Delay mechanisms of non-Fickian transport in heterogeneous media

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Received 30 May 2006; revised 6 July 2006; accepted 17 July 2006; published 29 August 2006.

[1] Fickian models of diffusion often fail to describe transport phenomena in heterogeneous environments due to their inability to capture the sub-scale fluctuations. We present an effective description of non-Fickian behavior that reflects the dichotomy between the continuum nature of Fick's law and the finite (effective) observation scale associated with experimental studies of transport phenomena in heterogeneous systems. This dichotomy gives rise to a time delay between the cause and effect, i.e. between the concentration gradient and the mass flux. Evolving scales of heterogeneity induce a spectrum of such delay times that can lead to anomalous behavior. The presented model is a direct generalization of Fick's law and the well-established delay diffusion model. It complements effective modeling frameworks based on stochastic non-local theories and continuous time random walks. **Citation:** Dentz, M., and D. M. Tartakovsky (2006), Delay mechanisms of non-Fickian trans-port in heterogeneous media, *Geophys. Res. Lett.*, *33*, L16406, doi:10.1029/2006GL027054.

1. Introduction

[2] Very few classical laws of physics are as ubiquitous as Fick's law of diffusion. Known by different names in various disciplines ranging from electromagnetics (Ohm's law) to heat conduction (Fourier's law) to flow in porous media (Darcy's law), it postulates a direct, instantaneous relationship between the cause (the system state gradient ∇c) and effect (the flux $\mathbf{J}_d$).

[3] Despite apparent successes of Fick's law of diffusion, it fails to capture such key transport characteristics as long tails and skewness, which are often observed in heterogeneous environments. Examples of the non-Fickian behavior of diffusive transport in heterogeneous environments can be found in almost every discipline in the natural sciences ranging from biology [e.g., *Upadhyaya et al.*, 2001], to atmospheric physics and oceanography [e.g., *Frisch*, 1995]. Here we focus on the effective modeling of non-Fickian contaminant transport in groundwater flow. Concentration distributions observed in field and laboratory experiments are in general non-Fickian and show spatial and temporal tailing [e.g., *Silliman and Simpson*, 1987; *Levy and Berkowitz*, 2003; *Berkowitz et al.*, 2006].

[4] Both microscopic and macroscopic models of anomalous transport have been proposed to account for the non-Fickian nature of transport in heterogeneous environments. Microscopic models, which include continuous time random walk (CTRW) [e.g., *Berkowitz et al.*, 2006], multirate mass transfer (MRMT) [e.g., *Haggerty and Gorelick*, 1995; *Carrera et al.*, 1998; *Haggerty et al.*, 2000] and fractional diffusion models [e.g., *Schumer et al.*, 2003], question the validity of Fickian diffusion by re-examining Brownian particle dynamics as a foundation of diffusion in heterogeneous environments. In particular, CTRW describes diffusive transport as a random walk in both space and time [e.g., *Berkowitz and Scher*, 1997; *Metzler and Klafter*, 2000; *Berkowitz et al.*, 2006]; MRMT and time-fractional advection-diffusion models can be formally seen as subsets of CTRW [e.g., *Dentz and Berkowitz*, 2003; *Berkowitz et al.*, 2006]. In these modeling frameworks, the impact of subscale heterogeneity on macroscale transport is quantified in terms of a generally unknown distribution of typical transport times.

[5] Macroscopic models of non-Fickian transport in heterogeneous media generally result from averaging the Fickian-diffusion-based local scale advection-dispersion equation [e.g., *Koch and Shaqfeh*, 1992; *Neuman*, 1993; *Cushman*, 1997; *Dykhne et al.*, 2005]. Such approaches typically require closure approximations for the average concentration.

[6] We present an alternative macroscopic model of non-Fickian anomalous diffusion that explicitly accounts for the finite (and often quite large) support volume of a typical transport experiment in porous media. This is in contrast with standard diffusion models of contaminant migration, which disregard inertia effects caused by both the finite support volume and the effect of subscale heterogeneities.

[7] Delayed diffusion models have been proposed to account for such inertia effects in reaction-diffusion systems [see, e.g., *Horsthemke*, 1999; *Fort and Méndez*, 2002, and references therein]. These and similar models postulate the existence of a time delay (finite relaxation or response time) τ_d between the cause and effect in Fick's law,

$$\mathbf{J}_d(\mathbf{x}, t) = -D\nabla c(\mathbf{x}, t - \tau_d), \tag{1}$$

where D denotes the diffusion coefficient and $c(\mathbf{x}, t)$ is the solute concentration. The standard Fick's law $\mathbf{J}_d(\mathbf{x}, t) = -D\nabla c(\mathbf{x}, t)$ can be viewed as an approximation that is valid for delay times τ_d that are much smaller than the observation time scale.

[8] Heterogeneity can also lead to a delay in the advective flux, which is expressed by

$$\mathbf{J}_a(\mathbf{x}, t) = \mathbf{u}c(\mathbf{x}, t - \tau_a), \tag{2}$$

where $\mathbf{u}(\mathbf{x}, t)$ is the macroscopic fluid velocity. A time delay τ_a in the advective flux can be caused by variable porosity and adsorption properties of the porous medium, as well as

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0094-8276/06/2006GL027054$05.00

by trapping mechanisms including diffusive or first-order particle trapping in low flow zones and trapping in closed streamlines [e.g., *Frisch*, 1995; *Isichenko*, 1992]. Since the mechanisms leading to delay in the advective and diffusive fluxes can be different, in general $\tau_a \neq \tau_d$. If the medium can be characterized by a single length scale, effective transport can be described by a single delay time model in which the delay time should be related to the finite "solute particle" velocity and Lagrangian correlation time.

[9] In this letter, we explore the concept of delayed advection and diffusion for the effective modeling of non-Fickian transport in heterogeneous media, which are characterized by a spectrum of typical heterogeneity scales. We show that non-Fickian transport can be a consequence of a distribution of delay times, which reflects the evolving scales of the medium heterogeneities. Finally, we establish a connection between the proposed model and the CTRW and fractional diffusion models.

2. Delayed Transport

[10] In the absence of sources and sinks, mass balance requires that $\partial c(\mathbf{x}, t)/\partial t = \nabla \cdot \mathbf{J}_a(\mathbf{x}, t) + \nabla \cdot \mathbf{J}_d(\mathbf{x}, t)$. Substitution of (1) and (2) into this equation yields a delayed advection-diffusion equation

$$\frac{\partial c(\mathbf{x},t)}{\partial t} + \nabla \cdot [\mathbf{u}c(\mathbf{x}, t-\tau_a)] - D\nabla^2 c(\mathbf{x}, t-\tau_d) = 0. \qquad (3)$$

It is immediately clear that transport is Fickian as soon as the transport time is larger than the delay scales. In fact, a direct solution of (3) demonstrates that delayed advection-diffusion with a single time delay cannot account for anomalous transport.

[11] We argue that the key to understanding anomalous transport within this framework is to realize that (i) anomalous diffusion has been observed exclusively in heterogeneous systems [e.g., *Bouchaud and Georges*, 1990; *Cushman*, 1997], and (ii) the medium heterogeneity gives rise to a distribution of typical delay times. On this basis, we propose a natural generalization of the delay transport model (3) that replaces a single delay time with a delay time distribution by introducing advection and diffusion kernels $\boldsymbol{\nu}(t)$ and $\mathcal{D}(t)$, such that

$$\frac{\partial c(\mathbf{x},t)}{\partial t} + \nabla \cdot \int_0^t [\boldsymbol{\nu}(t-t') - \mathcal{D}(t-t')\nabla] c(\mathbf{x}, t')\mathrm{d}t' = 0. \qquad (4)$$

The single delay model (3) is recovered for the Dirac-delta kernels $\boldsymbol{\nu}(t) = \mathbf{u}\tau_a^{-1}\delta(t/\tau_a - 1)$ and $\mathcal{D}(t) = D\tau_d^{-1}\delta(t/\tau_d - 1)$. The diffusion and advection kernels are written as

$$\mathcal{D}(t) = D\mathcal{P}_d(t) \qquad \boldsymbol{\nu}(t) = \mathbf{u}\mathcal{P}_a(t), \qquad (5)$$

where D represents a diffusion scale and $\mathbf{u}$ is a drift coefficient. The $\mathcal{P}_d(t)$ and $\mathcal{P}_a(t)$ denote the distribution densities of typical diffusion and advection time scales, respectively, which are normalized according to $\mathcal{P}_d(0) = \tau_d^{-1}$ and $\mathcal{P}_a(0) = \tau_a^{-1}$; and τ_d and τ_a denote characteristic diffusion and advection time scales. A typical advection length scale is given by $l = \|\mathbf{u}\|\tau_a$ and a (microscopic) Peclet number is defined by $Pe = \|\mathbf{u}\|l/D$.

[12] Note that the proposed time-delay models can be readily generalized to account for the anisotropy of an ambient environment by introducing a directional dependence of delay. Then, the d-dimensional Fick's law with a delay time distribution takes the form

$$J_{d_i}(\mathbf{x}, t) = -\int \mathcal{D}_{ij}(t-t')\frac{\partial c(\mathbf{x}, t')}{\partial x_j}\mathrm{d}t', \qquad (6)$$

for $i, j = 1, \ldots, d$.

3. Resident Concentration and Effective Dispersion

[13] Consider the response of a one-dimensional system of infinite extent to an instantaneous point source $c(x_1, 0) = \delta(x_1)$ at time $t = 0$, and prescribe $c(x_1 = \pm\infty, t) = 0$. A solution of the delay transport equation (4) in Laplace space is

$$\hat{c}(x_1, s) = \frac{\exp\left[-\frac{Pe\hat{\mathcal{P}}_a}{2\hat{\mathcal{P}}_d}\left(\frac{|x_1|}{l}\sqrt{1+4\frac{s\tau_a\hat{\mathcal{P}}_d}{Pe\hat{\mathcal{P}}_a^2}} - \frac{x_1}{l}\right)\right]}{u\sqrt{1+4\frac{s\tau_a\hat{\mathcal{P}}_d}{Pe\hat{\mathcal{P}}_a^2}}}. \qquad (7)$$

The Laplace transform is defined as by *Abramowitz and Stegun* [1972], s denotes the Laplace variable, and Laplace transformed quantities are marked by the hat.

[14] The center of mass of the solute distribution $c(x_1, t)$ is defined by

$$m(t) = \int_{-\infty}^{\infty} x_1 c(x_1, t)\mathrm{d}x_1, \qquad (8)$$

the dispersion of $c(x_1, t)$ is quantified by the variance

$$\kappa(t) = \int_{-\infty}^{\infty} x_1^2 c(x_1, t)\mathrm{d}x_1 - \left[\int_{-\infty}^{\infty} x_1 c(x_1, t)\mathrm{d}x_1\right]^2. \qquad (9)$$

These quantities are readily obtained by multiplying (4) with x_1 and x_1^2, respectively, and integrating over space,

$$m(t) = \int_0^t \int_0^{t'} \nu(t'')\mathrm{d}t'\mathrm{d}t'', \qquad \kappa(t) = \kappa_d(t) + \kappa_a(t). \qquad (10)$$

The contributions κ_d and κ_a due to diffusion and advection delay, respectively, are given by

$$\kappa_d(t) = 2\int_0^t \mathrm{d}t' \int_0^{t'} \mathcal{D}(t'')\mathrm{d}t'' \qquad (11)$$

$$\kappa_a(t) = \int_0^t \int_0^{t'} \nu(t'-t'')[2m(t'') - m(t)]\mathrm{d}t'\mathrm{d}t''. \qquad (12)$$

The latter can be attributed to the stretching of $c(x_1, t)$ due to retardation effects along the particle trajectory.

4. Solute Arrival Time Distribution

[15] The distribution of solute arrival times at a control plane at location x_1 is defined by the flux of $c(x_1, t)$,

$$f(x_1,t) = \int_0^t \left[\nu(t-t') - \mathcal{D}(t-t')\frac{\partial}{\partial x_1}\right] c(x_1,t')\mathrm{d}t', \qquad (13)$$

which, for $c(x_1, 0) = 0$, satisfies the transport equation

$$\frac{\partial f}{\partial t} + \int_0^t \left[\nu(t-t') - \mathcal{D}(t-t')\frac{\partial}{\partial x_1}\right] \frac{\partial f(x_1,t')}{\partial x_1}\mathrm{d}t' = 0, \qquad (14)$$

as can be verified by inspection. For a semi-infinite domain ($x_1 \geq 0$), and subject to the boundary conditions $f(0, t) = \delta(t)$ and $f(\infty, t) = 0$, and the initial condition $f(x_1, 0) = 0$, a solution of (14) in Laplace space is

$$\hat{f} = \exp\left[-\frac{Pe\hat{\mathcal{P}}_a}{2\hat{\mathcal{P}}_d}\left(\frac{|x_1|}{l}\sqrt{1+4\frac{s\tau_a\hat{\mathcal{P}}_d}{Pe\hat{\mathcal{P}}_a^2}} - \frac{x_1}{l}\right)\right]. \qquad (15)$$

The mean arrival time $T(x_1)$ is defined by

$$T(x_1) = \int_0^\infty tf(x_1,t)\mathrm{d}t = -\frac{\partial\hat{f}(x_1,s)}{\partial s}\bigg|_{s=0}. \qquad (16)$$

The latter equality can be shown by using the definition of the Laplace transform. The mean arrival time is given by $T = T_d + T_a$, where

$$T_d = \frac{x_1}{l}\frac{s\tau_a}{\hat{\mathcal{P}}_a}\frac{\mathrm{d}\log\hat{\mathcal{P}}_d}{\mathrm{d}s}\bigg|_{s=0}, \qquad (17)$$

and

$$T_a = \frac{x_1}{l}\frac{\tau_a}{\hat{\mathcal{P}}_a}\left(1 - 2\frac{\mathrm{d}\log\hat{\mathcal{P}}_a}{\mathrm{d}s}\right)\bigg|_{s=0}, \qquad (18)$$

correspond to the diffusive and advective fluxes, respectively. In the absence of advective delay, the latter reduces to $T_a = x_1/u$. Note that at large times, i.e., for $s\tau \ll 1$ with $\tau = \max(\tau_a, \tau_d)$, advective delay can lead to an increase of the mean arrival time.

5. Delayed Diffusion in Quasi-Fractal Media

[16] Of particular interest is transport in random environments with a continuous hierarchy of heterogeneity scales. These environments are characterized by long-range spatial correlations. In such media one often observes super-diffusion [e.g., *Bouchaud and Georges*, 1990; *Dykhne et al.*, 2005], which manifests itself by a power-law growth of the variance

$$\kappa(t) \propto t^{1+\alpha}, \qquad 0 < \alpha < 1. \qquad (19)$$

Such a behavior is typically observed in a certain time regime $\tau_1 \ll t \ll \tau_2$ (where τ_1 and τ_2 are related to the smallest and largest heterogeneity length scales) and can be modeled by a diffusion kernel given in terms of a truncated power-law distribution

$$\mathcal{P}_d(t) = \tau_d^{-1}\frac{\exp(-t/\tau_2)}{(1+t/\tau_1)^{1-\alpha}}, \qquad (20)$$

whose Laplace transform is $\hat{\mathcal{P}}_d = \tau_1/\tau_d(s\tau_1 + \epsilon)^{-\alpha}\exp(s\tau_1 + \epsilon)\Gamma(\alpha, s\tau_1 + \epsilon)$. Here $\epsilon \equiv \tau_1/\tau_2$, $\Gamma(\alpha, s)$ is the incomplete Gamma function [*Abramowitz and Stegun*, 1972], and τ_d is the normalization scale.

[17] To eliminate the effects of trapping and retardation, we focus on systems that exhibit time delay in the diffusive flux only, i.e., set $\mathcal{P}_a = \tau_a^{-1}\delta(t/\tau_a)$. We define a typical advection length by $l = u\tau_1$ and the Peclet number by $Pe = ul/D$. Substituting $\hat{\mathcal{P}}_d$ and $\hat{\mathcal{P}}_a = 1$ into (15) and expanding the resulting expression into a Taylor series for $\tau_1^{-1} \ll s \ll \tau_2^{-1}$, we obtain

$$\hat{f} = 1 - \frac{x_1}{l}s\tau_1 + \frac{x_1}{l}\frac{\tau_1}{\tau_d}\frac{\Gamma(1+\alpha)(s\tau_1)^{2-\alpha}}{Pe} + \mathcal{O}(s^2). \qquad (21)$$

Using a Tauberian theorem to invert this expression, we find the power-law behavior

$$f(t) \propto (t/\tau_1)^{\alpha-3} \qquad (22)$$

in the time regime $\tau_1 \ll t \ll \tau_2$. The resident concentration and the solute arrival time distribution—computed via Laplace inversion of (7) and (15) with $\mathcal{P}_d$ given by (20)—are displayed in Figure 1 for $\alpha = 0.5$. Both exhibit clear non-Fickian behavior.

6. Relation to Alternative Models

[18] We conclude our analysis of delay mechanisms of non-Fickian transport by analyzing the relationships between the proposed model and fractional advection-diffusion equations as well as CTRW. We focus on delayed diffusion only by ignoring a possible time delay in the advective flux.

[19] The connection to time fractional transport models is obvious. Indeed, for $\tau_1 \ll t \ll \tau_2$ and $\alpha > 0$, the transport equation (4) together with the truncated power-law diffusion kernel can be written as a time-fractional advection-diffusion equation

$$\frac{\partial c}{\partial t} + u \cdot \nabla c = D\tau_1^{-\alpha}\Gamma(\alpha)\nabla^2 D_t^{(-\alpha)}c, \qquad (23)$$

where $D_t^{(-\alpha)}f(t) = \Gamma(\alpha)^{-1}\int_0^t f(t')(t-t')^{\alpha-1}\mathrm{d}t'$ is the so-called Riemann-Liouville fractional integral [e.g., *Metzler and Klafter*, 2000] and Γ is the Gamma function [e.g., *Abramowitz and Stegun*, 1972]. The equivalence between (23) and (4) with (20) can be shown by Laplace transforming the respective equations.

[20] A fully coupled CTRW results in the integro partial differential equation [e.g., *Berkowitz et al.*, 2006]

$$\frac{\partial c}{\partial t} + \int_0^t \left[\mathbf{v}_\psi(t-t')\cdot\nabla c - \nabla\mathbf{D}_\psi(t-t')\nabla c\right]\mathrm{d}t' = 0, \qquad (24)$$

where the Laplace transforms of the advection and diffusion kernels are defined as $\hat{v}_{\psi_i} = s\int x_i\hat{\psi}(1-\hat{\psi}_m)^{-1}\,\mathrm{d}\mathbf{x}$ and $\hat{D}_{\psi_{ij}} =$

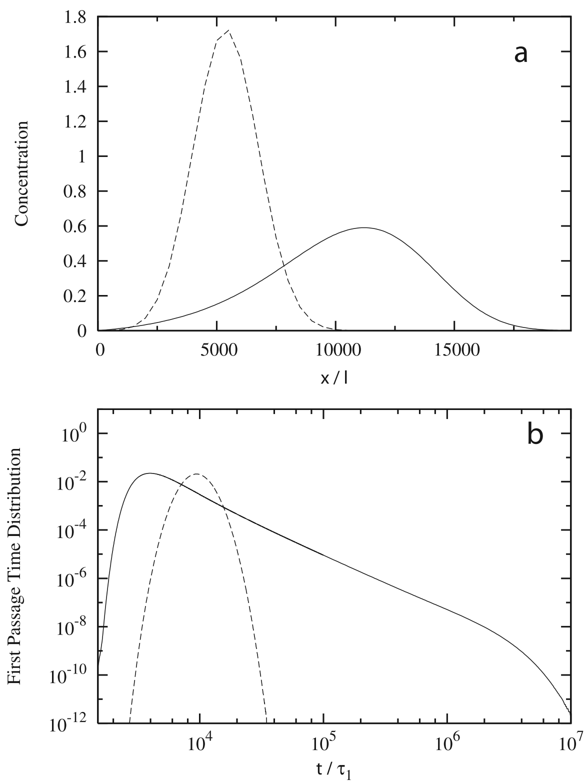

Figure 1. (a) Concentration profiles and (b) first passage time distributions computed by numerical inversion of (7) and (15) with the delay time distribution (20), respectively, for $Pe = 10^{-2}\tau_1/\tau_d$, $\alpha = 0.5$ and $\epsilon \equiv \tau_1/\tau_2 = 10^{-6}$ (solid lines) and 10^{-1} (dashed lines). The wider the range of delay times (the smaller the parameter ϵ), the more pronounced are the non-Fickian transport features. Note in Figure 1a that a front builds up for the spatial distribution in case of a broader distribution of delay times. This is also reflected in Figure 1b where the peak arrives faster for a broader delay time distribution while in the simulated case the mean arrival time is the same for both cases.

$s \int x_i x_j \hat{\psi}(1 - \hat{\psi}_m)^{-1}\, d\mathbf{x}$, respectively. Here $i, j = 1, \ldots, d$ with d denoting spatial dimension, $\hat{\psi}(\mathbf{x}, s)$ is the Laplace transform of the coupled transition length and time distribution $\psi(\mathbf{x}, t)$, and $\hat{\psi}_m(s)$ is the Laplace transform of the marginal transition time distribution

$$\psi_m(t) \equiv \int \psi(\mathbf{x}, t) \mathrm{d}\mathbf{x}. \quad (25)$$

The proposed delayed diffusion model and CTRW are equivalent if

$$\hat{v}_{\psi_1} = \|\mathbf{u}\|, \qquad \hat{v}_{\psi_i} = 0, \quad (26)$$

where $i > 1$. These conditions yields a hitherto unexplored constraint for the coupled transition length and time distribution

$$\int \mathbf{x}\psi(\mathbf{x}, t)\mathrm{d}\mathbf{x} = \mathbf{u} - \mathbf{u}\int_0^t \psi_m(t')\mathrm{d}t'. \quad (27)$$

For the decoupled CTRW (i.e., $\psi(\mathbf{x}, t) = p(\mathbf{x})\psi_m(t)$,), which includes linear MRMT models [e.g., *Dentz and Berkowitz*, 2003], this constraint is fulfilled for an exponential transition time distribution, $\psi_m(t) = \bar{x}_1 \exp(-\|\mathbf{u}\| t/\bar{x}_1)/\|\mathbf{u}\|$ where $\bar{x}_1 \equiv \int x_1 p(\mathbf{x})\mathrm{d}\mathbf{x}$. Nevertheless, the exponential transition time distribution describes Fickian transport [e.g., *Berkowitz et al.*, 2006].

[21] Despite the similarity between the time-delay model of non-Fickian diffusion and the approaches described above, fundamental differences exist. Specifically, the time-delay model accounts for the existence of genuine sub- and super-diffusive fluxes as the sole property of diffusion, i.e., in the absence of other transport mechanisms. This is in contrast to the decoupled CTRW framework and MRMT models which, in the absence of advection, describe sub-diffusive behavior that reflects the combined effects of retardation and diffusion of particles. In these models, super-diffusion occurs in the presence of advection [e.g., *Metzler and Klafter*, 2000; *Berkowitz et al.*, 2006], which gives rise to a dispersion term of the form κ^a in (11).

7. Concluding Remarks

[22] We presented a macroscopic model to account for non-Fickian transport in heterogeneous media based on delay mechanisms for the advective and diffusive solute fluxes. The phenomenological motivation for such a modeling approach is the observation that a macroscopic effective transport framework must account for inertia effects that are caused by the coarse resolution (i.e., large support volume) in conjunction with unresolved subscale heterogeneities. The model generalizes the well-established delay diffusion model by introducing a distribution of delay times reflecting the spatial scales of heterogeneity. The delay advection-dispersion equation (4) complements existing transport frameworks such as CTRW and MRMT and fractional advection dispersion equations, which model non-Fickian solute transport in heterogeneous media.

[23] **Acknowledgments.** M.D. acknowledges the support of the program 'Ramon y Cajal' of the Spanish Ministry of Education and Science (MEC), ENRESA, the European Union IP FUNMIG (contract 516514) and the MEC project 'MODEST' (project CGL-2005-05171). The work of D.M.T. was supported in part by the U.S. Department of Energy under the DOE/BES Program in the Applied Mathematical Sciences, contract KC-07-01-01, and in part by the LDRD Program at Los Alamos National Laboratory. This work made use of shared facilities supported by SAHRA (Sustainability of semi-Arid Hydrology and Riparian Areas) under the STC Program of the National Science Foundation under agreement EAR-9876800.

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