A Numerical Analysis of Wetting Front Instability Induced by Infiltration of Highly Saline Fluids

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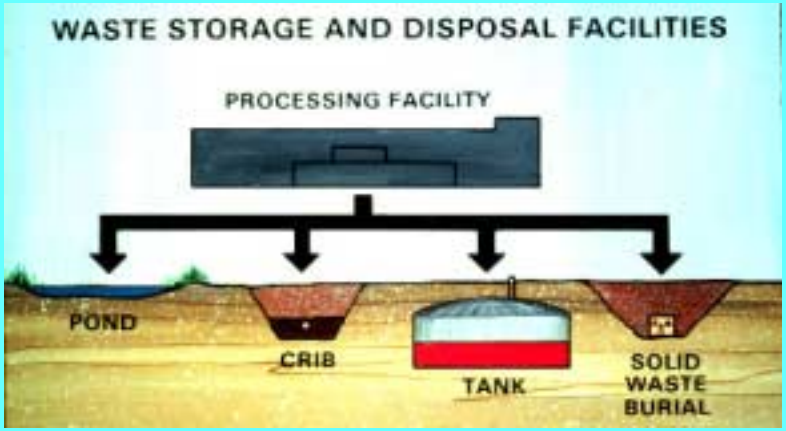
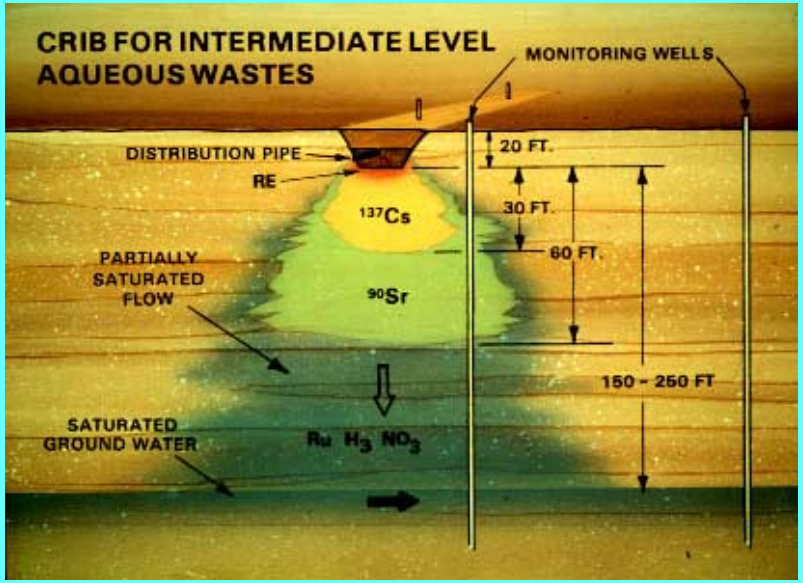
Background

- 67 of 149 single-shelled high level waste tanks are known or suspected leakers
- contaminants “deeper than would be expected” under ideal conditions
- non ideal wastes:
  - caustic, $10 < pH < 14$
  - saline, $6 \leq N \leq 12$
  - dense, specific gravity ≥ 1.4





Objectives:

- **Identify factors controlling the transport of highly saline fluids through the vadose zone**
  - relative effects of surface tension
  - do these factors contribute to fingering?
- **develop and incorporate theory into a numerical model for prediction at realistic spatial and temporal scales**

Effect of Fluid Properties on Contaminant Mobility

Usually neglected under low concentrations but at high concentrations could affect contaminant location and distribution via:

- **viscosity and density**
- **wettability**
  - capillary pressure
  - relative permeability
  - residual saturations
  - fluid displacement potential

Thermodynamic Model for Surface Tension of Concentrated Electrolytes

Gibbs thermodynamic, or phenomenological surface-phase, method (Butler, 1932). For salt MX in water (component i = M, X, w):

$$\mu_i^{\,B} = \mu_i^{\,BO} + RT \ln a_i^{\,B}$$

$$\mu_i^{\,S} = \mu_i^{\,SO} + RT \ln a_i^{\,S} - \overline{A}_i \gamma_{sol}$$

Vapor Phase

Surface Phase
MX + H_2O

Liquid Phase
MX + H_2O

Assumptions:

$$\mu_w^{\,B} = \mu_w^{\,S} \text{ at equilibrium}$$

$$A_w = \overline{A}_w = V_w^{\,2/3} N^{1/3}$$

$$m^S = g\, m^B$$

Electrolyte Surface Tension

$$\gamma_{sol} = \gamma_w + \frac{RT\nu}{A_w M_w}\left[m^B \phi^B - m^S \phi^S\right]$$

Osmotic Coefficient, Pitzer (1973):

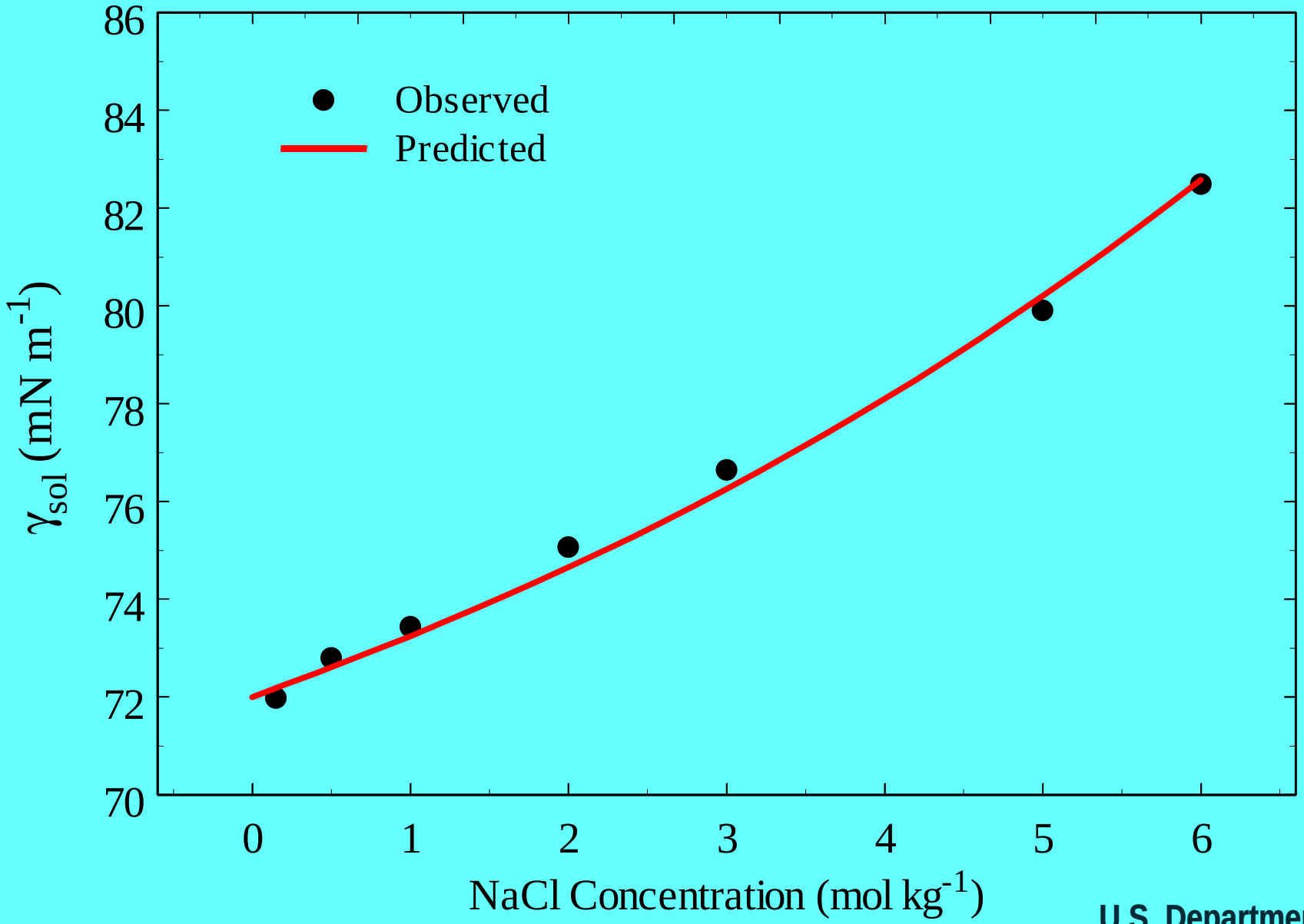
$$\phi = 1 - \left|z_M z_X\right| f^{\phi} + m(2\nu_M \nu_X / \nu) B^{\phi}_{MX} \ldots$$

$$+ m^2 [2(\nu_M \nu_X)^{3/2} / \nu] C^{\phi}_{MX}$$

$$f^{\phi} = -A_{\phi} I^{0.5} / (1 + bI^{0.5})$$

$$A_{\phi}(m,T,P) = \frac{1}{3}\left(\frac{2\pi N_o \rho_w}{1000}\right)^{0.5}\left(\frac{e^2}{4\pi\varepsilon_0 \varepsilon_r kT}\right)^{1.5}$$

Observed vs Predicted γ_{sol} for NaCl



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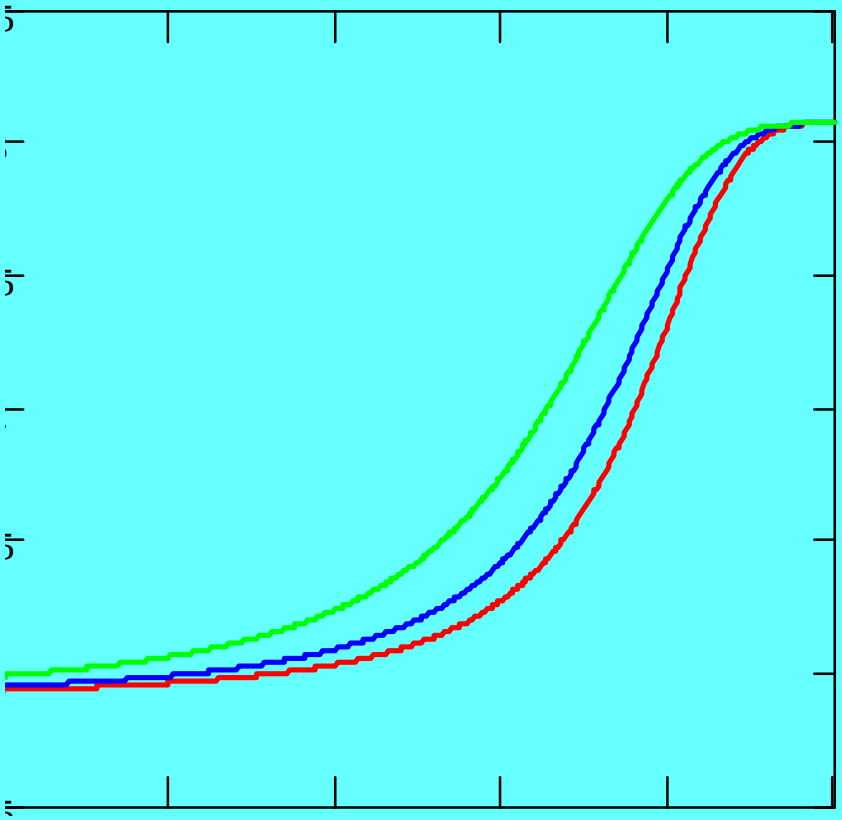
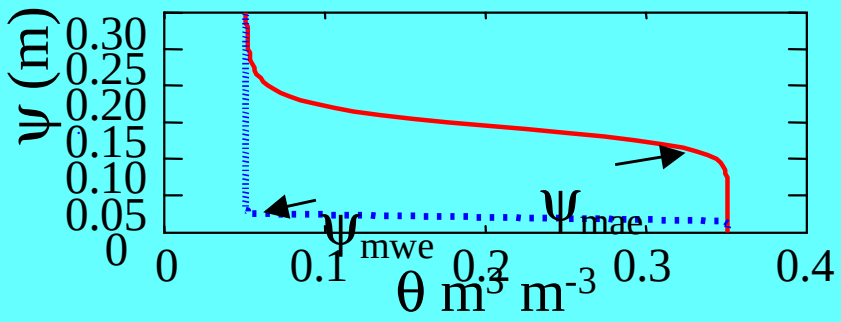
Effect of γ_{sol} on Hydraulic Properties

- Drainage is less sensitive to wetting angle, W
- effect of wetting angle on initial imbibition is minor for W < 30°
- spontaneous imbibition is arrested at W > 55°, which corresponds to a molality of about 9 mol kg^{-1}

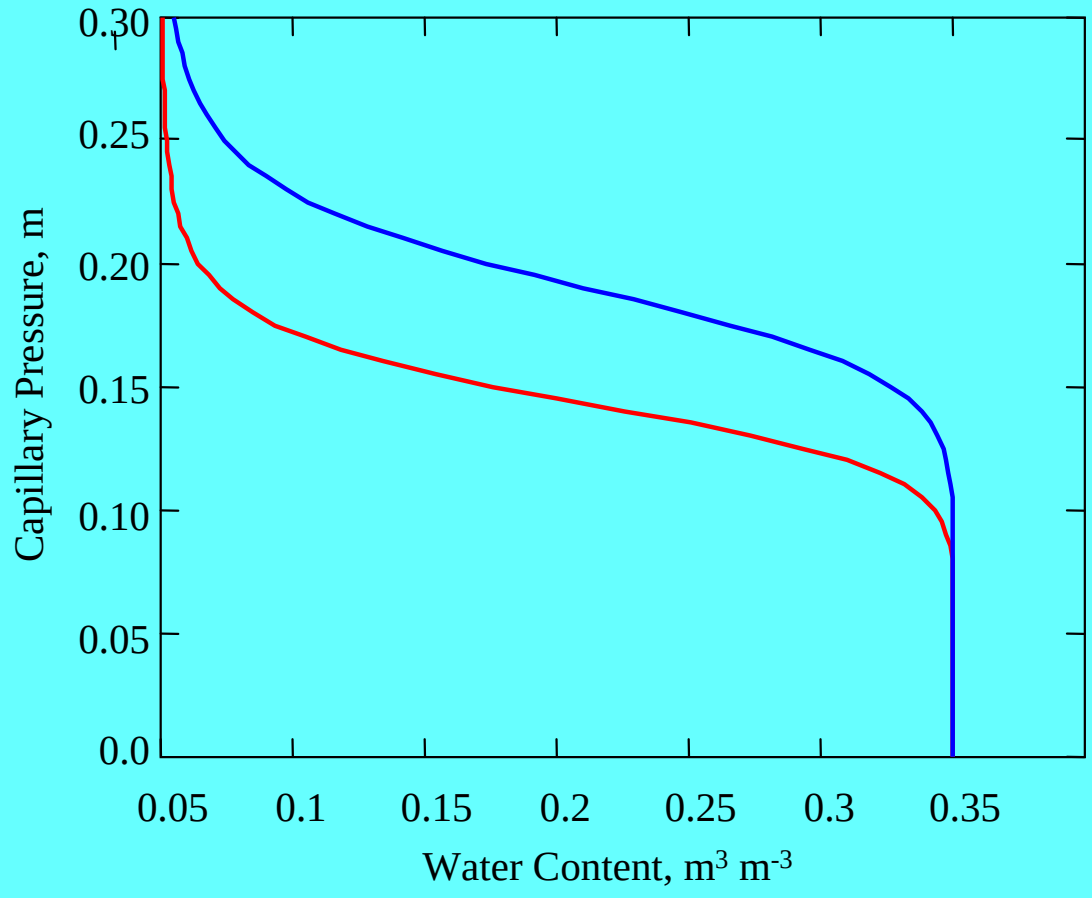


Modeling Finger Formation

- Must consider capillary hysteresis (Nieber, 1996)
- observed in soils and soils with steep wetting curves (hydrophobic)
- Water flux < K_{sat}
- if $\psi_{mwe} < \psi_{mae}$

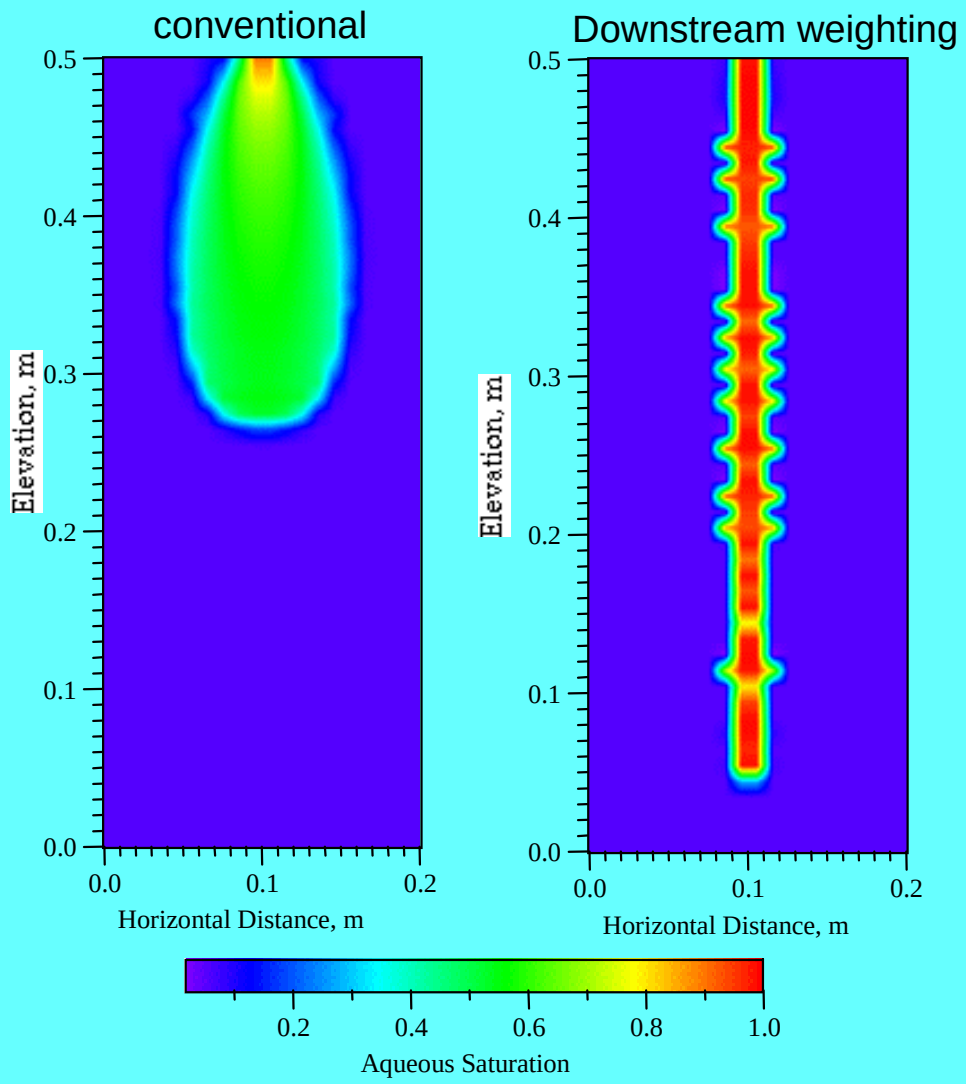


Effect of Surface Tension on Moisture Release

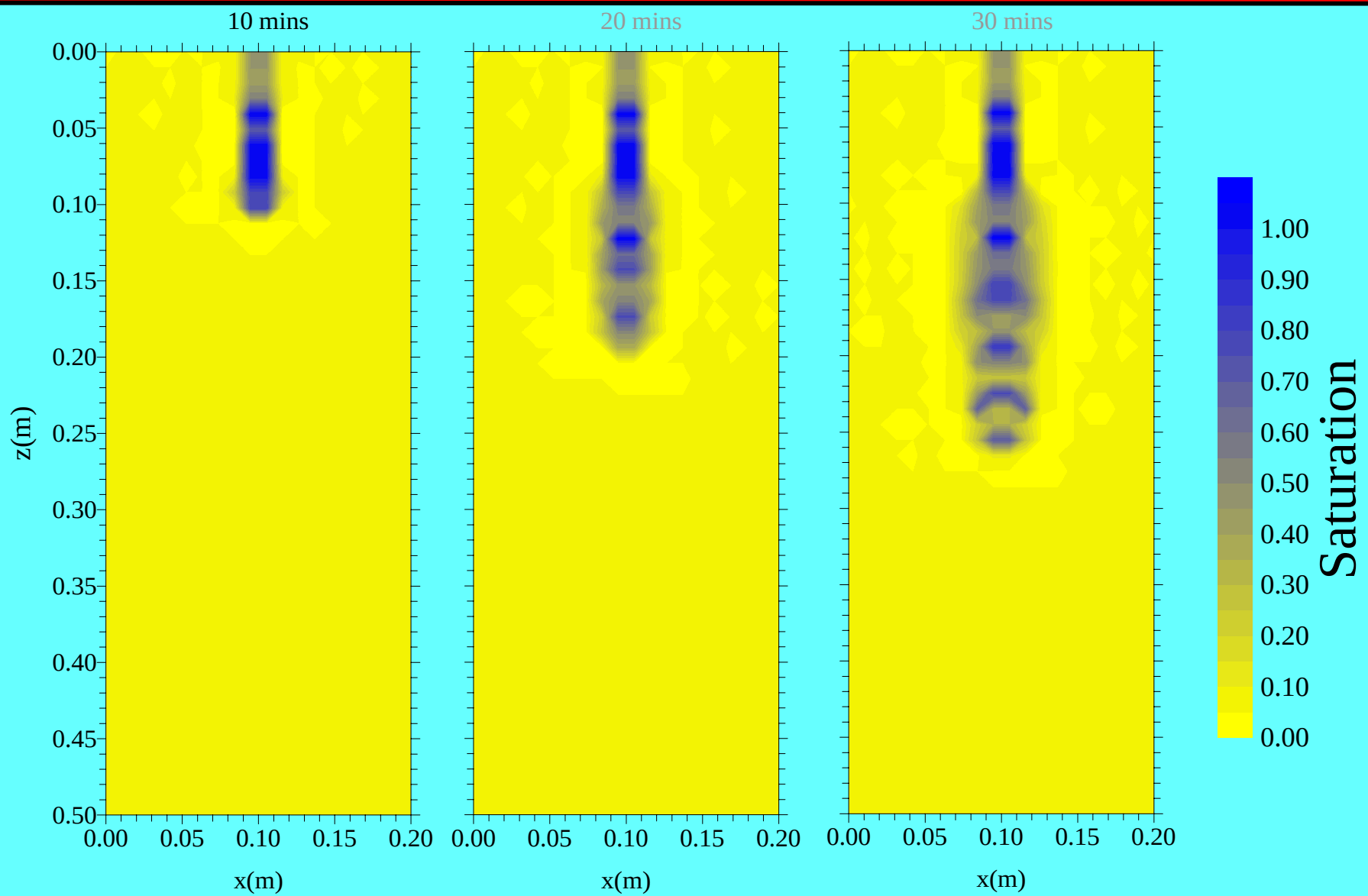


Preliminary Modeling Results

- **Results from upgraded STOMP model using**
  - improved interfacial averaging of K(ψ)

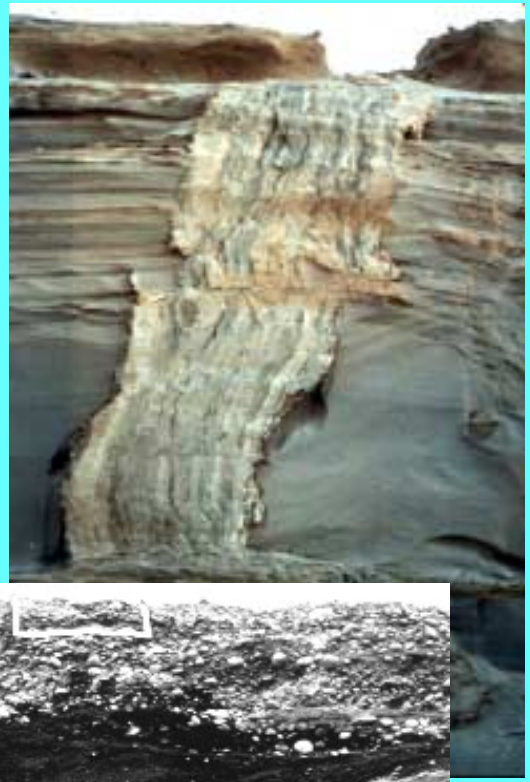


STOMP Simulations-Homogenous Column

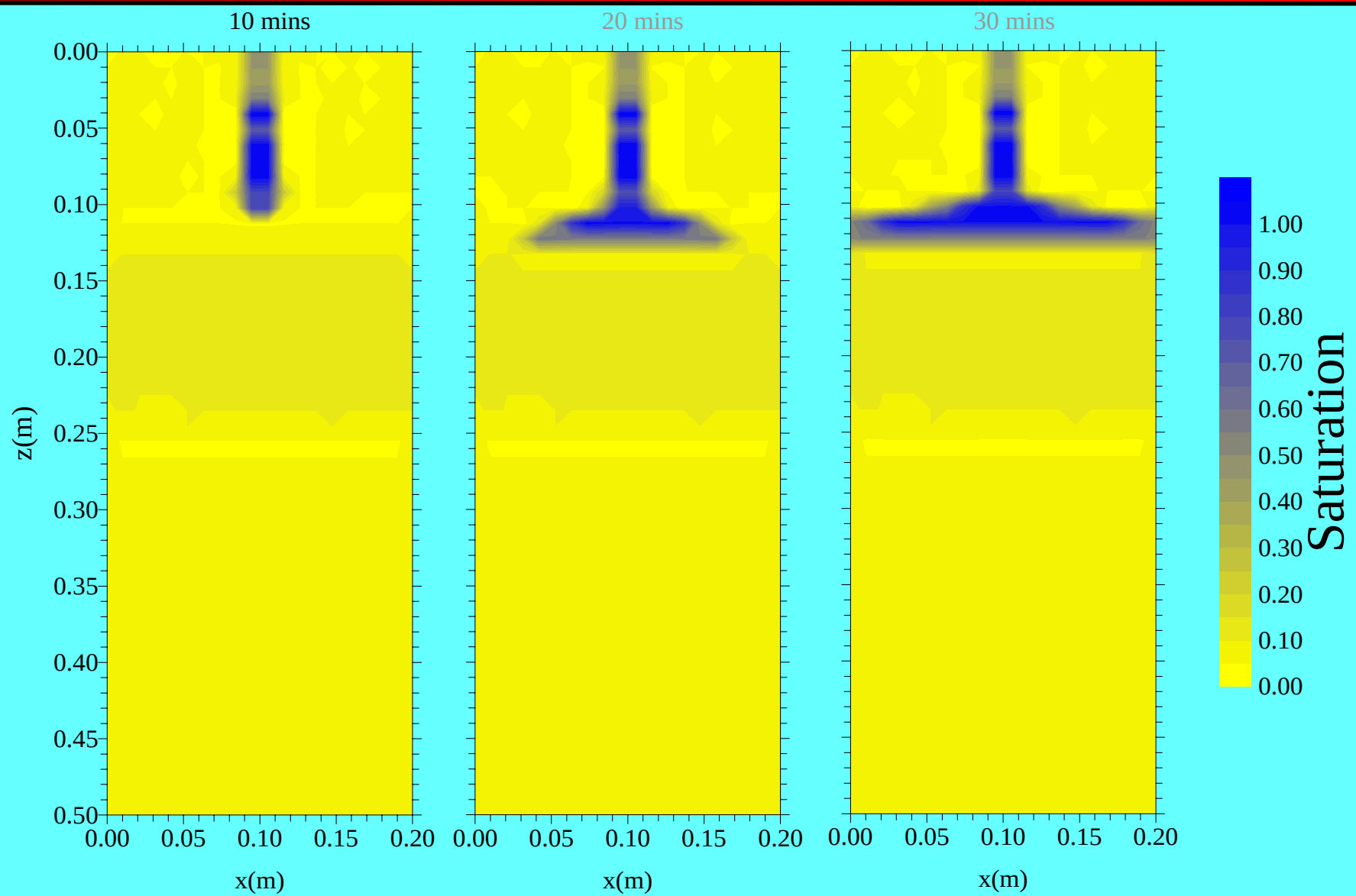


Hydrostratigraphy at Hanford-

Hanford Sands
Hanford Gravels
Plio-Pleistocene Unit
Ringold Conglomerate



STOMP Simulations- Layered Column



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Summary and Conclusions

- Traditional transport zone theory does not explain field observations
- hypothesized that fluid properties may influence contaminant migration
- thermodynamic model for electrolyte surface tension allows scaling of water retention function
- concentrated electrolytes causes initial change in wettability, which may cause fingering in early time
- eventual dilution by antecedent moisture causes some dissipation of finger