A semi-analytical method for simulating matrix diffusion in numerical transport models

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ABSTRACT

A semi-analytical approximation for transient matrix diffusion is developed for use in numerical contaminant transport simulators. This method is an adaptation and extension of the heat conduction method of Vinsome and Westerveld (1980) used to simulate heat losses during thermally enhanced oil recovery. The semi-analytical method is used in place of discretization of the low permeability materials, and it represents the concentration profile in the low permeability materials with a fitting function that is adjusted in each element at each time-step. The resulting matrix diffusion fluxes are added to the numerical model as linear concentration-dependent source/sink terms. Since only the high permeability zones need to be discretized, the numerical formulation is extremely efficient compared to traditional approaches that require discretization of both the high and low permeability zones. The semi-analytical method compares favorably with the analytical solution for transient one-dimensional diffusion with first order decay, with a two-layer aquifer/aquitard solution, with the solution for transport in a fracture with matrix diffusion and decay, and with a fully numerical solution for transport in a thin sand zone bounded by clay with variable decay rates.

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1. Introduction

There are thousands of sites around the world where groundwater has been contaminated by industrial activities. Many of these sites have been contaminated by chlorinated volatile organic compounds (CVOCs), often in the form of dense nonaqueous phase liquids (DNAPLs). DNAPLs serve as a concentrated source of groundwater contamination, and most dissolved plumes of CVOCs can be traced back to concentrated source zones. Considering that source concentrations can be four or five orders of magnitude greater than maximum contaminant levels (MCLs), restoration of source zones to pristine conditions is difficult; however, reduction of CVOC plumes is a realistic goal that may be achieved through various combinations of source and plume remediation. Much research over the past three decades has focused on technology development for both source and plume remediation at sites contaminated by CVOCs.

Unfortunately, some CVOC sites have proven to be exceedingly difficult to address due to the phenomenon of matrix diffusion. Matrix diffusion, also called "back diffusion," occurs when contaminants diffuse from high permeability zones into adjacent low permeability zones during a "loading period". During the "release period", the contamination may be removed from the high permeability zones, but contaminants in the low permeability zones gradually diffuse back into the high

permeability zones at significant levels (Parker et al., 1994, 1997; Ross and Lu, 1999; Slough et al., 1999; Espositio and Thomson, 1999; O'Hara et al., 2000; Reynolds and Kueper, 2001, 2002, 2004; Liu and Ball, 2002; Parker et al., 2004; Falta, 2005; Chapman and Parker, 2005; Parker et al., 2008; Sale et al., 2008; Sale et al., 2013). This process may occur in any heterogeneous setting, but it is particularly important in certain fractured bedrock sites, and in sites with extensive clay lenses or layers. These types of complex site conditions tend to lead to plumes that are long lived, requiring extensive long-term monitoring.

Analytical solutions have been developed for modeling matrix diffusion in single or parallel fractures (Neretnieks, 1980; Rasmuson and Neretnieks, 1981, Tang et al., 1981, Sudicky and Frind, 1982, West et al., 2004), in thick aquitards underlying permeable aquifers (Parker et al., 2004; Liu and Ball, 2002; Seyedabbasi et al., 2012), in coupled aquifer-aquitard systems (Sale et al., 2008), and in thin low permeability zones (Yang et al., 2015). The analytical solutions are widely used (see, for example Farhat et al., 2012), but they are restricted to idealized conditions.

Several numerical simulations of the back diffusion process have been performed. These simulations have used conventional contaminant transport modeling approaches where the transport occurs in high permeability zones due to advection and dispersion, and in the low permeability zones due to molecular diffusion (Parker et al., 2004; Chapman and Parker, 2005; Parker et al., 2008; Chapman et al., 2012; and Chapman and Parker, 2013). These studies have demonstrated the ability of existing numerical transport models to

simulate the transient matrix diffusion process. As noted by Chapman et al. (2012), a primary challenge in modeling the matrix diffusion process is that the diffusive fluxes at the high permeability/low permeability interfaces are controlled by local concentration gradients at a scale of cm. Resolving these gradients requires very high grid resolution, resulting in computationally intensive models. For this reason, few if any full scale three-dimensional simulations of matrix diffusion have been reported to date.

A hybrid method that fits between fully analytical and fully numerical modeling approaches employs embedded analytical or semi-analytical functions to increase local scale model resolution without greatly increasing computational effort. This approach has been successful in modeling transient heat conduction in reservoirs bounded by confining units (Vinsome and Westerveld, 1980) and in fractured reservoirs (Pruess and Wu, 1988, 1993). Since the governing differential equation for matrix diffusion is analogous to the heat conduction equation, this approach could be an attractive option for simulating contaminant matrix diffusion.

2. Heat conduction method of Vinsome and Westerveld

Steam and hot water injection are commonly used to enhance oil recovery from permeable oil reservoirs. Thermal conduction into adjacent low permeability zones can result in significant heat losses from the target reservoir. Conventional numerical simulation approaches for modeling the oil reservoir and adjacent confining units requires discretization of the confining units as well as the reservoir. This can result in greatly increased simulation run times compared to models that only consider the oil reservoir itself. Recognizing that the heat conduction process in confining units is primarily one-dimensional, Vinsome and Westerveld (1980) developed an elegant semi-analytical approximation for the heat conduction losses. This approach replaces discretization of the confining units with a temperature dependent heat source/sink term that is added to gridblocks that are adjacent to the confining layers. Their method has been shown to be both efficient and accurate, and it is widely used in thermal enhanced oil recovery and geothermal reservoir simulators (Pruess and Wu, 1993; Pope et al., 1999; Pruess et al., 1999; Pruess and Battistelli, 2002; Varavei and Sepehrnoori, 2009; Shi et al., 2009; Motley and Prevost, 2010).

Vinsome and Westerveld represent the temperature profile in the confining units with a fitting function of the form.

$$T_l(z_l, t) = \left(T^{t+\Delta t} + pz_l + qz_l^2\right)e^{-z_l/d} \tag{1}$$

where z_l is the distance into the low permeability material, $T^{t+\Delta t}$ is the current temperature at the interface between the reservoir and the confining unit, and d is a time dependent penetration depth for heat conduction:

$$d = \frac{\sqrt{\kappa_h t}}{2} \tag{2}$$

where $\kappa_h = k_h/\rho C_r$ is the thermal diffusivity, k_h is the thermal conductivity, ρ is the density and C_r is the heat capacity. The parameters p and q in Eq. (1) are time varying fitting parameters. The temperatures in Eq. (1) are defined with a zero level that corresponds to the initial uniform temperature in the confining layer. Algebraic expressions for parameters p and q are developed from two conservation of energy laws. First, the partial differential equation for heat conduction must be satisfied at the reservoir/caprock interface:

$$\rho C_r \frac{\partial T}{\partial t} = k_h \frac{\partial^2 T}{\partial z_l^2}\bigg|_{z_l=0} \tag{3}$$

and second, the rate of change of energy in the caprock must be equal to the conductive heat flux across the interface:

$$\frac{\partial}{\partial t}\int_0^\infty \rho C_r T dz_l = -k_h \frac{\partial T}{\partial z_l}\bigg|_{z_l=0} \tag{4}$$

Unique values of the parameters p and q are associated with each reservoir gridblock adjacent to the interface, and these parameters are updated during each time-step to represent the changing temperature profile in the confining units. The conductive heat flux into the confining unit is added to the reservoir gridblock energy balance as a linear temperature-dependent source/sink term using Fourier's Law with Eq. (1):

$$F_h = -k_h \frac{\partial T}{\partial z_l}\bigg|_{z_l=0} = k_h \left(\frac{T^{t+\Delta t}}{d} - p\right) \tag{5}$$

Since this method simulates the conductive response in the confining units, only the reservoir is discretized, greatly reducing the computation effort needed to simulate the process. In a multidimensional flow simulation, only those gridblocks adjacent to the confining units would use the semi-analytical conduction terms.

3. Matrix diffusion method

The governing partial differential equation for transient matrix diffusion is analogous to the differential equation for heat conduction, except that the matrix diffusion equation contains a first order decay term. Assuming that decay occurs only in the aqueous phase, one-dimensional matrix diffusion is governed by:

$$R_l \frac{\partial C_l}{\partial t} = \tau_l D \frac{\partial^2 C_l}{\partial z_l^2} - \lambda_l C_l \tag{6}$$

The subscript l in this equation denotes properties that correspond to the low permeability matrix or aquitard. Here, C_l is the aqueous concentration, R_l is the retardation factor, τ_l is the tortuosity, D is the molecular diffusion coefficient, and λ_l is the first order decay rate.

3.1. Calculation of matrix diffusion flux

Following Vinsome and Westerveld (1980) we assume a fitting function for the concentration in the low permeability zone:

$$C_l(z_l, t) = \left(C^{t+\Delta t} + pz_l + qz_l^2\right)e^{-z_l/d} \tag{7}$$

where $C^{t+\Delta t}$ is the current concentration at the interface between the high permeability and low permeability zones, the concentration penetration depth is defined by:

$$d = \frac{\sqrt{\kappa_l t}}{2} \tag{8}$$

and $\kappa_l = \tau_l D/R_l$. The concentrations in Eq. (7) are defined with a zero level that corresponds to the initial (uniform) concentration in the low permeability zone. This initial concentration would normally be zero. The parameters p and q are found by forcing the fitting function to satisfy the governing equation at the high permeability/low permeability interface, and from an expression of mass conservation in the low permeability material. The constraint at the interface is:

$$R_l \frac{\partial C_l}{\partial t} = \tau_l D \frac{\partial^2 C_l}{\partial z_l^2}\bigg|_{z_l=0} - \lambda_l C_l|_{z_l=0} \tag{9}$$

Using a first-order finite difference approximation for time derivative at the interface, and substituting the fitting function into the right