Rapid Migration of Radionuclides Leaked from High-Level Waste Tanks: Effects of Salinity Gradients, Wetted Path Geometry and Water Vapor Transport

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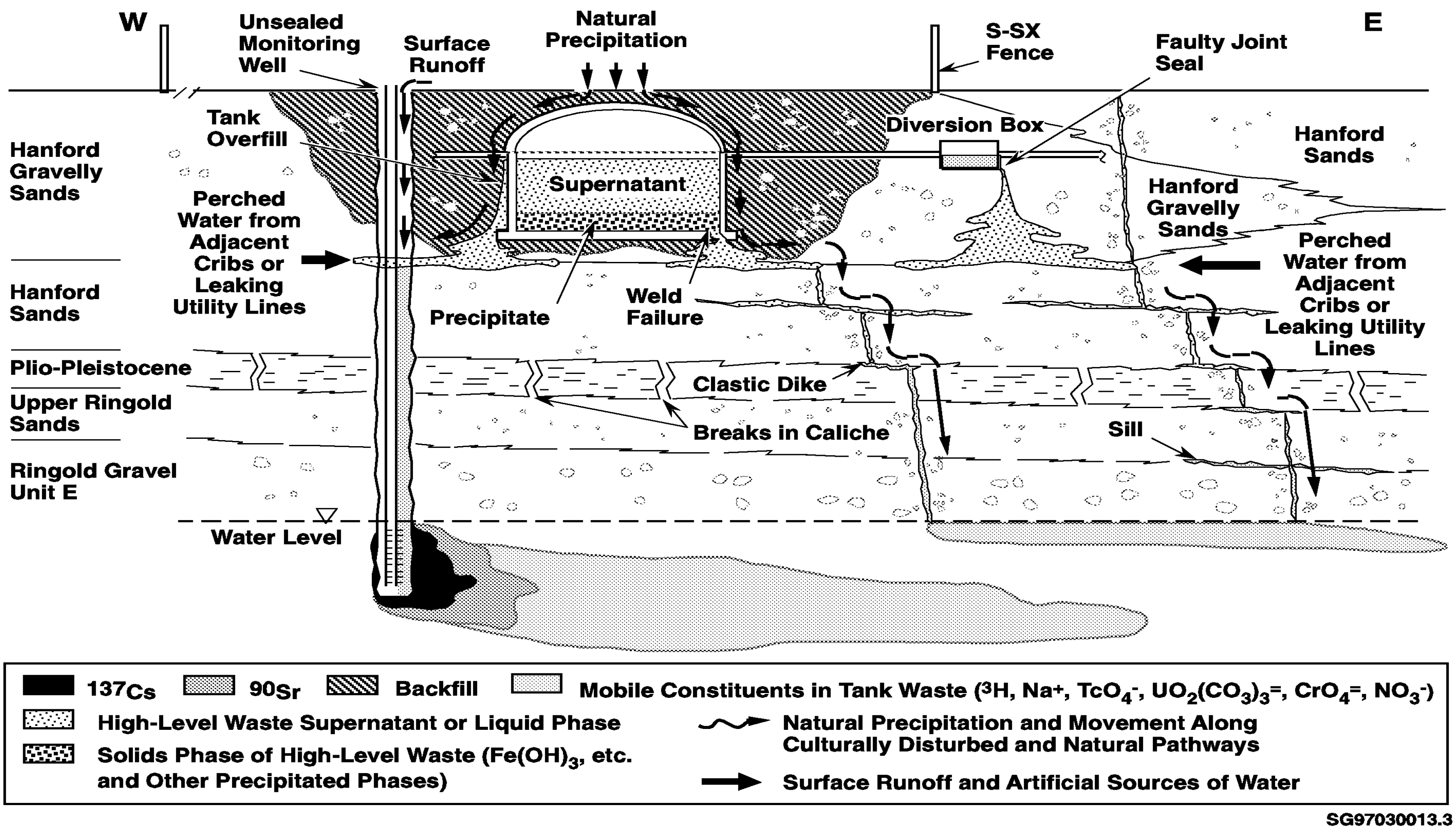
Background

- 177 High Level Waste Tanks (67 of 146 Single Shell Tanks Known or Suspected to Have Leaked)
- much uncertainty in leak volumes
- monitoring accounts for < 1% of total inventory
- ^{137}Cs “deeper than expected”
- some mobile contaminants in the ground water

Problem

- Traditional vadose zone transport theory and modeling approaches appear inadequate for explaining observed distribution of contaminants
  - episodic infiltration events
  - fluid properties
  - small-scale variability in lithology
  - enhanced vapor flux
  - unstable wetting fronts

Hypothetical Sources and Potential Pathways



Need

- basic research in vadose zone hydrology
  - understand subsurface flow/transport
  - improve effectiveness of remediation
  - calculate risk associated with different clean up strategies

Scope This Study

To investigate the Effects of Fluid Properties

- 54 million gallons of radioactive and hazardous waste
- caustic: $10 < pH < 14$
- saline: $6 < N < 10$
- dense: specific gravity ≥ 1.4

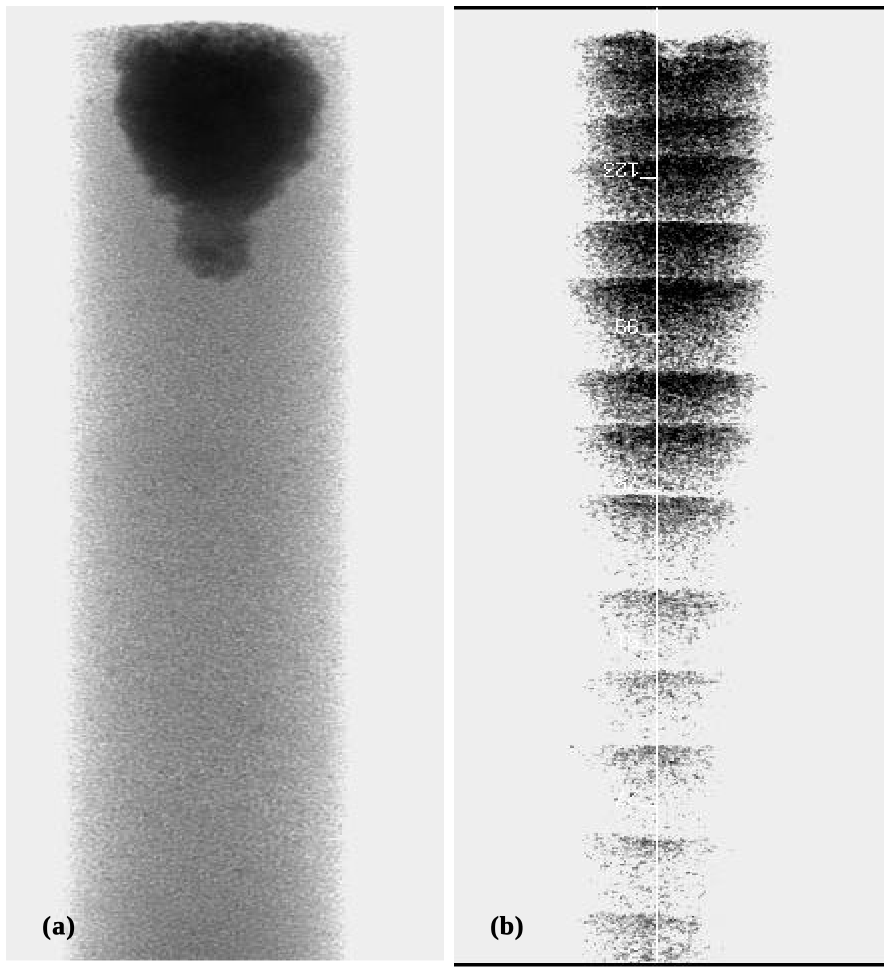
Objectives

- study the effect of elevated surface tension of caustic brines on flow and transport
- determine the conditions under which osmotically-enhanced vapor flux operate and quantify impact on transport processes
- develop and incorporate theory describing these processes into STOMP for prediction at realistic spatial and temporal scales

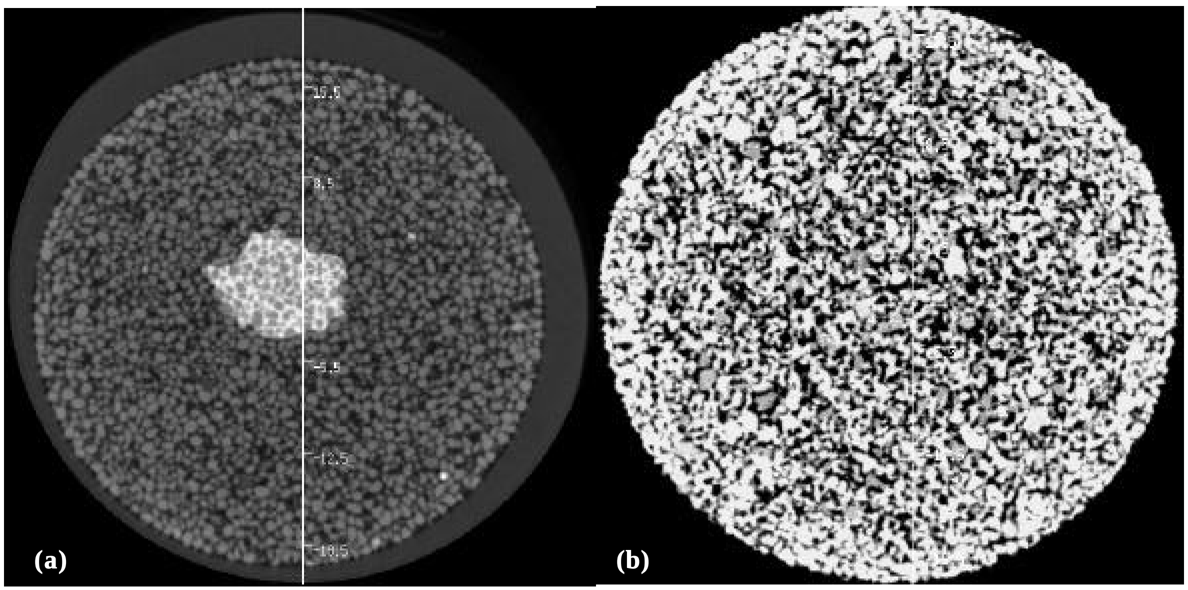
Collaborators

- Oregon State University (OSU)
  - laboratory scale experiments with Miller Sands
- Desert Research Institute (DRI)
  - effects of hypersaline fluids on hydraulic properties
- Pacific NW National Laboratory (PNNL)
  - laboratory experiments with Hanford Sediments
  - field experiments
  - modeling

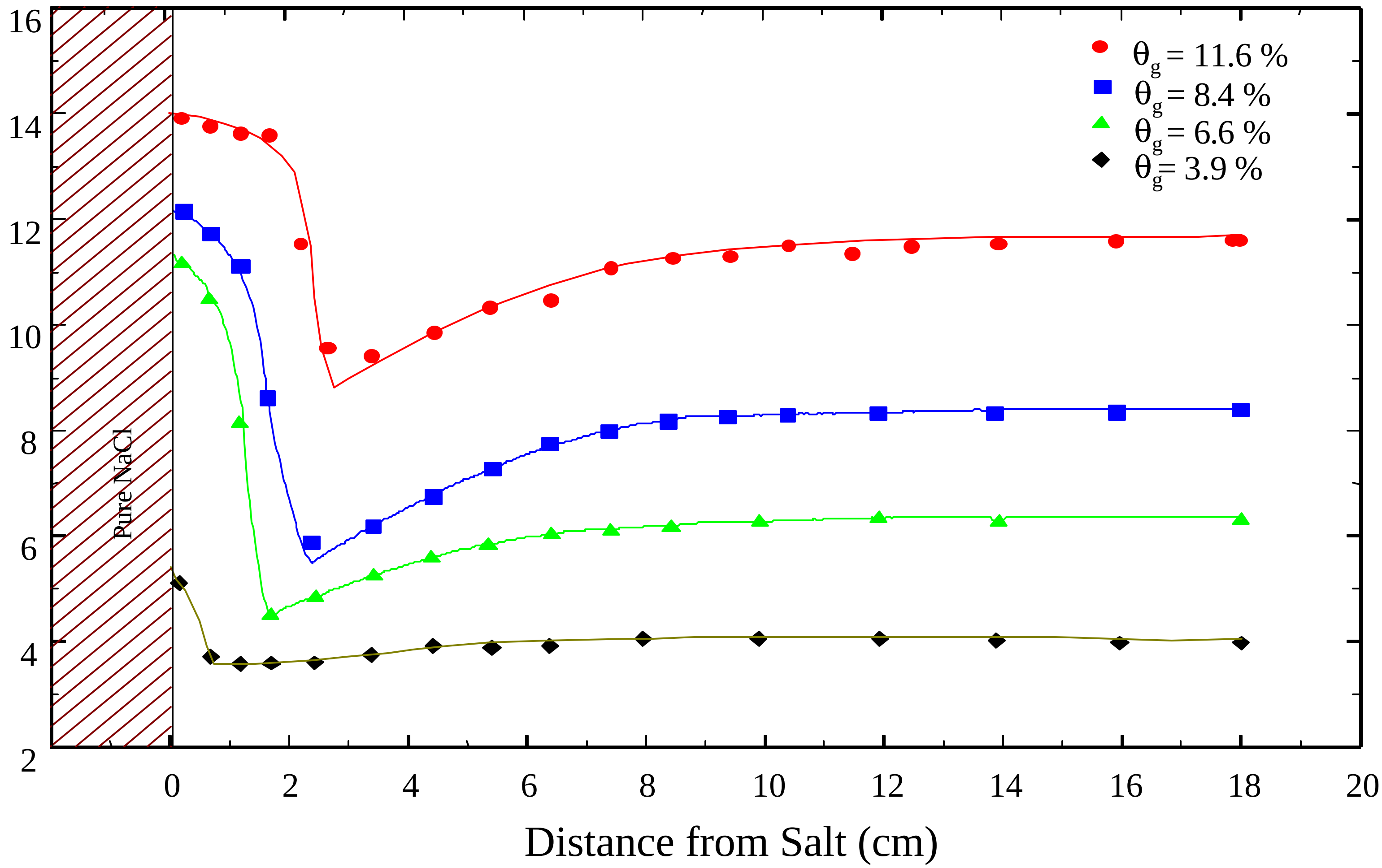
Xray CT Studies- Unstable Flow



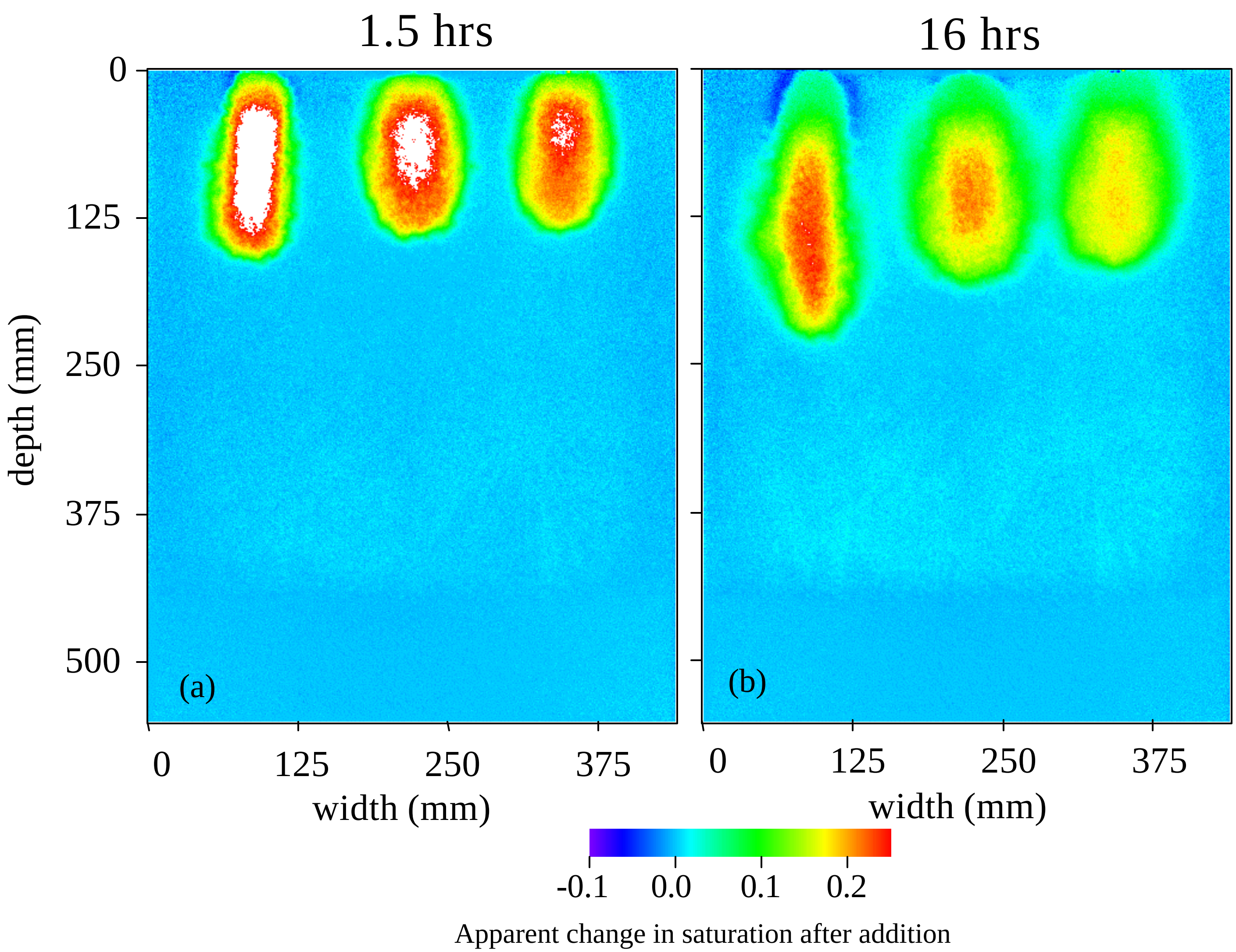
X-ray CT- Unstable Flow



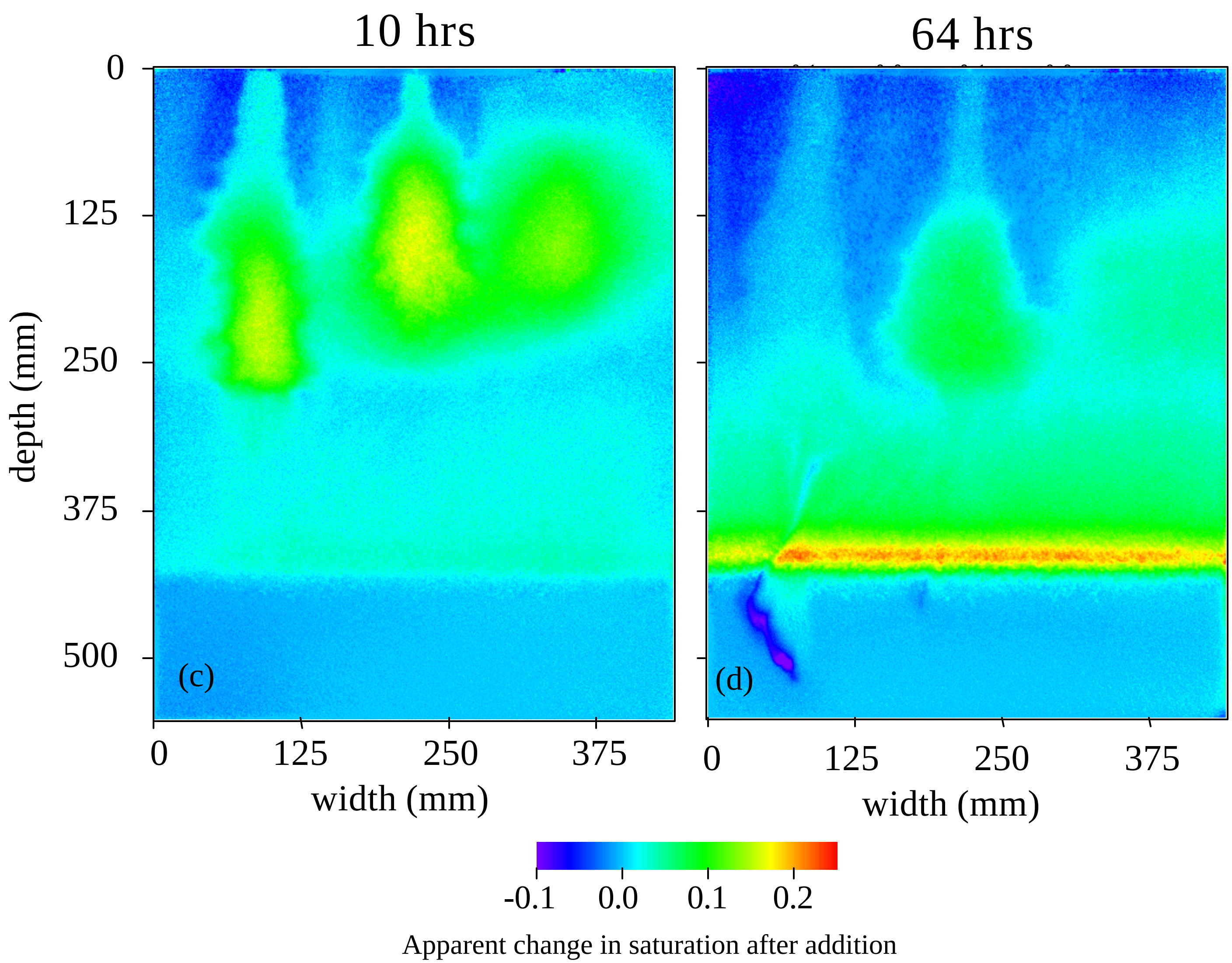
Osmotic Effects of Moisture Distribution



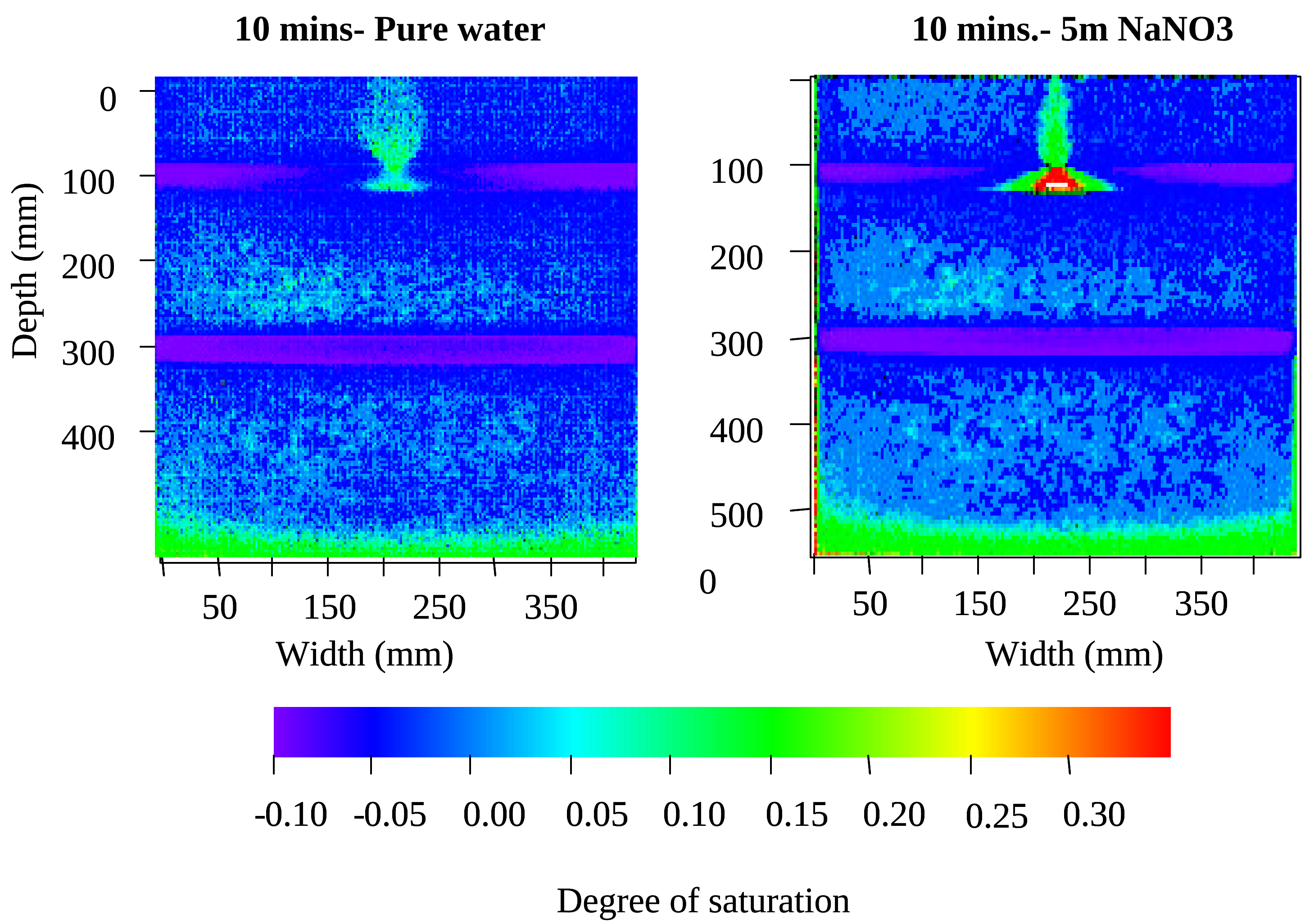
Laboratory Studies- Homogeneous Sands



Laboratory Studies- Homogeneous Sands



Laboratory Studies- Layered Sands

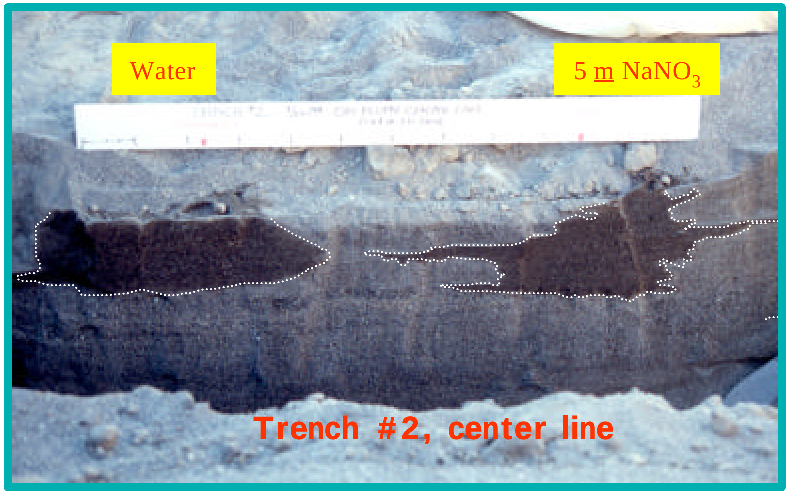


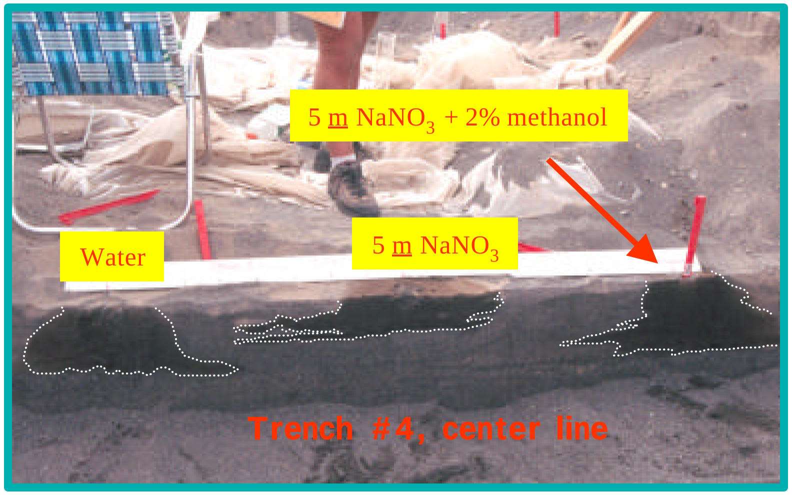
Field Infiltration Tests

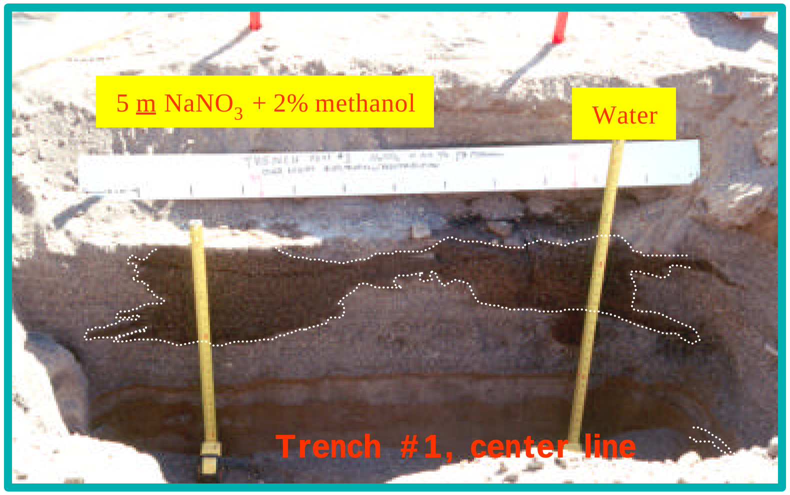
- Six reps
- Solutions applied at 0.4 L/ hr over 4 hrs with a tension infiltrometer
- Trench excavated between 6 and 12 hrs after application

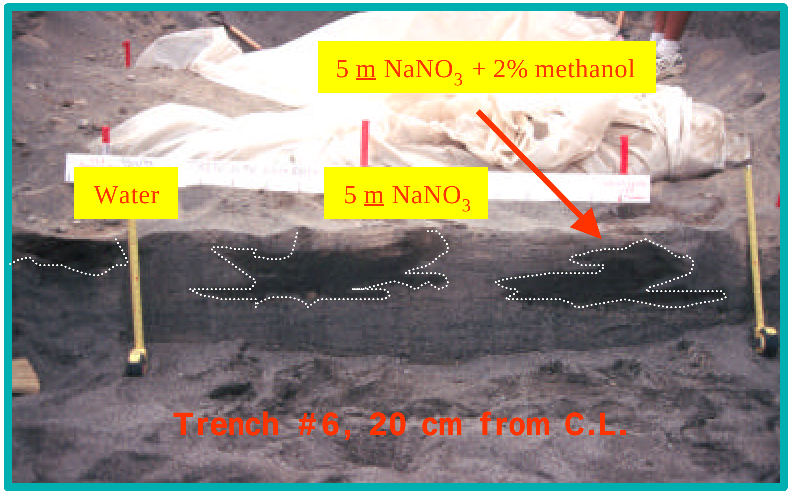
Typical Profile at Field Site

Field Studies









Plume Characteristics

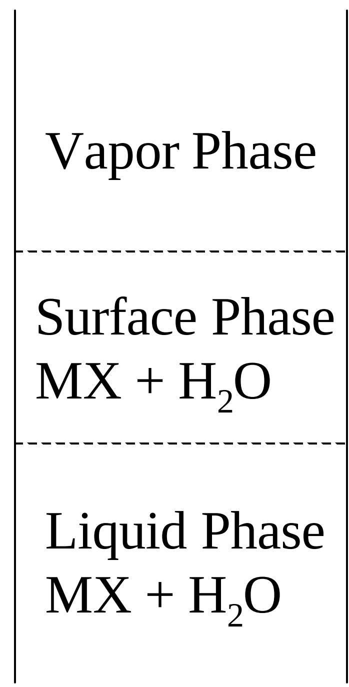
Solution	Mean depth (cm)	Mean width (cm)	Aspect ratio
5 m NaNO3	17.5 [3.7]	75.2 [6.5]	4.43 [0.87]
5 m NaNO3 + 2% methanol	22.75 [2.4]	67.5 [3.5]	2.99 [0.48]
Water	20.25 [5.9]	58.3 [10.3]	3.16 [1.20]

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):

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

$$m_i^{S} = m_i^{SO} + RT \ln a_i^{S} - \overline{A}_i g_{sol}$$



Assumptions:

$$m_w^{B} = m_w^{S} \text{ at equilibrium}$$

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

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

Electrolyte Surface Tension

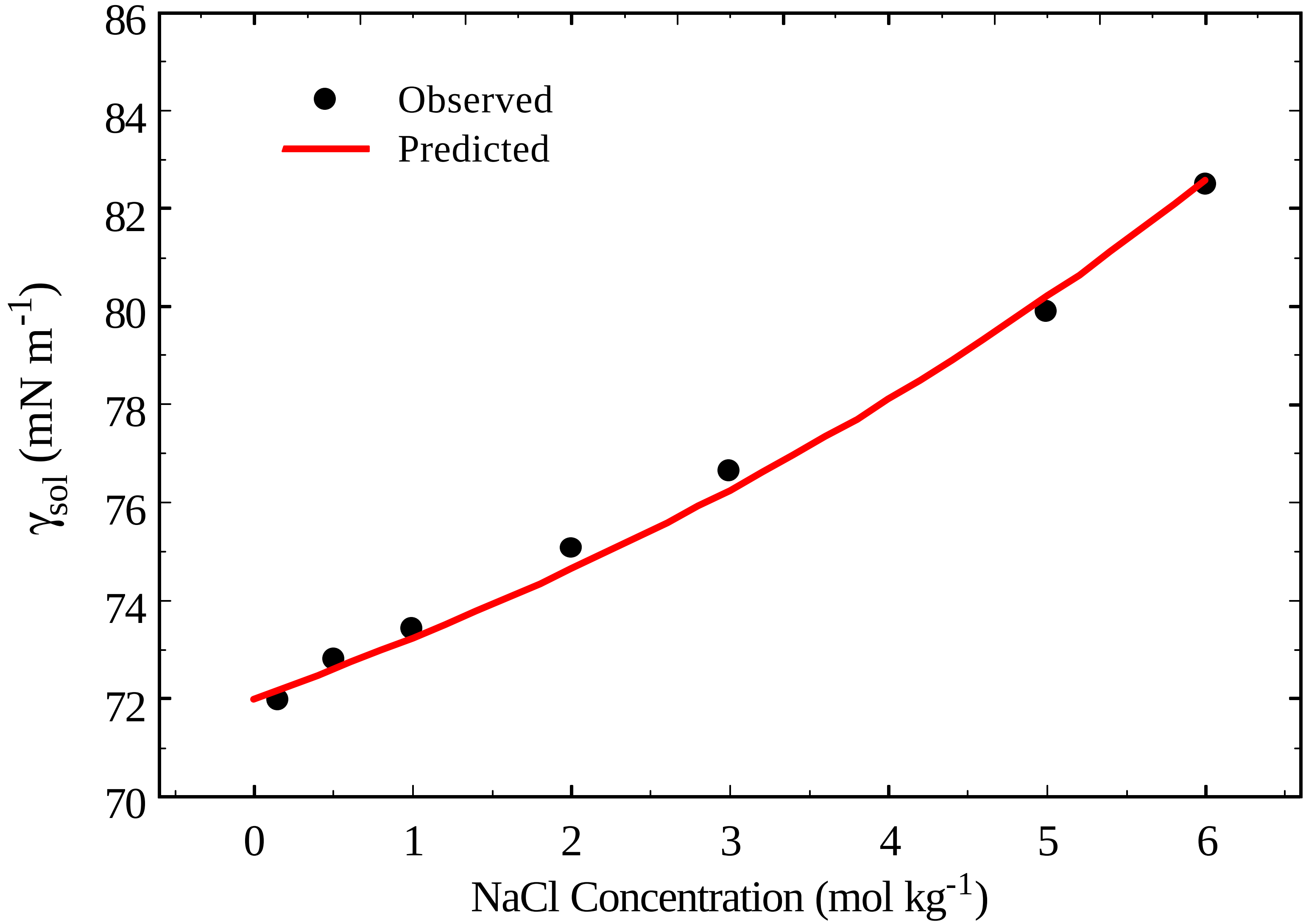
$$g_{so\ln} = g_w + \frac{RT}{A_w M_w}\left(f^B \sum v_i m_i^B - f^S \sum g_i v_i m_i^B\right)$$

Osmotic Coefficient, Pitzer (1973):

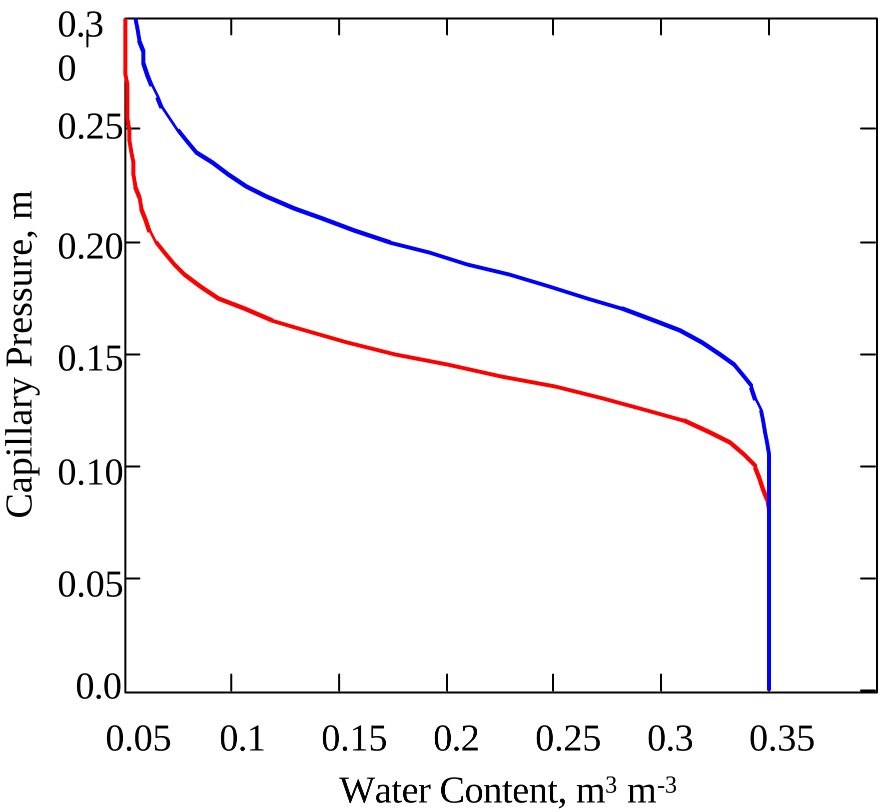
$$f = 1 - \left|z_M z_X\right| f^f + m(2v_M v_X / v)B_{MX}^f \ldots$$

$$+ m^2[2(v_M v_X)^{3/2} / v]C_{MX}^f$$

Observed vs Predicted γ_{sol} for NaCl

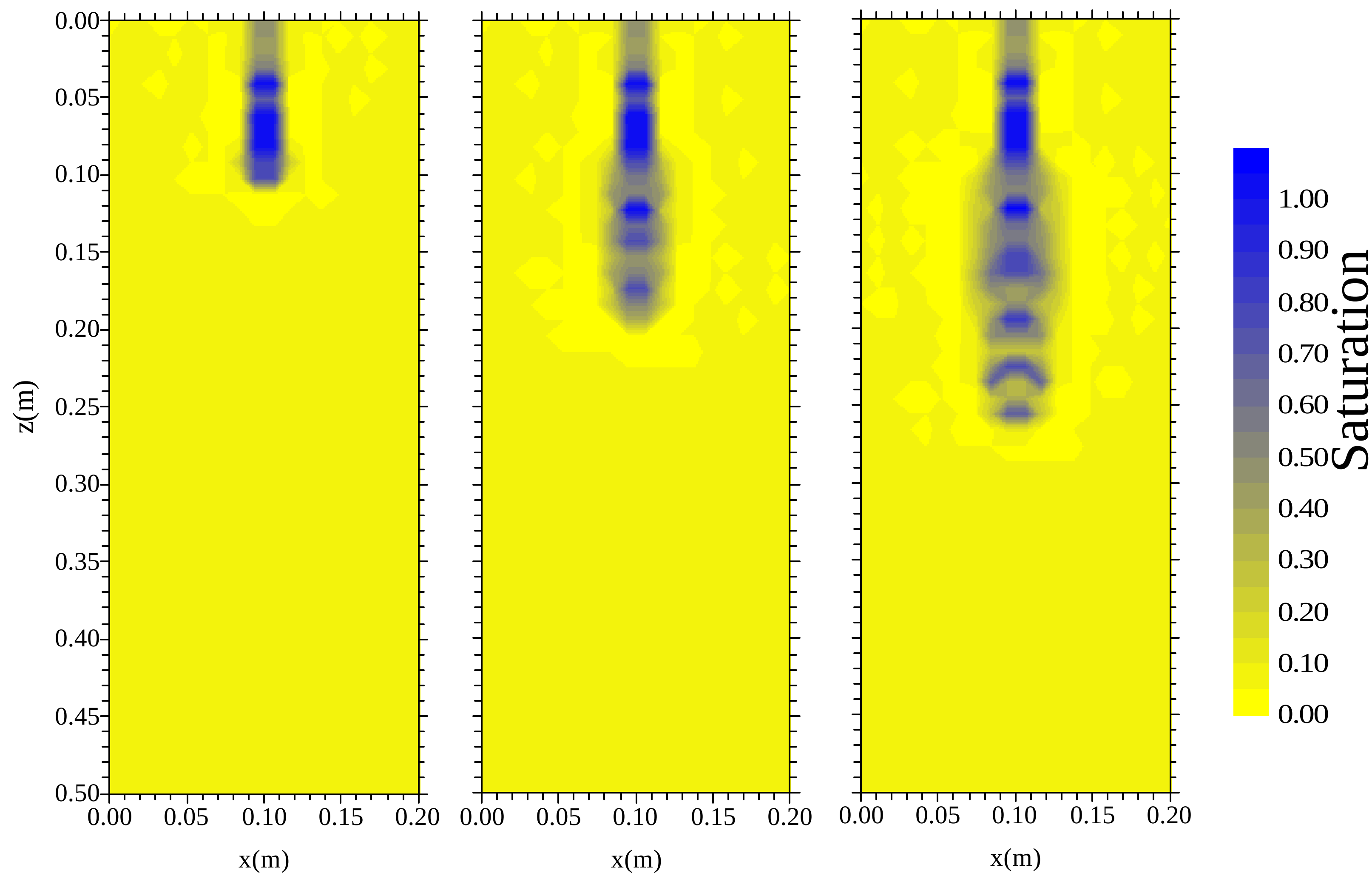


Effect of γ_{sol} on $\psi(\theta)$

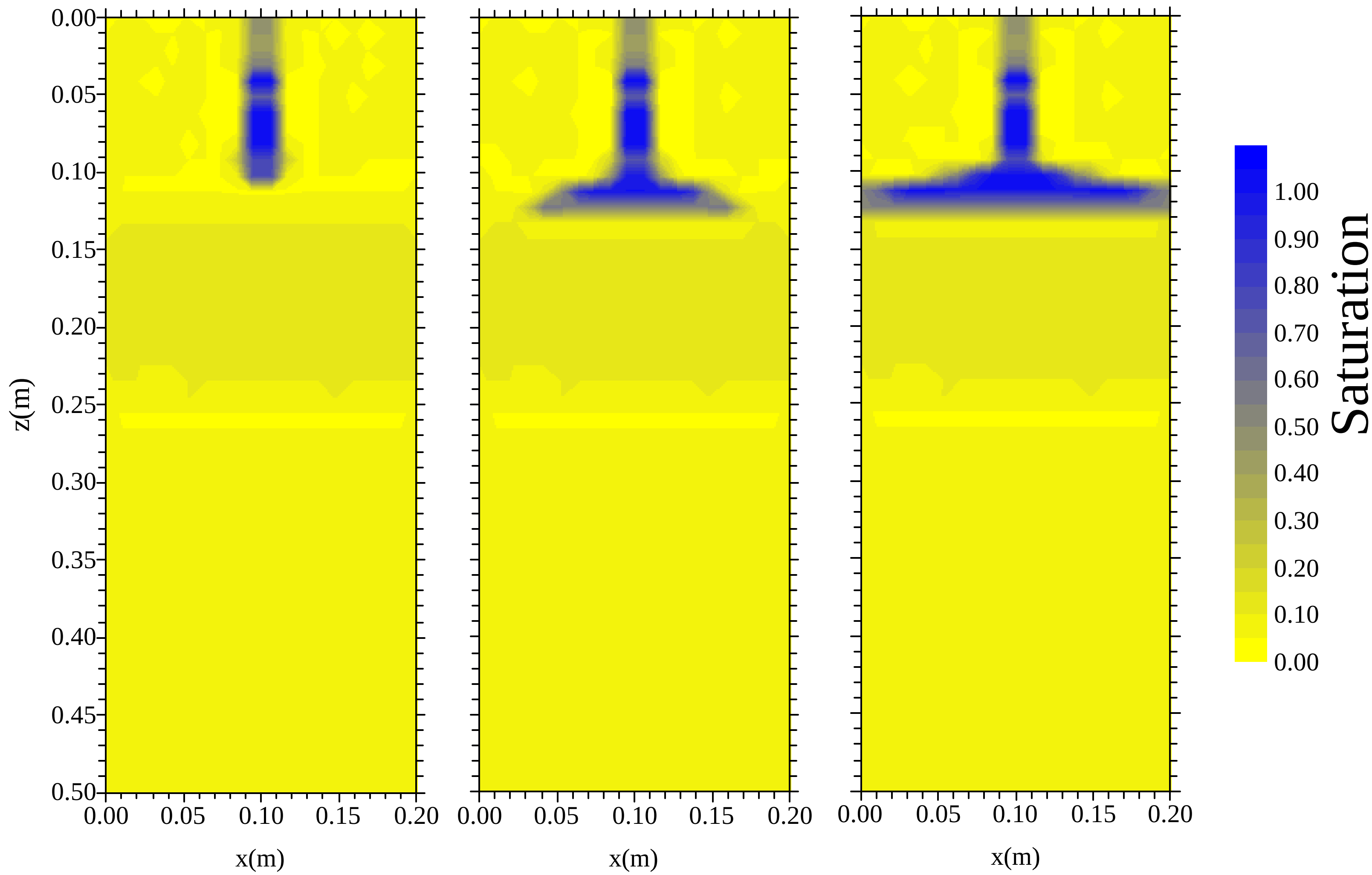


- Linear scaling in $\psi(\theta)$ cannot reproduce lab observations
- Contact angle approach in which $\alpha_a > \alpha_r$ producing desired results
  - Differences not constant over entire θ but greatest at low θ
  - Little effect on main drainage branch
  - Effect appear to be greatest on main imbibition branch

STOMP Simulations-Homogenous Column



STOMP Simulations- Layered Column



Vapor Flux Contribution

$$J_v = - D t q_a \frac{\partial r_v}{\partial T} \nabla T - D t q_a \frac{\partial r_v}{\partial y_m} \nabla y_m - D t q_a \frac{\partial r_v}{\partial y_o} \nabla y_o$$

J_v= mass flow rate of vapor

D = molecular diffusion coefficient of water vapor in air

τ = tortuosity of air-filled pore space

ρ_v = water vapor density

T = Temperature

ψ_m = matric pressure head

ψ_o = solution osmotic pressure head

Status

- Laboratory Studies
  - Homogeneous Miller Sands √
  - Heterogeneous Miller Sands- in progress
  - Hanford sediments- in progress
  - Effect of hypersaline fluids on $\psi(\theta)$- in progress
- Incorporate effects of electrolytes on surface tension √
- Implement adaptive gridding √
- Effect of Organics √
- Vapor flux due to osmotic potential gradient- in progress

Summary

- Traditional vadose zone theory inadequate for explaining laboratory and field observations
- research being conducted to determine effects of fluid properties on vadose zone transport
- Laboratory observations suggest impact of surface tension and density; theory incorporated into STOMP
- Product- a tool that DOE can use for prediction and evaluation of different SST retrieval/remediation strategies