Modeling Reactive Flows in Porous Media

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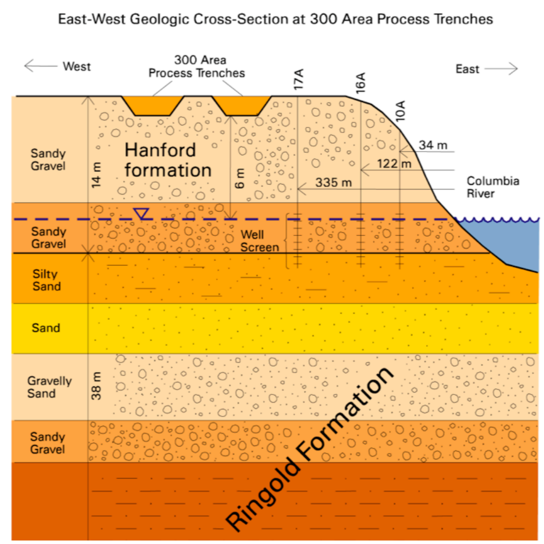
Introduction

- Companion to SciDAC-II project, "Modeling Multiscale-Multiphase-Multicomponent Subsurface Reactive Flows using Advanced Computing", involving several institutions:
  - LANL: Peter Lichtner (PI), Chuan Lu, Bobby Philip, David Moulton
  - ORNL: Richard Mills
  - ANL: Barry Smith
  - PNNL: Glenn Hammond, Steve Yabusaki
  - U. Illinois: Al Valocchi

- Project goals:
  - Develop a next-generation code (PFLOTRAN) for simulation of multiscale, multiphase, multicomponent flow and reactive transport in porous media.
  - Apply it to field-scale studies of
    - Geologic CO2 sequestration,
    - Radionuclide migration at Hanford site, Nevada Test Site,
    - Others…

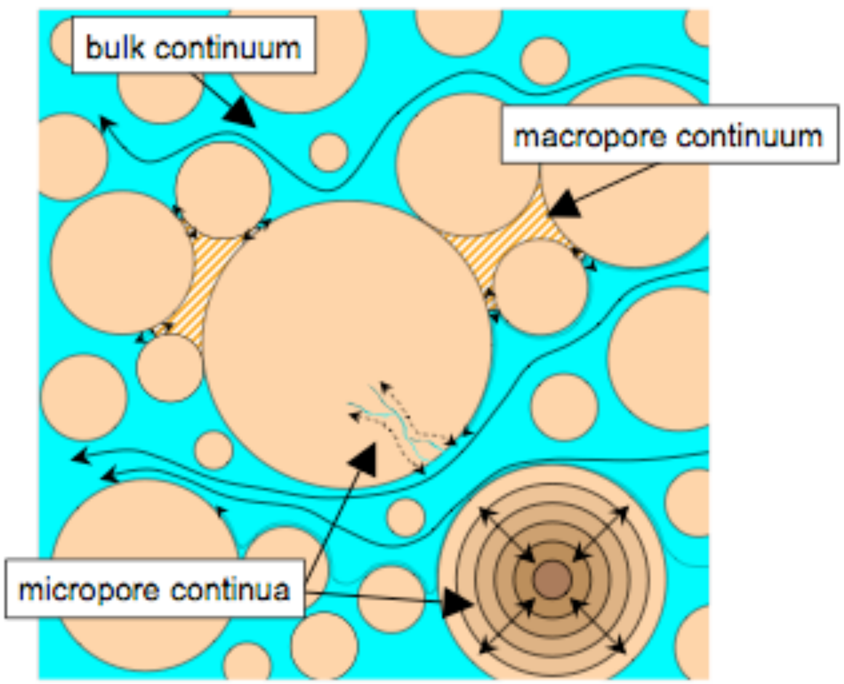
Motivating example -- Hanford 300 area

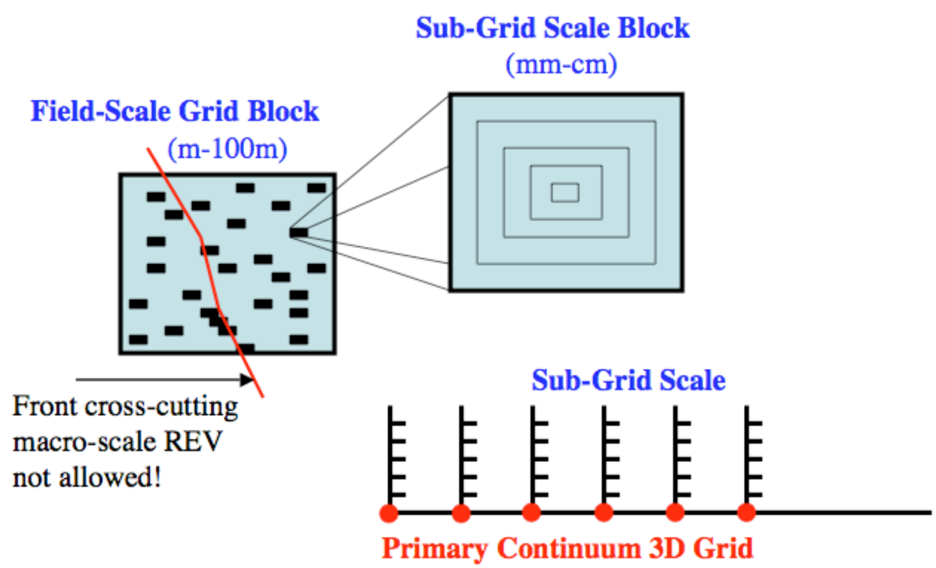


- At the 300 area, U(VI) plumes continue to exceed drinking standards.
- Calculations predicted cleanup by natural attenuation years ago!
- Due to long in-ground residence times, U(VI) is present in complex, microscopic inter-grain fractures, secondary grain coatings, and micro-porous aggregates. (Zachara et al., 2005).
- Constant K_d models do not account for slow release of U(VI) from sediment grain interiors through mineral dissolution and diffusion along tortuous pathways.
- In fact, the K_d approach implies behavior opposite to observations!
- We must accurately incorporate millimeter scale effects over a domain measuring approximately 2000 x 1200 x 50 meters!

Modeling multiscale processes

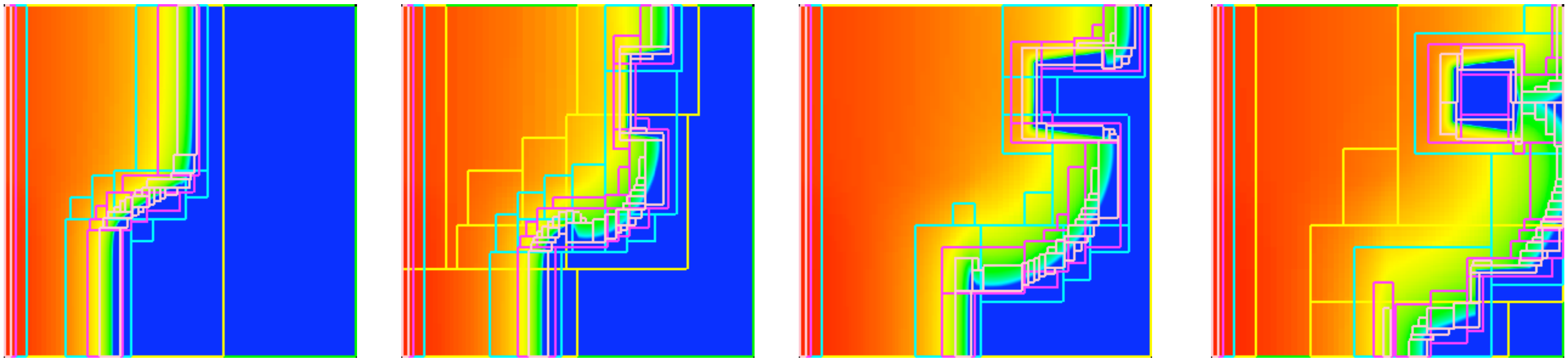
- Represent system through multiple interacting continua with a single primary continuum coupled to sub-grid scale continua.
- Associate sub-grid scale model with node in primary continuum
  - 1D computational domain
  - Multiple sub-grid models can be associated w/ primary continuum nodes
  - Degrees of freedom: $N \times N_K \times N_{\mathrm{DCM}} \times N_c$





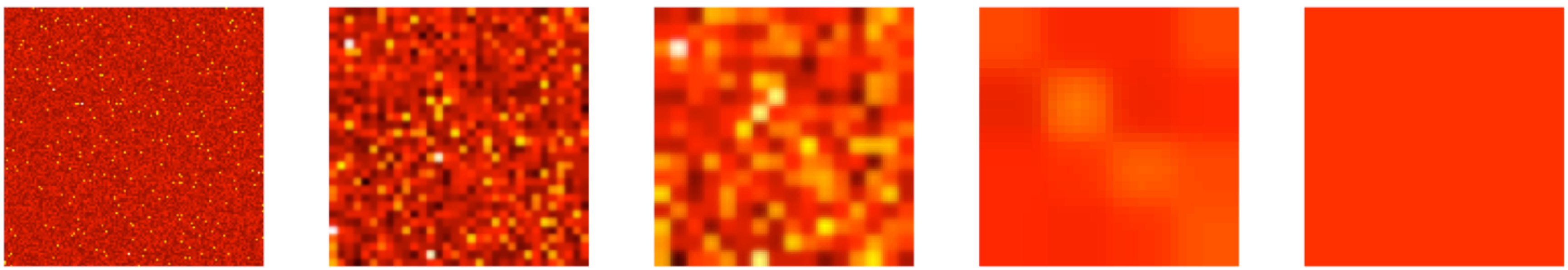
Adaptive mesh refinement (AMR)

- AMR introduces local fine resolution only in regions where needed.
- Significant reduction in memory and computational costs for simulating complex physical processes exhibiting localized fine scale features.
- AMR provides front tracking capability in the primary grid that can range from centimeter to tens of meters.
- Sub-grid scale models can be introduced in regions of significant activity and not at every node within the 3D domain.
- It is not necessary to include the sub-grid model equations in the primary continuum Jacobian even though these equations are solved in a fully coupled manner.



Upscaling

- Governing equations depend on averages of highly variable properties (e.g., permeability) averaged over a sampling window (REV).
- Upscaling and ARM go hand-in-hand: as the grid is refined/coarsened, material properties such as permeability must be calculated at the new scale in a self-consistent manner.



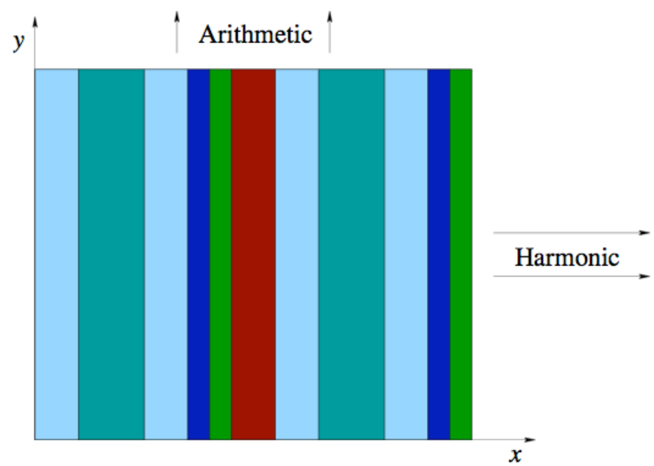
Above: A fine-scale realization (128 x 128) of a random permeability field,

$$\kappa(x,y) = \zeta^{-\ln(\alpha)}, \ \zeta \text{ uniformly distributed in } (0,1), \ \alpha = 5$$

followed by successively upscaled fields (*N* x *N*, *N* = 32, 16, 4, 1) obtained with Multigrid Homogenization (Moulton et al., 1998)

Upscaling

- Coarse-Scale Anisotropy: permeability must, in general, be considered as a tensor at larger scales even if it is a scalar (i.e., isotropic) at the finest scale.
- A single multi-dimensional average is inadequate for modeling flow (MacLachlan and Moulton, 2006)



- Uniform flow from left to right governed by harmonic mean.
- Uniform flow from bottom to top governed by arithmetic mean.
- Suggests a diagonal permability tensor; HOWEVER, if stripes not aligned with coordinate axes, equivalent permeability must be described by a full tensor.

- Upscaling that captures full-tensor permeability includes multigrid homogenization, and asymptotic theory for periodic media.
- Theory is limited to periodic two-scale media (well separated scales)
- Upscaling reactions poses a significant challenge as well. In some aspects of this work volume averaging will suffice, while in others new multiscale models will be required.

PFLOTRAN governing equations

Mass Conservation: Flow Equations

$$\frac{\partial}{\partial t}(\phi s_\alpha \rho_\alpha X_i^\alpha) + \nabla \cdot \left[q_\alpha \rho_\alpha X_i^\alpha - \phi s_\alpha D_i^\alpha \rho_\alpha \nabla X_i^\alpha \right] = Q_i^\alpha$$

$$q_\alpha = -\frac{k k_\alpha}{\mu_\pi} \nabla (p_\alpha - W_\alpha \rho_\alpha g z) \qquad p_\alpha = p_\beta - p_{c,\alpha\beta}$$

Energy Conservation Equation

$$\frac{\partial}{\partial t}\left[\phi \sum_\alpha s_\alpha \rho_\alpha U_\alpha + (1-\phi)\rho_r c_r T \right] + \nabla \cdot \left[\sum_\alpha q_\alpha \rho_\alpha H_\alpha - \kappa \nabla T \right] = Q_e$$

Multicomponent Reactive Transport Equations

$$\frac{\partial}{\partial t}\left[\phi \sum_\alpha s_\alpha \Psi_j^\alpha \right] + \nabla \cdot \left[\sum_\alpha \Omega_\alpha \right] = -\sum_m v_{jm} I_m + Q_j$$

Total Concentration

$$\Psi_j^\alpha = \delta_{\alpha l} C_j^\alpha + \sum_i v_{ji} C_i^\alpha$$

Total Solute Flux

$$\Omega_j^\alpha = (-\tau \phi s_\alpha D_\alpha \nabla + q_\alpha) \Psi_j^\alpha$$

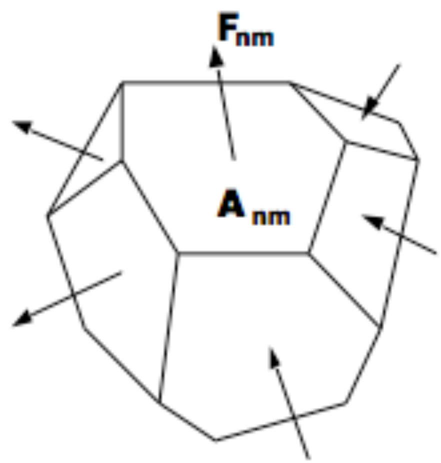
Mineral Mass Transfer Equation

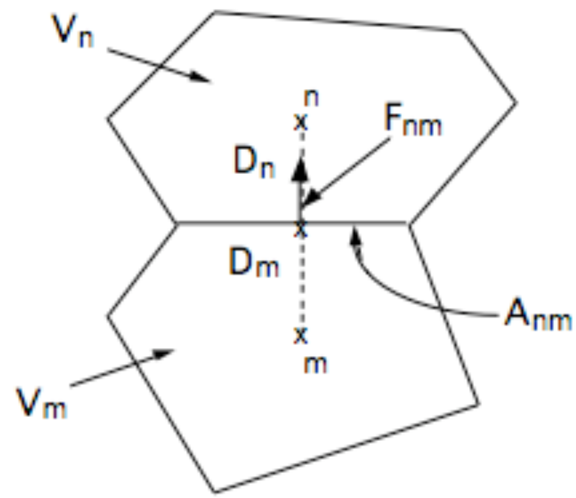
$$\frac{\partial \phi_m}{\partial t} = V_m I_m \qquad \phi + \sum_m \phi_m = 1$$

Integrated Finite-Volume Discretization

Form of governing equation: $$\frac{\partial A}{\partial t} + \nabla \cdot F = S \qquad F = qpX - \phi D p \nabla X$$

Integrated finite-volume discretization



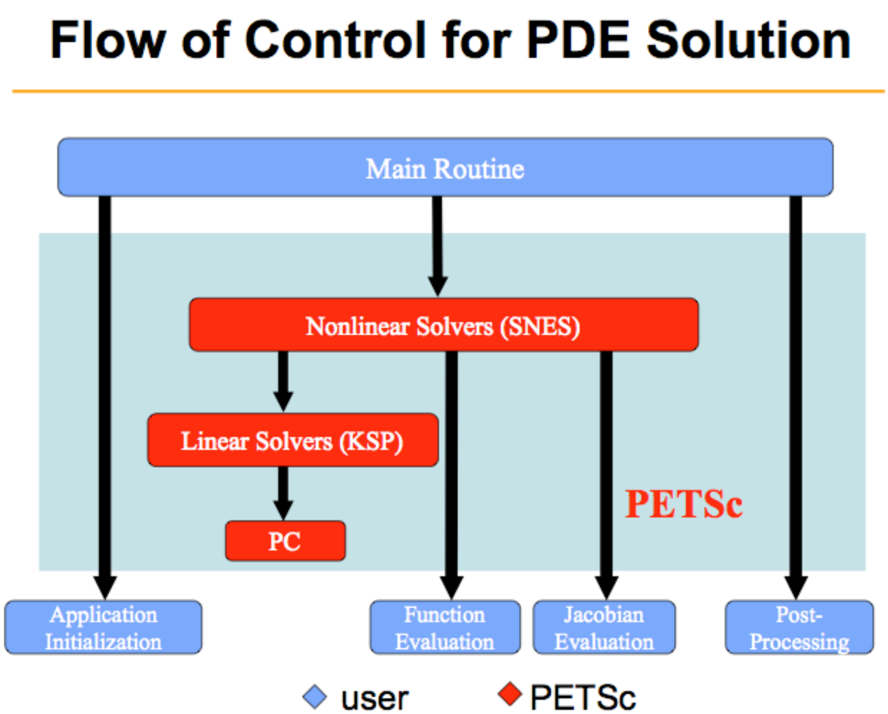


Discretized residual equation: $$R_n = \left(A_n^{k+1} - A_n^k\right)\frac{V_n}{\Delta t} + \sum_{n'} F_{nn'} A_{nn'} - S_n V_n$$

(Quasi-) Newton iteration: $$\sum_{n'} J_{nn'}^i \delta x_{n'}^{i+1} = -R_n^i \qquad J_{nn'}^i = \frac{\partial R_n^i}{\partial x_{n'}^i}$$

PFLOTRAN architecture

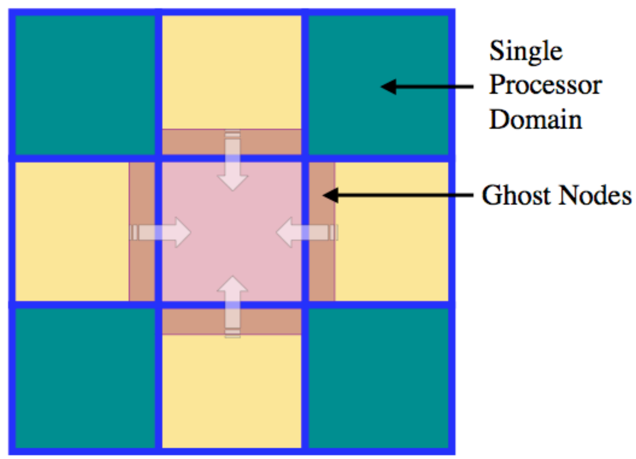
- PFLOTRAN designed from the ground up for parallel scalability.
- Built on top of PETSc, which provides
  - Management of parallel data structures,
  - Parallel solvers and preconditioners,
  - Efficient parallel construction of Jacobian and residuals,



- AMR capability being built on top of SAMRAI.

Parallelization of the multi-scale model

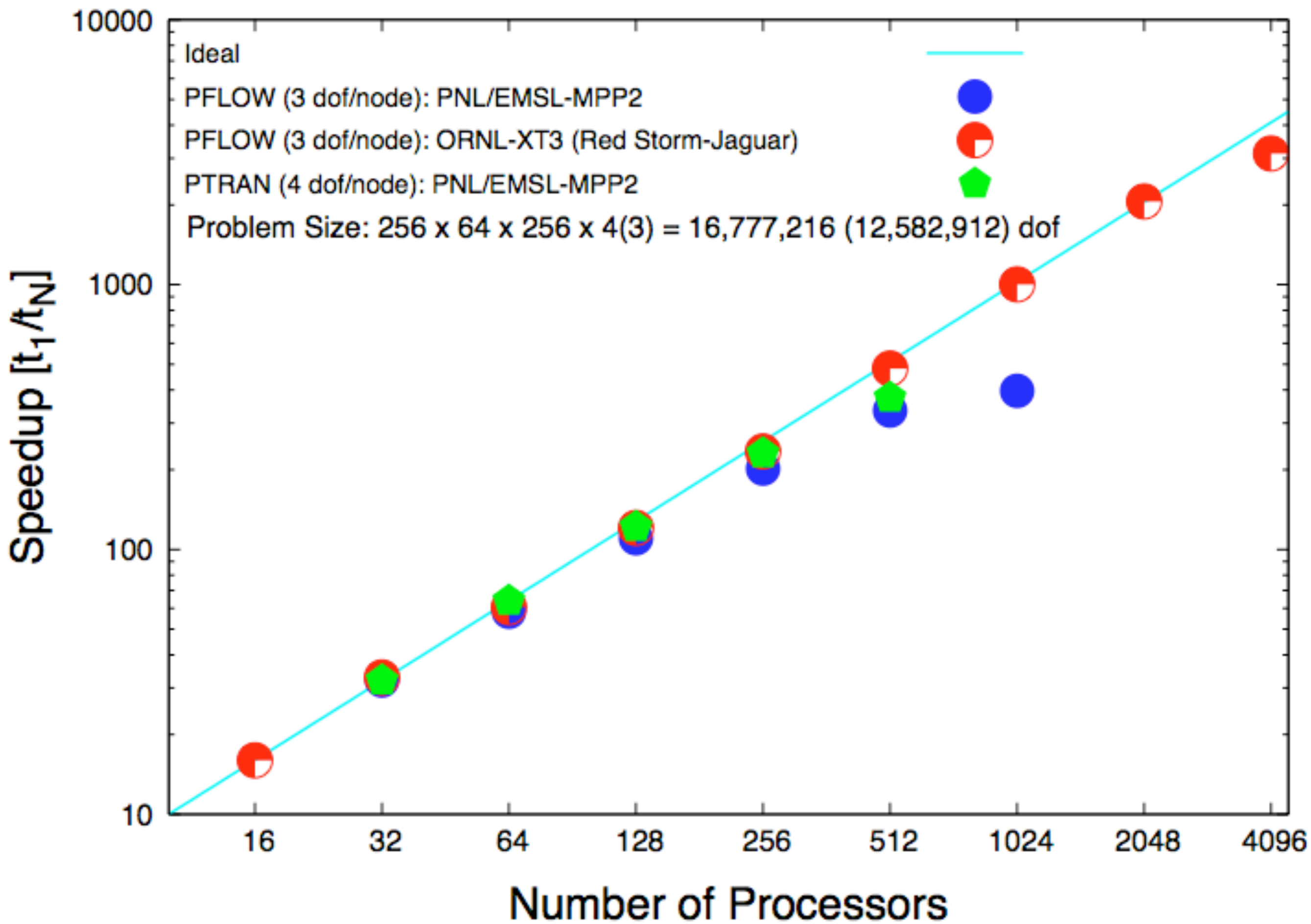
- Rigorously decouple primary and sub-grid scale equations over a Newton iteration (time step in linear case)
- Eliminate sub-grid scale boundary concentration from primary continuum equation (forward “embarrassingly” parallel solve).
- Solve primary equations in parallel using domain decomposition.



- Obtain sub-grid scale concentration (backward “embarrassingly” parallel solve).

Parallel scalability

So far, PFLOTRAN has exhibited excellent strong scaling on Jaguar:



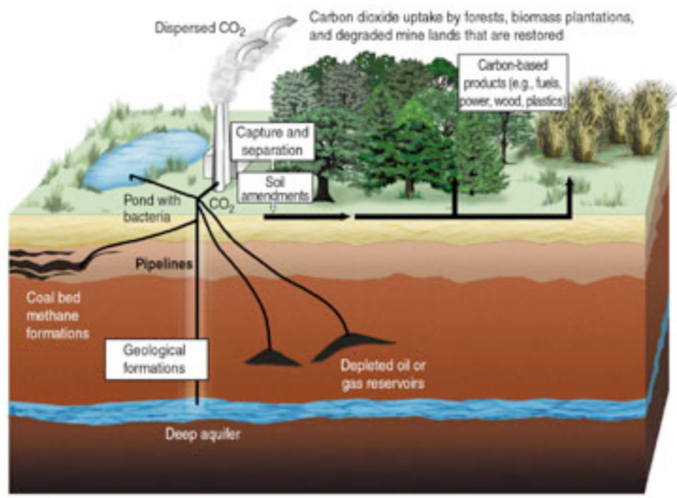
Application: Hanford 300 Area

- Lab experiments (Zachara et al., 2005) indicate that presence of pore structures that limit mass transfer is key to U(VI) persistence.
- Accurate characterization of pore scale effects and effective subgrid parameterizations needed for scientifically defensible decision making.

- Apply PFLOTRAN to a site-wide model of U(VI) migration, including:
  - Transport in both vadose zone (where source is located) and saturated zone (groundwater flow to Columbia River).
  - Surface complexation and ion exchange reactions, and kinetic phenomena caused by intra-grain diffusion and precipitation/dissolution of U(VI) solid phases to account for observed slow leaching of U(VI) from source zone.
  - Robust model for remobilization of U(VI) as river stage rises and falls, causing mixing of river water w/ ambient groundwater in vadose zone.
    - Must track river stage on daily basis.
    - AMR is key to track transient behavior induced by stage fluctuations.

Application: Geologic CO2 sequestration

- Capture CO2 from power production plants, and inject it as supercritical liquid in abandoned oil wells, saline aquifers, etc.



LeJean Hardin and Jamie Payne, ORNL Review, v.33.3.

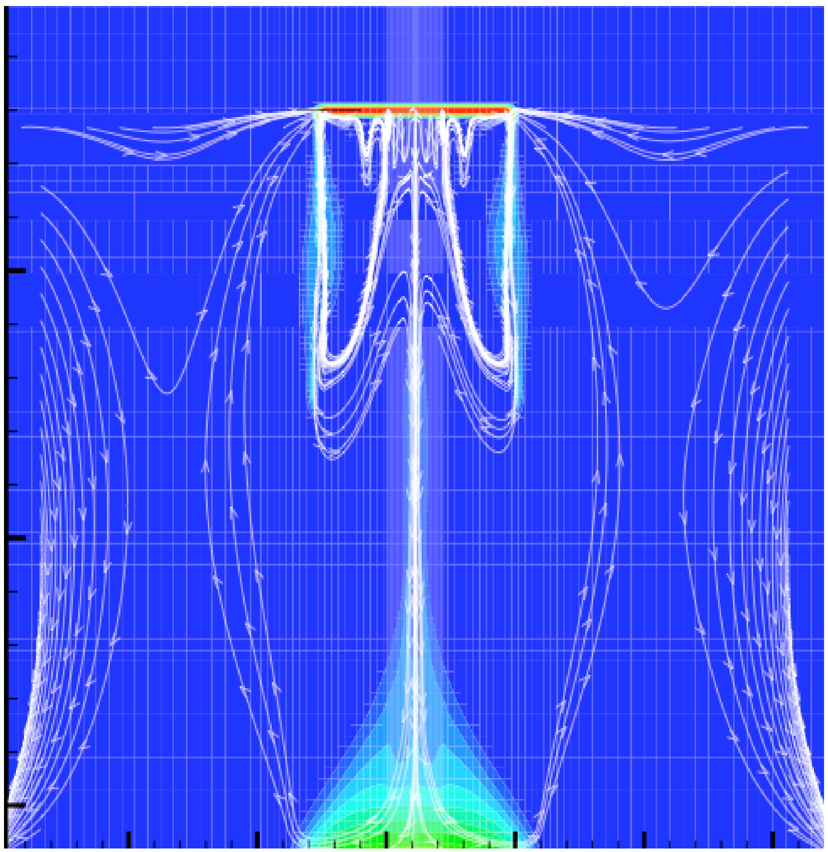
- Must be able to predict long-term fate:
  - Slow leakage defeats the point.
  - Fast leakage could kill people!

- Many associated phenomena are very poorly understood.

Application: Geologic CO_2 sequestration

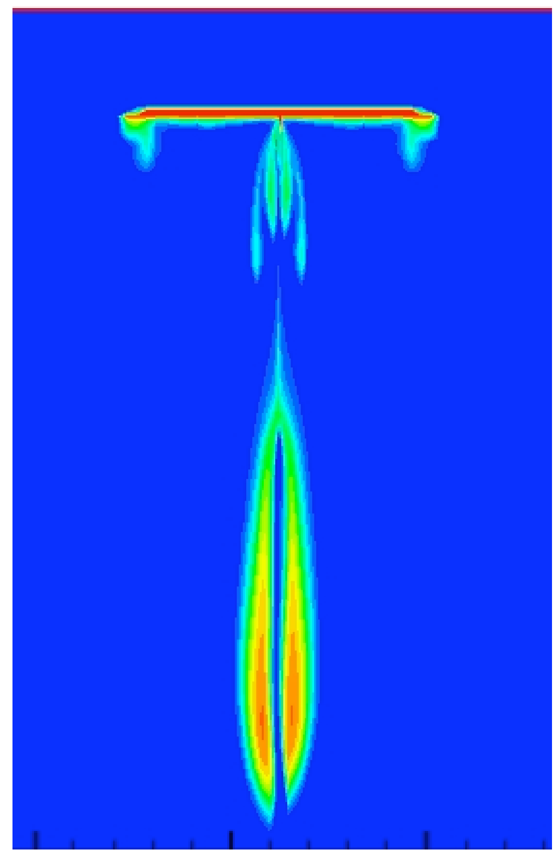
Density driven fingering is one feature of interest:

- Density increases as supercritical CO_2 dissolves into formation brine.
- Buoyancy effects result in fingering.
- Widths may be on the order of meters or smaller.



Left: Density-driven vortex made the fluid with higher CO_2 concentration "snap-off" from the source -- the supercritical CO_2 plume.

Right: Enlarged center part of this domain at earlier time, illustrating two sequential snap-off, the secondary is much weaker than the first one. The detailed mechanisms behind these behavior are under investigation.



CO2 sequestration: pH fingering

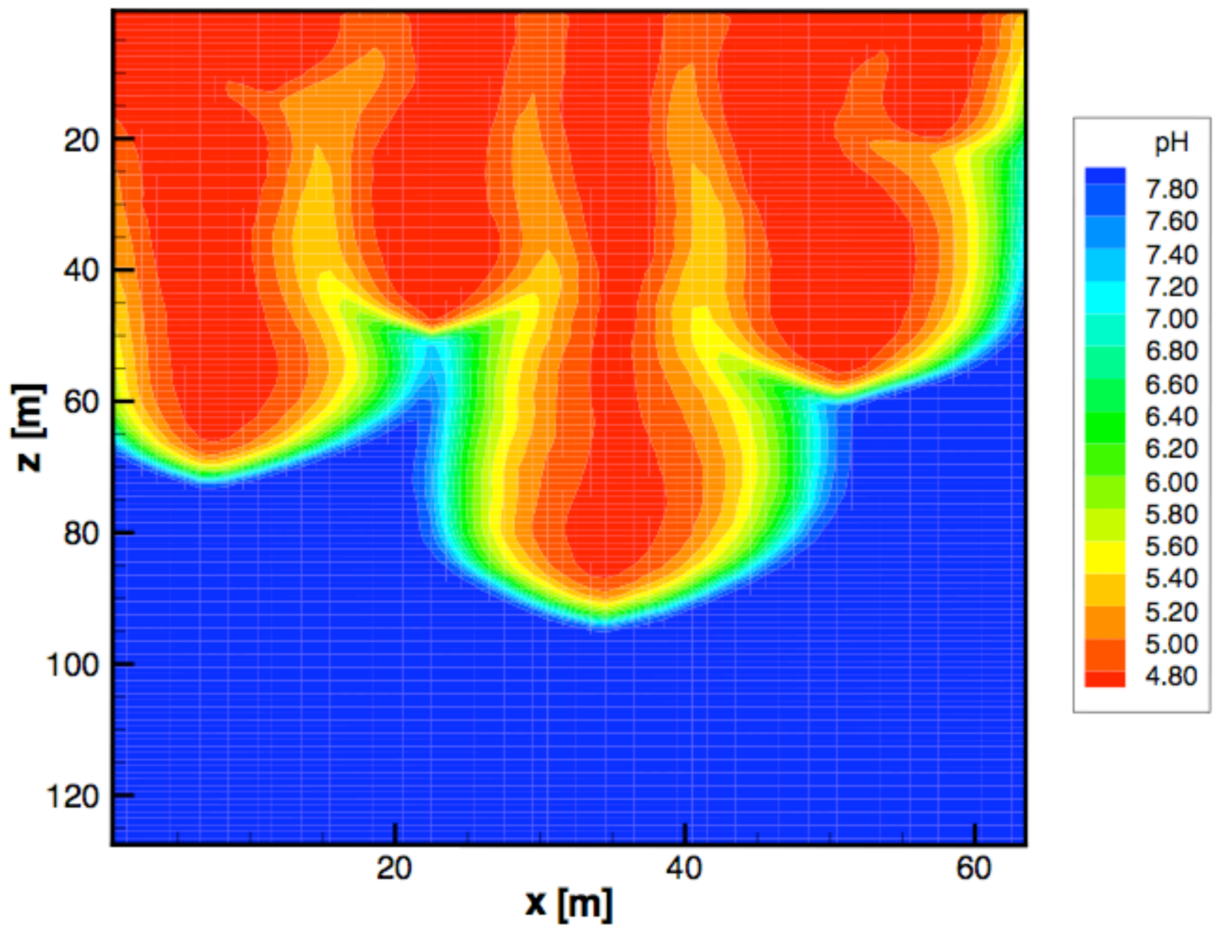


Figure: pH fingering due to density instabilities, 200 years after injection

Planned CO_2 sequestration studies with LCF

- We will study the SACROC unit in the Permian Basin of West Texas.
- CO_2 flooding for enhanced oil recovery began in 1972.
  - Since then, 68 MT CO_2 have been sequestered.
  - 30 MT are anthropogenic,
    derived by separation from Val Verde natural gas field.

- We have a 9-million node logically structured grid for SACROC.
- We will use ~10 degrees of freedom per node to represent the chemical system.

- One task is to investigate CO_2 density-driven fingering:
  - Characterize finger widths for typical reservoir properties.
  - Characterize critical time for fingering to occur.
  - Examine conditions where theoretical stability analysis yields ambiguous results.

Acknowledgements

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