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Testing high resolution numerical models for analysis of contaminant storage and release from low permeability zones

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ABSTRACT

It is now widely recognized that contaminant release from low permeability zones can sustain plumes long after primary sources are depleted, particularly for chlorinated solvents where regulatory limits are orders of magnitude below source concentrations. This has led to efforts to appropriately characterize sites and apply models for prediction incorporating these effects. A primary challenge is that diffusion processes are controlled by small-scale concentration gradients and capturing mass distribution in low permeability zones requires much higher resolution than commonly practiced. This paper explores validity of using numerical models (HydroGeoSphere, FEFLOW, MODFLOW/MT3DMS) in high resolution mode to simulate scenarios involving diffusion into and out of low permeability zones: 1) a laboratory tank study involving a continuous sand body with suspended clay layers which was 'loaded' with bromide and fluorescein (for visualization) tracers followed by clean water flushing, and 2) the two-layer analytical solution of Sale et al. (2008) involving a relatively simple scenario with an aquifer and underlying low permeability layer. All three models are shown to provide close agreement when adequate spatial and temporal discretization are applied to represent problem geometry, resolve flow fields and capture advective transport in the sands and diffusive transfer with low permeability layers and minimize numerical dispersion. The challenge for application at field sites then becomes appropriate site characterization to inform the models, capturing the style of the low permeability zone geometry and incorporating reasonable hydrogeologic parameters and estimates of source history, for scenario testing and more accurate prediction of plume response, leading to better site decision making.

1. Introduction

Attention was first drawn decades ago to back diffusion as a potential impediment to restoration of aquifers. Mutch et al. (1993) introduced the idea for fractured sedimentary rock and Wilson (1997) for low permeability lenses in sandy aquifers with modeling for hypothetical scenarios. The first field verification of back diffusion reported in the literature was provided by Liu and Ball (2002) who used core sampling of a clayey aquitard inside a sheet piling cell at a remediation experiment site within a PCE and TCE plume at Dover Air Force Base, Delaware. The inward and back diffusion that produced the concentration profiles at the bottom of the aquifer into the aquitard were closely represented by a 1-D analytical solution. Parker et al. (2004) and Chapman and Parker (2005) also used high resolution core sampling to define concentration profiles in a clayey aquitard underlying a TCE plume at an industrial site in Connecticut and 1-D analytical and numerical solutions to simulate the contaminant profiles in the source area and a 2-D finite element numerical model (HydroGeoSphere) to simulate the evolution of the TCE profiles in the aquitard below the plume and diffusive mass fluxes into and out of the aquitard that strongly influence aquifer concentrations. Back diffusion was shown to be the cause of plume tailing after the primary

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DNAPL source was isolated with a sheet pile enclosure, governing the long time scale of aquifer restoration. This scenario is similar to those examined conceptually by Sale et al. (2008) using a two-layer analytical solution. However use of numerical models is necessary for the more complex conditions typical of most sites.

A major challenge of diffusion as a governing process, both in terms of field characterization efforts and incorporation in numerical models, is that it occurs over small scales relative to the typical dimensions of plumes. The response of plumes at the scale of 100s to 1000s of meters can be dependent on concentration gradients that evolve over dimensions of centimeters or less. While upgradient loading to plumes is steady or increasing, concentration gradients drive accumulation of immobile contaminants in low permeability zones within the source and plume as dissolved and sorbed phase. With a reduction in upgradient loading, due to natural source depletion or source treatment or isolation from the active flow system, aqueous concentrations in transmissive zones decrease, reversing concentration gradients and driving release of the stored mass from the low permeability zones back into transmissive zones via back diffusion.

A key controlling factor is the surface area of the high/low permeability zone interfaces (i.e. geometry of the low permeability zones) relative to the source zone and plume concentration distribution. The consequence of release of contaminants from low permeability zones is a reduction in the magnitude and an increase in the timing of downgradient water quality improvements associated with upgradient flux reduction. The back diffusion problem for chlorinated solvents is magnified since in most cases these were initially released as DNAPLs and concentration levels of concern (regulatory limits or action levels) can be several orders of magnitude (OoM) lower than initial source concentrations (aqueous solubility).

Besides analytical solutions for simple scenarios (e.g. Gillham et al., 1984; Sale et al., 2008; Sudicky et al., 1985), other numerical techniques offer potential to examine back diffusion effects in heterogeneous porous media. These include hybrid analytical–numerical models (e.g. Dentz and Berkowitz, 2003), particle based methods (e.g. Herrera et al., 2008, 2010; LaBolle et al., 2000; Salamon et al., 2006) and highly discretized finite element or finite difference numerical models. Chapman and Parker (2005) used a finite element numerical model (HydroGeoSphere) to examine TCE back diffusion from an aquitard to an overlying aquifer after source zone isolation at an industrial site in Connecticut, and Parker et al. (2008) used the same model to simulate TCE evolution in a sandy aquifer in Florida, where back diffusion from thin clayey beds cause plume persistence after source zone flux control by pump-and-treat. These two model applications provided close representation of the field results; however much effort was directed at capturing the diffusion processes and minimizing effects of numerical dispersion via appropriate spatial and temporal discretization. These studies demonstrate that numerical models can be used to simulate inward and back diffusion over time scales of many decades or longer at contaminated sites subjected to source remediation and provide a rigorous basis for expectations concerning the slow rate of improvement of aquifer concentrations governed by back diffusion long into the future, showing that back diffusion from even thin low permeability layers within sandy aquifers can govern aquifer water quality for long periods. Validation is dependent on temporal monitoring data that is obtained over sufficiently long time periods (i.e. decades) which is not available at most sites.

Improved prediction capability is essential for decisions regarding management of subsurface releases of chlorinated solvents and other persistent groundwater contaminants, by providing better understanding of how reduced contaminant loading to plumes in unconsolidated porous media will improve downgradient water quality, which is a subject of broad debate (e.g. EPA, 2003; ITRC, 2002; NRC, 2005; Sale and Newell, 2010; Stroo et al., 2003). Use of numerical models that provide more accurate assessment of downgradient water quality response to reduced contaminant loading incorporating back diffusion effects can allow better decision making with costs balanced against benefits. The key is employing numerical models that 1) address the primary governing processes, 2) accurately solve the governing equations and minimize numerical dispersion, and 3) employ determinable input parameters at appropriate scales consistent with the geometry of low permeability zones and their influence on plume evolution.

Besides HydroGeoSphere, other numerical models have capability for high resolution simulations incorporating diffusion processes, such as MODFLOW with associated transport codes (e.g. MT3DMS) and FEFLOW. The goal of this paper is to compare these models with HydroGeoSphere for simulating forward and back diffusion effects from low permeability zones in stratified or layered sandy aquifers containing low permeability zones, which may occur at scales of several centimeters to decimeters (e.g. silty or clayey layers or lenses) or larger (e.g. aquitards), that can be identified with appropriate site investigation techniques. This comparison is benchmarked by experimental observations from a well-controlled laboratory sand tank experiment, where the full cycle of transport was assessed via injection of tracers into a system with suspended low permeability zones with known geometry followed by their removal via flushing. This represents a challenge for the numerical models because of the discreteness and number of the low permeability zones into which the tracer mass diffuses inward and then outward in response to source removal and flushing and the small scales over which these processes occur. Also in the supporting information the three numerical models are tested in their ability to simulate results from the two-layer analytical solution of Sale et al. (2008) representing field-scale conditions for a relatively simple scenario of a sand aquifer overlying a low permeability layer, similar to that examined by Chapman and Parker (2005). In addition to determining whether each model can successfully simulate the experimental and analytical solution results, a goal was to determine guidance for applying these models. If the applicability of numerical models can be established for assessing back diffusion for well-controlled lab experiments and simpler field-scale scenarios, the challenge for application at real sites becomes appropriate high resolution site characterization required to define the style of the low permeability zone geometry and determine appropriate input parameters to inform such numerical models in an advection–diffusion context. While capturing the full complexity of field site conditions may not be realistic, even stylistic simulations that incorporate diffusion processes with reasonable representations of the low permeability zone geometry will allow improved prediction of plume behavior and