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

Project ID: 65410

Dr. Anderson L. Ward, Pacific Northwest National Laboratory

Dr. Glendon W. Gee, Pacific Northwest National Laboratory
Dr. Scott W. Tyler, Desert Research Institute
Dr. John S. Selker, Oregon State University

Research Objective

The primary objective of this project is to develop a basic understanding of the fate and transport of caustic radioactive brines through the vadose zone. Research is focused primarily on migration of high-level waste leaked from single-shell tanks under environmental conditions, and over temporal and spatial scales relevant to the Hanford Site. Understanding the fate and transport of these wastes through the vadose zone is critical to the development of a framework for evaluating different waste retrieval/ remediation strategies and the associated health risks. The hypothesis underlying this project is that elevated surface tension of the leaked tank wastes will strongly inhibit lateral contaminant spreading, giving rise to narrow fingers of infiltration through the vadose zone. The extent and persistence of these fingers will be enhanced by the water migrating into the saline zone in response to the osmotic potential gradient. To validate this hypothesis, this project combines a series of laboratory, field, and numerical experiments with the following specific objectives:

1. investigate the effect of elevated surface tension of highly saline fluids on wetting front instability, finger formation, and contaminant mobility
2. investigate the conditions under which osmotically driven vapor flux is operative and quantify its impact on plume transport
3. develop and incorporate a theory describing these processes into an existing DOE-developed, numerical simulator to allow prediction of contaminant migration at realistic spatial and temporal scales.

Previous attempts to predict field-scale contaminant transport through the vadose zone have often neglected driving forces and mechanisms that can lead to non-uniform flow. Nonuniform flow has been shown capable of producing erratic transport patterns and can cause contaminants to bypass much of the unsaturated soil matrix. These processes not only lead to fast transport of contaminants to underlying groundwater, but the resulting lack of interchange between the main soil matrix and the fast pathways reduces the potential for natural attenuation.

Two mechanisms that could strongly influence flow non-uniformity and speed the movement of tank wastes in the vadose zone are 1) wetting front instability, due to the high interfacial energy of the saline waste and 2) osmotically driven vapor fluxes, due to the high ionic strength of the waste. Although wetting front instability has been studied extensively in the laboratory, little is known about the formation and persistence of gravity-driven fingers under hydro-geological conditions characteristic of tank farms. The contribution of gravity-driven unstable flow to vadose zone transport could also be enhanced by the migration of water vapor from the surrounding soil into the saline plume. Studies of osmotically driven vapor flux, which have been mostly limited to agricultural fertilizers, suggest that this mechanism can significantly increase the resident water content of the saline soil zone. This mechanism, when coupled with formation of gravity-driven fingers, has the potential to significantly increase contaminant migration rates through the vadose zone. Our strategy is to combine innovative laboratory, field, and numerical experiments to identify the conditions under which these mechanisms are active and to quantify the impact of vadose zone transport processes. The product will be a tool that DOE can use to perform more realistic analyses to predict fate and transport of vadose zone contaminants, evaluate different tank waste retrieval strategies and their impact on the vadose zone, and assess the associated health risks.

Research Progress and Implications

This project combines a theoretical, experimental, and numerical approach to developing an understanding of the driving forces and complex mechanisms affecting the migration of caustic, radioactive brines through the vadose zone. The project is divided into three main components: 1) controlled laboratory experiments, 2) controlled field experiments in the 200 Area of the Hanford Site, and 3) numerical modeling studies. Controlled laboratory experiments are being conducted at the Desert Research Institute, Pacific Northwest National Laboratory (PNNL), and Oregon State University (OSU). The numerical component is being performed at PNNL, and the controlled field experiments are being conducted at the Hanford Site. Although the project has been in operation for only six months, progress has been made in each of these areas and is summarized in the following sections.

Controlled Laboratory Experiments

Traditional approaches to interpreting and predicting contaminant transport distributions generally assume ideal properties for water and the water-solid-air interfaces. However, the solute concentrations found in Hanford tank wastes may invalidate this simplification. Density- and osmotically driven flows studied in this project must be coupled with measured fluid and porous media properties. To accommodate this data need, a detailed laboratory analysis of fluid-soil interactions is being conducted to determine the effect of concentration of salt in the imbibing solution, surface tension of imbibing and resident fluids, osmotic potential of imbibing fluid, soil texture, and antecedent moisture content.

Laboratory Analysis of Fluid Transport Properties

To date, work has focused on the design of fluids to mimic the ionic strengths and surface tensions of the Hanford tank fluids. These fluids are to be used in the field experiment phase of the research and will be used in the laboratory on a smaller scale. A review of the literature on these physical properties and the properties of the Hanford tank fluids has been completed to choose appropriate field and laboratory fluids for testing. Increases in surface tension above that of pure water are easily obtained using common salts such as $NaCl$, $NaNO_3$, and $MgCl_2$. In each of these cases, surface tension may be extended to 10–30% over pure water. In addition, at concentrations required for surface tension augmentation, the fluid ionic strengths and water activity are sufficient to induce steep osmotic gradients between the fluid and vapor phases present in soil water.

To achieve much higher surface tensions than those above, it was determined that adding $NaOH$ would be most appropriate. While such compounds are present in many of the Hanford tank fluids, their chemical and safety attributes are less than desirable. This is especially true for the field setting. The strong base developed with an $NaOH$ solution may be highly reactive in the soils found at Hanford; in particular, reaction of basalt-derived sediments and the fluid phase will result in the release of large volumes of silica. As fluids infiltrate deeper into the profile, the solution pH will decline due to these reactions and silica is likely to then precipitate into the pore spaces, perhaps as amorphous silica. The resulting changes in soil porosity and permeability due to silica translocation could be extreme. Such reactions would significantly confound the objectives of both the field and laboratory analysis of the processes called out in our objectives.

We have concluded that for the field experimental portion, a concentrated $NaNO_3$ solution will be used as the permeant ($\sigma = 87.05$ at 47% by weight). The field experiments will mimic the infiltration of high surface tension, low water activity solutions into typical Hanford sediments with relatively pure water forming the antecedent moisture. Significant degradation reactions are unlikely to occur in the subsurface at the concentrations to be used. In addition, microbial uptake of the NO_3 is unlikely, again due to the high ionic strength of the solution.

In the laboratory, we have developed two procedures. We will conduct water retention and imbibition experiments using Hanford sediments and $NaNO_3$ solutions to confirm behavior in the field. These experiments will look at the effects of surface tension and high ionic strength on water retention and conductivity. To assess the effects of surface tension only on wetting front instability, we have developed an alternative to the concepts of significantly raising the surface tension of the imbibing fluid. We will produce the same net effect, i.e., imbibition of high surface tension fluid into low surface tension fluid, by first wetting the soil samples with a solution of water and 1-butanol, which can dramatically lower the surface tension at very low concentrations (Smith and Gillham 1999). Pure water will then be used as the imbibing fluid. In this manner, we can study the effects of high surface-tension imbibition (pure water) into low surface-tension antecedent moisture (1-butanol). The processes will be identical to those found with Hanford fluids but will be free of geochemical and osmotic effects. Soil core samplers have been prepared for the field to obtain intact core samples at the field site for laboratory analysis. Construction of a surface tension measurement system (Sugden 1922, Maximum Bubble Pressure Method) is under way.

Flow and Transport in Miller Similar Sands

Work at OSU's Flow Visualization Laboratory involves the use of light transmission chambers and digital camera systems to study the migration of dense fluids through well-characterized Miller similar porous sands. This technique provides a powerful system for exploring the effects of the large number of variables that can influence transport behavior.

The project required a post-doctoral fellow to conduct the laboratory and field experiments. Following immediately upon receipt of the grant, advertisements were written for a post-doc with a closing date of December 31, 1998. A short list was developed on January 31, 1999, and full applications and references were received in February. Dr. Noam Weisbrod was selected in March and will begin work on the project on June 1, 1999.

In the interim, tests were conducted with Hanford sediments on light transmission systems. The Hanford sands, being basaltic in nature, are black and have proven not to be amenable to the conventional light transmission method. Over the past months, a variety of techniques have been tried for enhancing the visualization system. It is now apparent that the nature of the Hanford sediments may prove to be a limitation in the use of this technology. This problem will be overcome using blended quartz sands as surrogates to the site sands. These sands have already been painstakingly characterized for hydraulic and chemical properties at OSU. The comparison of flow behavior in the various textural blends will allow discrimination of the controlling flow processes through hydrodynamic scaling and will allow for ready interpretation of results.

Flow and Transport in Hanford Sediments

The main purpose of this component of the project is to obtain a data set from real Hanford soils to complement those obtained from the Miller similar sands. It is unclear to what extent wetted path geometry is influenced by minor soil morphological heterogeneity and how the combined effects of osmotically driven vapor flux will affect transport in real systems. Thus, this task will also seek to measure and compare the effects of the salt concentration, osmotic potential, and surface tension of the infiltrating fluid.

Early tests at the OSU Flow Visualization Laboratory show that the opacity of the Hanford sediments disqualifies the use of light-based flow visualization techniques. Furthermore, this technique is not applicable to the coarser gravel-dominated series of the Hanford Formation (discussed below). Over the past few months, a series of experiments has been conducted with the goal of adapting existing techniques for monitoring controlled flow and transport in these soils at scales relevant to the understanding of finger formation and persistence.

One technique that shows promise is x-ray computed tomography (XRCT), on which testing was initiated in early 1999 at PNNL. In the tests, four columns (15 cm long x 3.8 cm) were packed with soil. One-half of the columns were packed at air-dry moisture content while the other half was packed 10% by volume of water. Each column received a 5-mL application of saturated NaI solution (1.4 g L^{-1}), to simulate a dense saline fluid, and was monitored over time by XRCT to evaluate its efficacy for monitoring flow in opaque soils and identifying the conditions under which gravity-driven flow instability occurred.

Figure 1 shows an example of a transverse 2-dimensional image from the opaque basaltic sand. The volume displayed is representative of the attenuation values in gray scale. These attenuation values, corresponding to the density within the volume, display rocks or dense material as white and air as black. Not only is the pore space, consisting of various channels and voids, clearly seen in these images, but the plume of NaI is also quite clear. Finger formation is clearly seen in Figure 1a, while the solute front is somewhat more diffused in the wet soil (Figure 1b). Finger formation and persistence in the wetter soil may have been overcome by the heterogeneities caused by packing.

Figure 2 shows cross-sectional views of the same two columns at about 4 cm from the surface. In Figure 2a, a compact, well-defined outline of the NaI plume is clearly seen as a denser (white) zone. In the wetter soil, the solute front spread laterally along the stratification, essentially wetting most of the pore space, hence the more widely distributed, high-density zones (Figure 2b).

These images clearly show that the XRCT technique has potential for monitoring finger formation and persistence in opaque soils. These micro-lysimeter studies are being used to provide information on the initiation and persistence of fingers over a range of conditions. A major advantage of the XRCT system is that the transverse images can be assembled and rendered as 3-dimensional images from which the multidimensional features of fingers can be studied. Thus far, studies of fingers have been limited to thin, 2-dimensional columns that can be monitored using light-based transmission techniques. The results of these experiments will be used as a basis to design an intermediate scale (2 m x 1 m flow cell) laboratory experiment in which fingering phenomena will be monitored, including a fully automated dual-energy gamma radiation system and time-domain reflectometry (TDR) systems.

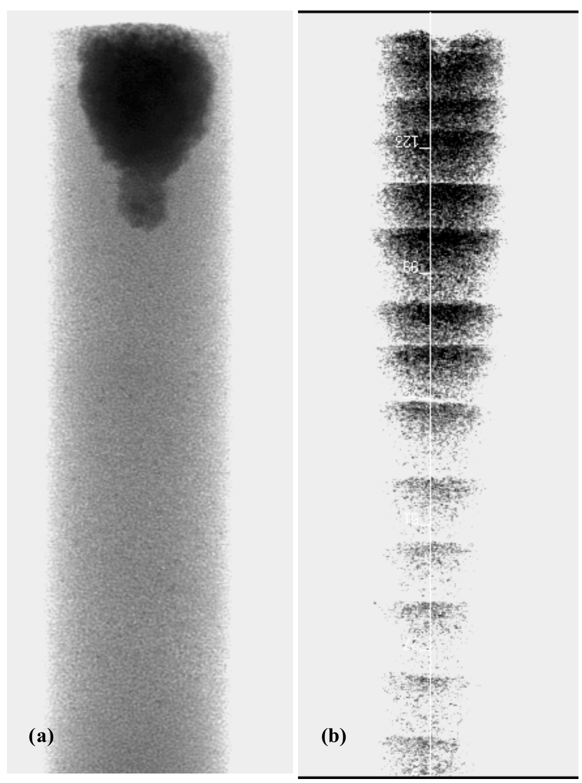

Figure 1. Sagittal CT Image Displaying Pore Space and Distribution of a Dense NaI Plume After 24 Hours of Redistribution (a) initially air-dry soil, (b) soil wet to a water content of 0.10 m^3 m^{-3}

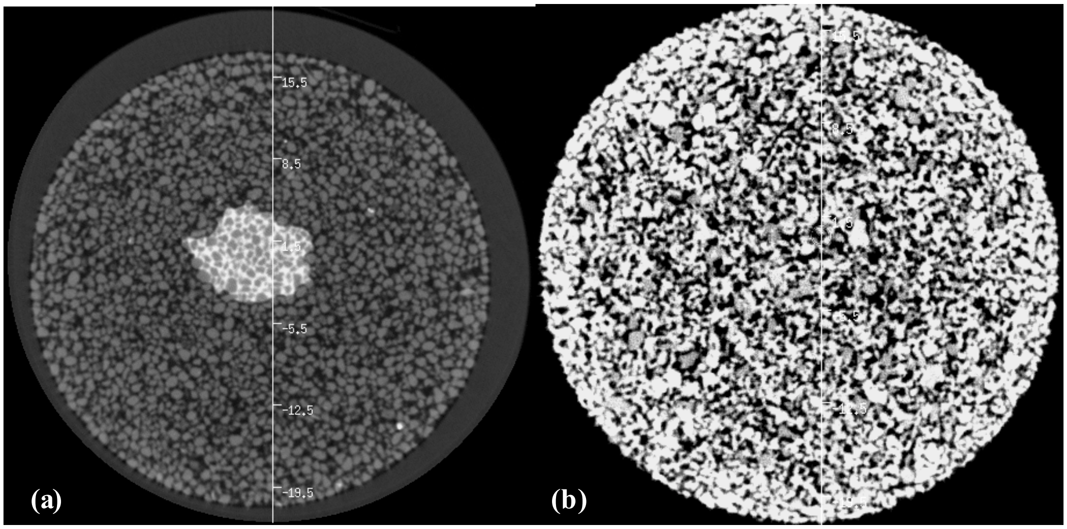

Figure 2. Transverse XRCT Image Displaying Pore Space and Distribution of a Dense NaI Plume After 24 Hours of Redistribution (a) the compact finger is maintained in the initially air-dry soil, (b) finger is dispersed due to layering in soil initially wet to a water content of 0.10 $m^3\ m^{-3}$

Field Experiments

The field experiments are intended to explore saline plume transport at two time scales, a reflection of the different time scales at which initial imbibition and vapor migration processes proceed. The objectives of the field component of this project are to

1. identify the degree to which field conditions mimic the flow processes observed in the laboratory
2. provide a validation data set to establish the degree to which our conceptual models, embodied in a numerical simulator, explain the observed field behavior.

We toured the Hanford Site in the fall of 1998 to investigate possible field sites. Based on the findings of the tour and subsequent discussions, planning and preparation for field activities were started. Two sites were selected to represent the extreme in porous media properties representative of the Hanford tank farms and the paperwork started to obtain the necessary permits. The permits were obtained in May 1999, and instrumentation has commenced.

Most of the tanks at Hanford were placed in the upper Hanford formation, and much of the site outside of the tank farms is covered with a 1–2-m layer of Holocene eolian deposits. Therefore, before the field experiments are conducted, the top 1–2-m of deposits had to be removed to expose the Hanford formation. The first site is located near the 200 West Area of the Hanford Site and is representative of the gravel-dominated Hanford formation (Figure 3a), the unit in which most of the waste resides.

The second site is located in the 200 East Area and is representative of the sand-dominated Hanford formation (Figure 3b). Both sites have been excavated in preparation for instrumentation and conducts the characterization and long-term plume migration studies.

The field experiment requires a 48 probe, multiplexed five-level TDR system. The entire system is designed to be fully automated with remote download capacity via cellular phone. The system will document the movement of water and salt along the flow paths of six separate plumes on a twice-daily basis. All system components have been acquired and the system is largely assembled (Figure 4a). The system is to be installed in the field in early June 1999. Each probe has been calibrated before installation (Figure 4b shows the method outside of calibration fixture).

Numerical Studies

Numerical modeling of gravity-driven flow instabilities using a continuum approach has only recently been successful through adjustment of the upstream weighting coefficient of permeability in a finite

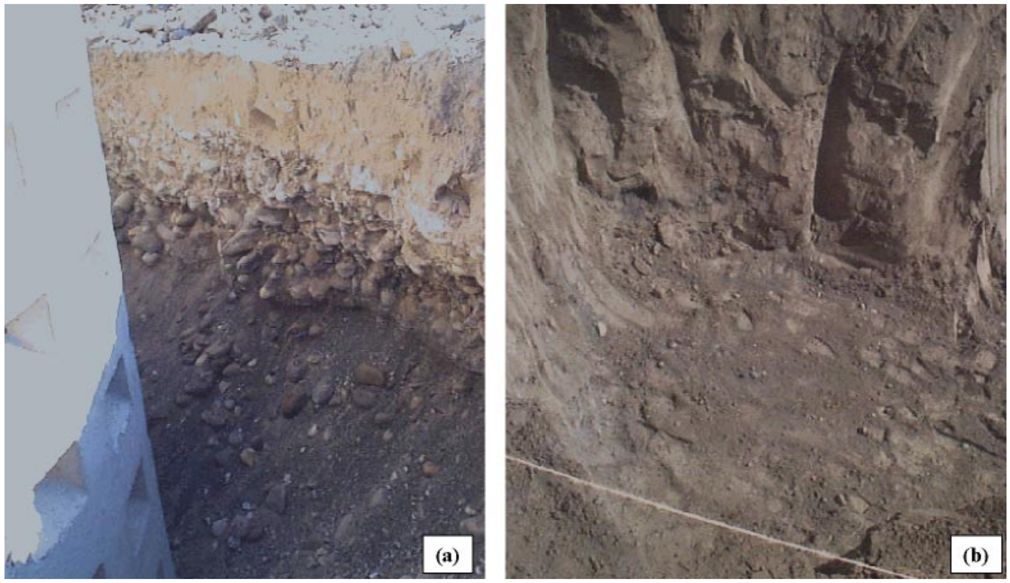

Figure 3. Photograph of (a) Typical Soil Profile in the Upper Hanford Formation at the 200 West Area Site and (b) the Sand-Dominated Site in the 200 East Area

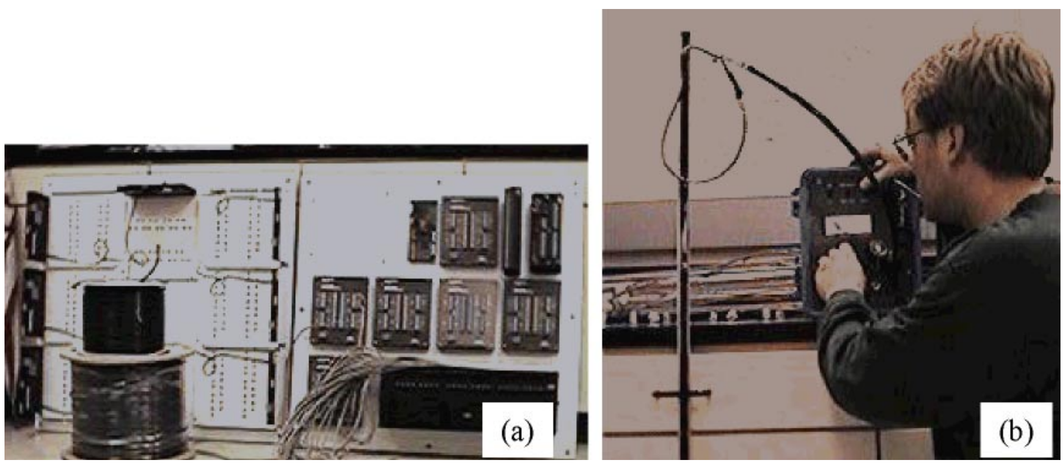

Figure 4. Photograph of Moisture Monitoring System for the Field Experiment. Each probe has been calibrated before installation (a) assembly of system, (b) system outside of calibration fixture

element model (Nieber 1996). It has also been suggested that the mechanism for the initial formation of flow instabilities and their persistence is best described in terms of the water retention _(_) and hydraulic conductivity K(_) relations for the given soil. Although the nature of the problem means that the solutions to the flow and transport equations may possess highly localized properties in both space and time, application of these techniques has been limited to relatively small domains (≤2 m x 2 m) with uniform 2-dimensional grids.

It has become increasingly clear that multigrid methods represent the most efficient approach for iteratively solving the large system of algebraic equations that arise from large-scale numerical simulations modeled by partial differential equations (Bramble et al. 1996). To scale the highly localized behavior of fine-scale fingering generated by heterogeneous porous media or flow instabilities up to computational and field scales, work has started on techniques for obtaining effective parameters for coarse-grid models that are consistent with fine-grid simulations. Over the past months, work has been ongoing to modify the multidimensional Subsurface Transport Over Multiple Phases (STOMP) computer software program to incorporate an adaptive mesh refinement. The approach is to use adaptive mesh refinement to cluster grid points in regions where they are most needed, e.g., around a developing flow instability or other regions where the solution has steep gradients. This strategy is being developed for both temporal and spatial refinements on Cartesian grids and will ultimately take advantage of the vectorization of the STOMP simulator.

The time-stepping scheme is an integral part of the adaptive gridding solution technique; the first step was to implement a second-order, two-step implicit backward differentiation formula (BDF) method for solving the differential-algebraic equations into STOMP. A series of tests to compare the default Euler scheme in STOMP with the new BDF scheme has been successful. In a test involving a one-dimensional infiltration problem, the Euler scheme executed the problem in 65 time steps with one convergence failure, while the BDF scheme executed the same problem in 49 time steps with no convergence failures and produced essentially the same results. Schemes to allow description of a variable upstream weighting parameter and definition of the hysteretic water retention and hydraulic conductivity models, critical to modeling flow instability, have also been implemented.

Planned Activities

During the next year, work will continue at the Desert Research Institute to obtain water retention and unsaturated hydraulic conductivity measurements on samples recovered from the field beginning in the summer of 1999. Pure water, $NaNO_3$, and 1-butanol solutions will be used. Following completion of this characterization, imbibition experiments will be conducted using both pure water imbibing into pure water (as a control) and 1-butanol imbibing into pure water. It is anticipated that these experiments will begin at the end of summer 1999 and will be completed by winter 1999-2000.

Work at the OSU Flow Visualization Laboratory will continue to study the effects of heterogeneity of finger formation and persistence using simple horizontally layered systems of pairs of Miller similar sands. These experiments will attempt to determine the ability of the surface tension effects to overcome the attenuating effect on fingered flow observed in naturally occurring layered systems. These tests are expected to start in July 1999.

In the PNNL-controlled laboratory studies, work will proceed on calibration of the XRCT system to permit conversation of attenuation values into soil water content/solute concentration. We will further investigate the conditions that lead to finger formation and the characteristics that influence persistence during wetting and drying cycles. The medium-scale flow experiment will begin in late summer 1999.

Starting in June 1999, and before the field-scale experiments at the two sites begin, detailed hydraulic and stratigraphic analysis will be performed. Based on these analyses, experimental design parameters (e.g., water application rates, sensor location, sampling frequency) will be developed using predictive models. Site instrumentation should be completed by the end of July 1999, after which the long-term plume migration studies should begin. Following the long-term plume migration experiments, the plots will be excavated to further investigate the tracer transport as well as to characterize the hydraulic properties directly beneath the field site.

Theoretical analysis and model simulations of pure water entering initially dry soils show that finger formation results from hysteresis in the water retention function and that the characteristics of the finger depends on the shape of the main imbibition and drainage branches. During the next year, work will continue to incorporate the effects of surface tension inferred from the laboratory water retention and unsaturated hydraulic conductivity measurements using tank waste simulants. Work will also continue toward the implementation of the adaptive mesh refinement strategy. Numerical experiments will continue with the goal of elucidating the regime that leads to the onset of fingering and to determine how finger characteristics are influenced by vapor flux in response to osmotic potential gradients.

Information Access

References

Bramble JH, RE Ewing, JE Pasciak, and J Shen. 1996. "The analysis of multigrid algorithms for cell centered finite difference methods." *Advances in Computational Mathematics* 5(1):15-19.

Nieber JL. 1996. "Modeling finger development and persistence in initially dry porous media." *Geoderma* 70:207-229.

Smith J, and RW Gillham. 1999. "Effects of solute concentration-dependent surface tension on unsaturated flow: Laboratory sand column experiments." *Water Resources Research* 35(4):973-982.

Sugden J. 1922. *Journal of the American Chemical Society* 121:858.

Presentations

Selker JS. December 5-10, 1998. "Fingered flow from high salinity sources." Presented at the AGU Fall Meetings, San Francisco.

Ward AL and GW Gee. October 31-November 4 1999. "A Numerical Analysis of Wetting Front Instability Induced by Infiltration of Highly Saline Fluids." Symposium on Preferential Flow, Soil Science Society of America Annual Meeting, Salt Lake City.