



Solute source depletion control of forward and back diffusion through low-permeability zones



Minjune Yang^a, Michael D. Annable^b, James W. Jawitz^{a,*}

^a Soil and Water Science Department, University of Florida Gainesville, FL 32611, United States

^b Department of Environmental Engineering Sciences, University of Florida Gainesville, FL 32611, United States

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ABSTRACT

Solute diffusive exchange between low-permeability aquitards and high-permeability aquifers acts as a significant mediator of long-term contaminant fate. Aquifer contaminants diffuse into aquitards, but as contaminant sources are depleted, aquifer concentrations decline, triggering back diffusion from aquitards. The dynamics of the contaminant source depletion, or the source strength function, controls the timing of the transition of aquitards from sinks to sources. Here, we experimentally evaluate three archetypical transient source depletion models (step-change, linear, and exponential), and we use novel analytical solutions to accurately account for dynamic aquitard-aquifer diffusive transfer. Laboratory diffusion experiments were conducted using a well-controlled flow chamber to assess solute exchange between sand aquifer and kaolinite aquitard layers. Solute concentration profiles in the aquitard were measured in situ using electrical conductivity. Back diffusion was shown to begin earlier and produce larger mass flux for rapidly depleting sources. The analytical models showed very good correspondence with measured aquifer breakthrough curves and aquitard concentration profiles. The modeling approach links source dissolution and back diffusion, enabling assessment of human exposure risk and calculation of the back diffusion initiation time, as well as the resulting plume persistence.

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1. Introduction

Storage and release of aquifer contaminants, including dissolved constituents of dense non-aqueous phase liquids (DNAPLs) from low-permeability zones is considered to be a critical limitation to contaminated site management (Cavanagh et al., 2014; Mackay and Cherry, 1989; Stroo et al., 2012; Stroo et al., 2003; Travis and Doty, 1990). In particular, the solute release process (back diffusion) from low-permeability zones leads to sustaining plume longevity with low-level contamination in the aquifer after the source zone has been isolated or remediated. Several field studies have demonstrated back diffusion from low-permeability zones as a significant contributor to plume persistence at contaminated sites (Ball et al., 1997; Chapman and Parker, 2005; Liu and Ball, 2002; Parker et al., 2004). Fundamental laboratory and modeling studies have been conducted to investigate back diffusion mechanisms and to develop predictive methods (Chapman et al., 2012; Sale et al., 2008b; Yang et al., 2015). In these studies, the aquifer concentration change that triggered a reversal from forward to back diffusion occurred instantaneously. However, aquifer concentrations in real source zones decrease gradually rather than instantaneously, because of aquifer and contaminant heterogeneity (Rao and Jawitz, 2003).

The dynamic behavior of source dissolution has been shown to be strongly influenced by the DNAPL architecture, with the pattern of concentration reduction with time referred to as source strength function, SSF (Chen and Jawitz, 2008; Chen and Jawitz, 2009; Christ et al., 2006; Falta et al., 2005a; Falta et al., 2005b; Fure et al., 2006; Jawitz et al., 2005; Parker and Park, 2004; Rao and Jawitz, 2003; Suchomel and Pennell, 2006). Plume persistence is thus affected both by SSF and back diffusion, and an important question is the relative contribution of these two factors. A recent modeling study addressed this question, finding that the persistence of higher-solubility contaminant source zones is supported more from back diffusion than from dissolution, while both processes contribute to persistence of contaminants with lower solubility (Seyedabbasi et al., 2012). That study focused on source mass discharge but did not consider the evolution of aquitard contaminant concentration profiles during forward and back diffusion. The concentration profiles in aquitards represent the integrated history of the source zone, and these data may be used to infer information about the SSF (Chapman and Parker, 2005; Liu and Ball, 2002). Another modeling study investigated the influence of DNAPL source architecture on the risk of back diffusion in plume zones (Brown et al., 2012). Source zone dissolution was generated by a power law source depletion model, and the results suggested that the greatest back diffusion risk occurs from a constant SSF whereas less risk was expected with an increase in aquifer and contaminant heterogeneity. However, that study did not compare fluxes from back diffusion to those resulting from source

* Corresponding author.

E-mail address: jawitz@ufl.edu (J.W. Jawitz).

dissolution dynamics, leaving a gap in understanding about the combined effects of SSFs on plume persistence.

In this study we extend the work of previous investigators by assessing the relative contributions to plume persistence of source dissolution dynamics (characterized by SSF) and back diffusion in controlled laboratory experiments where both breakthrough curves and aquitard solute concentration profiles were monitored. We implemented three archetypical transient source depletion patterns (step-change, linear, and exponential) in laboratory experiments in a two-dimensional flow chamber with high- and low-permeability layers representing an aquifer and aquitard, and with these data we validate analytical solutions for aquifer solute flux and aquitard concentrations. We also classify stages in plume evolution based on relative importance of dissolution and back diffusion, and provide guidance for the application of analytical solutions to verify timing of back diffusion initiation and the magnitude of back diffusion flux from low-permeability layers, based on SSFs.

2. Material and methods

2.1. Experimental method

2.1.1. Flow chamber boundary conditions

Three experiments were conducted in a well-controlled two-dimensional flow chamber to generate forward and back diffusion with three SSFs: (1) constant source with a step-change in strength, (2) linear source depletion, and (3) exponential source depletion. The flow chamber consisted of aluminum tubing frames (28 × 12 × 1.2 cm) and two glass sheets, similar to previous studies (Yang et al., 2014; Yang et al., 2015) and was filled with 20/30 mesh Accusand (4 cm) (Unimin Corp.) with an upper layer of kaolinite (7 cm) (Fluka) under saturated conditions (Fig. 1). Teflon tape was applied at the top of the clay layer to minimize evaporation. Steady-state flow was maintained with variable source concentrations delivered to an injection well using a syringe pump (Harvard Apparatus Syringe Infusion Pump 22) and outflow controlled using a piston pump (Fluid Metering Corp. “Q” Pump).

In each experiment bromide was displaced through the aquifer for a finite duration to facilitate forward diffusion to the aquitard, followed by different transitions to zero-concentration water flooding. After the loading periods in all experiments, water flushing was maintained without bromide for an additional 30 days to trigger back diffusion. In experiment I, a solution of 10 g/L of bromide was displaced for 7.5 d. In experiment II, for reproducing linear source depletion, bromide concentrations were reduced linearly from 10 to 0 g/L in 15 daily increments. In experiment III, a continuously stirred tank reactor (CSTR) was used to generate an exponential SSF (Fig. 1). The effluent concentration, $C(t)$, from a CSTR is defined as (Schmidt, 1998)

$$C(t) = C_0 e^{-t/\tau} \tag{1}$$

The CSTR volume (V) was maintained at 230 mL and the initial bromide concentration, C_0 , was 10 g/L. The effluent bromide concentration decreased from 10 to 0 g/L over 40 days. All experiments were conducted at a steady flow rate, $Q = 0.02$ mL/min. The experimental pore volume size was 40.3 mL and the estimated mean travel time from the inlet to the outlet of the flow chamber was 1.4 d. The timing and rate of the solute boundary condition transitions were designed such that an equal mass of bromide (2160 mg) was injected in each experiment.

2.1.2. In situ measurement of aquitard concentration

Bromide concentrations were measured in the kaolinite layer non-destructively by electrical conductivity analysis. Fourteen steel electrodes were installed in two columns of seven rows at 1 cm intervals on one side of the flow chamber (Fig. 1). The pore water electrical conductivity in the kaolinite was measured every 24 h by connecting the electrodes in each row using a digital conductivity meter (Fisher Scientific). The conductivity of the flow chamber influent and effluent was also monitored continuously. Measured conductivities were converted to relative bromide concentration, $C_r(t)$,

$$C_r(t) = \frac{EC(t) - EC_i}{EC_0 - EC_i} \tag{2}$$

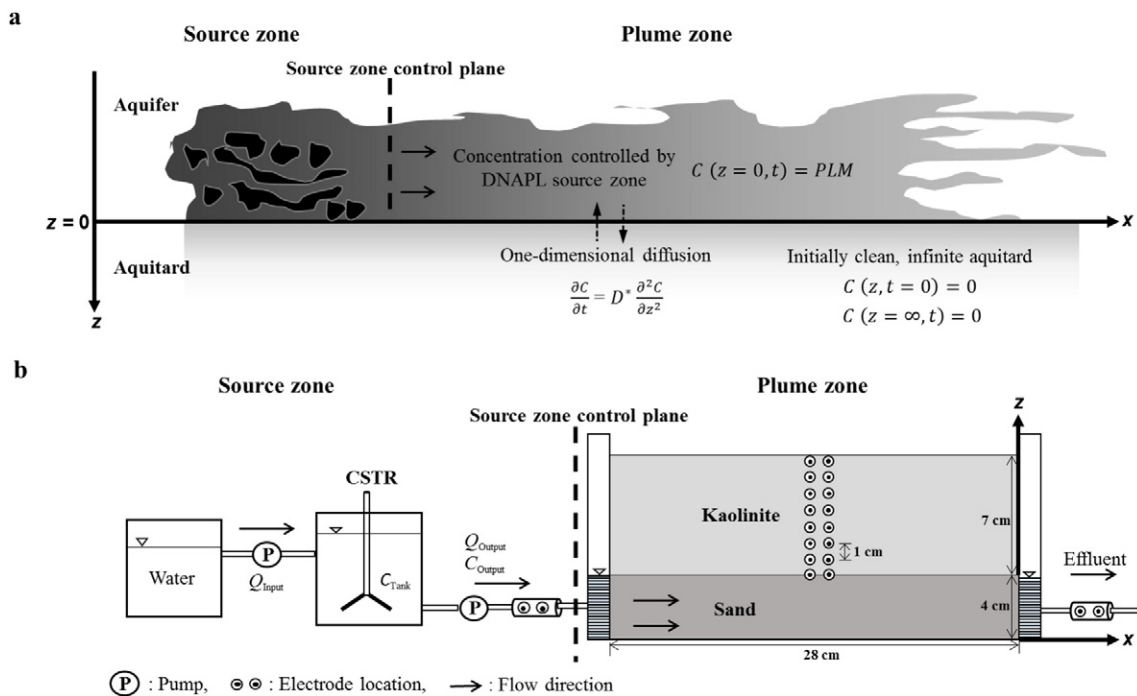


Fig. 1. Conceptual illustration (a) of DNAPL source and plume zones where the dissolved contaminant plume (shaded area) is crossing the source zone control plane, and schematic of laboratory experimental apparatus (b), with a CSTR as a model of DNAPL source zone dissolution dynamics, and a flow chamber as a model of plume transport through an aquifer with an adjacent aquitard.

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