Coupled Processes Influencing the Transport of Uranium over Multiple Scales

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DOE Hanford Reservation and nuclear waste

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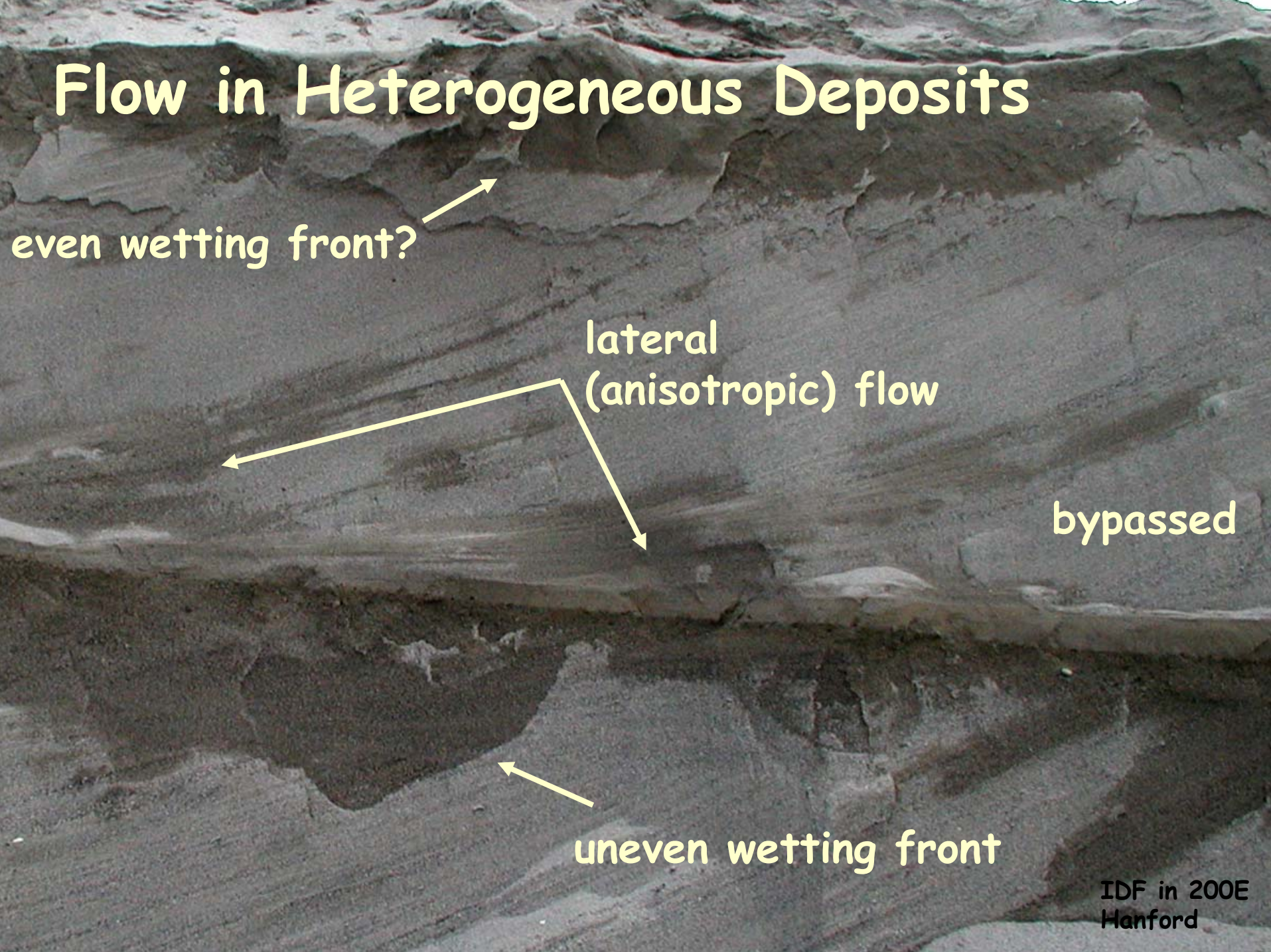
Heterogeneous Subsurface Deposits

- Grain (pore) size
- Sedimentary structures and arrangement of sedimentary layers
- Mineralogy and geochemistry

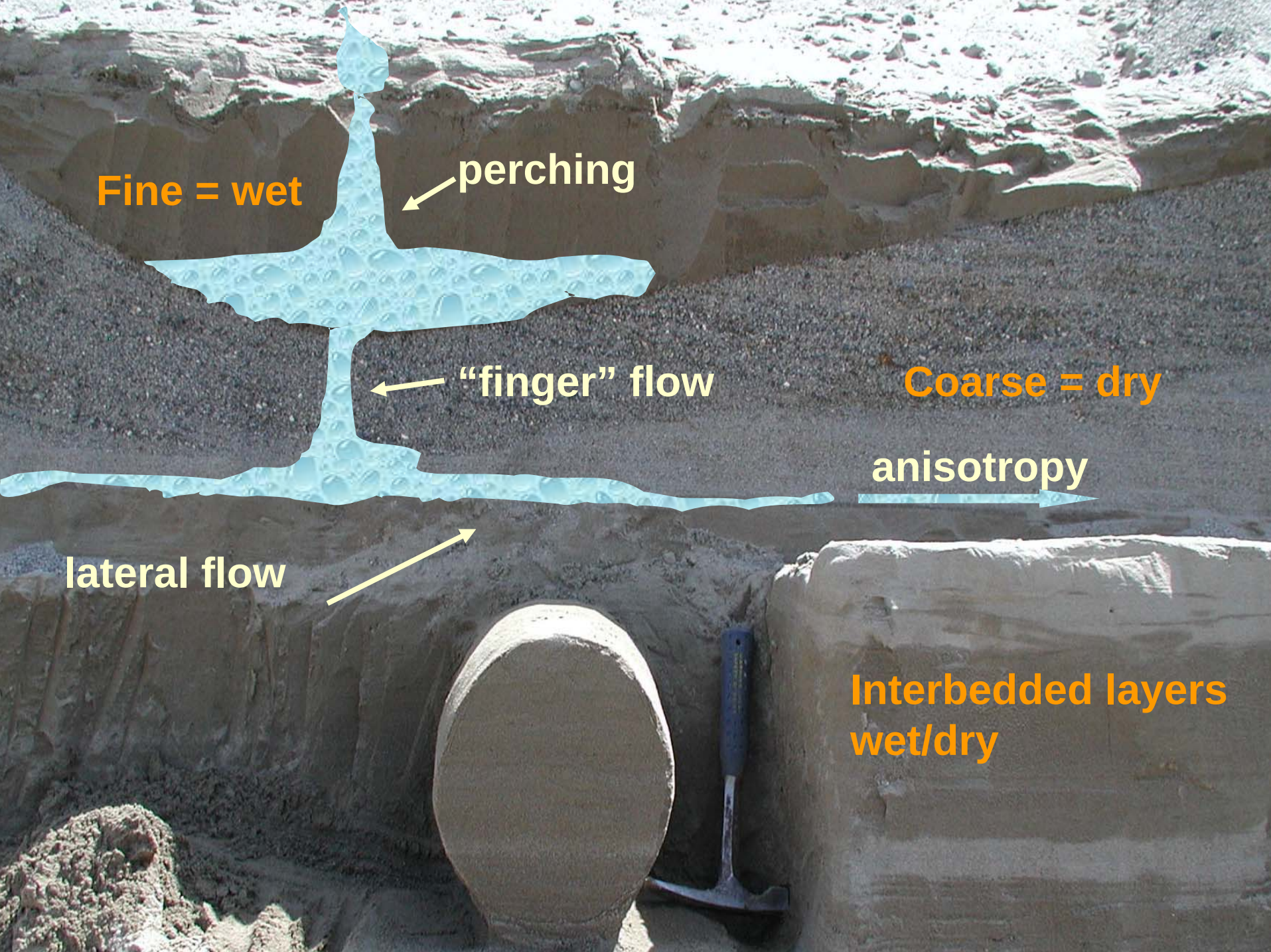
IDF in 200E
Hanford

Flow in Heterogeneous Deposits

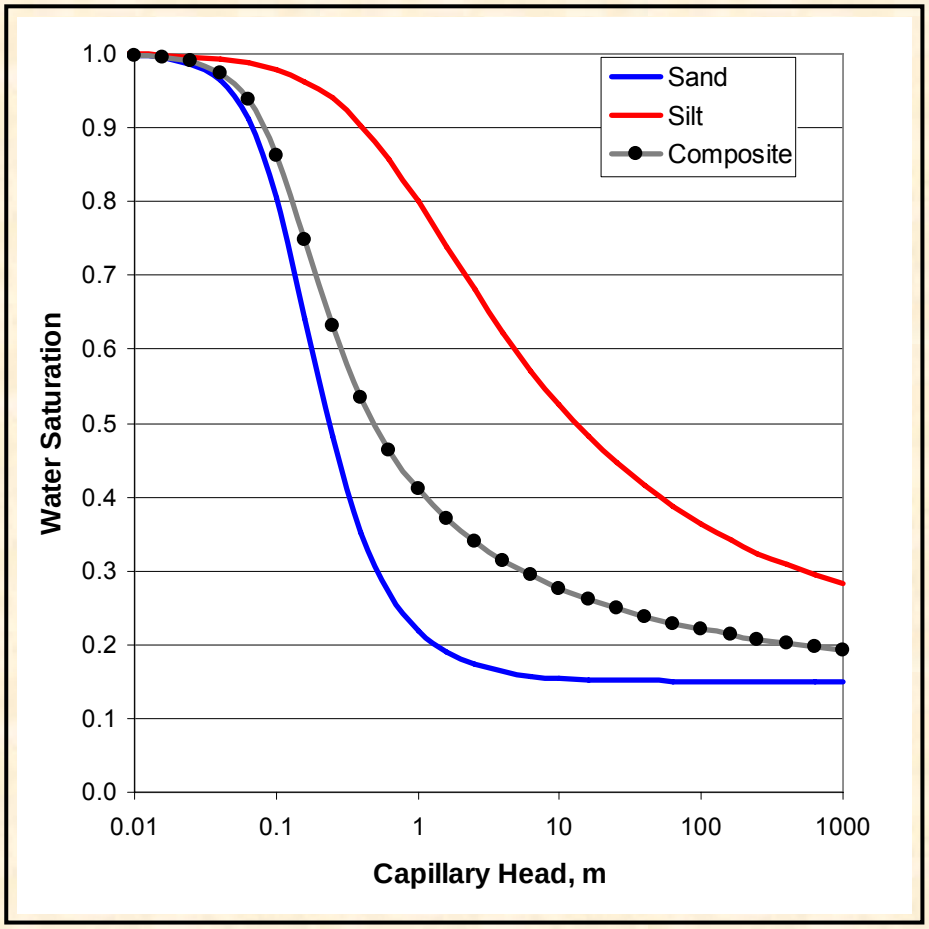


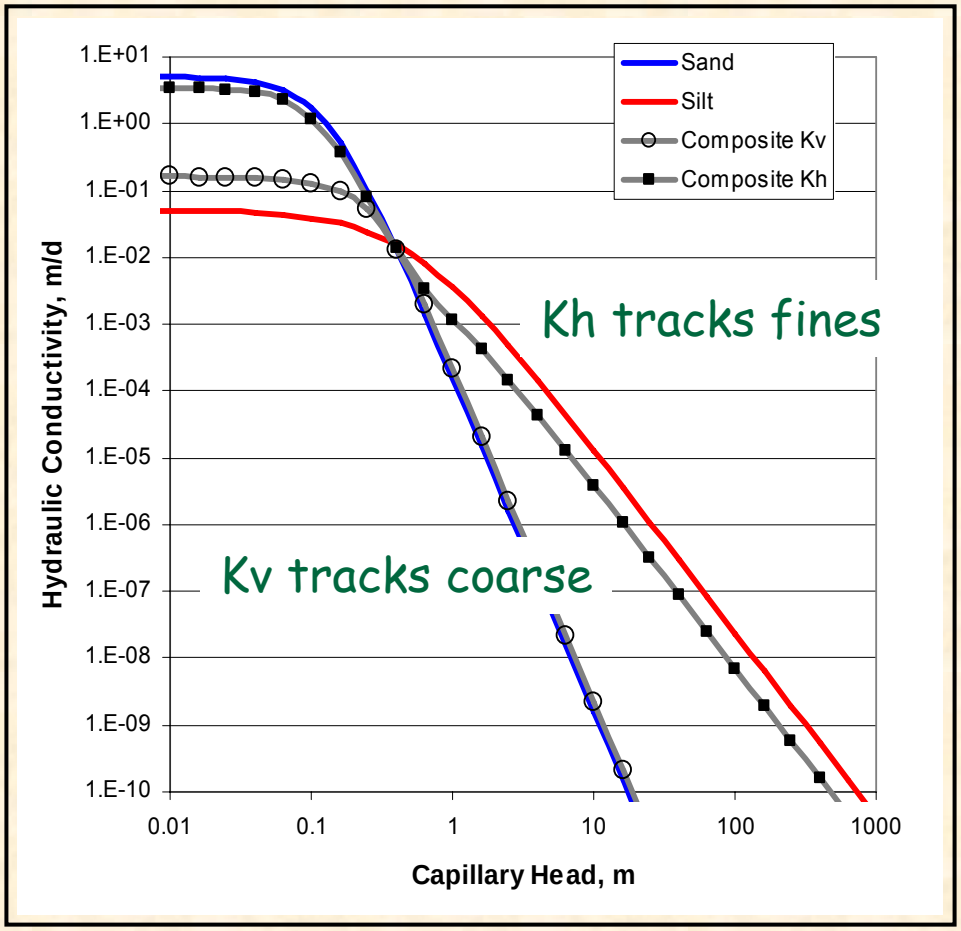
Heterogeneity and Scaling in Vadose Zone

- Conductivity varies with water content
- Heterogeneity of adjacent sedimentary beds
  - pore-scale interactions
  - extent of <u>vertical v. lateral</u> infiltration
- Results in anisotropy (directionality) of flow
- Scale-dependent physical hydraulic parameters (λ)
- Influence on reactivity (R, K_d)?
  - mineralogical heterogeneities


Fine = wet
perching
"finger" flow
Coarse = dry
anisotropy
lateral flow
Interbedded layers
wet/dry

Up-scaling:
Case of alternating sand and silt layers





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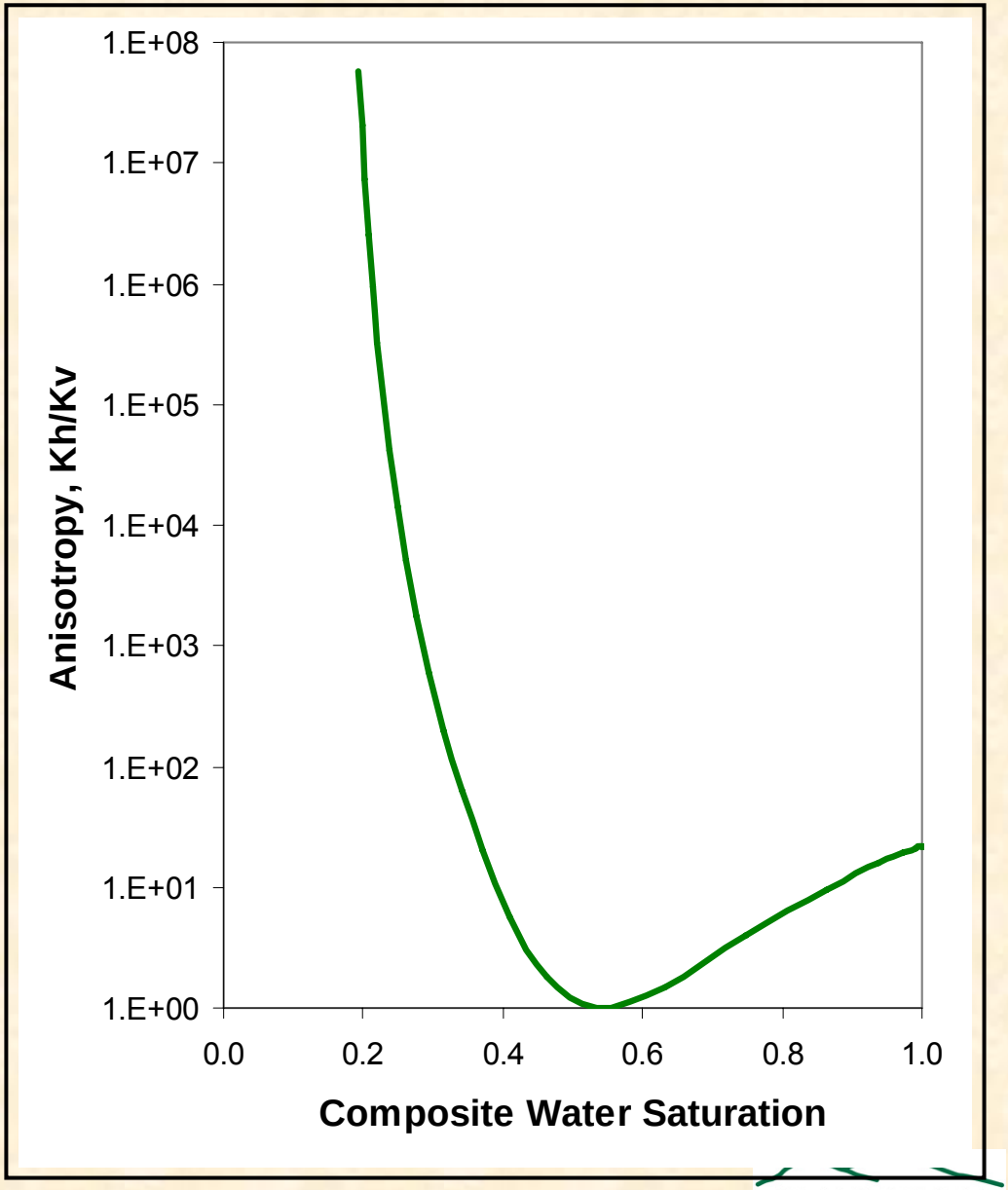
Heterogeneity, Scaling, and Water Content

Up-scaled conductivity exhibits anisotropy that is sensitive to:

- average capillary pressure
- water content
- heterogeneity



Evidence?

Jardine, Fendorf, and Mayes:
EMSP 1999-2005

Mayes and Jardine:
Tank Farm Vadose Zone Group,
CH2M Hill, Inc. 2004-present

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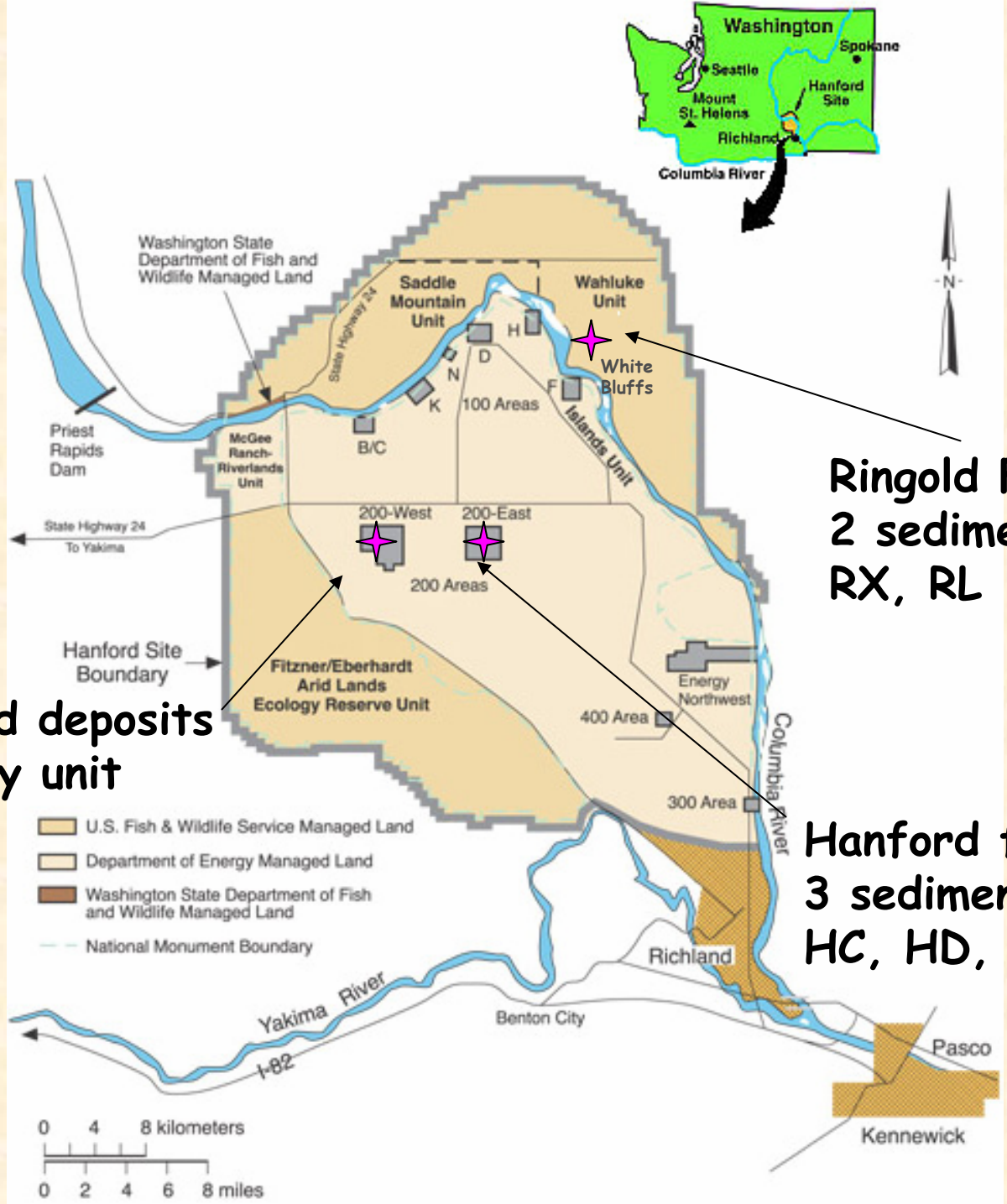


Ringold Formation
2 sedimentary units
RX, RL

Hanford flood deposits
1 sedimentary unit
HL

Hanford flood deposits
3 sedimentary units
HC, HD, HI

Intact Core Samples (25 x 25 cm)

5 cm

massive coarse sand

interbedded sand/silt

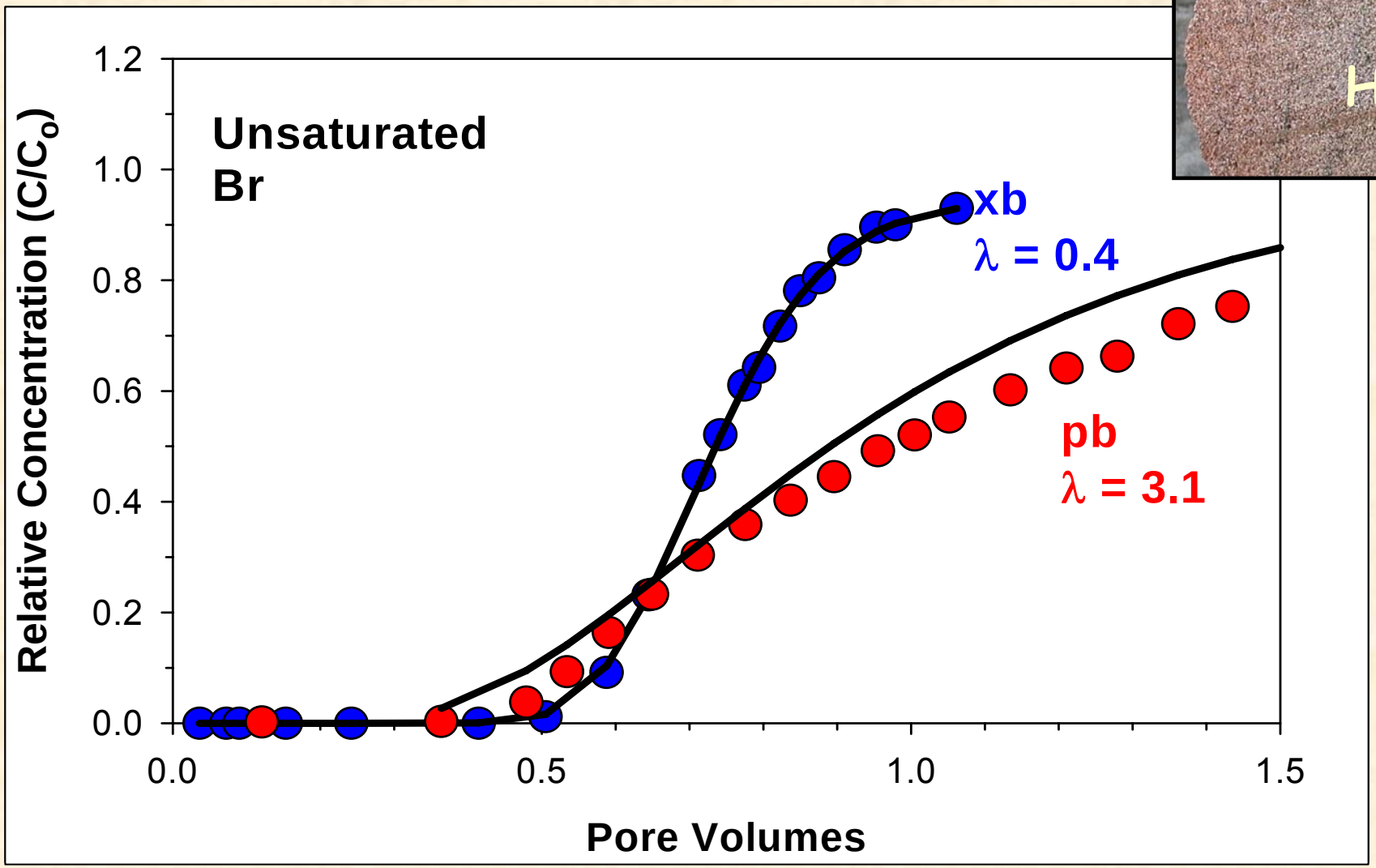
fine sand, clay laminations

clastic dike

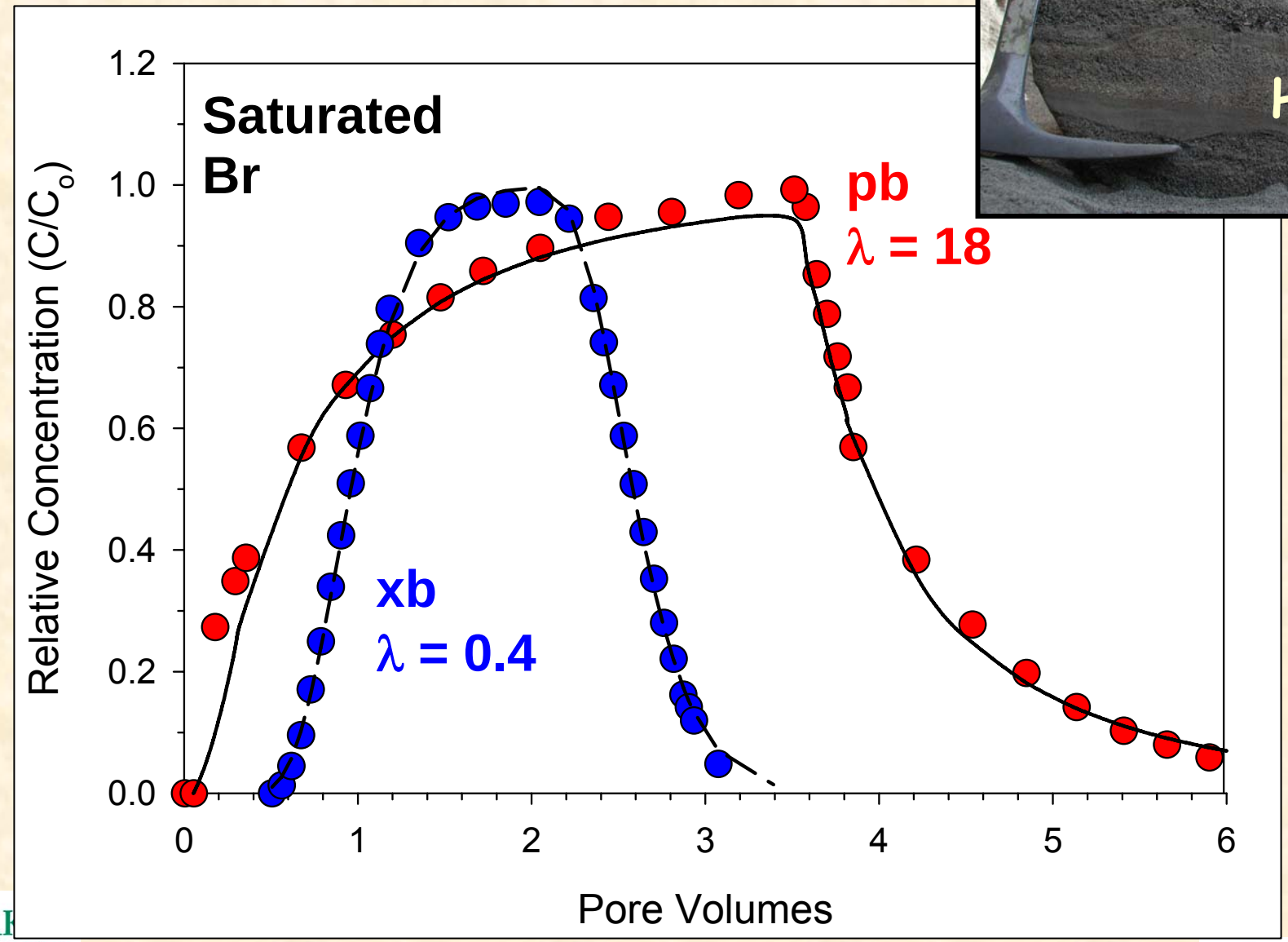
cross-bedded sandy loam

laminated silt loam

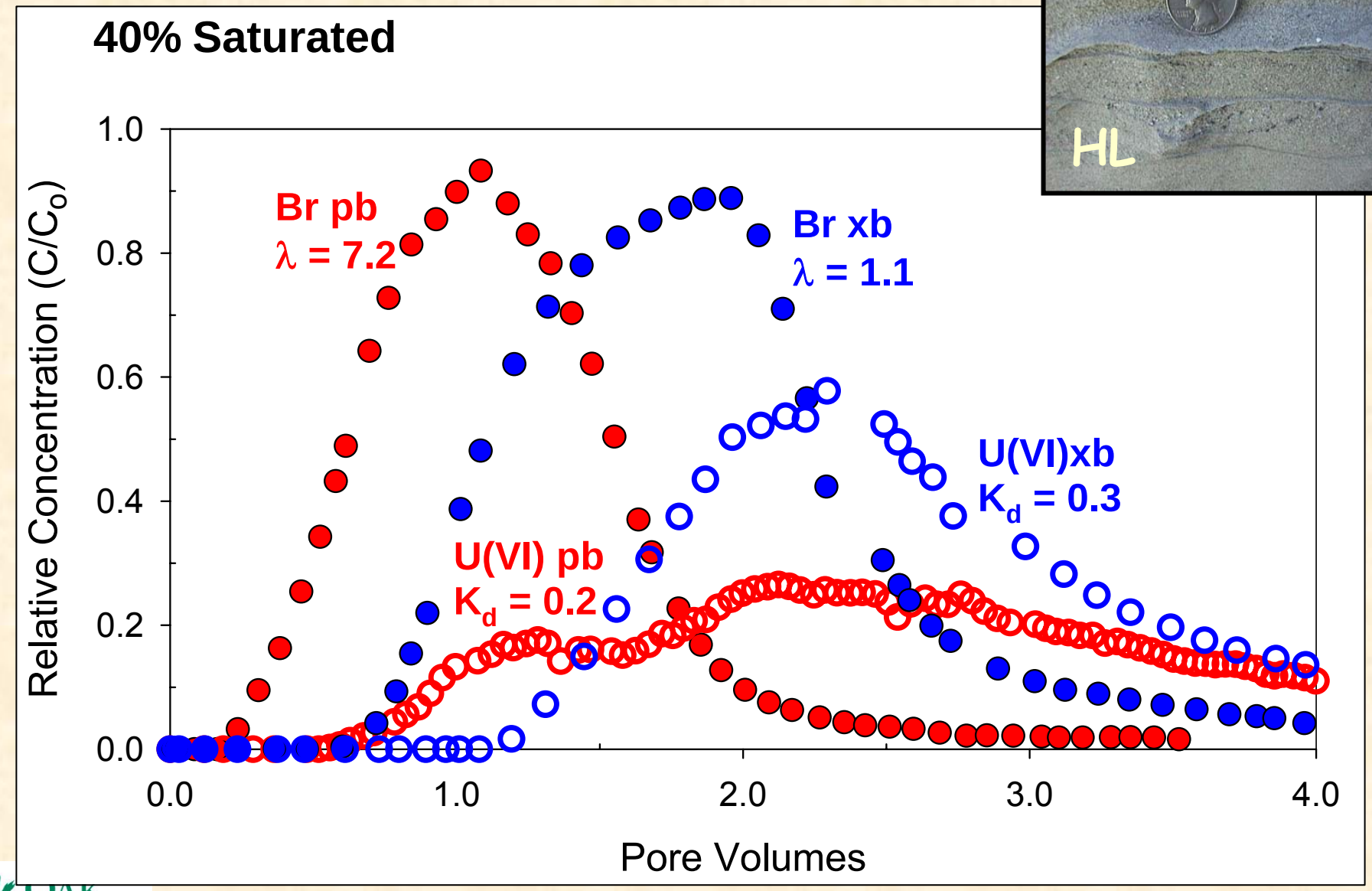
Anisotropy and Water Content



Anisotropy and Heterogeneity



Anisotropy and U(VI) Mobility



Heterogeneity, Water content, Anisotropy, and Scaling

- Higher dispersivity when flow is parallel to bedding
- Higher dispersivity at lower water content
- Higher dispersivity in heterogeneous samples
- U(VI) more mobile with higher dispersivity

New Research Goals

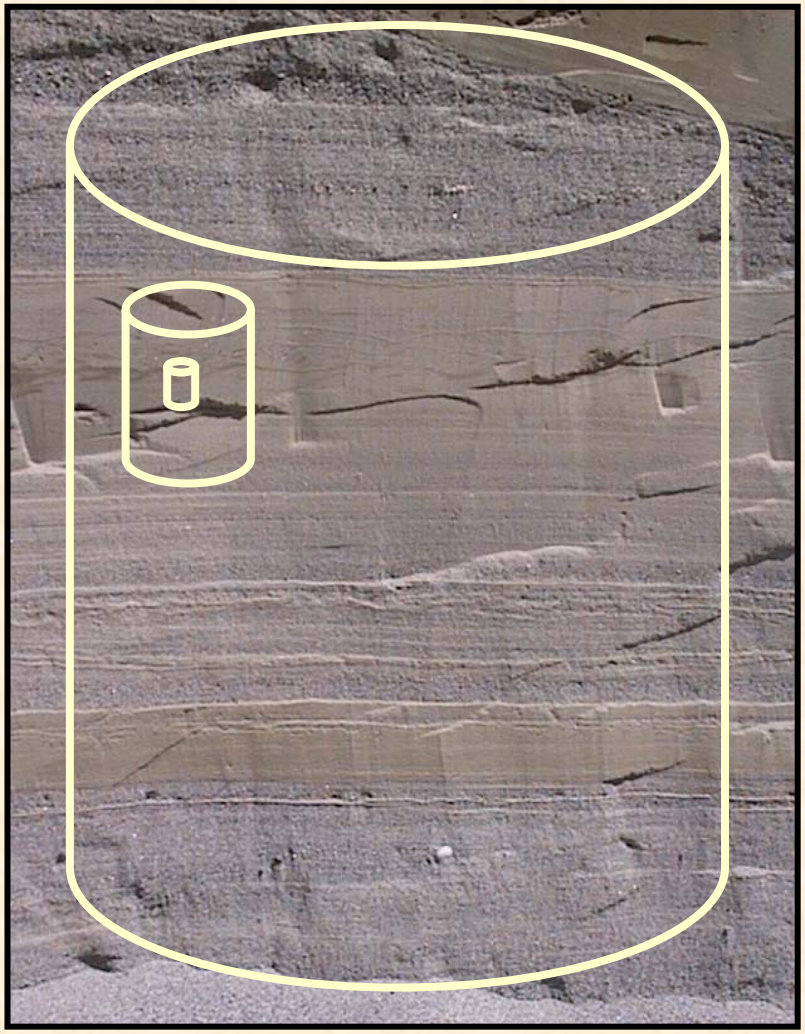
To provide validated scaling strategies which can be applied to existing contaminant distributions and migration scenarios at Hanford and similar sites

Research Questions

- Contribution of individual layers to bulk K(θ) ?
- Contribution of individual layers to U(VI) transport?
- How are reactive mineral phases spatially arranged?
- Interaction of competing hydrological, geochemical, and mineralogical processes in multi-layered systems?
- Are measurements of hydraulics and reactivity of individual layers sufficient to predict contaminant transport in large heterogeneous systems of multiple complex layers?

Multi-Scale Experimental Strategy

1. Single Layers
2. Sedimentary Unit
   - Previous data
   - 1-D
   - Flow parallel to beds (pb)
   - Flow crosses beds (xb)
3. Multi-Layered System
   - 2-D: pb <u>and</u> xb



Objectives

1. Layer Scale: Separate quantification of hydraulic, geochemical, and mineralogical factors influencing U(VI) transport
2. Up Scale: Apply numerical, composite medium, and fractal approaches to compute effective coupled hydraulic and reactive transport parameters
3. Validate: Apply Up Scaled parameters to U(VI) transport through progressively larger scales of intact samples that encompass both lateral and vertical U(VI) transport

Layer Scale: Hydraulic Factors

POSTER TONIGHT

Ultra Rock Core Centrifuge (URC): Transient flow method for determining moisture retention properties and predicting unsaturated hydraulic conductivity

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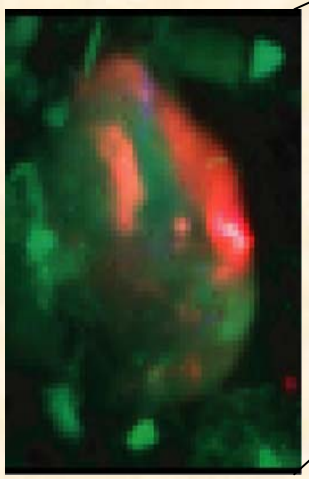
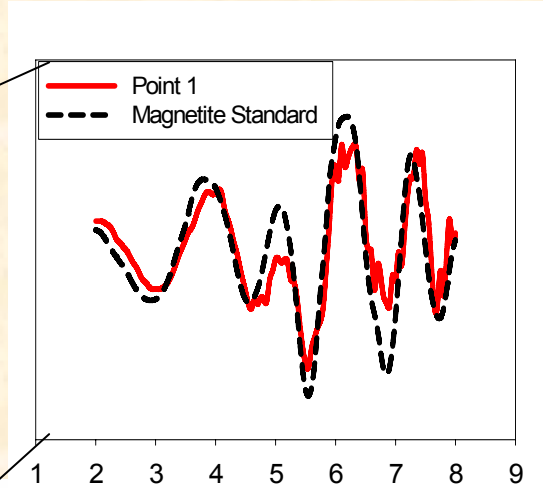
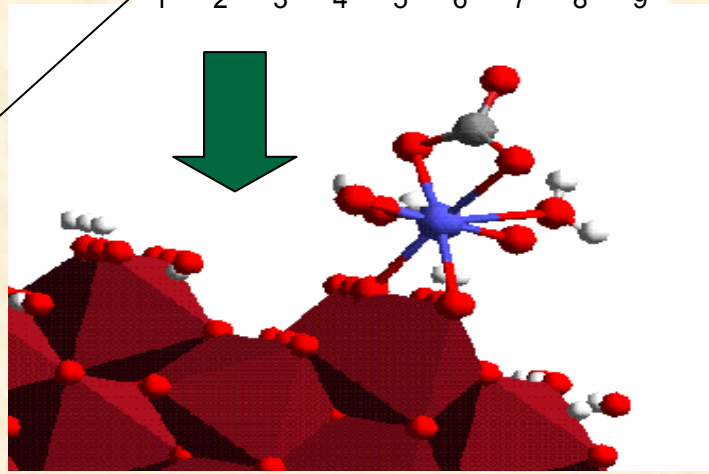
Layer Scale: Mineralogical Factors

Adsorbent/Adsorbant Speciation

- Element (U) mapping
- XANES
- EXAFS
- Mossbauer
- Raman
- Micro-focused XRD







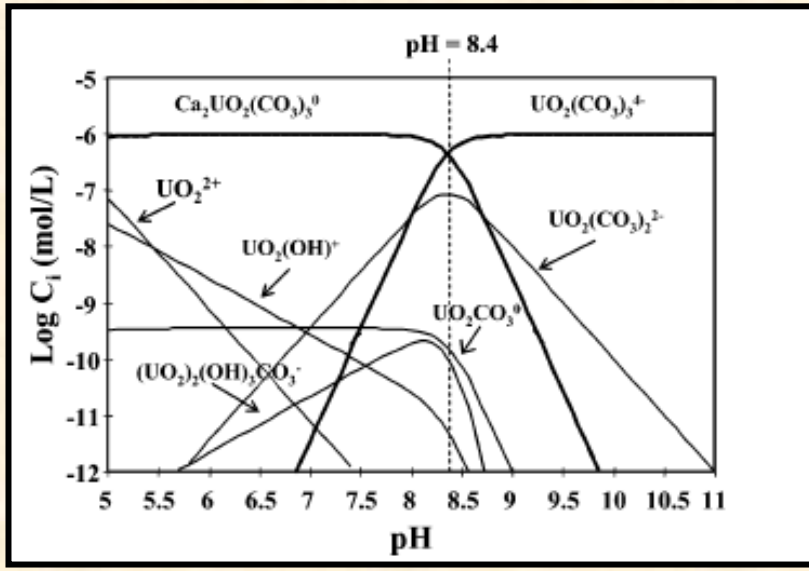
Importance of Particle Size

- Sand, silt, clay mineral reactivity

Layer Scale: Geochemical Factors

U(VI) Speciation

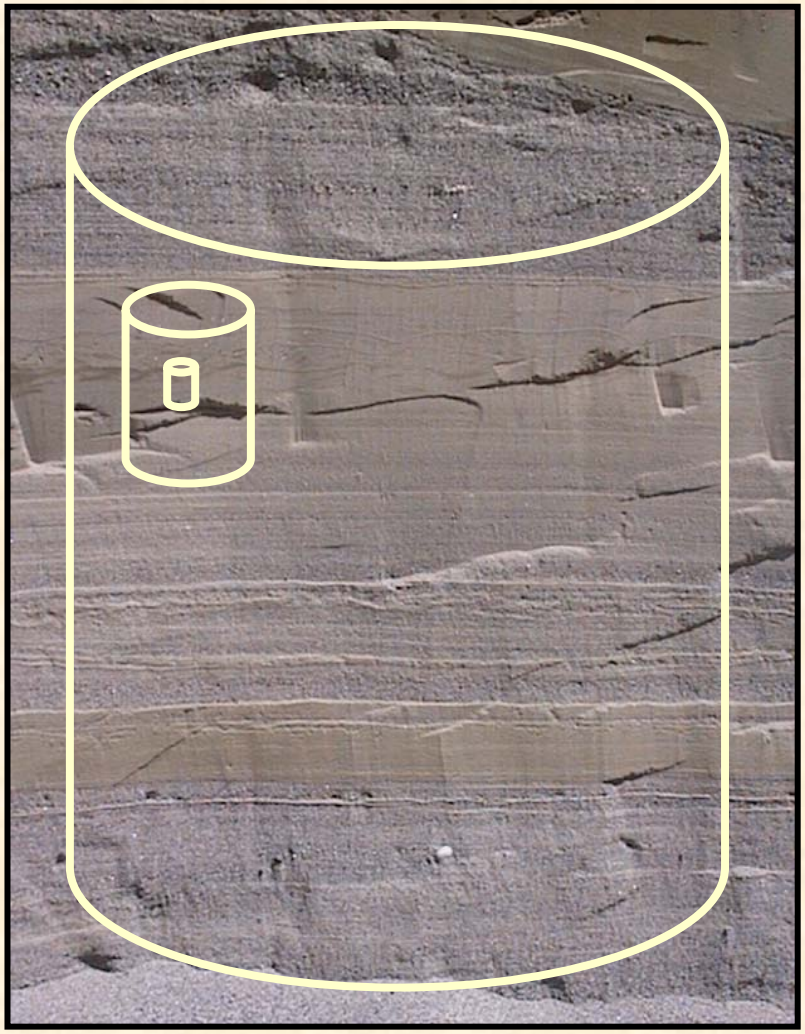
- Influence of solid-phase calcite
- Cryogenic Laser-Induced Fluorescence Spectroscopy (LIFS at EMSL)
- Link with **Mineralogical Factors**



Dong et al., 2005

Multi-Layered System

- Intact sample 2D/3D
  - statistical distribution of material properties
- Instrument individual layers
  - solution samplers, matric potential
- Transient application of nonreactive tracers, U(VI)
- Up-scaling approaches applied to Layer-Scale data
  - is the whole a sum of parts?

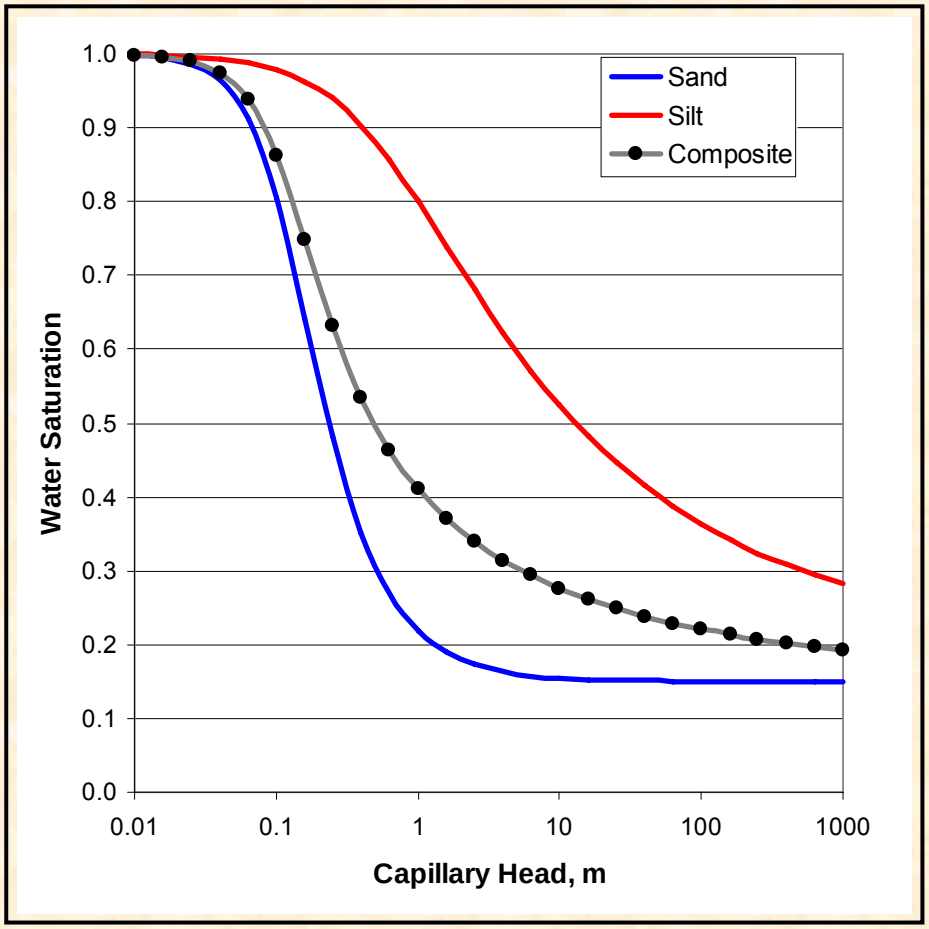


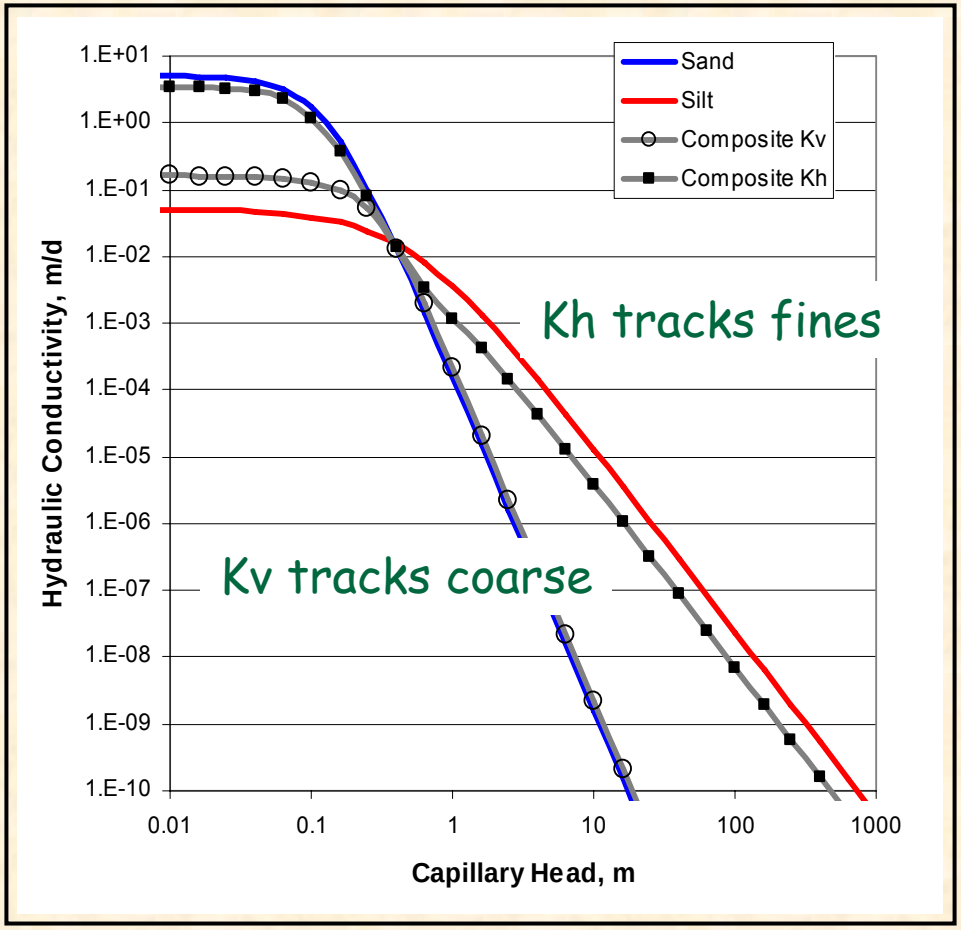
From Layer-Scale to Multi-Layer System

Can unsaturated flow in heterogeneous media be described by "effective" upscaled properties?

- Apply numerical, composite medium, and fractal approaches to Layer-scale data to compute effective coupled hydraulic and reactive transport parameters
  - HydroGeoChem (HGC) 5.0 (Yeh et al., 2004)
  - Composite Medium Approach (COMA) (Pruess, 2004)
  - Cantor Bar fractal model (Perfect et al., manuscript in preparation for special issue of VZJ)
- Compare with observed results in multi-layer system

Up-scaling:
Case of alternating sand and silt layers





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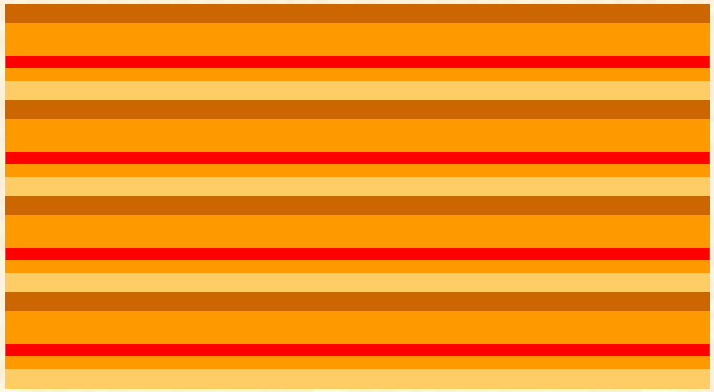
Up-scale: Example of COMA approach

Consider a soil composed of i=1,…,N material types with volume fractions f_i in randomly distributed horizontal layers.



Objective: Predict average fluxes, concentrations, etc. across multiple layers

Composite capillary pressure-water content function:

$$\theta_{comp}(h) = \sum_{i=1}^{N} f_i \theta_i(h)$$

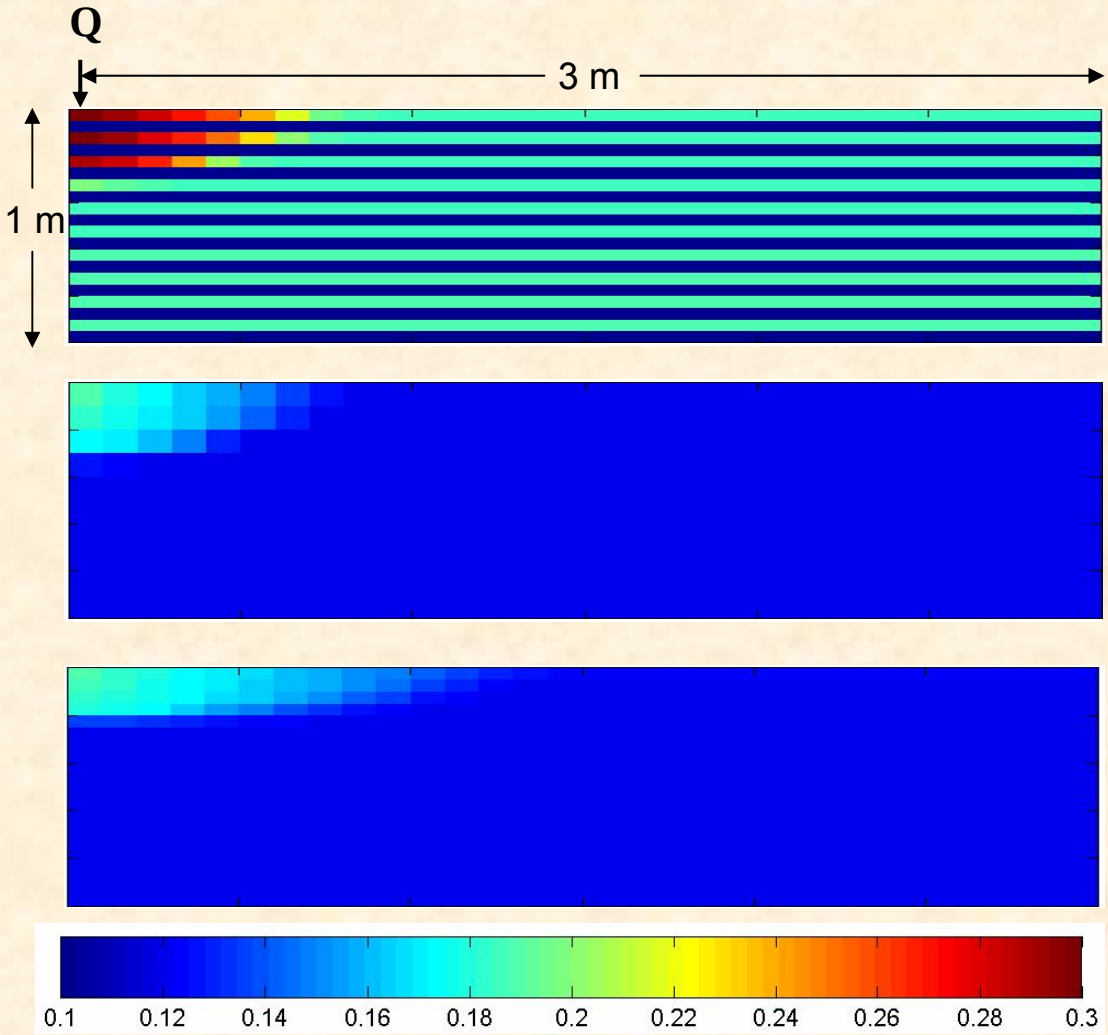
Horizontal conductivity will upscale as a weighted arithmetic mean:

$$K_{H\ comp}(h) = \sum_{i=1}^{N} f_i K_i(h)$$

Vertical conductivity will upscale as the harmonic mean:

$$K_{V\ comp}(h) = \frac{1}{\sum_{i=1}^{N} \frac{f_i}{K_i(h)}}$$

Infiltration on line-source in layered soil with alternating layers



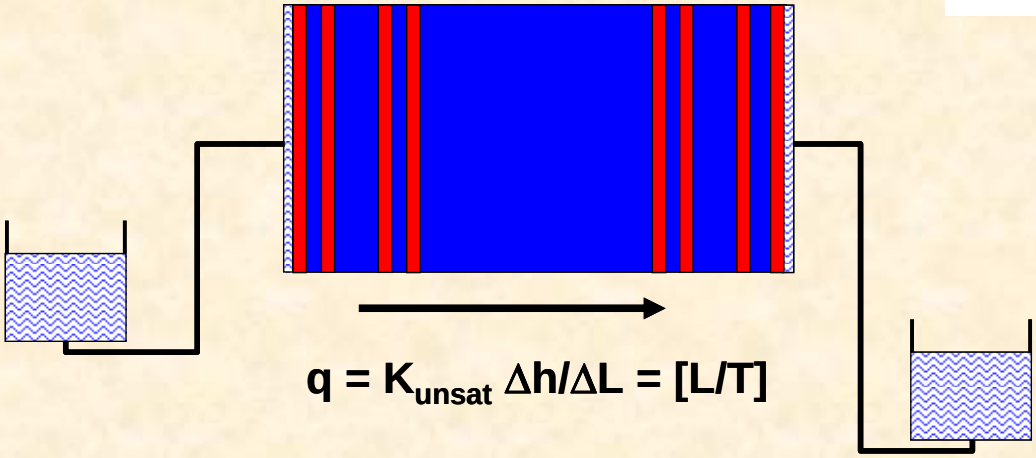
Layered numerical simulation – resolution at layer scale

Numerical simulation with layers – averaged over adjacent layers

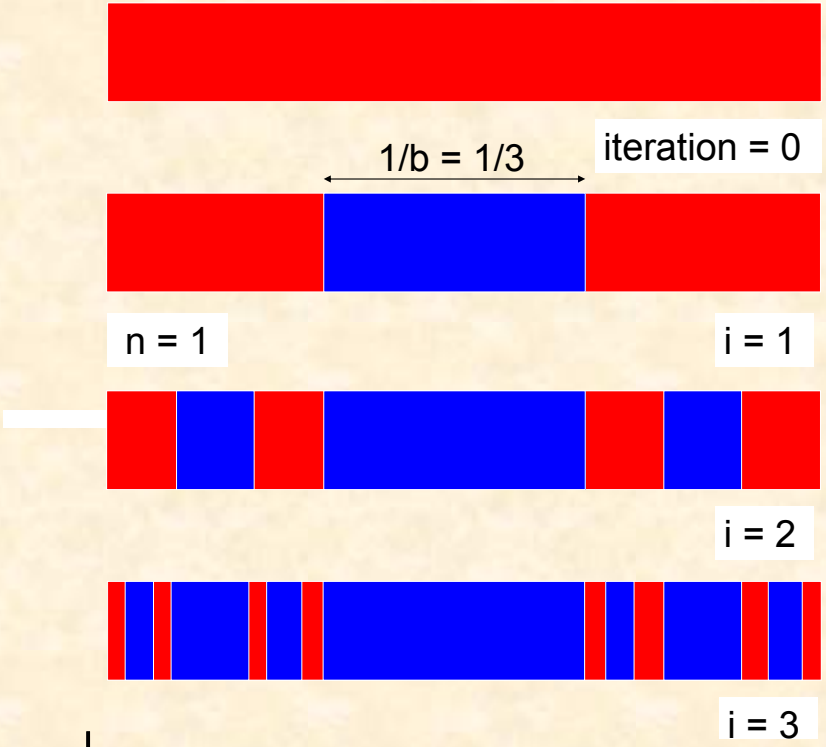
"Equivalent" homogeneous upscaled properties

Up-scale: Fractal Methods

- Cantor Bar fractal model (Perfect et al., in prep)



$q = K_{unsat}\ \Delta h/\Delta L = [L/T]$



$K_{unsat} \rightarrow K(h) \rightarrow$ Layered system $\rightarrow$ Effective K $\rightarrow$ <K(h)>

Up-scale: Future Directions

- Incorporate up-scaled models into HGC
  - Couple numerical model with parameter estimation code
  - Up-scaling of dispersive and mass transfer processes
  - Up-scaling of reaction processes for U(VI) reaction network
- Extend monofractal Cantor bar model (based on 2 contrasting media) to multifractal case (with many contrasting media)
- Effects of heterogeneities within layers
- Statistical distribution of physical properties $\Rightarrow$ stochastic approaches?

Anticipated Research Products

1. New insights into the nature of coupled hydrological, mineralogical, and geochemical processes in partially-saturated, heterogeneously-layered systems
2. Validated approaches for applying laboratory-scale coupled reactive transport parameters to progressively larger and increasingly complex geologic systems

thank you!

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