DNAPL migration through interbedded clay-sand sequences

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Abstract This paper examines the migration of DNAPL and the associated aqueous phase plume through clay-sand sequences. Numerical simulations are carried out examining contaminant distribution and migration rates in single fractures, simple layered clay-sand sequences, and spatially-correlated random fields where all low permeability lenses are assigned fractures. The results of this work illustrate that matrix diffusion does not significantly affect the rate of DNAPL migration in fractured clay, and that fracture aperture is the most influential parameter governing the rate of migration through fractures. In all simulations, the arrival of the aqueous phase plume at depth was approximately coincident with the arrival of DNAPL. Time periods for redistribution to residual were in excess of 15 years in the spatially correlated random field. Factors influencing the vertical and horizontal degree of spreading include the variance of the permeability field, the mean permeability, and the percentage of fractures assigned. In general, the role of fractures was more pronounced in the simple layered systems as compared to the spatially correlated random fields.

Key words clay; contaminant migration; DNAPL; fractures; remediation; sand

INTRODUCTION

Groundwater contamination derived from dense, non-aqueous phase liquids (DNAPLs) is a common occurrence throughout the industrialized areas of the world. DNAPLs of environmental concern include PCB oils, chlorinated solvents, creosote, coal tar, and some crude oils. It is now recognized that the volume of DNAPL released to the subsurface at many sites was sufficient to penetrate bedrock and fractured clay aquitards underlying surficial overburden deposits. Some of these geological environments are not simple layered systems, but comprise interbedded sand-clay sequences where DNAPL migration can take place concomitantly through porous and fractured porous media. To date, the evolution of DNAPL saturations and associated aqueous phase plumes in such complex geological environments has not received large amounts of attention. This stems primarily from the fact that controlled experiments at the field scale are prohibitive, and numerical multi-phase flow simulators have to date focused largely on porous media issues.

The objective of this paper is to examine the migration of DNAPL in fractured clay-sand sequences using numerical simulation. A three-step approach is taken, beginning with DNAPL migration in a single fracture, followed by migration in idealized layered clay-sand sequences, and finally DNAPL migration in spatially-correlated random fields consisting of both fractured and unfractured media. All simulations were conducted using the Queen’s University Multi-Phase Flow Simulator.
(QUMPFS) as described by Reynolds & Kueper (2001a). This model is a three-dimensional, multiphase compositional simulator that allows for explicit simulation of matrix-fracture interactions, non-equilibrium phase partitioning, and hysteresis in the relative permeability and capillary pressure–saturation relationships. The governing equations are discretized through the finite volume method using a lumped mass time derivative.

DNAPL MIGRATION IN A SINGLE FRACTURE

Past studies examining DNAPL migration in a single fracture include Kueper & McWhorter (1991) in which the rate of DNAPL migration through a single clay aquitard was examined in the absence of dissolution and matrix diffusion. Ross & Lu (1999) presented an analytical solution to investigate the effects of mass transfer to the matrix on the advancement of DNAPL down a vertical fracture. Although allowing for dynamic flow behaviour, the study did not account for relative permeability or capillary pressure gradients. Slough et al. (1999) simulated DNAPL migration through a single fracture subject to matrix diffusion, and found that increasing the matrix porosity dramatically increased the time required for DNAPL to penetrate a 10 m long fracture. This finding was influenced, however, by the use of a constant non-wetting flux boundary condition at the top of the fracture.

The physical system employed here consists of a single fracture dipping at 60°, embedded in a porous clay matrix. The fracture is either 3 m or 10 m long, and has a unit depth into the third dimension. Flow boundary conditions consist of no-flow along all vertical faces, and a constant wetting phase pressure along the top and bottom faces such that the pressure distribution is hydrostatic. DNAPL is introduced into the top of the fracture at a constant capillary pressure of 4295 Pa, equivalent to 0.3 m of pooled trichloroethylene (TCE) situated at the water table (which is located at the top of the domain). Transport boundary conditions were set such that the aqueous phase contaminant was unable to leave the domain except at the fracture outlet. The presented simulations employ a Brooks-Corey representation of the $P_c(S_w)$ and $k_r(S_w)$ relationships for the fracture with $\lambda = 2.0$ and $S_r = 0.1$. The interfacial tension was set to 0.020 N m$^{-1}$, the matrix permeability to $1.0 \times 10^{-17}$ m$^2$, the matrix $f_{oc}$ to 0.01, the matrix porosity to 0.3, the longitudinal dispersivity in the fracture to 0.01 m, and the transverse dispersivity to 0.001 m. The simulations examine the rate of migration of five particular DNAPLs which differ significantly with respect to density, solubility, and sorptive characteristics: chlorobenzene (CB), 1,2-dichloroethylene (1,2-DCE), TCE, tetrachloroethylene (PCE), and 1,2-dibromoethane (EDB). The chemical properties (density, solubility, $K_{oc}$, viscosity, and diffusion coefficient) were taken from Pankow & Cherry (1996).

Figure 1 presents a plot of DNAPL arrival time at the base of the fracture as a function of fracture aperture for all DNAPLs of interest in both the 3 m and 10 m long fractures. The differences in arrival time for a particular aperture are explained primarily by differences in density and viscosity. It is important to note, however, that the change in arrival time with aperture is much more influential than the change in arrival time with DNAPL properties. Additional simulations (not shown) also showed that moderate changes in matrix porosity and matrix $f_{oc}$ had relatively little influence.
on DNAPL migration rate compared to the influence of aperture. These results are in contradiction to those of Slough et al. (1999) where matrix diffusion was shown to significantly influence the rate of DNAPL migration. The discrepancy arises from the different boundary conditions employed. Slough et al. (1999) employed a constant DNAPL flux boundary condition at the top of the fracture such that any mass loss due to matrix diffusion would have a direct impact on slowing the rate of DNAPL migration. The current study uses a constant capillary pressure boundary condition, in which case the flux into the top of the fracture can adjust itself in response to gravity and capillary gradients, and the loss of mass to the matrix through diffusion. The net result is that matrix diffusion does not significantly influence the rate of DNAPL migration in clay fractures.

To examine the persistence of aqueous phase contamination in the system, five simulations were performed in which 5 ml of each of the DNAPLs was allowed to enter a 30 μm, 3 m long vertical fracture (matrix porosity = 0.3, matrix \( f_{oc} = 0.01 \)). After the 5 ml of DNAPL had entered (less than 1 h in each case, volume insufficient to reach bottom of fracture), the DNAPL source to the fracture was removed and a wetting phase gradient of 0.01 was applied to flush clean water through the fracture (0.64 m day\(^{-1}\) groundwater velocity in fracture). The simulations allowed for equilibrium dissolution of DNAPL into the flowing groundwater, as well as matrix diffusion. The lifespan of the DNAPL in the fracture ranged from 15 h (following start of injection) for DCE, to greater than 140 days for PCE. Figure 2 presents a plot of dissolved phase concentration exiting the bottom of the fracture vs time for each of the five DNAPLs. The maximum concentrations typically occur after 1 year because of the attenuating effect of matrix diffusion on plume migration. With respect to persistence, concentrations are still above 5 ppb (typical USA based clean-up goal) for up to 1000 years following the start of injection. The longevity of aqueous phase contamination is the result of reverse diffusion back out of the porous matrix in response to the clean water injection.
Two-dimensional simulations were performed in the domain illustrated in Fig. 3 to examine the rate of both DNAPL and plume migration through layered sand-clay sequences. The domain of interest contains an upper, middle, and lower sand layer separated by fractured clay units. In certain simulations, the upper and middle sand layers were replaced by fractured clay units to produce a "fractured only" domain for comparison to simulations in the layered system. The domain is initially 100% water saturated with the water table located at the top boundary, and constant wetting phase pressure boundary conditions at the sides and bottom. All porous media boundaries were assigned a free-exit condition for both the non-wetting and dissolved phases.

The DNAPL of interest (TCE) was applied along a 3 m wide area in the centre of the top boundary at a constant capillary pressure of 14 317 Pa. In total, 10 m$^3$ of TCE
was released in the finite volume simulations. Certain simulations involved a con­tinuous source of TCE. All fractures were assigned an aperture of 35 μm. The clay matrix was assigned a permeability of $1.0 \times 10^{-17}$ m$^2$ and a porosity of 0.35. The sand layers were assigned a permeability of $1.0 \times 10^{-14}$ m$^2$ and a porosity of 0.25. The DNAPL–water interfacial tension was set to 0.020 N m$^{-1}$. Additional details regarding parameter selection can be found in Reynolds & Kueper (2001a).

Figure 4 presents a plot of DNAPL and dissolved plume arrival time in the lower sand aquifer for six particular simulations. In Fig. 4, a negative gradient corresponds to downward groundwater flow at a gradient of 0.25, while a positive gradient corresponds to upward groundwater flow at a gradient of 0.10. The upper three simulations (LD5, LD3, and LD2) correspond to the domain illustrated in Fig. 3, while the lower three simulations (FD1, FD2, and FD3) correspond to the “fractured only” domain described above.

All simulations show nearly identical arrival times in the lower sand aquifer for the DNAPL and the associated dissolved phase plume, implying that the presence of aqueous phase contamination in the lower sand unit will always be associated with DNAPL presence in that unit (note, however, that the volume of DNAPL released in each of the illustrated six simulations was sufficient to reach the lower sand unit). A second characteristic feature of Fig. 4 is that the DNAPL and aqueous plume arrival times are much greater in the layered sequence domain than in the “fractured only” domain. The presence of sand layers clearly slows that rate of contaminant migration. While DNAPL velocities were found to be very high in any invaded fractures, the DNAPL flux is very low. Despite the high rate of travel through fractures, it takes a relatively longer time for DNAPL to traverse the sand layers because of this flux limitation (i.e. the sand layers are supplied DNAPL from the fractures only, and have a much higher storage capacity for the non-wetting phase). A further observation from Fig. 4 is that downward groundwater flow accelerates the rate of DNAPL migration, while upward groundwater flow slows the rate of DNAPL migration. Further details can be found in Reynolds & Kueper (2001a).

![Fig. 4 DNAPL and dissolved plume arrival times in the lower aquifer.](image-url)
SPATIALLY CORRELATED RANDOM FIELDS

The spatially correlated random field simulations presented here were carried out in a 120 m wide, 40 m high two-dimensional domain with the water table located at the upper boundary. DNAPL (30 m$^3$ of TCE having an interfacial tension of 0.01 N m$^{-1}$) was introduced over a 20 m area in the centre of the top boundary at a constant capillary pressure of 7158 Pa. The random field is characterized by an exponential autocorrelation function having vertical and horizontal correlation lengths of 1.0 m and 20.0 m, respectively. The mean permeability is $1.02 \times 10^{-13}$ m$^2$, and the variance of the log-transformed permeability is 5.5. The high variance results in local permeability values ranging from those characteristic of clay, to those characteristic of medium sand. Vertical fractures were assigned (2 m spacing) to all nodes exhibiting a permeability equal to or less than $1.0 \times 10^{-12}$ m$^2$. This resulted in a total cumulative fracture length of 728.5 m. If fractures were assigned to the entire domain, the total cumulative fracture length would be 2400 m. Further details can be found in Reynolds & Kueper (2001b).

Figure 5 presents a plot of wetting phase saturations at a time of 15 years. The 30 m$^3$ of DNAPL required approximately 2.2 years to enter the domain. A moment analysis indicated that the DNAPL was still migrating downwards at 15 years time, moving through both porous media and fractured lenses. DNAPL velocities were high in fractures, but flow rates were very low. The bulk of the downward DNAPL migration therefore occurred through the porous media lenses. DNAPL migration occurred over and around the fractured lenses while at the same time occurring through fractures. In comparison to the simple layered systems described above, DNAPL migration through the spatially correlated random field was less influenced by the presence of fractures. Examination of the aqueous phase plume formed in the spatially correlated random field showed that its vertical migration is approximately coincident with the vertical migration of DNAPL, similar to what was observed in the simple layered systems.

![Figure 5](image_url)

Fig. 5 DNAPL distribution at $t = 15$ years. White vertical lines depict fractures in low permeability lenses. Highest DNAPL saturations (lowest wetting phase saturations) shown in black.
REFERENCES


