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Tracer travel and residence time distributions in highly heterogeneous aquifers: Coupled effect of flow variability and mass transfer

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SUMMARY

The driving mechanism of tracer transport in aquifers is groundwater flow which is controlled by the heterogeneity of hydraulic properties. We show how hydrodynamics and mass transfer are coupled in a general analytical manner to derive a physically-based (or process-based) residence time distribution for a given integral scale of the hydraulic conductivity; the result can be applied for a broad class of linear mass transfer processes. The derived tracer residence time distribution is a transfer function with parameters to be inferred from combined field and laboratory measurements. It is scalable relative to the correlation length and applicable for an arbitrary statistical distribution of the hydraulic conductivity. Based on the derived residence time distribution, the coefficient of variation and skewness of residence time are illustrated assuming a log-normal hydraulic conductivity field and first-order mass transfer. We show that for a low Damkohler number the coefficient of variation is more strongly influenced by mass transfer than by heterogeneity, whereas skewness is more strongly influenced by heterogeneity.

1. Introduction

An important challenge of subsurface hydrology is modeling advective transport in aquifers of high heterogeneity. Although two-dimensional cases have been extensively studied numerically (e.g., Bellin et al., 1992; Cvetkovic et al., 1996; Salandin and Fiorotto, 1998; Hassan et al., 1998; Jankovic et al., 2003; de Dreuzy et al., 2007; Gotovac et al., 2008; Gotovac et al., 2010), solutions for three-dimensional cases are still difficult and rare. Furthermore, mass transfer processes can affect transport significantly; these are known to be rate-limiting due to diffusion, yet their modeling is not straightforward. The most important consequence of mass transfer is retention – the slowing down of advective transport – and enhanced macro-dispersion. Hence there is a need to better understand the *combined* effect of advection in three-dimensional aquifers with moderate to high heterogeneity, and diffusive mass transfer, in view of the prevalence of reactive contaminants in many applications.

The approach followed here is the Lagrangean one we used in the past for weakly heterogeneous formations, where we adopted a first-order approximation in the log-conductivity variance σ_Y^2 (e.g., Dagan, 1984; Cvetkovic and Dagan, 1994). Advective transport of a tracer was modeled in terms of the travel time τ along streamlines of the steady velocity field. By incorporating a source term in the advective transport equation we were able to derive solutions for the mass arrival (the BTC) of reactive solutes at control planes normal to the mean uniform flow. The procedure was illustrated for spatially variable permeability of lognormal distribution and linear first order rate reaction; the latter was characterized by the constant partition coefficient A and the backward rate coefficient k_0. Due to the variability of τ among streamlines, the effect of reaction upon the arrival of a pulse at control planes varied between a constant retardation coefficient $R = 1 + A$ (for $k_0\tau \gg 1$) to pure advection (for $k_0\tau \ll 1$) (e.g., Cvetkovic and Dagan, 1994, Fig. 2a). By averaging over streamlines, the various spatial moments of solute plumes were determined at first order in σ_Y^2 as function of the flow and transport parameters. The results could be used in order to analyze field measurements for the mass and centroid position time variations of the bromoform and 1,2-dichlorobenzene plumes in the Borden Site experiment (Cvetkovic and Dagan, 1994, Fig. 6).

The aim of the present study is to extend the approach to aquifers of moderate to high heterogeneity, as encountered in many applications. This is achieved by using the semi-analytical solution of advective transport based on a multi indicator or multi inclusions model (MIM) structure (e.g., Fiori et al., 2003; Fiori et al.,

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2006; Dagan et al., 2003; Jankovic et al., 2006; Fiori et al., 2007), which has been recently cast as a time-domain random walk (TDRW) by Cvetkovic et al. (2014). The TDRW approach is currently a method of choice e.g., in modeling radionuclide transport for performance and safety assessment in advanced nuclear waste isolation programs (Painter et al., 2008; Painter and Mancillas, 2012; Smith et al., 2012) and has a number of advantages. Our recent work (Cvetkovic et al., 2014) provided a new tool for studying tracer residence time for advective transport in aquifers of arbitrary hydraulic conductivity distribution; when cast in the Laplace Transform domain, the TDRW solution is shown to be suitable for incorporating mass transfer processes.

In this paper we derive the expected tracer residence time distribution that combines the semi-analytical TDRW solution for advective transport in a three-dimensional aquifer with generalized linear mass transfer (Villermaux, 1987); first-order mass transfer is used for illustration. Relative effects of hydrodynamic and exchange processes on the coefficient of variation and skewness of expected tracer discharge are illustrated for different variances of hydraulic conductivity and different characteristic rates of mass transfer.

The paper is organized as follows. In the next two sections the problem is defined and the relevant theory presented. Based on the theory, a semi-analytical solution for the expected reactive solute BTC is developed in Section 4, including the temporal moments. Their relative dependencies on heterogeneity and mass transfer are illustrated and discussed in Section 5 while the main conclusions are summarized in Section 6.

2. Problem formulation

Groundwater flow, driven by a constant mean head gradient J in the x direction, takes place in a three-dimensional domain Ω. We consider transport between two aquifer cross-sections, the injection plane at $x = 0$ and a control plane at x (Fig. 1a). In this work we focus on two major physical mechanisms of transport: (i) hydrodynamic mechanism due to groundwater flow of steady velocity field $\mathbf{V}(\mathbf{x})$ and (ii) mass transfer process between mobile solute advected by the fluid and the immobilized one.

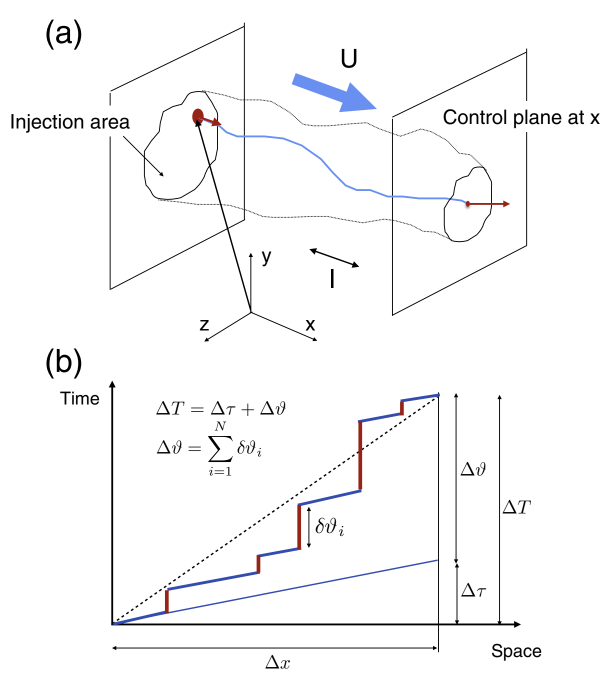

Fig. 1. Conceptualisation: (a) Tracer particle trajectory through a heterogeneous porous medium. (b) Transition (or residence) time for a segment Δx, ΔT, composed of the advective travel time $\Delta\tau$ and retention time $\Delta\vartheta$ (Painter et al., 2008). The retention time $\Delta\vartheta$ is a sum of return times (or duration of temporary trapping) $\delta\vartheta_k$ with the PDF $g(t)$, i.e., $\Delta\vartheta = \sum_{k=1}^{N} \delta\vartheta_k$ where the number of trappings N is a random variable. The PDF of $\Delta\vartheta$ is deduced from (11) as $\mathscr{L}^{-1}\{\exp[-pA\Delta\tau g(p)]\}$ based on tracer mass balance.

The formation is heterogeneous and of random stationary conductivity $K(\mathbf{x})$, of finite horizontal and vertical integral scale such that $\mathbf{V}(\mathbf{x})$, solution of the flow problem, is a stationary random space function in the transport domain of Fig. 1a. The mean velocity in the x direction is given by $U = K_{ef}J/\theta$, where K_{ef} is the effective conductivity and the porosity θ is assumed constant. A thin plume of total mass M_0 is injected at $t = 0$ in an area A_0 at the injection plane (IP) $x = 0$, of large dimension compared to the heterogeneity scale. Cartesian coordinates are defined as $\mathbf{x}(x, y, z)$, whereas coordinates in the plane $x = 0$ are denoted by $\mathbf{y}(y, z)$, $\mathbf{b}(b, c)$ such that $\mathbf{y} = \mathbf{b}$ is a coordinate within A_0. Toward quantification of transport by the mass arrival (the BTC) at the control plane, the hydrodynamic transport mechanism accounted for is advection by the velocity field $\mathbf{V}$, with neglect of local pore scale dispersion. Indeed, it was shown in Fiori et al. (2011) by accurate numerical simulations that pore scale dispersion has a minor effect on the BTC for the usual high values of Peclet numbers $Pe > 100$.

For most applications, retention takes place in some form, typically combining physical (diffusion) and chemical (sorption) mechanisms. A partitioning coefficient A (once equilibrium is reached) is a basic parameter for mass transfer between the immobile and mobile solute. A convenient and general means of quantifying the kinetics of mass transfer is through a memory function g (denoted as W in Cvetkovic and Dagan (1994)), as defined in the sequel. Its most basic parameter is a characteristic kinetic rate k_0 (the return rate), related to the rate of mass transfer.

Our focus in this work is to quantify the expected tracer residence time distribution between the injection and control planes (Fig. 1a). The residence time distribution is defined as normalized tracer mass discharge $\mu(t, x)$ $[1/T]$ (for a pulse of unit mass) between the two sections, given as a function of scale x. This is also the transfer function of the system that depends on the tracer (or water) travel time by advection on the one hand, and on the mechanism of mass transfer as characterized by the memory function g on the other. The goal of this work is to quantify μ as a function of the underlying structural/flow and mass transfer/retention properties that reflect controlling physical mechanisms of hydrodynamic transport and mass transfer. Specifically, we are set to derive $\mu(t, x)$ as a function of the flow and mass transfer parameters for an arbitrary distribution of K, and study its basic properties through moment analysis.

3. Theory

3.1. Advective tracer transport.

For the sake of completeness we recapitulate briefly the definitions and approach developed in the past (e.g., Cvetkovic and Dagan, 1994). The *travel time* τ of a fluid particle from the injection plane to the control plane at x is given by

$$\tau(x, \mathbf{b}) = \int_0^x \frac{dx'}{V_x[x', \eta(x', \mathbf{b}), \zeta(x', \mathbf{b})]} \tag{1}$$

where $y = \eta(x, \mathbf{b})$, $z = \zeta(x, \mathbf{b})$ is the equation of the streamline originating at $\mathbf{y} = \mathbf{b}$. The contribution of a solute (tracer) particle moving along the infinitesimal streamtube originating at the area $d\mathbf{b}$ to the relative mass M_r arrival at the control plane (the cumulative BTC) is given by

$$dM_r(x, t) = m_0(\mathbf{b})H[t - \tau(x, \mathbf{b})]\,d\mathbf{b} \tag{2}$$