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Characterisation of a DNAPL source zone in a porous aquifer using the Partitioning Interwell Tracer Test and an inverse modelling approach

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ABSTRACT

In this paper, we discuss the results of a Partitioning Interwell Tracer Test (PITT) performed in a large scale experiment with a well-defined TCE spill, and present a novel combined analytical–numerical inverse modelling approach using measured concentration profiles within a TCE plume to predict the distribution of the DNAPL in a virtual vertical plane of the source. The proposed inverse modelling approach assumes local thermodynamic equilibrium of the distribution of TCE between the NAPL phase and the aqueous phase and no decay or sorption of the dissolved TCE concentrations downstream of the spill area.

The analytical part of the inverse modelling approach contains two steps. As a first step, the location of the contaminant in a virtual vertical plane of a porous medium is fixed by using measured concentration profiles and considering the dissolution of the organic phase under equilibrium conditions. In the second step, the volume of contaminant entrapped in the source cells is estimated. A multiphase advective–dispersive transport model is used in the final step to adjust the volumes quantified in the second step. The predictions are highly dependent on the quantity and quality of the data in space and time.

From the PITT-breakthrough curves measured at the pumping well, a mean TCE saturation in the sweep zone of 0.0004 was derived, which is very low compared to that determined at the local scale. In a second analysis, tracer breakthrough curves available at measuring points placed closely downstream and upstream of the presumed source zone, were used to explain why the globally obtained DNAPL saturation was very low compared to the “real”, locally evaluated TCE saturations in the source zone. This was principally caused by the overall travel time compared to the short travel time of the tracers in the source zone. Another reason is that due to bypassing, only part of the volume of tracer injected had been in contact and had eventually interacted with the DNAPL. Furthermore, the quantified TCE volume was nearly 30% higher than the spilled volume; this agrees with the conclusions from other studies emphasizing that calculated volumes can overestimate the measured volumes, particularly in the case of an inhomogeneous distribution of the DNAPL within the soil.

A good agreement of the measured and inversed concentration profiles was obtained, highlighting that it is possible to determine the length-averaged distribution of a residual pollution source from dissolved concentration profiles measured downstream of the source zone. The numerically obtained non-uniform distribution of DNAPL entrapped in the vertical plane of the source zone was experimentally confirmed by the TCE saturation values derived from PITT-breakthrough curves at measuring points located 0.75 m downstream of the source

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zone. However, the sensitivity study showed that the inverse modelling approach provided a rather non-unique solution. More data available may reduce the number of possible representations of the estimated source zone.

1. Introduction

Dense Non-Aqueous Phase Liquids (DNAPL) such as chlorinated solvents, e.g., trichloroethylene (TCE) commonly used in industry, have often been found to be the source of contamination of underground drinking water supplies subsequent to their accidental release (Cohen and Mercer, 1993). In the subsurface, chlorinated solvents migrate vertically depending upon gravity and capillary forces through the unsaturated zone, where a large vapour plume appears as a result of their high volatility. After rainfall, they can strongly contaminate the groundwater by dissolution of vapours (Mendoza and Frind, 1990; Sleep and Sykes, 1992). If the released volume is significant, the contaminant reaches and penetrates the groundwater table as a result of its high density, dissolves and forms a solute plume. Within the aquifer, a certain quantity of solvents is entrapped as blobs or forms pools over low-permeability geological structures. This constitutes a long-term source of contamination of drinkable groundwater resources, given the low solubilities of the solvents (Schwille, 1988; Brusseau, 1992; Kennedy and Lennox, 1997; Eberhardt and Grathwohl, 2002). Furthermore, infiltration of DNAPLs is not uniform, but is influenced by both the heterogeneity of the porous medium and the contrast of viscosity between water and solvents, generating fingering. Moreover, dissolution of DNAPL is not homogeneous in space and seems to be rate-limited: depending on the groundwater flow rate, the shape of the source zone, the interfacial area between organic and aqueous phase and finally the composition of the DNAPL (Kueper and Gerhard, 1995; Wiedemeier et al., 1999, Imhoff et al., 2003; Bradford et al., 2003; Soga et al., 2004).

Knowledge of spatial and mass distribution of organic contaminants is a key problem in groundwater contamination by chlorinated solvents. To remediate efficiently a polluted site, one needs to characterise appropriately the source zone. Kram et al. (2001a,b) gave a complete review of DNAPL site characterisation methods that included the advantages and disadvantages of each method. One of the traditional techniques consists of extracting soil samples (Jellali et al., 2001) and using an interpolation technique. However, this kind of method is intrusive, and is sensitive to the choice of both the interpolation technique and the number of samples. It is also expensive, and the results obtained are very uncertain due to the high risk of mobilisation of the DNAPL during the process (Pankow and Cherry, 1996). Thus, new experimental methods have been developed for source zone characterisation. While methods such as geophysical screening (Azaria et al., 2000) or isotopic tracer techniques (Höhener et al., 2005) have been proposed, they need to be further tested before application to real contaminated sites.

Currently, the most commonly used tracer technique is the so-called Partitioning Interwell Tracer Test (PITT) (Jin et al., 1995; Dwarakanath et al., 1999). It is non-intrusive and consists of injecting simultaneously a conservative and a reactive tracer upstream of the source zone within the saturated zone, and their subsequent recovery downstream of the supposed location of the source. The delay between the two tracer signals is measured and is the basis for estimating the hydrodynamically accessible DNAPL quantity, i.e., the volume and average saturation in the swept zone (Rao et al., 2000). The advantage of this method is that the swept DNAPL volume is larger than that taken into account in soil core sampling or geophysical measurements (Mayer and Miller, 1992). Theoretical details of the PITT have been discussed in many papers (Jin et al., 1995; Rao et al., 2000; Brooks et al., 2002; Meinardus et al., 2002; Bohy et al., 2004).

The performance of this technique has been reported in the literature. While some studies have shown a good agreement between measurements and calculations (Jin et al., 1995; Meinardus et al., 2002), several other research works have exposed the limitations of this technique. Namely, the calculated volumes overestimated measured values (Brooks et al., 2002; Dai et al., 2001) due to the mass transfer resistance (Imhoff and Pirestani, 2004), the properties of the porous medium and the inhomogeneous distribution of the DNAPL within the soil (Dai et al., 2001; Jalbert et al., 2003; Moreno-Barbero et al., 2004). Under these conditions, the sampling method can also reduce the accuracy of the partitioning tracer test, which can be minimised by using multi-level sampling (Nelson et al., 1999).

Recently, with the development of numerical multiphase codes, several authors have been interested in mathematical approaches coupled with measurements in order to characterise DNAPL sources. Michalak and Kitanidis (2002) presented a geostatistical inverse modelling method using an analytical solution given by Liu and Ball (1999). This approach allows determining the historical evolution of the source zone with reasonable precision, but needs more additional information about the physical boundary conditions of the polluted zone. Saenton and Illangasekare (2003, 2004) developed a concentration-flux matching method, which consists of an inverse modelling approach based on vertical profiles of equilibrium concentrations and dissolved mass flux. However, the uncertainty of their results is quite high.

In this paper, the results of a controlled large scale experiment with a known amount of released TCE in the saturated zone of a model aquifer are used in order to (i) perform the PITT under well known boundary conditions, (ii) compare both the estimated DNAPL saturations and volume with the initially released quantity, and (iii) apply a novel combined analytical and numerical inverse modelling approach to measured concentration profiles in the plume downstream of the source zone to obtain possible locations of the DNAPL in a virtual vertical plane of the TCE spill.

2. Theory of the inverse modelling approach used

To determine one or more possible DNAPL distributions in the source zone, the modelling approach is based on an inversion of measured dissolved TCE concentrations downstream of the