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Continuous time random walks for non-local radial solute transport

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ABSTRACT

This study formulates and analyzes continuous time random walk (CTRW) models in radial flow geometries for the quantification of non-local solute transport induced by heterogeneous flow distributions and by mobile–immobile mass transfer processes. To this end we derive a general CTRW framework in radial coordinates starting from the random walk equations for radial particle positions and times. The particle density, or solute concentration is governed by a non-local radial advection–dispersion equation (ADE). Unlike in CTRWs for uniform flow scenarios, particle transition times here depend on the radial particle position, which renders the CTRW non-stationary. As a consequence, the memory kernel characterizing the non-local ADE, is radially dependent. Based on this general formulation, we derive radial CTRW implementations that (i) emulate non-local radial transport due to heterogeneous advection, (ii) model multirate mass transfer (MRMT) between mobile and immobile continua, and (iii) quantify both heterogeneous advection in a mobile region and mass transfer between mobile and immobile regions. The expected solute breakthrough behavior is studied using numerical random walk particle tracking simulations. This behavior is analyzed by explicit analytical expressions for the asymptotic solute breakthrough curves. We observe clear power-law tails of the solute breakthrough for broad (power-law) distributions of particle transit times (heterogeneous advection) and particle trapping times (MRMT model). The combined model displays two distinct time regimes. An intermediate regime, in which the solute breakthrough is dominated by the particle transit times in the mobile zones, and a late time regime that is governed by the distribution of particle trapping times in immobile zones. These radial CTRW formulations allow for the identification of heterogeneous advection and mobile-immobile processes as drivers of anomalous transport, under conditions relevant for field tracer tests.

1. Introduction

Solute transport in heterogeneous porous media displays behaviors that cannot be captured by transport models based on an equivalent advection dispersion equation (ADE) parameterized by (constant) effective transport parameters. Such behaviors range from the non-linear evolution of solute dispersion to power-law tails in solute breakthrough curves [1,2]. The last three decades have seen intense research to quantify these behaviors in terms of effective transport models that can be obtained by moment equation approaches [3], and projector formalisms [4], for example, and include time and space fractional ADEs [5,6], multirate mass transfer (MRMT) models [7,8], as well as continuous time random walks [9,10], see also the reviews in [2,11,12].

In this paper, we focus on the CTRW approach to modeling non-Fickian solute transport in heterogeneous media. Classical random walks model particle movements by using variable spatial steps which are taken within constant time increments at equidistant times [13,14]. A CTRW, in contrast, models particle movements in a heterogeneous medium effectively as a random walk in which both space and time increments are variable. The spatial transitions may reflect the geometry of the underlying medium and flow heterogeneity, while particle transition and waiting times reflect persistent particle velocities over given transition distances, or particle retention due to adsorption or diffusion into immobile zones, for example [9,15–18]. The medium heterogeneity is mapped into the probability distribution density (PDF) of characteristic particle transition times. The evolution of the particle density, or, equivalently the solute concentration is governed by a temporally non-local ADE whose memory kernel is given in terms of the PDF of transition times [10,19].

The MRMT approach is phenomenologically similar to the CTRW modeling framework as it models the impact of medium heterogeneity on large scale transport through a distribution of typical solute retention times in immobile regions. In fact, it can be shown [20,21] that one model can under certain conditions be

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mapped onto the other. The latter amounts essentially to identifying the relation between the PDF of particle transition times and particle retention times in immobile regions [22–24].

As pointed out above, the CTRW model is a random walk approach in that particle movements are governed by random walk equations for the space and time coordinates. Therefore the solution of CTRW and equivalent models is directly accessible to numerical solution through random walk particle tracking simulations [10]. This provides an avenue for the efficient simulation of transport in the presence of mobile–immobile processes [23–25], for example, and for temporally non-local transport in general [26].

Many formulations of the above models are for transport situations under uniform mean flow. Thus, for the interpretation of tracer tests under forced conditions they are only of limited applicability because the non-stationarity of the underlying flow field is not accounted for. Haggerty et al. [27] used a Eulerian radial MRMT implementation to interpret radial single-well injection-withdrawal (SWIW) tracer tests in fractured dolomite. Le Borgne and Gouze [25] used a CTRW based random walk implementation of MRMT to model tracer breakthrough data from SWIW tracer tests. Benson et al. [6] developed a fractional-order dispersion model in radial coordinates to model tracer tests under forced conditions. A general issue when interpreting field tracer data is to decipher the origin of the observed non-local transport behavior, which may range from mobile–immobile diffusive mass transfer processes to highly heterogeneous advective transport [28,29]. In the latter case, non-Fickian transport may be caused by a broad distribution of flow and transport velocities; the distribution of particle transit times depends on the flow rate and heterogeneity in the flow properties. In the former case, anomalous transport features are due to mass transfer between mobile and immobile zones; particle transition times may depend on the retention properties and geometries of the immobile regions. Testing these different hypothesis requires non-local transport models, that integrate both diffusive and advective mass transfer processes in non-uniform flow conditions.

In this paper, we develop a general CTRW approach that allows for the modeling of non-local solute transport under radial conditions, which are relevant for field tracer experiments under forced conditions. The derivation from the space–time random walk equations gives directly the particle tracking method for its numerical solution. We present three non-local CTRW based radial transport implementations, for the modeling of heterogeneous advection, mobile–immobile mass transfer (MRMT), and the combination of both. To this end we review in Section 2 briefly the random walk formulation of general radial advective–dispersive transport. Section 3 then derives the general radial CTRW framework and defines the specific CTRW models. The model breakthrough curves then are analyzed in Section 4 using numerical random walk simulations and explicit analytical expressions for the asymptotic breakthrough behavior developed in Appendix B. In particular, we discuss the expected differences in non-Fickian transport behaviors induced by purely advective processes, purely diffusive processes, and the combination of these processes.

2. Radial random walks

The classical advection–dispersion equation (ADE) for the solute concentration $c(r,t)$ in radial coordinates can be written as

$$\frac{\partial c(r,t)}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r} v(r) r c(r,t) - \frac{1}{r}\frac{\partial}{\partial r} r D(r) \frac{\partial c}{\partial r} = 0, \tag{1}$$

where $v(r)$ and $D(r)$ are the radially dependent transport velocity and dispersion coefficient; r denotes the radial distance, t denotes time. We set the constant porosity equal to one, which is equivalent to rescaling time. The equivalent random walk particle tracking formulation is obtained by rewriting (1) in mass conservative form. Therefore, we define the conserved radial concentration as

$$p(r,t) = 2\pi r c(r,t). \tag{2}$$

Notice that $p(r,t)$ denotes the concentration per unit radial distance. Inserting the latter into (1) and rearranging terms we obtain the radial Fokker–Planck equation

$$\frac{\partial p(r,t)}{\partial t} + \frac{\partial}{\partial r}\left[v(r) + \frac{D(r)}{r} + D'(r)\right] p(r,t) - \frac{\partial^2}{\partial r^2} D(r) p(r,t) = 0, \tag{3}$$

where $D'(r)$ denotes the derivative of $D(r)$ with respect to r. The equivalent Langevin equation is given by [30]

$$\frac{dr(t)}{dt} = v[r(t)] + \frac{D[r(t)]}{r(t)} + D'[r(t)] + \sqrt{2D[r(t)]}\xi_r(t), \tag{4}$$

where $\xi_r(t)$ is a Gaussian white noise of zero mean and the correlation function $\langle \xi_r(t)\xi_r(t')\rangle = \delta(t-t')$. Here and in the following, we employ the Ito interpretation [30] of the Langevin equation (4). The particle density is given in terms of the radial trajectories as $p(r,t) = \langle \delta[r - r(t)]\rangle$, and by virtue of (2), we obtain for the concentration distribution

$$c(r,t) = \frac{\langle \delta[r - r(t)]\rangle}{2\pi r}. \tag{5}$$

In the following, we will consider the case of [31]

$$v(r) = \frac{k_v}{r}, \quad D(r) = \frac{\alpha k_v}{r}, \tag{6}$$

where α is dispersivity, and $k_v = Q/(2\pi)$ with Q the flow rate. Notice that more general radial dependences of flow velocity and dispersion can be considered within the approaches developed in the following. Here, we focus on the choice (6). With these definitions, the Langevin equation (4) simplifies to

$$\frac{dr(t)}{dt} = \frac{k_v}{r(t)} + \sqrt{\frac{2\alpha k_v}{r(t)}}\xi_r(t). \tag{7}$$

The temporally discretized version of the radial Langevin equation is given by

$$r_{n+1} = r_n + \frac{k_v \Delta t}{r_n} + \sqrt{\frac{2\alpha k_v \Delta t}{r_n}}\xi_n, \tag{8}$$

where $r_n = r(t_n)$, $t_n = n\Delta t$, and ξ_n is a Gaussian random variable with zero mean and unit variance.

3. Radial continuous time random walks

The radial random walk particle tracking formulations developed in the following are based on the generalization of the radial random walk process (8) in terms of the continuous time random walk

$$r_{n+1} = r_n + \ell + \sqrt{2\alpha\ell}\xi_n \tag{9a}$$

$$t_{n+1} = t_n + \tau_n(r), \tag{9b}$$

where ℓ is a constant transition length, and $\tau(r)$ a radially dependent, independently distributed random transition time with the probability density function (PDF) $\psi_\tau(\tau, r)$. Notice that the classical formulation (8) is obtained by setting $\tau_n(r) = \ell r/k_v$ in (9). The distribution of the spatial transition lengths $\Delta r = \ell + \sqrt{2\alpha\ell}\xi_n$ is denoted by $\psi_r(\Delta r)$. The mean and mean square displacements are given by $\langle \Delta r\rangle = \ell$ and $\langle \Delta r^2\rangle = 2\alpha\ell$, where we disregard contributions of order ℓ^2. Notice that the transition length ℓ are chosen such that $\ell \ll \alpha$.