Physics of DNAPL Migration and Remediation in the Presence of Heterogeneities
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Lead Principal Investigator:
Dr. Stephen H. Conrad
Sandia National Laboratories - Albuquerque
P.O. Box 5800-1345
Albuquerque, New Mexico 87185
505-844-5267, shconra@sandia.gov

Co-Investigator:
Dr. Robert J. Glass
Sandia National Laboratories - Albuquerque
P.O. Box 5800
Albuquerque, New Mexico 87185
505-844-5606, rjglass@sandia.gov

Research Objective

Spilled solvents have created pervasive groundwater contamination problems across the DOE complex because of their ubiquitous use, their toxicity and persistence in the environment, combined with the difficulty of recovering them from the subsurface. Because organic solvents are more dense than water and immiscible with water, they are commonly referred to as DNAPLs (dense non-aqueous phase liquids). They migrate below the water table downward and laterally under the influence of gravity, capillary, and viscous forces. Variations in media texture that the DNAPLs encounter as they migrate can have a profound influence on the migration path. This interplay between textural heterogeneities and driving forces complicates the migration of the DNAPLs and therefore it is not straightforward to predict the locations in the aquifer at which the spilled DNAPLs may ultimately reside. Uncertainties in the region of solvent contamination translate into higher remediation costs as the remedial system must be designed in light of these uncertainties. In an effort to clean up spilled DNAPLs, several remediation approaches are currently under development. Chemically enhanced solubilization, alcohol displacement, in situ oxidation, and air sparging are among the most promising. Many of these techniques have already undergone preliminary field demonstrations. However, results from such field demonstrations cannot be extrapolated to predict remedial performance under the wide range of field conditions to be encountered at spill sites across the DOE complex. Indeed, these techniques have not yet had the opportunity to be sufficiently tested and quantitatively compared in well-controlled laboratory experiments under heterogeneous conditions indicative of what can be expected in the field. In addition, the numerical simulation techniques used to predict DNAPL migration and remediation treatments have yet to be adequately verified through comparison against laboratory experiments conducted in heterogeneous media.

Our research effort has been designed as broad and crosscutting. The goal of our research is to develop a fundamental quantitative understanding of the role of physical heterogeneities on DNAPL migration and remediation in aquifers. Such understanding is critical to cost effectively identify the location of the subsurface zone of contamination and design remediation schemes focused on removing the source of the contamination, the DNAPL itself. There are two major aspects to the DNAPL problem: finding them (migration) and cleaning them up (remediation). We have been working on both aspects. By designing lab experiments within heterogeneous porous media analogous to field conditions, we have been able to directly observe DNAPL initial migration and subsequent interactions between injected remedial agents and the DNAPL. In these experiments we have identified critical mechanisms having important implications affecting both the initial migration and the successfulness of remedial processes.

For migration, we have found the influence of heterogeneities to yield high DNAPL saturation "pools" of a wide range of sizes, corresponding to the effects imposed by textural variations between geologic facies. These pools are interconnected by fingers where very little DNAPL resides. When viscous forces are low, we discovered the DNAPL structure to pulsate at both the pore scale within fingers and at the unit scale within large pools due to a capillary-gravity pulsation mechanism. We have built pore scale invasion mechanisms into a fundamentally new modeling approach, a form of Modified Invasion Percolation or MIP model. Our simulations yield results that closely track the migration behavior seen in our experiments. In the continuation of this work we will further explore DNAPL migration in the presence of both macro- and micro-layered heterogeneities. We will closely consider the effects imposed on migration by capillary-gravity pulsation and when significant viscous forces are present as each of these has been found experimentally to yield multiple DNAPL migration pathways. For remediation, we performed both micro-model experiments to elucidate surfactant enhanced mobilization and dissolution mechanisms as well as bench-scale remediation demonstration experiments for two different surfactants and an in situ oxidizer (potassium permanganate). From our results *we emphasize caution and advocate a restrained approach to site remediation at this time.* In all cases considered so far, the initial configuration of the DNAPL in pools and fingers dramatically influenced the efficacy of the remediation method. In the continuation of this work we will conduct further demonstrations for remediation techniques. However, we intend to focus our effort using the insights gained and models developed from our work on migration to better understanding the conditions in which inadvertent re-mobilization can occur during remediation efforts. We will also focus on improving our representativeness by working with so-called "dirty DNAPLs" -- less ideal, more realistic multi-component organic liquids.

Research Progress

This report summarizes work after 8 months of a 36-month project. Since the beginning of the fiscal year we have accomplished quite a lot. We have handled revisions associated with two papers to Water Resources Research – one on migration and one on percolation modeling. Both papers have now been published. We have submitted a paper describing our remediation experiments to the Journal of Contaminant Hydrology. This is a big manuscript that covers a lot of ground (2 surfactant experiments, a permanganate experiment, and an extensive analysis discussing the onset of DNAPL mobilization). We have also completed extensive work to update, improve, and reconfigure our work environment. We have reorganized and backed up the extremely large data sets from our experiments and simulation output from our modeling efforts from over the past 4 years. We replaced the computer we will use to do the bulk of our modeling. We are in the process of reworking percolation code to run on a PC-based architecture. In our experimental apparatus, we replaced experimental control computer and rewired the entire setup. In May, we disassembled the experimental chamber and we are in the process of refurbishing it in preparation for conducting a series of collaborative experiments.

We gave an invited presentation and a contributed presentation at GSA in November and we gave another invited presentation at Spring AGU in May. We also participated in the EMSP conference at Hanford in November.

Planned Activities

We continue to work collaboratively with researchers developing promising DNAPL remediation technologies. We have established a collaborative relationship with researchers at Laval University in Quebec working on surfactant remediation techniques. We plan to conduct joint experiments with them at our facilities beginning this summer.

We are in the process of reworking our percolation code to run on a PC-based architecture. This will allow wide dissemination of the code.

Information Access

Conrad, S.H., R.J. Glass, and W.J. Peplinski. 2001. Bench-scale visualization experiments of DNAPL remediation processes in analog heterogeneous aquifers: surfactant floods and in situ oxidation using permanganate. Journal of Contaminant Hydrology (in review). Manuscript available at The Enviro-Science e-Print Service web site at http://esn.osti.gov/enviroscience.html

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