward vertical advection, lateral spreading, and physical nonequilibrium.

- Initiate unsaturated transport experiments involving ^{134}Cs, ^{238}U, ^{85}Sr, and ^{95}Tc to determine how physical (stratification, pore regime connectivity) and mineralogic heterogeneities influence the retardation and degree of geochemical nonequilibrium during transport.

- Continue to use X-ray Absorption Spectroscopy and Laser Induced Hyperquenching Fluorescence to quantify the sorption mechanisms of Cs, U, and Sr in sediments beneath the Hanford tank farms.

- Technology transfer to support EM's performance/risk assessment and decision-making process for tank farm restoration.