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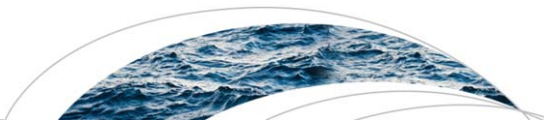
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TECHNICAL REPORTS: METHODS

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Key Points:

- New multidimensional time-domain random walk (TDRW) algorithms are derived
- The stochastic travel time of particles in 2-D and 3-D is calculated in one step
- The approach may be applicable to a variety of mathematical transport models

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From analytical solutions of solute transport equations to multidimensional time-domain random walk (TDRW) algorithms

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Abstract In this study, new multi-dimensional time-domain random walk (TDRW) algorithms are derived from approximate one-dimensional (1-D), two-dimensional (2-D), and three-dimensional (3-D) analytical solutions of the advection-dispersion equation and from exact 1-D, 2-D, and 3-D analytical solutions of the pure-diffusion equation. These algorithms enable the calculation of both the time required for a particle to travel a specified distance in a homogeneous medium and the mass recovery at the observation point, which may be incomplete due to 2-D or 3-D transverse dispersion or diffusion. The method is extended to heterogeneous media, represented as a piecewise collection of homogeneous media. The particle motion is then decomposed along a series of intermediate checkpoints located on the medium interface boundaries. The accuracy of the multi-dimensional TDRW method is verified against (i) exact analytical solutions of solute transport in homogeneous media and (ii) finite-difference simulations in a synthetic 2-D heterogeneous medium of simple geometry. The results demonstrate that the method is ideally suited to purely diffusive transport and to advection-dispersion transport problems dominated by advection. Conversely, the method is not recommended for highly dispersive transport problems because the accuracy of the advection-dispersion TDRW algorithms degrades rapidly for a low Péclet number, consistent with the accuracy limit of the approximate analytical solutions. The proposed approach provides a unified methodology for deriving multi-dimensional time-domain particle equations and may be applicable to other mathematical transport models, provided that appropriate analytical solutions are available.

1. Introduction

Lagrangian (particle-tracking) methods are among the most popular techniques for simulating solute transport in porous media. The primary advantage of these methods is the absence of numerical dispersion compared with other alternatives such as Eulerian or mixed Eulerian-Lagrangian methods. For this reason, Lagrangian methods are well suited to advection-dominated transport problems. A central feature of Lagrangian methods is the representation of the solute mass by a large number of particles. Depending on how these particles move, two distinct approaches may be used: (i) space-based particle-tracking methods and (ii) time-domain particle methods. In the first approach, which may be regarded as “traditional,” the particles are moved in fixed time steps. The particle displacement during a given time step includes a deterministic displacement along flow-path lines to simulate advection and may also include an additional stochastic displacement to simulate dispersion (or diffusion), as in the well-known random walk (RW) method. The reader is referred to *Delay et al.* [2005] and *Salamon et al.* [2006] for comprehensive reviews of space-based particle-tracking methods, including theoretical, conceptual, and numerical developments in the field of subsurface hydrology since the early works of *Ahlstrom et al.* [1977] and *Prickett et al.* [1981]. Additional recent publications of relevance include *Bechtold et al.* [2011] and *Lejay and Pichot* [2012].

As an alternative to space-based algorithms, various time-domain particle methods have been developed since the early 1990s [*Yamashita and Kimura*, 1990; *Banton et al.*, 1997; *Delay and Bodin*, 2001; *James and Chrysikopoulos*, 2001; *Delay et al.*, 2002; *Reimus and James*, 2002; *Bodin et al.*, 2003; *Bodin et al.*, 2007; *Delay et al.*, 2008; *Painter et al.*, 2008; *Dentz et al.*, 2012]. In contrast to space-based particle tracking, time-domain algorithms first establish fixed checkpoints along the particle transport path and then directly calculate the time required for the particle to move from one checkpoint to the next. This time may be deterministic, in the case of purely advective transport, or stochastic, if dispersion (and/or diffusion) is considered.

The presently available time-domain particle methods have been developed based on different conceptual and theoretical backgrounds, which are briefly reviewed below. *Yamashita and Kimura* [1990] examined the problem of solute transport in a single fracture embedded in a porous rock matrix. Assuming one-dimensional advective transport (neglecting dispersion) in the fracture and one-dimensional diffusive transport (normal to the fracture) into the matrix, the authors derived an expression for the cumulative probability density function of the particle travel times in the fracture-matrix system from an analytical solution of the corresponding transport equations. Applying the rejection method to this probability density function allows for single-step computations of the stochastic travel time of particles through a fracture-matrix system of a given length. *Banton et al.* [1997] were the first to develop a time-domain particle method for simulating advection-dispersion transport problems. Considering the well-known log-normal shape of solute breakthrough curves in advection-dominated transport problems, the authors proposed a stochastic algorithm to compute the logarithms of particle travel times that consistently obey a Gaussian distribution. The algorithm, termed the time-domain random walk (TDRW), was inspired by the RW method that simulates Gaussian spreading in the space of solute particles subject to advection and dispersion. The TDRW method was further developed to address diffusion into nonflowing pore spaces adjacent to advective-dispersive 1-D flow channels (rock matrix and/or stagnant zones in fracture planes [*Delay and Bodin*, 2001; *Bodin et al.*, 2007]) as well as sorption with first-order kinetics [*Delay et al.*, 2008]. Starting from a different theoretical framework based on a Monte Carlo sampling of particle transit times, *Painter et al.* [2008] proposed an alternative TDRW method capable of accommodating a broad range of retention models and first-order decay chains. As noted by the various aforementioned authors, the two major advantages of the TDRW approach over the RW method are (i) a higher computational efficiency (only one calculation step is required to move each particle between two distant locations, whereas the RW method requires several particle jumps to accurately simulate the dispersion process) and (ii) a more facile management of particle transitions through media with sharp discontinuities in their advection-dispersion properties, as the particle checkpoints may be conveniently placed at the interfaces at which such discontinuities arise. Thus far, the primary limitation of TDRW methods is that they are suited only to 1-D transport problems because it appears that it is very difficult to incorporate transverse dispersion into the existing algorithms. Because of this limitation, TDRW methods have been primarily applied to fractured media conceptualized as networks of interconnected 1-D transport pathways [e.g., *Bodin et al.*, 2003; *Painter et al.*, 2008].

Along with methods for advection and dispersion, various algorithms have been specifically developed for purely diffusive transport. *James and Chrysikopoulos* [2001] proposed 1-D, 2-D, and 3-D algorithms based on empirical expressions of the mean and standard deviation of travel-time probability densities derived from traditional RW simulations. *Reimus and James* [2002] proposed a more exact algorithm based on a truncated infinite-series solution of the one-dimensional diffusion equation and extended their algorithm to account for both advection and diffusion (dispersion), but the resulting method appears to be less numerically efficient than the TDRW method of *Banton et al.* [1997] because it requires more computational steps. Following a different approach based on a finite-volume formulation of the diffusion equation, *Delay et al.* [2002] developed a time-domain particle method to simulate diffusion in a 2-D heterogeneous porous medium discretized as a regular grid. *Dentz et al.* [2012] formalized this method within the continuous-time random walk (CTRW) framework and further developed it to handle spatially variable multirate mass-transfer properties.

The above literature review shows that none of the currently available time-domain particle methods are sufficiently flexible to address both multidimensional advection-dispersion transport problems and purely diffusive transport problems. The purpose of this technical note is to propose such a unified approach. This note is structured as follows. Following this introduction, section 2 presents the general methodology for the derivation of new TDRW equations from suitable analytical solutions of solute transport equations. In section 3, we elaborate the method for 2-D advective-dispersive transport and then extend the approach to multidimensional advective-dispersive and purely diffusive transport. In section 4, the accuracy of the new TDRW algorithms is verified against exact analytical solutions of solute transport in homogeneous media. Section 5 discusses the extension of the method to heterogeneous media, and the conclusions are presented in section 6.

2. Methodological Approach

The proposed TDRW method does not attempt to solve the partial differential equation and associated initial and boundary conditions describing the transport problem of interest. Instead, the strategy is to

simulate (mimic) the concentration-time curve at the observation point as given by the corresponding analytical solution, if existing. For this purpose, the proposed approach is inspired by the rejection method initially suggested by *Yamashita and Kimura* [1990] and subsequently used by a number of authors [e.g., *Tsang and Tsang*, 2001; *Zhang et al.*, 2006; *Zafarani and Detwiler*, 2013]. We generalize this method to multidimensional purely diffusive and advective-dispersive transport by identifying two key parameters of suitable analytical solutions of the corresponding transport equations: first, the probability density function of the particle travel times, and second, a recovery-ratio function accounting for the complete or partial mass recovery at the observation point resulting from 2-D or 3-D dispersion/diffusion. The proposed method can be applied to any transport problem provided that the following three conditions are fulfilled. First, an analytical solution exists (or can be derived) either for the case of first-type (or Dirichlet) boundary condition with constant concentration C_0 (ML^{-3}) or for the case of third-type (or Cauchy) boundary condition of constant strength QC_0 (MT^{-1}). Note that in the latter case, the physical meaning of Q differs between (i) advection-dispersion (flowing) transport problems, in which it represents a volumetric injection rate, and (ii) purely diffusive (non flowing) transport problems, in which Q is the rate at which the diffusing substance is continuously liberated, e.g., due to the fixed dissolution rate of a solid pollutant.

Second, this analytical solution can be written in the following generic form:

$$C(\mathbf{x}, t) = QC_0 \Gamma(\mathbf{x}) \Psi(\mathbf{x}, t) \quad (1)$$

where C (ML^{-3}) is the solute concentration; t (T) is the time variable; \mathbf{x} (L) represents the one, two, or three-dimensional spatial coordinate(s); and $\Psi(\mathbf{x}, t)$ is a mathematical expression that tends toward 1 as time approaches infinity. This condition permits $\Psi(\mathbf{x}, t)$ to be treated as the cumulative probability density function of the particle travel times and $\Gamma(\mathbf{x})$ to be the recovery-ratio function that controls the infinite-time asymptotic value of the concentration at the observation point (\mathbf{x}) for a fixed boundary condition. Note that the identification between equation (1) and analytical solutions corresponding to the first-type boundary condition in which the term Q does not exist may be performed by canceling this term through the function Γ , e.g., by defining $\Gamma(\mathbf{x}) = 1/Q$.

Third, the following equation can be solved for the time variable:

$$\Psi(\mathbf{x}, t) = U_{01} \quad (2)$$

where U_{01} is a random number drawn from a uniform distribution between 0 and 1. The solution to equation (2) with respect to time gives the stochastic travel time of the particle, as suggested by *Yamashita and Kimura* [1990], whereas $\Gamma(\mathbf{x})$ is used to calculate the Lagrangian concentration at a given time from the histogram of particle arrival times.

The complete TDRW algorithm can be summarized by the following steps:

1. Release N particles at the source location, which defines the origin of the spatial coordinate system in which the location \mathbf{x} of the observation point is specified.
2. For each particle, generate a random number U_{01} and solve equation (2) for the particle's travel time.
3. Determine the minimum and maximum travel times t_{\min} and t_{\max} for the entire set of particles.
- 4a. Calculate the Lagrangian concentration for \mathbf{x} at time t as follows:

$$C(\mathbf{x}, t) = \frac{QC_0}{N} \Gamma(\mathbf{x}) M_c(t) \quad (3)$$

where $M_c(t)$ is the number of particles with a travel time less than t .

- 4b. Alternatively, the concentration corresponding to an instantaneous release of mass m_0 at $t=0$ can be calculated as follows:

$$C(\mathbf{x}, t) = \frac{m_0}{N\Delta t} \Gamma(\mathbf{x}) M_i(t, \Delta t) \quad (4)$$

where Δt is a time step and $M_i(t, \Delta t)$ is the number of particles that arrived at \mathbf{x} between $t - \Delta t/2$ and $t + \Delta t/2$.

5. Repeat step 4a or 4b for a series of time values t within the time interval $[t_{\min} \leftrightarrow t_{\max}]$ to obtain the breakthrough curve at \mathbf{x} .

As generally discussed in the literature [Kreft and Zuber, 1978; Parker and van Genuchten, 1984], two concepts may be used to define the concentration of a solute in a flowing fluid: (i) resident concentration, which "expresses the mass of solute per unit volume of fluid contained in an elementary volume of the system at a given instant," and (ii) flux concentration, which "expresses the mass of solute per unit volume of fluid passing through a given cross section at an elementary time interval" [Kreft and Zuber, 1978]. Obviously, the concept of flux concentration has no physical relevance in the case of purely diffusive transport and the distinction between resident and flux concentration is only appropriate for advection-dispersion transport problems. Another case in which this distinction is not meaningful is that of advection-dominated transport problems, as the numerical difference between resident and flux concentration can be considered negligible for high Péclet numbers [Parker and van Genuchten, 1984]. According to the above definitions, the concentration-time curves generated by the present TDRW algorithm clearly represent flux concentration in the case of advection-dispersion transport. However, as discussed later in section 4, the multidimensional TDRW method is mainly intended for advection-dominated transport problems and the calculated concentrations may therefore also be treated as resident concentrations.

3. Multidimensional TDRW Equations

3.1. Transport Equations

The advection-dispersion equation (ADE) for solute transport in a homogeneous porous medium with a uniform and steady flow can be formulated as follows [e.g., Fetter, 1993]:

$$\frac{\partial C(\mathbf{x}, t)}{\partial t} = -\mathbf{u} \cdot \nabla C(\mathbf{x}, t) + \mathbf{D} \cdot \nabla^2 C(\mathbf{x}, t) \quad (5)$$

where \mathbf{u} is the advection velocity vector and \mathbf{D} is the dispersion tensor. Additional terms describing the effects of reactions and sorption are commonly included for noninert solutes but are neglected here for simplicity. For two-dimensional (x, y) and three-dimensional (x, y, z) problems, the x axis is generally oriented along the flow direction, which simplifies the forms of \mathbf{u} and \mathbf{D} as follows:

$$\begin{aligned} \text{in 2-D : } \mathbf{u} &= \begin{pmatrix} u \\ 0 \end{pmatrix} \text{ and } \mathbf{D} = \begin{pmatrix} D_x & 0 \\ 0 & D_y \end{pmatrix} \\ \text{in 3-D : } \mathbf{u} &= \begin{pmatrix} u \\ 0 \\ 0 \end{pmatrix} \text{ and } \mathbf{D} = \begin{pmatrix} D_x & 0 & 0 \\ 0 & D_y & 0 \\ 0 & 0 & D_z \end{pmatrix} \end{aligned} \quad (6)$$

where $u(\text{LT}^{-1})$ is the scalar value of the advection velocity, $D_x(\text{L}^2\text{T}^{-1})$ is the longitudinal dispersion coefficient and $D_y(\text{L}^2\text{T}^{-1})$ and $D_z(\text{L}^2\text{T}^{-1})$ are the transverse dispersion coefficients.

Considering now the case of purely diffusive transport, the basic transport equation is Fick's second law, which can be written in the following form:

$$\frac{\partial C(\mathbf{x}, t)}{\partial t} = D_e \nabla^2 C(\mathbf{x}, t) \quad (7)$$

where $D_e(\text{L}^2\text{T}^{-1})$ is the effective diffusion coefficient, which accounts for the effects of pore geometry.

3.2. Example Derivation of TDRW Equations for 2-D Advective-Dispersive Transport

The analytical solution of equation (5) for the case of continuous injection in a 2-D infinite medium was derived by Wilson and Miller [1978] as follows:

$$C(x, y, t) = \frac{QC_0}{4\pi nb\sqrt{D_x D_y}} \exp\left(\frac{x}{B}\right) W\left(\eta, \frac{r}{B}\right) \quad (8)$$

where n (dimensionless) is the effective porosity of the porous medium; b (L) is the thickness of the two-dimensional system, in which the solute is assumed to be uniformly injected along the z axis at the point $(x=0, y=0)$; Q ($L^3 T^{-1}$) is the volumetric injection rate; and

$$B = \frac{2D_x}{u} \quad (9)$$

$$\eta = \frac{r^2}{4D_x t} \quad (10)$$

$$r = \sqrt{x^2 + \frac{D_x}{D_y} y^2} \quad (11)$$

$$W\left(\eta, \frac{r}{B}\right) = \int_{\eta}^{\infty} \frac{1}{\theta} \exp\left[-\left(\theta + \frac{r^2}{4B^2\theta}\right)\right] d\theta \quad (12)$$

Unfortunately, the exact analytical solution expressed by equations (8–12) cannot be used for the method described in section 2 because equation (8) cannot be cast in the general form of equation (1). However, *Wilson and Miller* [1978] also provided an approximation of the function $W(\eta, r/B)$ for the case of advection-dominated problems:

$$W\left(\eta, \frac{r}{B}\right) \cong \sqrt{\frac{\pi B}{2r}} \exp\left(-\frac{r}{B}\right) \operatorname{erfc}\left(\frac{2\eta - r/B}{2\sqrt{\eta}}\right) \quad (13)$$

According to the authors, the error incurred by this approximation is on the order of 10% for $r/B > 1$ and 1% for $r/B > 10$. The resulting approximate analytical solution expressed by equations (8–11) and (13) is well suited to the proposed approach, as the following identifications can easily be determined through comparison to equation (1):

$$\Psi(x, y, t) = \frac{1}{2} \operatorname{erfc}\left(\frac{2\eta - r/B}{2\sqrt{\eta}}\right) \quad (14)$$

$$\Gamma(x, y) = \frac{1}{2nb\sqrt{\pi ru D_y}} \exp\left(\frac{u(x-r)}{2D_x}\right) \quad (15)$$

Substituting equation (14) in equation (2), we obtain

$$\frac{r-ut}{\sqrt{t}} = 2\sqrt{D_x} \operatorname{erfc}^{-1}(2U_{01}) \quad (16)$$

where erfc^{-1} is the inverse complementary error function. Solving equation (16) for the time variable yields the following solution:

$$t = \frac{r}{u} + \frac{Z^2 + Z\sqrt{Z^2 + 4ur}}{2u^2} \quad (17)$$

where

$$Z = 2\sqrt{D_x} \operatorname{erfc}^{-1}(2U_{01}) \quad (18)$$

Substituting equation (15) into equation (3) yields the expression for the Lagrangian concentration for continuous injection:

$$C(x, y, t) = \frac{QC_0}{2nbN\sqrt{\pi ru D_y}} \exp\left(\frac{u(x-r)}{2D_x}\right) M_c(t) \quad (19)$$

The following expression for the case of instantaneous injection can be obtained by substituting equation (15) into equation (4):

Table 1. Approximate ($C(x, t) \cong$) and Exact ($C(x, t) =$) Analytical Solutions and TDRW Equations for 1-D, 2-D, and 3-D Advective-Dispersive Transport (ADE) and for 1-D, 2-D, and 3-D Purely Diffusive Transport (Diff)

Transport	Analytical Solution [Reference]	Mass Recovery Ratio Function $\Gamma(x)$	TDRW Transit Time
1-D ADE, first-type boundary condition	$C(x, t) \cong \frac{C_0}{2} \operatorname{erfc}\left(\frac{x-ut}{2\sqrt{D_x t}}\right)$ [Ogata and Banks, 1961]	$\Gamma(x) = \frac{1}{Q}$	$t = \frac{x}{u} + \frac{Z^2 + Z\sqrt{Z^2 + 4ux}}{2u^2}$ where $Z = 2\sqrt{D_x} \operatorname{erfc}^{-1}(2U_{01})$
2-D ADE, third-type boundary condition	$C(x, y, t) \cong \frac{QC_0}{4nb\sqrt{\pi ru D_y}} \exp\left(\frac{u(x-r)}{2D_x}\right) \operatorname{erfc}\left(\frac{2\eta-r/B}{2\sqrt{\eta}}\right)$ where $r = \sqrt{x^2 + \frac{D_x}{D_y} y^2}$; $\eta = \frac{r^2}{4D_x t}$; $B = \frac{2D_x}{u}$ [Wilson and Miller, 1978]	$\Gamma(x, y) = \frac{1}{2nb\sqrt{\pi ru D_y}} \exp\left(\frac{u(x-r)}{2D_x}\right)$	$t = \frac{x}{u} + \frac{Z^2 + Z\sqrt{Z^2 + 4ux}}{2u^2}$ where $Z = 2\sqrt{D_x} \operatorname{erfc}^{-1}(2U_{01})$
3-D ADE, third-type boundary condition	$C(x, y, z, t) \cong \frac{QC_0}{8\pi nr\sqrt{D_x D_z}} \exp\left(\frac{u(x-r)}{2D_x}\right) \operatorname{erfc}\left(\frac{r-ut}{2\sqrt{D_x t}}\right)$ where $r = \sqrt{x^2 + \frac{D_x}{D_y} y^2 + \frac{D_x}{D_z} z^2}$ [Hunt, 1978]	$\Gamma(x, y, z) = \frac{1}{4\pi nr\sqrt{D_x D_z}} \exp\left(\frac{u(x-r)}{2D_x}\right)$	$t = \frac{x}{u} + \frac{Z^2 + Z\sqrt{Z^2 + 4ux}}{2u^2}$ where $Z = 2\sqrt{D_x} \operatorname{erfc}^{-1}(2U_{01})$
1-D Diff, first-type boundary condition	$C(x, t) = C_0 \operatorname{erfc}\left(\frac{x}{2\sqrt{D_x t}}\right)$ [Crank, 1975]	$\Gamma(x) = \frac{1}{Q}$	$t = \frac{1}{D_x} \left(\frac{x}{2 \operatorname{erfc}^{-1}(U_{01})}\right)^2$
2-D Diff, first-type boundary condition	$C(x, y, t) = C_0 \operatorname{erfc}\left(\frac{x}{2\sqrt{D_x t}}\right) \operatorname{erfc}\left(\frac{ y }{2\sqrt{D_y t}}\right)$ [Etori, 1992]	$\Gamma(x) = \frac{1}{Q}$	$t =$ numerical root of equation $\left[\operatorname{erfc}\left(\frac{x}{2\sqrt{D_x t}}\right) \operatorname{erfc}\left(\frac{ y }{2\sqrt{D_y t}}\right) = U_{01}\right]$
3-D Diff, third-type boundary condition	$C(x, y, z, t) = \frac{QC_0}{4\pi n D_e \sqrt{x^2 + y^2 + z^2}} \operatorname{erfc}\left(\frac{\sqrt{x^2 + y^2 + z^2}}{2\sqrt{D_e t}}\right)$ [Crank, 1975]	$\Gamma(x, y, z) = \frac{1}{4\pi n D_e \sqrt{x^2 + y^2 + z^2}}$	$t = \frac{x^2 + y^2 + z^2}{4D_e [\operatorname{erfc}^{-1}(U_{01})]^2}$

$$C(x, y, t) = \frac{m_0}{2nbN\Delta t \sqrt{\pi ru D_y}} \exp\left(\frac{u(x-r)}{2D_x}\right) M_i(t, \Delta t) \quad (20)$$

3.3. Extension to Multidimensional Advection-Dispersion and Pure-Diffusion TDRW Algorithms in Homogeneous Media

Following the same methodology described above, new TDRW equations can be derived from approximate analytical solutions of the 1-D and 3-D advection-dispersion equations and from exact analytical solutions of the 1-D, 2-D, and 3-D pure-diffusion transport equations. The analytical solutions that have been considered and the resulting TDRW equations are summarized in Table 1. The case of 2-D purely diffusive transport is somewhat specific because the equation $\Psi(x, y, t) = U_{01}$, where $\Psi(x, y, t) = \operatorname{erfc}\left(\frac{x}{2\sqrt{D_x t}}\right) \operatorname{erfc}\left(\frac{|y|}{2\sqrt{D_y t}}\right)$, cannot be analytically solved for the time variable. Fortunately, this equation is easy to solve numerically using standard iterative methods. In the present work, a Newton-Raphson algorithm [e.g., Press et al., 1993] is used.

Although not addressed in this technical note for the sake of clarity, the TDRW equations can be easily adapted to account for particle retardation caused by linear instantaneous sorption/desorption. As discussed in many textbooks [e.g., Fetter, 1993], the coupling of linear sorption with advection-dispersion can easily be mathematically modeled by applying a retardation factor to both the advection velocity and the dispersion coefficients. For purely diffusive transport, the effects of linear sorption may be similarly accounted for by substituting the effective diffusion coefficient with an apparent diffusion coefficient. Other transport processes that can be straightforwardly addressed include time-varying source conditions, by appropriately defining the particle-release times at the source, and first-order decay reactions, by decreasing the time-series concentration values in accordance with the decay constant.

4. Verification Cases

The accuracy of a numerical model can be verified through comparison with (exact) analytical and/or alternative numerical solutions. In the present case, good agreement is logically expected between the TDRW simulations and the analytical solutions on which the particle algorithms are based. The accuracy of the 1-D, 2-D, and 3-D purely diffusive algorithms, which are based on exact analytical solutions, is therefore expected to be stochastically exact, whereas the accuracy of the 1-D, 2-D, and 3-D advection-dispersion algorithms is expected to be comparable to that of the approximate analytical solutions on which they are

Table 2. Input Parameters for Test 1 (1-D Purely Diffusive Transport, Continuous Source), Test 2 (2-D Purely Diffusive Transport, Continuous Source), Test 3 (3-D Purely Diffusive Transport, Instantaneous Point Source), Test 4 (1-D Advective-Dispersive Transport, Continuous Injection), and Test 5 (2-D Advective-Dispersive Transport, Instantaneous Injection)

Test	Parameters	Values
1	Effective diffusion coefficient D_e	$1 \times 10^{-10} \text{ m}^2/\text{s}$
	Prescribed concentration C_0	1 g/m^3
	Travel distance x between the source and observation points	5 m
2	Effective diffusion coefficient D_e	$1 \times 10^{-10} \text{ m}^2/\text{s}$
	Prescribed concentration C_0	1 g/m^3
	Coordinates (x, y) of the source	(0, 0)
3	Coordinates (x, y) of the observation point	(5, 4) m
	Effective diffusion coefficient D_e	$1 \times 10^{-10} \text{ m}^2/\text{s}$
	Effective porosity n	0.05
4	Injected mass m_0	300 g
	Coordinates (x, y, z) of the source	(0, 0, 0)
	Coordinates (x, y, z) of the observation point	(10, 5, 2) m
5	Prescribed concentration C_0	1 g/m^3
	Travel distance x between the injection and observation points	5 m
	Advection velocity u	$2 \times 10^{-5} \text{ m/s}$
	Longitudinal dispersion coefficient D_x , test 4a ($Pe=100$)	$1 \times 10^{-6} \text{ m}^2/\text{s}$
	Longitudinal dispersion coefficient D_x , test 4b ($Pe=30$)	$3.3 \times 10^{-6} \text{ m}^2/\text{s}$
	Longitudinal dispersion coefficient D_x , test 4c ($Pe=10$)	$1 \times 10^{-5} \text{ m}^2/\text{s}$
	Longitudinal dispersion coefficient D_x , test 4d ($Pe=1$)	$1 \times 10^{-4} \text{ m}^2/\text{s}$
	Longitudinal dispersion coefficient D_x , test 4e ($Pe=0.1$)	$1 \times 10^{-3} \text{ m}^2/\text{s}$
	Injected mass m_0	20 g
	Aquifer thickness b	10 m
	Advection velocity u	$4 \times 10^{-5} \text{ m/s}$
	Effective porosity n	0.05
	Longitudinal dispersion coefficient D_x	$2 \times 10^{-5} \text{ m}^2/\text{s}$
	Transverse dispersion coefficient D_y	$1 \times 10^{-6} \text{ m}^2/\text{s}$
	Coordinates (x, y) of the injection point	(0, 0)
	Coordinates (x, y) of the observation point	(5, 1) m
	r/B ratio	6.71

based. To demonstrate these claims, 5 test cases are presented below to provide comparisons to exact analytical solutions. The values of the parameters used for the calculation of both the analytical curves and the TDRW simulations are listed in Table 2.

In test 1, a source is assumed to supply a continuous injection of constant concentration in a one-dimensional semi-infinite homogeneous porous medium in which transport is purely diffusive. Considering the following initial and boundary conditions,

$$C(x, t) = 0, \quad x > 0, \quad t = 0 \quad (21)$$

$$C(x, t) = C_0, \quad x = 0, \quad t > 0 \quad (22)$$

the exact analytical solution is [e.g., Crank, 1975]

$$C(x, t) = C_0 \operatorname{erfc} \left(\frac{x}{2\sqrt{D_e t}} \right) \quad (23)$$

Test 2 addresses the case of two-dimensional diffusion in a semi-infinite plane ($y, x > 0$) from a prescribed constant concentration at the injection point $C(x=0, y=0, t) = C_0$. The corresponding analytical solution can be written as follows [e.g., Etori, 1992]:

$$C(x, y, t) = C_0 \operatorname{erfc} \frac{x}{2\sqrt{D_e t}} \operatorname{erfc} \frac{|y|}{2\sqrt{D_e t}} \quad (24)$$

Test 3 addresses the case of an instantaneous point source in a three-dimensional medium. The corresponding analytical solution is [e.g., Charbeneau, 2000]

$$C(x, y, z, t) = \frac{m_0}{n(4\pi D_e t)^{3/2}} \exp \left(-\frac{x^2 + y^2 + z^2}{4D_e t} \right) \quad (25)$$

Test 4 addresses the case of advection-dispersion in a 1-D semi-infinite medium with a continuous-source boundary condition. The exact analytical solution for this transport scenario was derived by Ogata and Banks [1961] as follows:

$$C(x, t) = \frac{C_0}{2} \left[\operatorname{erfc} \left(\frac{x-ut}{2\sqrt{D_x t}} \right) + \exp \left(\frac{ux}{D_x} \right) \operatorname{erfc} \left(\frac{x+ut}{2\sqrt{D_x t}} \right) \right] \quad (26)$$

To assess the accuracy of the advection-dispersion algorithms, which are based on approximate analytical solutions, five different values of the longitudinal dispersion coefficient D_x are considered in test 4. These values correspond to Péclet numbers ($Pe=ux/D_x$) ranging from 0.1 to 100.

Test 5 addresses the case of an instantaneous injection in a two-dimensional medium. The corresponding analytical solution was derived by *Wilson and Miller* [1978] as follows:

$$C(x, y, t) = \frac{m_0}{4\pi nbt \sqrt{D_x D_y}} \exp \left(-\frac{(x-ut)^2}{4D_x t} - \frac{y^2}{4D_y t} \right) \quad (27)$$

As expected and as illustrated in Figure 1, the numerical and analytical results coincide very well for purely diffusive transport. Because the corresponding TDRW algorithms were derived from exact analytical solutions, these algorithms do not suffer from any particular application restriction. Conversely, as demonstrated in Figure 2, the advection-dispersion TDRW curves deviate from the exact analytical solutions for low Pe values while conforming to the approximate analytical solution in each case. This finding demonstrates that the inaccuracies for low Pe values do not originate from the proposed methodological approach but instead correspond to the inherent limitations of the approximate analytical solutions from which the advection-dispersion TRDW equations were derived. In other words, the accuracy of the TDRW algorithms derived following the general methodological approach described in section 2 is strictly comparable to that of the considered analytical solutions. For an exact analytical solution, the TDRW algorithms are stochastically exact. If the TDRW algorithms are based on approximate analytical solutions, their accuracy is on the same order as that of the analytical approximation. Therefore, the stochastic accuracy of the 1-D, 2-D, and 3-D advection-dispersion TDRW algorithms summarized in Table 1 are inherently equivalent to the relative accuracy of the approximate analytical solutions reported by *Ogata and Banks* [1961], *Wilson and Miller* [1978], and *Hunt* [1978], respectively, with respect to the exact analytical solutions provided by the same authors. Because of the analytical similarities between the 1-D, 2-D, and 3-D ADE solutions, the general error estimation provided by *Wilson and Miller* [1978] for the 2-D case (see discussion in section 3.2) can be generalized to 1-D and 3-D. The error of the 1-D, 2-D, and 3-D advection-dispersion TDRW algorithms is therefore less than 10% for $r/B > 1$, where $r = \sqrt{x^2 + \frac{D_x}{D_y} y^2 + \frac{D_x}{D_z} z^2}$ and $B = \frac{2D_x}{u}$, which is actually verified in tests 4a, 4b

and 4c. However, as illustrated by tests 4d and 4e, the error increases rapidly for r/B ratios less than 1, which indicates that the advection-dispersion TDRW algorithms are not suited to highly dispersive transport problems.

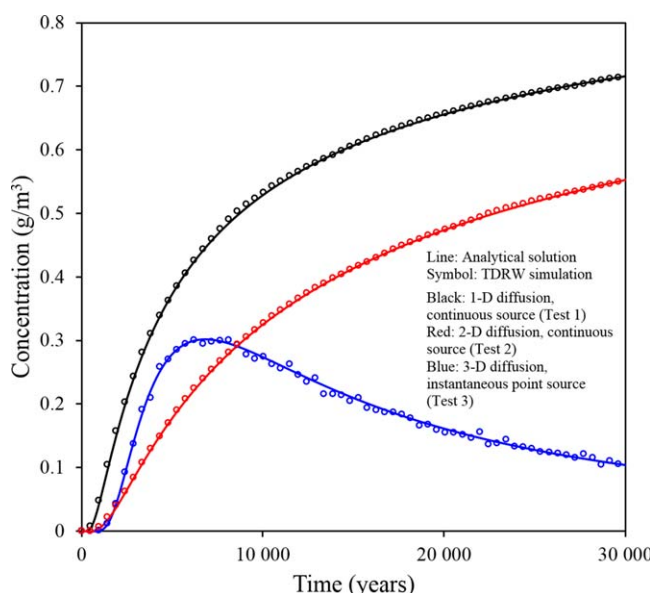


Figure 1. Comparison between TDRW simulations and analytical solutions for 1-D (test 1, continuous source), 2-D (test 2, continuous source), and 3-D (test 3, instantaneous point source) purely diffusive transport. The number of particles used in the TDRW simulations is 1×10^4 for test 1 and test 2, and 5×10^5 for test 3.

5. Extension to Multidimensional TDRW Simulations in Heterogeneous Media

Because of the homogeneity assumption inherent in the analytical solutions from which the algorithms given by equations (3) and (4) and Table 1 were derived, these algorithms can only be used to simulate transport in homogeneous porous media. However, contaminant

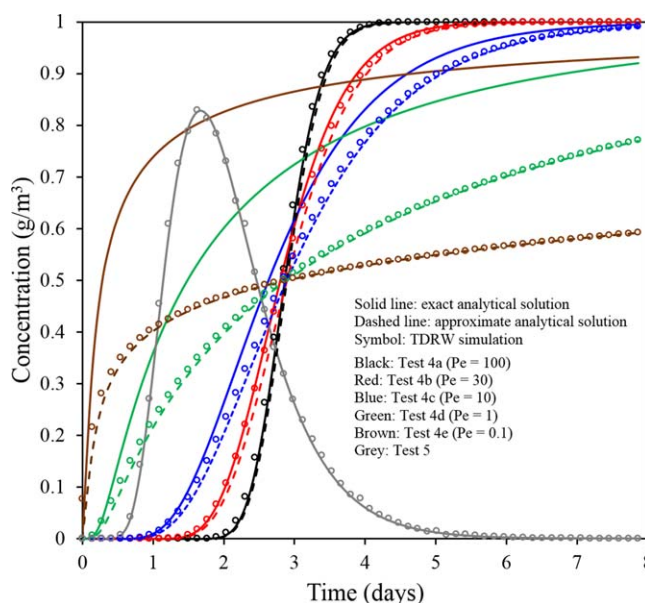


Figure 2. Comparison between TDRW simulations and exact analytical solutions for 1-D (test 4, continuous injection) and 2-D (test 5, instantaneous injection) advective-dispersive transport. The number of particles used in the TDRW simulations is 1×10^4 for test 4 and 5×10^5 for test 5.

transport problems in natural subsurface systems are typically influenced by spatial variability in the flow and transport properties. In this section, the extension of the algorithms to heterogeneous media conceptualized as a piecewise collection of homogeneous media separated by sharp interfaces is addressed. The key step is the determination of the location of particle transition points across the medium interfaces; these locations are used as intermediate checkpoints at which the particle travel time is updated. Ideally, the particle checkpoints should correspond to the intersections between the interface boundaries and the "natural" particle transport paths as determined by a conventional (i.e., space-based) random-walk simulation. This issue is straight-

forward for 1-D heterogeneous media, and the proposed 1-D TDRW algorithms can indeed be used to simulate advective-dispersive or purely diffusive transport in such media, similar to the application of the method of *Banton et al.* [1997] by *Bodin et al.* [2003] to interconnected 1-D transport pathways. For 2-D and 3-D heterogeneous media, the placement of the checkpoints depends on the underlying transport mechanisms. For purely diffusive transport, the particle motion may be considered to obey a purely random process. Therefore, starting from a known particle location \mathbf{x}_n , the next particle location \mathbf{x}_{n+1} can be defined as the intersection between a straight line of random direction starting from \mathbf{x}_n and the surrounding medium boundaries. For advective-dispersive transport, the proposed approach is to first compute the intersection \mathbf{x}_\perp between the advective pathline and the downstream interface boundary and then to compute a stochastic translation of \mathbf{x}_\perp along the boundary according to the conventional transverse random-walk equations $\Delta x_T = Z_1 \sqrt{2D_y \bar{t}}$ and $\Delta x_V = Z_2 \sqrt{2D_z \bar{t}}$, where Δx_T , Δx_V are the spatial translations in the transverse horizontal and vertical directions; $\bar{t} = L/u$ is the advective travel time, where L is the length of the advective pathline from \mathbf{x}_n to \mathbf{x}_\perp ; and Z_1 , Z_2 are two random numbers drawn from a standardized normal distribution. Note that the translation from \mathbf{x}_\perp to \mathbf{x}_{n+1} is in 1-D in a 2-D heterogeneous medium and in 2-D in 3-D heterogeneous media.

Regardless of the considered transport scenario and because the particle displacement is then decomposed into a series of jumps in a collection of homogeneous media, the TDRW transit time equations presented in Table 1 can be used to compute the particle travel time between two successive particle locations and therefore, by simple summation, the total travel time from the injection point to the observation point. The main difference between homogeneous and heterogeneous media is that particles may come from different locations for their last jump toward the observation point. Equations (3) and (4) must therefore be modified to account for the distinct Γ values relative to each particle transport path. Because the elementary mass associated with each particle is preserved during transport, only the last particle jump toward the observation point must be considered in the calculation of Γ . Therefore, the new equations that must be used in place of equations (3) and (4) can be written, respectively, as

$$C(\mathbf{x}, t) = \frac{QC_0}{N} \sum_{j=1}^N \Gamma_j(t) \quad (28)$$

where $\sum_{j=1}^N \Gamma_j(t)$ is the sum of the Γ values corresponding to the last jump of particles arriving at \mathbf{x} with a travel time less than t , and

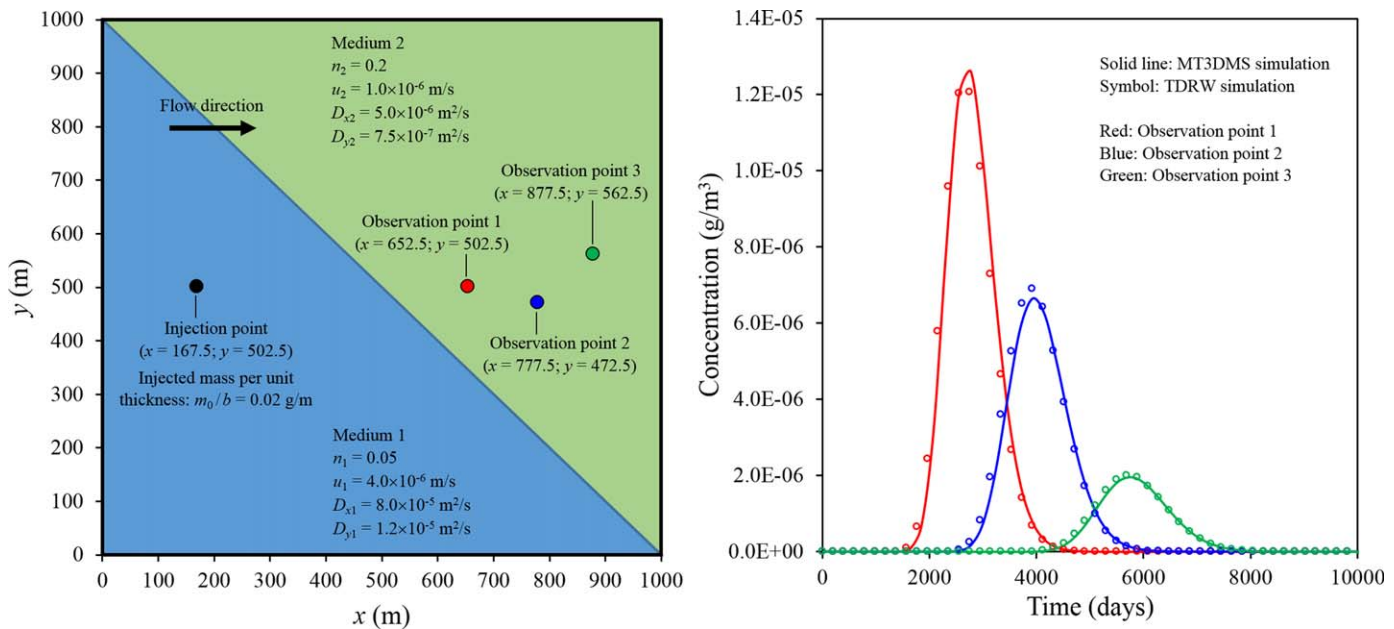


Figure 3. Comparison between TDRW and MT3DMS simulations of advective-dispersive transport in a synthetic 2-D heterogeneous medium. (left) description of the transport problem; (right) simulated breakthrough curves at three different observation points.

$$C(\mathbf{x}, t) = \frac{m_0}{N\Delta t} \sum_{j=1}^N \Gamma_j(t, \Delta t) \quad (29)$$

where $\sum_{j=1}^N \Gamma_j(t, \Delta t)$ is the sum of the Γ values corresponding to the last jump of particles arriving at \mathbf{x} between $t - \Delta t/2$ and $t + \Delta t/2$.

It must be acknowledged that the above-proposed approach for advection-dispersion TDRW simulations is supposed to be rigorously valid only when the medium interface boundaries are both infinite and perpendicular to the advective pathlines. To illustrate this point, let us consider the counter-case of a particle displacement between two locations within medium 1 close to the interface with medium 2 and with advective pathlines more or less parallel to the medium interface boundary. In this case, the real particle transport path may cross the interface between the two media because of transverse dispersion and be partially transported in medium 2 before returning to medium 1. The particle travel time between the two locations in medium 1 can therefore be partially affected by the transport properties of medium 2. However, the TDRW algorithm for 2-D and 3-D heterogeneous media will not "see" medium 2 because the advective pathline does not cross the medium interface boundary, and the TDRW particle travel time will therefore be biased. Further developments may therefore be needed to rigorously address this issue. However, as discussed in the previous section, the multidimensional TDRW method is mainly intended for advection-dominated transport problems. In these cases, accuracy errors due to lateral heterogeneities are expected to be negligible.

Figure 3 illustrates the synthetic transport problem employed to verify the validity of the proposed approach. In this problem, a finite mass of solute is assumed to be released instantaneously and then subjected to advection-dispersion in a 2-D heterogeneous medium. The flow direction is assumed to be uniform in the x direction, and the solute spreading in the x and y directions is affected by longitudinal and transverse dispersion, respectively. The purpose is to simulate the solute breakthrough curves at three observation points located downstream and beyond a discontinuity within the medium transport properties. The finite difference transport model MT3DMS [Zheng, 2009] was selected for comparison with the multidimensional TRDW method. The simulation domain was discretized using a $5 \text{ m} \times 5 \text{ m}$ finite-difference grid. The flow field was solved using MODFLOW [Harbaugh et al., 2000], assigning no-flow boundaries at the top and bottom and fixed-head boundaries on the left and right sides such that a lateral head gradient was produced. The third-order total-variation-diminishing (TVD) scheme available in the MT3DMS code was

selected as the advection solver package. As illustrated in Figure 3, the breakthrough curves from the TDRW method and MT3DMS simulations are in good agreement, which demonstrates the applicability of the TDRW method to multidimensional heterogeneous media.

The general methodology and resulting TDRW algorithms presented here represent a significant theoretical improvement over existing time-domain particle-tracking methods and broaden the potential applications of such methods. For example, the 2-D and 3-D TDRW equations may be advantageously incorporated into Lagrangian models of multispecies reactive transport [e.g., *Paster et al.*, 2013; *Ding et al.*, 2013] because they provide a simple method of calculating the probability density for two reactant particles at the same location at the same time; these equations may also be useful for routing particles through 2-D and 3-D fracture intersections [e.g., *Zafarani and Detwiler*, 2013].

6. Conclusions

A set of new time-domain particle-tracking algorithms has been derived from approximate analytical solutions of the advection-dispersion equation and from exact analytical solutions of the pure-diffusion equation. These algorithms allow one to compute both the time required for a particle to travel a specified distance in a homogeneous medium and the mass recovery at the observation point, which may be incomplete due to 2-D or 3-D transverse dispersion or diffusion. The method is extended to heterogeneous media represented as a piecewise collection of homogeneous media. The particle motion is then decomposed along a series of intermediate checkpoints located on the medium interface boundaries. The accuracy of the multidimensional TDRW method is verified against (i) exact analytical solutions of solute transport in homogeneous media and (ii) finite-difference simulations in a synthetic 2-D heterogeneous medium of simple geometry. The results demonstrate that the method is ideally suited to purely diffusive transport and to advection-dispersion transport problems dominated by advection. Conversely, the method is not recommended for highly dispersive transport problems because the accuracy of the advection-dispersion TDRW algorithms degrades rapidly for low Péclet number, consistent with the accuracy limit of the approximate analytical solutions.

The 1-D advection-dispersion algorithm proposed in this note may be regarded as an alternative to the TDRW method of *Banton et al.* [1997], and the new 2-D and 3-D advection-dispersion algorithms constitute a significant improvement over existing time-domain particle methods, which are strictly limited to 1-D transport problems. Furthermore, the proposed approach provides a unified methodology for deriving multi-dimensional time-domain particle equations and may be applicable to other mathematical transport models (e.g., reactive solute transport, multiple-porosity media, or non-Fickian transport), provided that appropriate analytical solutions are available.

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