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Highly parameterized inversion of groundwater reactive transport for a complex field site

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ABSTRACT

In this study a numerical groundwater reactive transport model of a shallow groundwater aquifer contaminated with volatile organic compounds is developed. In addition to advective–dispersive transport, the model includes contaminant release from source areas, natural attenuation, abiotic degradation by a permeable reactive barrier at the site, and dilution by infiltrating rain. Aquifer heterogeneity is parameterized using pilot points for hydraulic conductivity, specific yield and groundwater recharge. A methodology is developed and applied to estimate the large number of parameters from the limited data at the field site (groundwater levels, groundwater concentrations of multiple chemical species, point-scale measurements of soil hydraulic conductivity, and lab-scale derived information on chemical and biochemical reactions). The proposed methodology relies on pilot point parameterization of hydraulic parameters and groundwater recharge, a regularization procedure to reconcile the large number of spatially distributed model parameters with the limited field data, a step-wise approach for integrating the different data sets into the model, and high performance computing.

The methodology was proven to be effective in reproducing multiple contaminant plumes and in reducing the prior parameter uncertainty of hydraulic conductivity and groundwater recharge. Our results further indicate that contaminant transport predictions are strongly affected by the choice of the groundwater recharge model and flow parameters should be identified using both head and concentration measurements.

1. Introduction

Subsurface reactive transport models are increasingly applied to real contaminant sites. However, major challenges are present when applying such models to reproduce the observed concentration measurements and attempt to use them as predictive tools. One of these challenges is the correct conceptualization and description of the subsurface processes using a limited amount of measurements (Gupta et al., 2012; Matott and Rabideau, 2008) and the computational burden. In practical terms, simulation of solute transport over a period of years to decades may require hours or days to complete on modern personal computers (Konikow, 2011). For these reasons and to avoid complexity not justified by the available data, simplifying assumptions are often made regarding the spatial distribution of aquifer parameters, groundwater flow and transport dynamics, degradation rates and parameter inference procedures (Hill, 2006).

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In numerous studies homogeneous aquifer properties (within zones, layers or over the entire model domain) are used (D'Affonseca et al., 2011; Karlsen et al., 2012; Vandenbohede et al., 2013). Disadvantages of simple parameterization schemes are that the real response of the physical system might not be captured, introducing structural errors (Gallagher and Doherty, 2007) and hampering the ability to understand the natural system through model calibration (Hunt et al., 2007). Only a limited number of field-scale reactive transport studies directly inferred heterogeneity from available data (Fienen et al. (2009b); Tonkin and Doherty, 2009), whereas abundant literature is present for flow only problems (Hayley et al., 2014; Laloy et al., 2013; Yoon et al., 2013). Tonkin and Doherty (2005) applied the pilot point method (Marsily, 1984) to infer spatially variable flow and transport parameters (hydraulic conductivity, recharge and porosity) at the Hampton Bays Site, New York. In their approach a large number of model parameters were defined and the computational burden reduced through the definition of "super parameters". The method was used to reproduce the observed MTBE (Methyl tert-butyl ether) concentrations at the site, effectively demonstrating that a better reproduction of the observations was obtained when aquifer heterogeneity is accounted for. Kowalsky et al. (2012) studied the effect of different parameterizations of the hydraulic conductivity field (e.g. pilot point number and locations) in a synthetic tracer experiment, indicating the need for real applications where various data types are used in parameter inference. Fienen et al. (2009b) used different data types (head, oxygen and tritium isotopic measurements) in the geostatistical method (Hoeksema and Kitanidis, 1984; Kitanidis and Vomvoris, 1983) to infer hydraulic conductivities of an isthmus between two lakes in the Trout Lake watershed, northern Wisconsin, United States. They concluded that the flexible parameterization offered by the geostatistical method and the simultaneous use of multiple data sources improved the flow path delineation in the isthmus zone. In all cases, hundreds to thousands model parameters were used, combined with parameter regularization (Carrera et al., 2005; Yeh, 1986) to include prior parameter knowledge (Fienen et al. (2009a)). Pilot point and geostatistical methods allow the optimal level of complexity to be inferred directly from the data, provided that the model is correctly conceptualized and the correct assumptions about prior parameter knowledge are made.

Besides heterogeneity in hydraulic conductivity, spatial and temporal variation of groundwater recharge is important in modeling contaminant transport in shallow unconfined aquifers, due to the possible dilution effect by infiltrating rainwater. For example, Kowalsky et al. (2011) incorporated geochemical and time-lapse resistivity data in a hydrogeochemical model of the Oak Ridge Integrated Field Research Challenge (IFRC) site in Tennessee, United States. In their model it was assumed that groundwater recharge determines nitrate concentrations in the unconfined aquifer, in particular in the shallow part. Their study also points to the need for spatially extensive datasets (including chemical concentrations and geophysical measurements) to monitor recharge related concentration variations. Şengör and Ünlü (2013) developed a numerical model to determine the extension of acrylonitrile contamination at a spill site in Turkey. In their long term simulations (2001 to 2011), the size of the plume in a high permeability zone shrank significantly due to dilution by groundwater recharge. In these types of problems, flow and transport in the unsaturated zone should ideally also be modeled, further increasing the computational demand (Yabusaki et al., 2011). A simpler approach is to assume groundwater recharge equal to the average net balance at the water table (D'Affonseca et al., 2011) or a spatially variable fraction of this balance (Hayley et al., 2014; Tonkin and Doherty, 2005). Assefa and Woodbury (2013) estimated groundwater recharge in the Okanagan basin (Canada) by calibrating a HYDRUS-1D model (Simunek et al., 2005) using soil moisture data, and extrapolating the results over the entire basin (245 km^2) based on soil characteristics. Their results show significant spatial variability in recharge, varying from 12 to 170 mm y^{-1}. On a larger scale (3600 km^2), Hayley et al. (2014) estimated groundwater recharge and horizontal hydraulic conductivity from head and flow data, also obtaining variable groundwater recharge rates.

Another important issue concerns the calibration methodology. In most reactive transport studies, flow parameters are calibrated first against head and flow data to provide the flow field to be used in reactive transport calculations (D'Affonseca et al., 2011; Gorelick et al., 1983; Prommer et al., 2009). Afterwards, the remaining reactive transport parameters are calibrated on concentration data, using independent estimations (e.g. lab experiments or field observations) and refining the initial guesses by manual calibration. The limitation of this approach is that the information about flow parameters contained in the concentration measurements is not used. Many synthetic and lab-scale studies demonstrate that concentration data are more informative of the local subsurface heterogeneity than groundwater head data (Pollock and Cirpka, 2012; Wagner, 1992).

In this study, we explicitly tackle the challenges discussed above, by means of an application of existing methodologies to a complex real-world contaminated site. Our study goes beyond existing applications which typically are restricted to single reactive species or synthetic aquifers. Applications of highly parameterized inversion of multi-component reactive transport models are not widely reported, and our study illustrates both the benefits and limitations of such an approach for real-world applications. Specifically, we show that parameter estimation in shallow heterogeneous contaminated aquifers strongly influenced by recharge dynamics is possible by a combination of regularization, high-performance computing, and a step-wise approach of parameter refinement using laboratory and field data. The methodology is evaluated in terms of reproducing the observations, realism of the estimated parameters and reduction of the prior parameter uncertainty.

2. Methods

2.1. Study site

The studied contaminated site, depicted in Fig. 1, is located in Wilrijk, just south of Antwerp, Belgium. The factory shown in the Fig. 1 began production of compressors around 1951 and used chlorinated solvents mainly for painting and degreasing activities. The first use of chlorinated solvents (VOC) occurred in 1957, including tetrachloroethene (PCE), trichloroethene