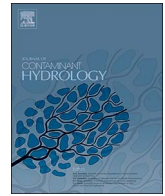




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Field-scale forward and back diffusion through low-permeability zones

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ABSTRACT

Understanding the effects of back diffusion of groundwater contaminants from low-permeability zones to aquifers is critical to making site management decisions related to remedial actions. Here, we combine aquifer and aquitard data to develop recommended site characterization strategies using a three-stage classification of plume life cycle based on the solute origins: aquifer source zone dissolution, source zone dissolution combined with back diffusion from an aquitard, and only back diffusion. We use measured aquitard concentration profile data from three field sites to identify signature shapes that are characteristic of these three stages. We find good fits to the measured data with analytical solutions that include the effects of advection and forward and back diffusion through low-permeability zones, and linearly and exponentially decreasing flux resulting from source dissolution in the aquifer. Aquifer contaminant time series data at monitoring wells from a mature site were well described using analytical solutions representing the combined case of source zone and back diffusion, while data from a site where the source had been isolated were well described solely by back diffusion. The modeling approach presented in this study is designed to enable site managers to implement appropriate remediation technologies at a proper timing for high- and low-permeability zones, considering estimated plume life cycle.

1. Introduction

An understanding of the relationship between dense non-aqueous phase liquid (DNAPL) source zone changes and down-gradient plume response is critical to evaluate human exposure risk and to make site management decisions for efficient remedial actions. The primary factor resulting in persistence of groundwater contaminant plumes after source remediation is aqueous phase contaminant released from low-permeability zones (back diffusion) with transfer of mass to high-permeability zones. For example, Ball et al. (1997) and Liu and Ball (2002) used DNAPL concentration distributions in low-permeability zones before and after treatment of the source zone at Dover Air Force Base (DAFB), Delaware to demonstrate that contaminant mass stored in low-permeability zones could serve as a long-term secondary source of contamination, sustaining groundwater plume longevity in high-permeability zones. Other field studies conducted by Parker et al. (2004) and Chapman and Parker (2005) at a chlorinated-solvent contaminated site in Connecticut underpinned these conclusions based on comparison of aquitard concentration profiles and plume behavior before and after source isolation. They observed plume persistence of trichloroethene (TCE) in the aquifer 3 years after source isolation and concluded that the aquifer TCE concentration will be orders of magnitude greater than the maximum contaminant level (MCL) of 5 µg/L for centuries. Parker

et al. (2008) verified that back diffusion from thin low-permeability zones several years after source isolation was a dominant cause of plume persistence at a TCE-contaminated site in Florida, supported by concentration data and model simulations.

The modeling approaches employed in these previous studies have reconstructed source histories assuming either that solute concentrations have remained constant, or that changes have occurred in discontinuous steps. Moreover, similar constant or step-change concentration boundaries have also been implemented in several recent laboratory and modeling studies of back diffusion from low-permeability zones (Chapman et al., 2012; Sale et al., 2008; Yang et al., 2014a; Yang et al., 2015). However, the source dissolution dynamics of real source zones result in continuously changing source concentrations. Theoretical considerations (Jawitz et al., 2005), laboratory and field experiments (Brusseau et al., 2008; Chen and Jawitz, 2009; Difilippo and Brusseau, 2008; Fure et al., 2006) and modeling studies (Adamson et al., 2016; Basu et al., 2008; Newell and Adamson, 2005; Parker and Falta, 2008; Parker and Park, 2004) have established that source dissolution combined with heterogeneous DNAPL architecture results in a convergence to exponentially decreasing source concentrations. This result is consistent with an empirical study where breakthrough curve data from 23 contaminated sites were well described by exponentially decreasing source concentrations (Newell et al., 2006).

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Recently, Adamson et al. (2015b) provided a simple modeling tool using one-dimensional (1D) diffusion equations to hindcast the concentration boundary condition history based on measured concentration profiles in low-permeability zones at the two sites described in Parker et al. (2004) and Liu and Ball (1998, 2002). Their modeling results showed good fits with the measured profile data using similar boundary conditions as reported in the original studies. They also concluded that the aquifer concentration history can be derived from high-resolution contaminant profiles in low-permeability zones. However, Adamson et al. (2015b) still used discontinuous step-changes in boundary conditions, and also did not use the simulated source histories or concentration profiles to estimate combined impacts of source dissolution and back diffusion on aquifer concentration time series and plume persistence. Yang et al. (2016) investigated in a laboratory and modeling study how different patterns of decreasing aquifer concentrations, or source strength functions (SSFs, step-change, linear, and exponential source depletion) impact forward and back diffusion. They used the concentration gradients at the interface between high- and low-permeability zones to identify forward and back diffusion timing, and provided signatures of the source strength function boundary condition. Here, we evaluate the hypothesis that SSF signatures can be found in historical site data, and that these aquitard concentration profile signatures can be used to evaluate the relative contributions of exponential source dissolution and back diffusion to long-term plume persistence at field sites.

Many contaminated sites have available information on concentration time series from monitoring wells to assess source and plume characteristics and design remediation options. However, accurate prediction of future long-term source dissolution dynamics is confounded by the relatively short-term data collection period compared to the total contamination period. But when combined with aquitard core samples, these historic data can provide a robust basis for estimating source history. Therefore, the specific objectives of this paper are to (1) test whether signature shapes of aquitard concentration profiles at selected field sites are characteristic of dissolution alone, dissolution plus back diffusion, or back diffusion alone, (2) use historical data coupled with analytical and numerical models using linearly and exponentially decreasing solute concentrations to develop SSFs for site management purposes, (3) characterize the near-source plume response to source-mass depletion at selected field sites to provide the understanding needed to predict long-term plume responses, and (4) link the characterization of the near-source responses and back diffusion profiles to likely long-term behavior of the dissolved plume.

In this study, we analyze aquitard concentration data from 14 cores collected at three field sites: Dover Air Force Base (DAFB) in Delaware (Liu and Ball, 2002), a contaminated site in Connecticut (Chapman and Parker, 2005; Parker et al., 2004), and Naval Air Station Jacksonville (NASJ) in Florida (Adamson et al., 2015a). We simulate DNAPL source depletion based on deductive reasoning from simulation of the aquitard concentration data from the field sites using 1D analytical solutions, and adequately predict long-term plume responses based on DNAPL source dissolution in the aquifer and back diffusion from the aquitard. We also assess the relative contributions of SSFs and back diffusion to the plume downgradient of the source based on a three-stage plume classification: Stage 1 consisting of source dissolution in the aquifer and forward diffusion into the aquitard, Stage 2 with continued dissolution but aquifer concentrations have decreased below those in the aquitard resulting in back diffusion, and Stage 3 where the plume concentrations are predominantly attributed to back diffusion. This information is critical for site management decisions regarding proper timing of aquifer versus aquitard remediation.

2. Modeling approach

The conceptual model in this study is considered to be a single aquifer and aquitard system (Fig. 1). Both layers are homogeneous and

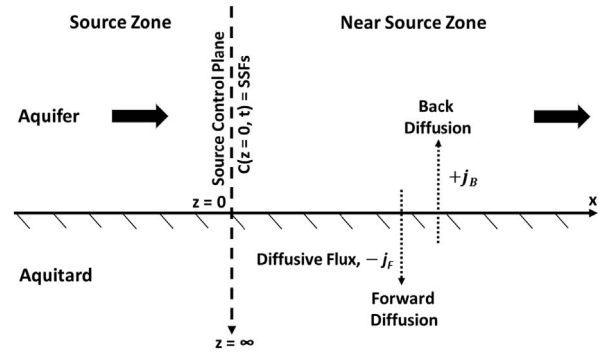


Fig. 1. Conceptual illustration of DNAPL source and near-source zones. The dissolved phase plume in the aquifer crosses the source zone control plane with different source strength functions (SSFs). Forward and back diffusion occur for negative and positive concentration gradients between the aquifer and aquitard.

isotropic and the interface between the aquifer and the aquitard is horizontal. The aquifer has groundwater flow at a constant rate in the x -direction. The aquitard thickness (z -direction) is assumed to be infinite. Advection in the aquifer is only in the x -direction. The initial contaminant concentration in the aquifer and aquitard is zero in the near-source zone before contaminant transport begins. Contaminants at the source zone control plane are controlled by source zone dissolution described by SSFs, and are transported to the near-source zone where the contaminants diffuse into the aquitard (forward diffusion) with negative diffusive flux in the z -direction. When source depletion decreases the near-source zone aquifer contaminant concentrations sufficiently for z -direction diffusive flux to become positive, contaminants in the aquitard are released through back diffusion. Note that we do not consider degradation reactions and transverse diffusion in the aquitard.

2.1. Source strength functions

The time-dependent relative concentration of dissolved DNAPL at the source zone control plane can be described by functions relating the source strength and the source mass (Brown et al. (2012); Falta et al. (2005); Parker and Park, 2004; Zhu and Sykes, 2004)

$$\frac{C_t}{C_0} = \left[1 + \frac{J_0}{M_0} \Gamma - 1t \right]^{\frac{1}{1-\Gamma}} \quad \Gamma > 0, \Gamma \neq 1 \quad (1)$$

where C_0 is the initial contaminant concentration [$M L^{-3}$] at the source zone control plane, M_0 is the initial mass [M] in the source zone, $J_0 = qAC_0$ is the initial mass discharge rate [$M L^{-2} T^{-1}$] over the cross sectional area A [L^2] perpendicular to the Darcy flux q [$L T^{-1}$], and Γ is a dimensionless empirical parameter for SSFs that expresses the degree of the combined heterogeneity of the aquifer and the contaminant source. The time t [T] is the difference between the time since the initial release and the arrival time of the contaminant at the down-gradient location of interest.

Based on Eq. (1), solute concentrations that decrease as step changes, linearly, or exponentially are represented with $\Gamma = 0, 0.5$, and 1 , respectively. Here we refer to the latter two as linear and exponential source depletion (LSD and ESD). For these SSFs, the time-dependent relative concentration at the control plane can be expressed as follows (Brown et al. (2012); Falta et al. (2005); Yang et al. (2016); Zhu and Sykes (2004))

$$\frac{C_t}{C_0} = \begin{cases} 1 & t < \frac{M_0}{J_0} \\ 0 & t \geq \frac{M_0}{J_0} \end{cases} \quad \Gamma = 0 \quad (2)$$

$$C_z = 0 \quad t = C_0 - kt \quad \Gamma = 0.5 \quad (3)$$

$$C_z = 0 \quad t = C_0 \exp(-\beta t) \quad \Gamma = 1.0 \quad (4)$$

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