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Diffusion processes in composite porous media and their numerical integration by random walks: Generalized stochastic differential equations with discontinuous coefficients

Eric M. LaBolle,[1] Jeremy Quastel,[2] Graham E. Fogg,[3] and Janko Gravner[4]

Abstract. Discontinuities in effective subsurface transport properties commonly arise (1) at abrupt contacts between geologic materials (i.e., in composite porous media) and (2) in discrete velocity fields of numerical groundwater-flow solutions. However, standard random-walk methods for simulating transport and the theory on which they are based (diffusion theory and the theory of stochastic differential equations (SDEs)) only apply when effective transport properties are sufficiently smooth. Limitations of standard theory have precluded development of random-walk methods (diffusion processes) that obey advection dispersion equations in composite porous media. In this paper we (1) generalize SDEs to the case of discontinuous coefficients (i.e., step functions) and (2) develop random-walk methods to numerically integrate these equations. The new random-walk methods obey advection-dispersion equations, even in composite media. The techniques retain many of the computational advantages of standard random-walk methods, including the ability to efficiently simulate solute-mass distributions and arrival times while suppressing errors such as numerical dispersion. Examples relevant to the simulation of subsurface transport demonstrate the new theory and methods. The results apply to problems found in many scientific disciplines and offer a unique contribution to diffusion theory and the theory of SDEs.

1. Introduction

Facilitated by geostatistical methods, detailed characterizations of the subsurface can capture the character of intricate heterogeneities that strongly control transport [*Copty and Rubin*, 1995; *Sheibe and Freyberg*, 1995; *McKenna and Poeter*, 1995; *Carle et al*., 1998]. Adequately resolving subsurface heterogeneity can yield immense computational grids, commonly with greater than 10^6 nodes [e.g., *Tompson*, 1993], that demand specialized numerical techniques to solve governing transport equations. In many cases, random-walk methods are favored over finite difference, finite element, and method of characteristic techniques for large problems of this type [e.g., see *Tompson et al*., 1987; *Tompson and Gelhar*, 1990; *LaBolle et al*., 1996] because of their ability to efficiently simulate solute-mass distributions and arrival times while suppressing errors such as numerical dispersion [*Prickett et al*., 1981; *Uffink*, 1985; *Ahlstrom et al*., 1977; *Kinzelbach*, 1988; *Tompson et al*., 1987].

Spatial averaging [*Plumb and Whitaker*, 1990] of pore-scale equations for transport by advection and molecular diffusion in porous media gives rise to advection-dispersion equations (ADEs) commonly used to model subsurface transport:

$$\frac{\partial}{\partial t}[\Theta(\mathbf{x}, t)c(\mathbf{x}, t)] = -\sum_i \frac{\partial}{\partial x_i}[v_i(\mathbf{x}, t)\Theta(\mathbf{x}, t)c(\mathbf{x}, t)] + \sum_{i,j} \frac{\partial}{\partial x_i}\left[\Theta(\mathbf{x}, t)D_{ij}(\mathbf{x}, t)\frac{\partial c(\mathbf{x}, t)}{\partial x_j}\right], \quad (1)$$

where t is time, c $[M\ L^{-3}]$ is concentration, v_i $[L\ T^{-1}]$ is velocity, Θ $[L^3\ L^{-3}]$ is effective porosity, and D_{ij} $[L^2\ T^{-1}]$ is a real symmetric dispersion tensor. Standard random-walk methods [e.g., *Kinzelbach*, 1988; *Tompson et al*., 1987] approximate solutions to (1) by simulating sample (particle) paths corresponding to diffusion processes described by stochastic differential equations (SDEs). Here "diffusion processes" refer to Markov processes with continuous sample paths as mathematical models of real subsurface-transport phenomena. Therefore, in the present context, the term "diffusion process" refers to "advection dispersion process."

Use of (1) as a model of transport in heterogeneous porous media poses problems in the context of random-walk simulation methods: Standard methods, and the theory on which they are based (i.e., diffusion theory and the theory of SDEs), only apply when coefficients, porosity and dispersion tensors are sufficiently smooth functions of space [*LaBolle et al*., 1996, 1998]. Discontinuities in effective transport properties, however, commonly arise (1) at abrupt contacts between geologic materials (i.e., in composite porous media) and (2) in discrete velocity fields of numerical groundwater-flow solutions. As a result, standard random-walk methods (and SDEs) cannot simulate transport in heterogeneous porous media with abrupt contacts between geologic materials.

Both interpolation [*LaBolle et al*., 1996] and "reflection" [*Uffink*, 1985; *Ackerer*, 1985; *Cordes et al*., 1991; *Semra et al*., 1993; *LaBolle et al*., 1998; *LaBolle and Fogg*, this issue]

[1]Hydrologic Sciences, University of California, Davis.
[2]Department of Mathematics, University of Toronto, Toronto, Ontario, Canada.
[3]Hydrologic Sciences and Department of Geology, University of California, Davis.
[4]Department of Mathematics, University of California, Davis.

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techniques have been proposed to address the aforementioned limitations of standard random-walk methods. By spatially interpolating coefficients, one can ensure they remain sufficiently smooth throughout the domain such that standard random-walk (or Itô-Euler integration) methods can be applied. Accuracy of this approach, however, suffers unless one refines the interpolation (i.e., the region over which coefficients are smoothed) and time step simultaneously [*LaBolle et al*., 1996], which commonly leads to undesirable increases in computational effort. Furthermore, in the limit, refining the interpolation ultimately gives rise to the original problem: discontinuous coefficients.

LaBolle et al. [1998] developed necessary conditions for the convergence of diffusion processes to ADEs in composite porous media and applied the new theory to test four one-dimensional "reflection" techniques [*Uffink*, 1985; *Ackerer*, 1985; *Cordes et al*., 1991; *Semra et al*., 1993]. The term "reflection" is derived from the usual method of reflecting particles to maintain no-flux boundary conditions in a random walk [see *Tompson et al*., 1987]. These techniques rely on either an analytical solution to the specific problem or the specialized numerical treatment of particle displacements to maintain mass balance at an interface between regions with constant, but different, diffusion coefficients. *LaBolle et al*. [1998] showed that two of these reflection techniques fail to solve the specified problem, while the methods of *Uffink* [1985] and *Semra et al*. [1993] succeed. One can show that these methods relate to an analytical solution to the problem of one-dimensional diffusion at an interface [*LaBolle and Fogg*, this issue]. The method of *Semra et al*. [1993] has been recently extended to three dimensions [*Semra*, 1994; *Ackerer*, 1999] for constant coefficients within subdomains. General mathematical representations of multidimensional diffusion processes obeying ADEs in composite porous media (i.e., porous media characterized by discontinuous coefficients) have remained undeveloped [*LaBolle et al*., 1998].

In this paper we generalize SDEs to the case of discontinuous coefficients to develop (1) new mathematical representations of diffusion processes that simulate advection and dispersion in composite porous media and (2) random-walk methods for numerical integration of these equations. The new methods retain many of the computational advantages of standard methods [e.g., *Kinzelbach*, 1988; *Tompson et al*., 1987]. Examples demonstrate application of the new theory and methods to problems of transport in porous media. However, our results apply to problems found in numerous scientific disciplines. Further, since the treatment of diffusion processes with discontinuous coefficients is presently not covered in stochastic theory (e.g., as described by *Arnold* [1992]), our results offer a unique contribution to diffusion theory and the theory of SDEs. Before considering the new approximations, we review standard stochastic methods for simulating diffusion processes.

2. Standard Methods for Simulating Diffusion Processes

Standard stochastic methods for simulating subsurface transport [e.g., *Kinzelbach*, 1988; *Tompson*, 1987] may be applied when effective transport properties vary smoothly in space. In this case, diffusions corresponding to (1) are commonly represented by an Itô SDE [*Itô and McKean*, 1961]:

$$\int_{t_0}^{t} \mathrm{d}X_i(t') = \int_{t_0}^{t} A_i(\mathbf{X}, t')\, \mathrm{d}t' + (\mathrm{I}) \int_{t_0}^{t} \sum_j B_{ij}(\mathbf{X}, t')\, \mathrm{d}W_j(t') \tag{2a}$$

$$A_i(\mathbf{X}, t) = v_i(\mathbf{X}, t) + \Theta^{-1}(\mathbf{X}, t) \sum_j \frac{\partial}{\partial x_j} [\Theta(\mathbf{X}, t) D_{ij}(\mathbf{X}, t)], \tag{2b}$$

where the last integral in (2a) is referred to as a stochastic integral, (I) denotes the Itô interpretation of this integral (defined below), $X_i(t)$ $[L]$ is a sample path in space, $B_{ik}B_{jk} = 2D_{ij}$, and $W_j(t)$ $[T^{-1/2}]$ is a Brownian motion process such that $\Delta W_j = W_j(t) - W_j(t_0)$ has mean zero and covariance $\Delta t \delta_{ij}$. Note that B_{ij} is generally not unique. The coefficients A_i $[L\ T^{-1}]$ and D_{ij} are referred to as the drift vector and diffusion tensor, respectively, and are defined as

$$\lim_{\Delta t \to 0} \frac{1}{\Delta t} \langle X_i(t) - X_i(t_0) \rangle = A_i = v_i + \Theta^{-1} \sum_j \frac{\partial}{\partial x_j} (\Theta D_{ij}) \tag{3a}$$

$$\lim_{\Delta t \to 0} \frac{1}{\Delta t} \langle [X_i(t) - X_i(t_0)][X_j(t) - X_j(t_0)] \rangle = \sum_k B_{ik}B_{jk} = 2D_{ij}, \tag{3b}$$

where angle brackets denote the expectation. We will refer to a component of the drift that involves gradient terms, such as the second term in the right-hand side of (3a), as a "gradient drift term."

In general, to arrive at a unique definition for a stochastic integral, it is necessary to specify how it is to be evaluated [see *Arnold*, 1992, chapter 2]. The Itô stochastic integral specified in (2a) is defined as

$$(\mathrm{I}) \int_{t_0}^{t} \sum_j B_{ij}(\mathbf{X}, t')\, dW_j(t')$$

$$= \mathrm{ms} - \lim_{n \to \infty} \sum_{k=1}^{n} \sum_j B_{ij}[\mathbf{X}(t_{k-1}), t_{k-1}][W_j(t_k) - W_j(t_{k-1})], \tag{4}$$

where ms-lim denotes the limit in the mean square [*Gardiner*, 1990]. The Itô definition given in (4) evaluates B_{ij} at location $\mathbf{X}(t_{k-1})$ rendering B_{ij} statistically independent of $\mathrm{d}W_j$ thus ensuring that the integral in (4) has mean zero. One well-known alternative to the Itô interpretation of the stochastic integral is that of *Stratonovich* [1963], in which $\mathbf{B}$ is evaluated at location $[\mathbf{X}(t_{k-1}) + \mathbf{X}(t_k)]/2$, that is,

$$(\mathrm{S}) \int_{t_0}^{t} \sum_j B_{ij}(\mathbf{X}, t')\, \mathrm{d}W_j(t')$$

$$= \mathrm{ms} - \lim_{n \to \infty} \sum_{k=1}^{n} \sum_j B_{ij}\left[\frac{\mathbf{X}(t_k) + \mathbf{X}(t_{k-1})}{2}, t_{k-1}\right]$$

$$\cdot [W_j(t_k) - W_j(t_{k-1})], \tag{5}$$

where the (S) denotes the Stratonovich interpretation. For convenience, herein we will adopt the following notation:

$$(\mathrm{I}) \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j(t) = \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j(t) \tag{6a}$$

$$(\mathrm{S}) \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j(t) = \sum_j B_{ij}(\mathbf{X} + \mathrm{d}\mathbf{Y}, t)\, \mathrm{d}W_j(t) \tag{6b}$$

$$\mathrm{d}\mathbf{Y}(t) = \tfrac{1}{2}\mathrm{d}\mathbf{X}(t), \tag{6c}$$

where we have written stochastic integrals as differentials, leaving all integration implied. We will say that B_{ij} is evaluated at location $\mathbf{X}$ and $\mathbf{X} + \mathrm{d}\mathbf{Y}$ in the Itô and Stratonovich integrals of (6a) and (6b), respectively. For $\mathrm{d}X_i = B_{ij}(\mathbf{X} + \mathrm{d}\mathbf{Y}, t) \cdot \mathrm{d}W_j$ and assuming that B_{ij} is sufficiently smooth, expanding the Stratonovich equation (6b) in a Taylor series shows the relationship between Itô and Stratonovich integrals is given as

$$\begin{aligned}
\mathrm{d}X_i &= (\mathrm{S}) \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j = \sum_j B_{ij}(\mathbf{X} + \tfrac{1}{2}\mathrm{d}\mathbf{X}, t)\, \mathrm{d}W_j \\
&= \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j + \frac{1}{2} \sum_{j,l} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l}\, \mathrm{d}X_l\, \mathrm{d}W_j \\
&= \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j + \frac{1}{2} \sum_{j,l,k} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l} B_{lk}(\mathbf{X}, t)\, \mathrm{d}W_k\, \mathrm{d}W_j \\
&= \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j + \frac{1}{2} \sum_{j,l} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l} B_{lj}(\mathbf{X}, t)\, \mathrm{d}t \\
&= (\mathrm{I}) \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j + \frac{1}{2} \sum_{j,l} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l} B_{lj}(\mathbf{X}, t)\, \mathrm{d}t,
\end{aligned} \tag{7}$$

where we have used $\mathrm{d}W_k \mathrm{d}W_j = \mathrm{d}t\, \delta_{kj}$. Assuming constant Θ, one can show that the following Stratonovich SDE obeys (1) and is equivalent to the Itô SDE (2a):

$$\begin{aligned}
\mathrm{d}X_i(t) = &\left[v_i(\mathbf{X}, t) + \frac{1}{2} \sum_{j,l} B_{ij} \frac{\partial}{\partial x_l} B_{lj}(\mathbf{X}, t) \right] \mathrm{d}t \\
&+ \sum_j B_{ij}(\mathbf{X} + \tfrac{1}{2}\mathrm{d}\mathbf{X}, t)\, \mathrm{d}W_j(t),
\end{aligned} \tag{8}$$

where B_{ij} of the stochastic integral is evaluated at time t and location given by the vector $\mathbf{X} + \frac{1}{2}\mathrm{d}\mathbf{X}$. Therefore evaluating B_{ij} at various locations, which differ infinitesimally from the current location, and adding or subtracting necessary gradient drift terms allows one to formulate a variety of equations that are mathematically equivalent to (2a).

Standard random-walk methods for simulating (1) are normally based on Euler integration of the Itô SDE in (2a) [e.g., see *Tompson et al.*, 1987]. However, equation (2a) only applies when coefficients Θ and D_{ij} are sufficiently smooth. Therefore, when either D_{ij} or Θ are discontinuous, these standard methods fail [*LaBolle*, 1996]. For example, Euler approximations to (2a) that either evaluate gradient drift terms by finite differences [*Tompson et al.*, 1987] or neglect gradient drift terms all together [*Prickett et al.*, 1981] cannot simulate (1) in composite media. Further, results from these methods cannot be improved by refining the time step of the integration scheme. In summary, standard stochastic theory fails to provide a definition for such equations in composite media in which Θ and D_{ij} are discontinuous. We address this problem in section 3.

3. Diffusion Processes in Composite Media

The methods developed here stem from the premise that SDEs may be generalized to consider discontinuous coefficients, yielding diffusion processes that obey (1) for both smooth and discontinuous transport properties. By inspection the principle objection to applying the SDE (2a) in composite media is the presence of the gradient drift term therein, not formally defined when Θ and/or D_{ij} are discontinuous, that is, step functions. We will show that one can formulate equations equivalent in meaning to (2a) but free of gradient drift terms. Numerical integration by random walks will demonstrate that these equations correspond to (1) in composite media, where Θ and D_{ij} are discontinuous. These new methods preserve many of the computational advantages of standard random-walk techniques. In Appendix A we consider the new methods in one dimension and show that they, indeed, correspond to (1) in composite media. In Appendix B we show that a similar result may be obtained by applying a stochastic calculus using generalized functions. Next we present these methods and demonstrate their application to subsurface transport problems beginning with the simple case of isotropic dispersion in composite media, and then we consider the case of anisotropic dispersion.

3.1. Isotropic Diffusions

The mathematical representation of isotropic diffusion processes arising from advection and dispersion in composite porous media is relevant to the simulation of transport in heterogeneous porous media. As we will demonstrate in a subsequent paper, the dispersion tensor can often be approximated as isotropic without loss of accuracy provided the pore-scale dispersion tensor can be approximated as isotropic with respect to its minor axes (assumed to be orthogonal to the velocity vector) and provided longitudinal spreading due to explicitly modeled heterogeneities is much greater than longitudinal spreading represented by the pore-scale dispersion process. Beginning with the case of constant Θ and isotropic D_{ij}, we develop an equation that is equivalent to (2a) and yet free of gradient terms. We seek an equation that takes the form of (2a) after expanding in Taylor series. By inspection we arrive at the following result:

$$\mathrm{d}X_i(t) = v_i(\mathbf{X}, t)\, \mathrm{d}t + \sum_j B_{ij}(\mathbf{X} + \mathrm{d}\mathbf{X}, t)\, \mathrm{d}W_j(t) \tag{9a}$$

$$B_{ij}(\mathbf{x}, t) = \sqrt{\lambda(\mathbf{x}, t)}\, \delta_{ij}, \tag{9b}$$

where λ is the eigenvalue of $2D_{ij}$ and B_{ij} is evaluated at time t and location given by the vector $\mathbf{X} + \mathrm{d}\mathbf{X}$. For smooth coefficients, (9a) has been referred to as the "backward Itô" stochastic integral [*Karatzas and Shreve*, 1991]. Expanding (9a) in a Taylor series shows that (9a) and (2a) are, indeed, equivalent for constant Θ and smooth isotropic D_{ij}. For all D_{ij} we have

$$\begin{aligned}
\mathrm{d}X_i &= v_i(\mathbf{X}, t)\, \mathrm{d}t + \sum_j B_{ij}(\mathbf{X} + \mathrm{d}\mathbf{X}, t)\, \mathrm{d}W_j = v_i(\mathbf{X}, t)\, \mathrm{d}t \\
&\quad + \sum_j \left[B_{ij}(\mathbf{X}, t) + \sum_{k,l} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l} B_{lk}(\mathbf{X}, t)\, \mathrm{d}W_k + \cdots \right] \mathrm{d}W_j \\
&= v_i(\mathbf{X}, t)\, \mathrm{d}t + \sum_{j,l} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l} B_{lj}(\mathbf{X}, t)\, \mathrm{d}t + \sum_j B_{ij}(\mathbf{X}, t)\, \mathrm{d}W_j,
\end{aligned} \tag{10a}$$

and when D_{ij} is diagonal,

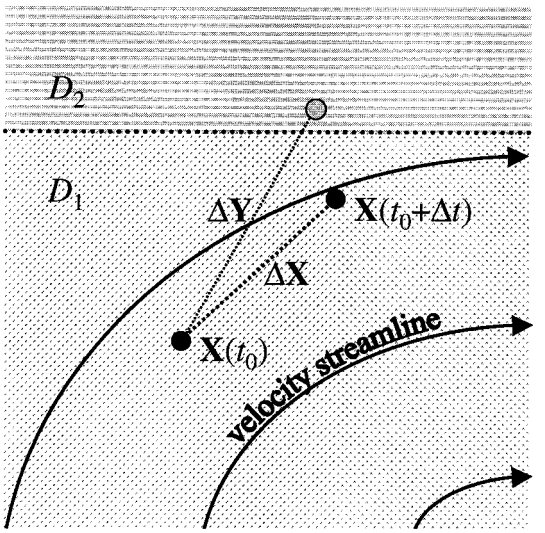

Figure 1. The two-step process of random-walk simulation of advection and isotropic dispersion in composite porous media for the algorithm given in equations (11a) and (11b).

$$\begin{aligned} \mathrm{d}X_i &= v_i(\mathbf{X}, t)\,\mathrm{d}t + \sum_{j,l} \frac{\partial B_{ij}(\mathbf{X}, t)}{\partial x_l} B_{lj}(\mathbf{X}, t)\,\mathrm{d}t \\ &\quad + \sum_j B_{ij}(\mathbf{X}, t)\,\mathrm{d}W_j \\ &= v_i(\mathbf{X}, t)\,\mathrm{d}t + \sum_j \frac{\partial D_{ij}(\mathbf{X}, t)}{\partial x_j}\,\mathrm{d}t \\ &\quad + \sum_j B_{ij}(\mathbf{X}, t)\,\mathrm{d}W_j. \end{aligned} \tag{10b}$$

Formally, this Taylor series expansion is not allowed for discontinuous D_{ij}; however, in Appendix B we show that one can expand (9a) for discontinuous D_{ij} using generalized functions. In the following sections we introduce a method for numerically integrating (9a) and apply this method to clearly demonstrate convergence to (1) in composite media.

3.1.1. Numerical integration. Equation (9a) may be integrated over a time step Δt by taking two particle displacements, ΔY_l and ΔX_i, as

$$\Delta X_i = v_i \Delta t + \sum_j B_{ij}(X_l + \Delta Y_l, t)\Delta W_j \tag{11a}$$

$$\Delta Y_l = \sum_k B_{lk}(\mathbf{X}, t)\Delta W_k. \tag{11b}$$

Note that advective transport is not included in (11b); here velocity only contributes to higher-order terms that can be neglected in the limit. To implement this algorithm, (11b) is first evaluated to determine the particle displacement ΔY_l. The result is used in (11a) to determine ΔX_l. This simulation method is illustrated in Figure 1. Boundary conditions must be implemented in the application of both (11a) and (11b). If a particle exits an absorbing or reflecting boundary in the application of (11a) or (11b), it is either removed from the simulation or reflected in the usual way, respectively [see *Tompson et al.*, 1987].

In Appendix A we show that distributions approximated by (11a) satisfy (1). In Appendix B we show that a similar result can be obtained through the use of a stochastic calculus using generalized functions. To our knowledge this result has not been described elsewhere in the literature. We do not, however, present a formal proof of convergence of these approximations. Such a lengthy proof usually demonstrates convergence only; it does not guarantee a robust approximation. To this end we will demonstrate convergence and applicability of these approximations through numerical examples. In all examples, Brownian motions will be simulated by uniformly distributed random variables with mean 0 and variance Δt.

3.1.2. Diffusion in one-dimensional composite media. In this example we consider one-dimensional diffusion in an unbounded domain with governing equation (1), constant Θ, $v = 0$, and initial and boundary conditions

$$c_i(x, 0) = \delta(x - x_0), \qquad x_0 \in \Omega_i \tag{12a}$$

$$c_1(-\infty, t) = c_2(+\infty, t) = 0 \tag{12b}$$

$$c_1(x, t) = c_2(x, t), \qquad x = 0 \tag{12c}$$

$$\lim_{x\to 0-} D_1 \frac{\partial c_1(x, t)}{\partial x} = \lim_{x\to 0+} D_2 \frac{\partial c_2(x, t)}{\partial x} \tag{12d}$$

$$D(x) = \begin{cases} D_1, & x < 0 \\ D_2, & x > 0 \end{cases} \tag{12e}$$

where $i = 1, 2$ denotes quantities within subdomains Ω_1 and Ω_2 to the left and right, respectively, of an interface located at $x = 0$.

First, we apply (11a) performing three simulations corresponding to $\Delta t = 100.0$, 10.0, and 1.0 to demonstrate convergence with decreasing time step of simulated moments to analytical moments of the distribution. In each simulation we use $D_1 = 10.0$ and $D_2 = 1.0$ and begin with 1000 particles located at $x_0 = -1.0$. Figure 2 plots the mean and standard deviation computed from an analytical solution [see *Carslaw and Jaeger*, 1959] against values computed from particle displacements, given as

$$\bar{X}(t) = \langle X(t) \rangle = \frac{1}{N_p} \sum_{p=1}^{N_p} X_p(t) \tag{13a}$$

$$\begin{aligned} \Sigma(t) &= \langle [X(t) - \bar{X}(t)]^2 \rangle^{1/2} \\ &= \left\{ \frac{1}{N_p} \sum_{p=1}^{N_p} [X_p(t) - \bar{X}(t)]^2 \right\}^{1/2} \end{aligned} \tag{13b}$$

respectively, where N_p is the total number of particles and $X_p(t)$ is the location of the pth particle at time t.

Second, using (11a), we perform three simulations corresponding to $D_2 = 2.5$, 0.5, and 0.05. In each simulation we use $D_1 = 5.0$, $\Delta t = 0.01$, $x_0 = -5.5$ and 10^5 particles. Figure 3 compares simulated density at time $t = 6$, computed by summing particle masses within unit lengths along the x axis, with analytical distributions [see *Carslaw and Jaeger*, 1959] to (1) with initial and boundary conditions (12a)–(12e).

Numerical simulation results presented here compare well with analytical solutions and demonstrate convergence of (11a) and (9a) to (1) in one dimension with discontinuous coefficients. As we will show, extension to (multidimensional) isotropic diffusions for hyperplane interfaces follows directly from

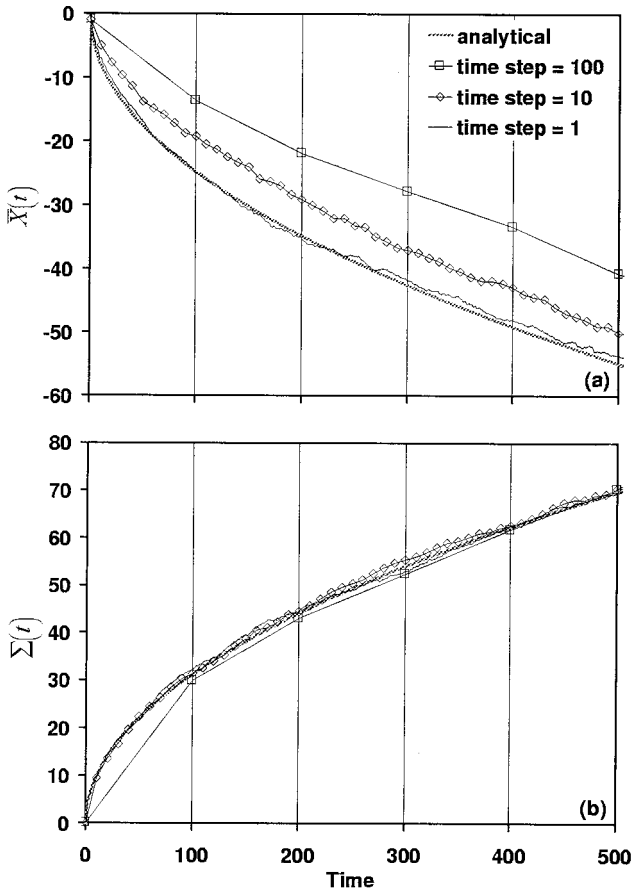

Figure 2. Simulation results for (a) $\bar{X}(t)$ and (b) $\Sigma(t)$ are compared with analytical moments for $\Delta t = 100$, 10, and 1.

this result by noting that diffusion processes in the different coordinate directions are independent in this case.

3.1.3. Effective diffusivity of composite media. In this example, we estimate effective diffusivity of composite media. The specific geometry considered here is that of circular cylinders packed in regular square arrays within a matrix of contrasting material as shown in Figure 4. Dispersion tensors of both materials are constant and isotropic. Transport is described by (1) with constant Θ and $\nu = 0$. This classic problem has received much attention [e.g., see *Keller*, 1963; *Sangani and Acrivos*, 1983; *Quintard and Whitaker*, 1993], and accurate experimental and analytical values of effective diffusivity are available [*Perrins et al.*, 1979].

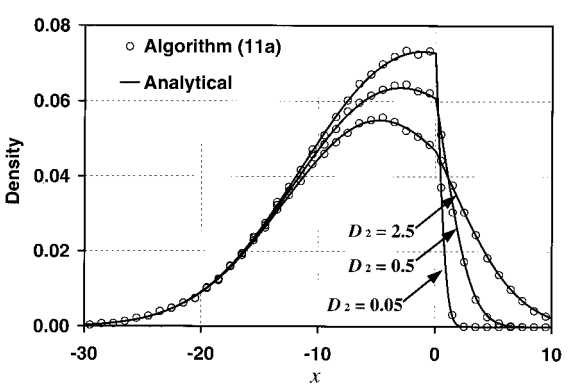

Figure 3. Simulation results for density are compared with analytical solutions for $D_1 = 5.0$ and $D_2 = 2.5$, 0.5, and 0.05.

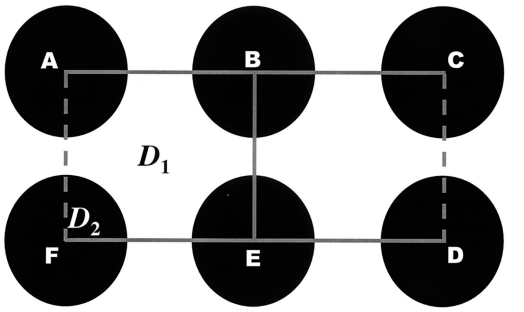

Figure 4. Square array of cylinders with known diffusivity D_2 embedded in a matrix with contrasting diffusivity D_1.

We estimate the effective diffusivity of the periodic array by specifying reflecting boundaries at lines ABC and DEF and absorbing boundaries at lines AF and CD as shown in Figure 4. Simulation proceeds by releasing particles at time $t = 0$ and random locations with uniform distribution along the line BE that bisects the system. Particle paths are simulated using (11a) until particles exit the system by crossing absorbing boundaries at lines AF or CD. Effective diffusivity is given by the relationship

$$\bar{D} = L_{\mathrm{AB}}^2 \left(\frac{2}{N_p} \sum_{p=1}^{N_p} \tau_p \right)^{-1}, \tag{14}$$

where τ_p is the elapsed time from release until particle p exits the system and L_{AB} is the length of line AB.

The matrix diffusivity is arbitrarily chosen as 1.0. Simulations are performed for a range of cylinder volume fractions, controlled by varying cylinder diameter, and diffusivities. In each simulation, 1000 particles are released; time step is dynamically controlled to ensure particles cannot bypass subdomains in any single step of the algorithm given by (11a). Figure 5 compares simulated effective diffusivities with values reported by *Perrins et al.* [1979] for a range of cylinder volume fractions and diffusivities. Simulation results compare well with reported values

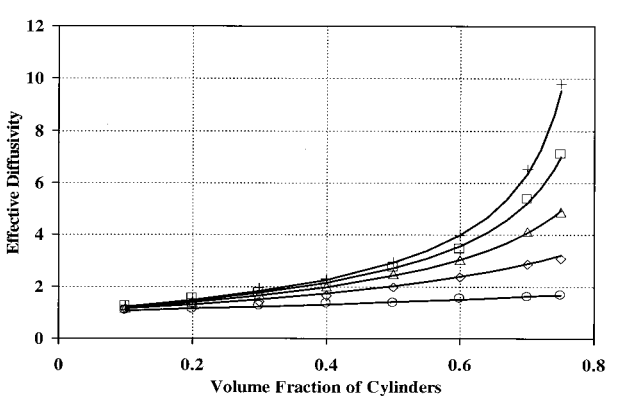

Figure 5. Simulated effective diffusivities are compared with values reported by *Perrins et al.* [1979] for cylinder volume fractions of 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, and 0.75, matrix diffusivity $D_1 = 1$, and cylinder diffusivities, $D_2 = 2$ (circles), 5 (diamonds), 10 (triangles), 20 (squares), and 50 (plus signs).

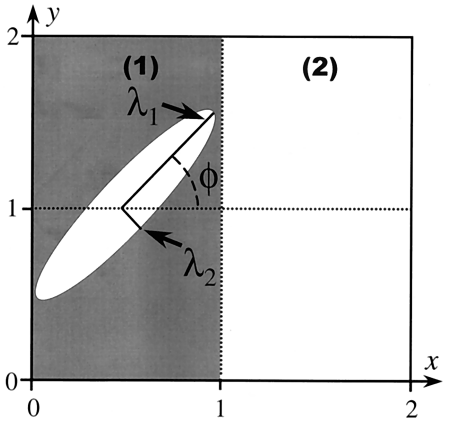

Figure 6. Composite media with anisotropic diffusion tensors.

as expected because of the previous success of (11a) in one dimension.

3.1.4. Discussion. Our results show that diffusions described by (9a) obey (1) in composite media with discontinuities in effective transport properties. Furthermore, the numerical integration method (11a) "accurately" solves this equation.

Convergence and analysis of errors associated with the numerical simulation of SDEs is a developing field [e.g., see *Kloeden and Platen*, 1992]. Verification of convergence and quantification of accuracy may generally be addressed by comparing numerical results with known analytical solutions, as we have done here, or by using a bench mark against established numerical methods as in sections 3.2.1 and 3.2.2. Since we cannot consider all of the many problems to which (11a), and the other approximations that follow, may be applied, we suggest verification by a similar procedure to assess accuracy and convergence when the methods are used under circumstances other than those considered herein.

The examples presented above were for isotropic diffusions with constant Θ and $\mathbf{v} = 0$. Next we present a more general multidimensional approximation for anisotropic diffusions. Examples that follow will consider advection and dispersion in composite porous media.

3.2. Anisotropic Diffusions

As with the isotropic case, we find ourselves faced with the following problem: eliminating the gradient drift term in (2a) in the anisotropic case. Note that this problem is not trivial. Nevertheless, with considerable effort we arrived at the following inspired result:

$$\mathrm{d}\mathbf{X}_i(t) = v_i(\mathbf{X}, t)\,\mathrm{d}t + \frac{1}{2}\sum_{j,k}\Theta^{-1/2}(\mathbf{X}, t)\hat{B}_{ijk} \cdot \left[X_l + \Theta^{-1/2}(\mathbf{X}, t)\sum_{m,n}\hat{B}_{lmn}(\mathbf{X}, t)\,\mathrm{d}W_m, t\right]\mathrm{d}W_j + \frac{1}{2}\sum_{j,k,m,n} Z_{imn}(\mathbf{X}, t)\Theta^{-1/2}(\mathbf{X}, t) \cdot \hat{B}_{mjk}(X_l + \lambda_n^{1/2}(\mathbf{X}, t)\,\mathrm{d}W_l, t)\,\mathrm{d}W_j \tag{15a}$$

$$\hat{B}_{ijk}(\mathbf{x}, t) = \Theta^{1/2}(\mathbf{x}, t)\lambda_k^{1/2}(\mathbf{x}, t)Z_{ijk}(\mathbf{x}, t) \tag{15b}$$

$$Z_{ijk}(\mathbf{x}, t) = [e_k(\mathbf{x}, t)]_i[e_k(\mathbf{x}, t)]_j, \tag{15c}$$

where $\mathbf{e}_k$ is the normalized eigenvector corresponding to the kth eigenvalue λ_k of $2D_{ij}$, and, for example, in the first term on the right-hand side of (15a), $\hat{B}_{ijk}$ is evaluated at time t and location given by the vector whose lth component is $X_l + \Theta^{-1/2}(\mathbf{X}, t)\,\Sigma_{m,n}\,\hat{B}_{lmn}(\mathbf{X}, t)\,\mathrm{d}W_m$. For isotropic D_{ij}, $\lambda = \lambda_k$, $\forall k$, and using relationships given in Appendix C, (15a) and (15b) simplify considerably:

$$\mathrm{d}X_i(t) = v_i(\mathbf{X}, t)\,\mathrm{d}t + \Theta^{-1/2}(\mathbf{X}, t) \cdot \sum_j B_{ij}[\mathbf{X} + \Theta^{-1/2}(\mathbf{X}, t)\,\mathrm{d}\mathbf{X}, t]\,\mathrm{d}W_j(t) \tag{16a}$$

$$B_{ij}(\mathbf{x}, t) = \sqrt{\Theta(\mathbf{x}, t)\lambda(\mathbf{x}, t)}\,\delta_{ij}. \tag{16b}$$

Further, for constant Θ, (16a) and (16b) reduce to (9a) and (9b). Therefore (9a) can be viewed as a special case of (15a).

Integration of (15a) may proceed according to a discrete-time random walk that includes the approximations

$$\Delta X_i = v_i(\mathbf{X}, t)\Delta t + \tfrac{1}{2}\Theta^{-1/2}(\mathbf{X}, t)\sum_{j,k}\hat{B}_{ijk}(X_l + \Delta\hat{Y}_l, t)\Delta W_j + \tfrac{1}{2}\Theta^{-1/2}(\mathbf{X}, t)\sum_{j,k,m,n} Z_{imn}(\mathbf{X}, t)\hat{B}_{mjk}(X_l + \Delta U_{nl}, t)\Delta W_j \tag{17a}$$

$$\Delta\hat{Y}_l = \Theta^{-1/2}(\mathbf{X}, t)\sum_{m,n}\hat{B}_{lmn}(\mathbf{X}, t)\Delta W_m \tag{17b}$$

$$\Delta U_{nl} = \lambda_n^{1/2}(\mathbf{X}, t)\Delta W_l. \tag{17c}$$

First, equations (17b) and (17c) are evaluated to determine $\Delta\hat{Y}_l$ and ΔU_{nl}, $\forall n$. These results are used in (17a) to determine $\Delta\mathbf{X}$. As with the (11a) and (11b), boundary conditions must be implemented in the application of each step of algorithm (17a)–(17c).

When coefficients are sufficiently smooth, (15a) is a SDE. In Appendix C we expand this SDE in Taylor series to show that it is, indeed, equivalent to (2a) in this case. Standard stochastic theory [e.g., see *Arnold*, 1992] shows that diffusions described by (2a) and therefore (15a) obey (1) when coefficients are smooth. Next we demonstrate that (17a) converges to (1) in composite media and is applicable to the simulation of subsurface transport.

3.2.1. Anisotropic diffusion in composite media. In this example we simulate anisotropic diffusion in composite media. The system illustrated in Figure 6 is a composite of two media with contrasting diffusion (dispersion) tensors whose principle axes, designated by λ_1, are oriented at an angle ϕ with the x axis; eigenvectors corresponding to λ_1 and λ_2 are given as $\mathbf{e}_1 = (\sin\phi, \cos\phi)$ and $\mathbf{e}_2 = (-\sin\phi, \cos\phi)$, respectively. Two problems are considered. In the first problem we test the ability of approximation (17a) to maintain the invariant distribution, that is, a uniform number density, for the system in Figure 6 given periodic boundaries in the x direction and reflective, no-flux boundaries at $y = 0$ and $y = 2$. In each of the four simulations a total of 5000 particles are initially distributed uniformly over the domain, and the system is evolved over time using the parameters specified in Table 1. Results for first moments in the x and y directions plotted in Figures 7a–7d indicate that approximation (17a) can successfully maintain the invariant distribution for these problems.

Table 1. Parameters Corresponding to Simulation Results Presented in Figures 7a–7d and 8a–8d

	Region 1			Region 2		
Figures	λ_1	λ_2	ϕ, deg	λ_1	λ_2	ϕ, deg
7a and 8a	2×10^{-2}	2×10^{-3}	90	2×10^{-2}	2×10^{-3}	0
7b and 8b	2×10^{-2}	2×10^{-3}	−45	2×10^{-2}	2×10^{-3}	45
7c and 8c	2×10^{-2}	2×10^{-3}	30	2×10^{-3}	2×10^{-4}	30
7d and 8d	2×10^{-2}	2×10^{-3}	70	2×10^{-3}	2×10^{-4}	0

In the second problem we compare predictions for evolution of concentration from approximation (17a) with finite difference numerical solutions to (1) for the system in Figure 6 with absorbing (zero concentration) boundaries on $x = 0$, $x = 2$, $y = 0$, and $y = 2$ and initial distribution $c(\mathbf{x}, 0) = c_0$ in the region $0.99 < x < 1.00$ and $0.99 < y < 1.00$ and $c(\mathbf{x}, 0) = 0$ outside this region. The finite difference algorithm is implemented using an explicit updating scheme. Choosing $\Delta x = \Delta y = 0.005$ and $\Delta t = 0.0005$ satisfies criteria for numerical stability [*Peaceman*, 1977]. Particle simulations are implemented using 10^6 particles. Coarse particle and finite difference solution contours of $c(\mathbf{x}, t)/c_0$ at $t = 5.0$ plotted in Figures 8a–8d for parameters given in Table 1 compare well. The results herein clearly demonstrate that diffusions described by (15a) and approximation (17a) obey (1) for the cases considered here.

3.2.2. Advective-dispersive transport in composite media. In this example we simulate advective-dispersive transport according to (1) for the system illustrated in Figure 6 with absorbing (zero concentration) boundaries on $x = 0$, $x = 2$, $y = 0$, and $y = 2$. The functional form of the hydrodynamic-dispersion tensor used here is given as

$$D_{ij} = (|\mathbf{v}|\alpha_{\mathrm{T}} + D^*)\delta_{ij} + (\alpha_{\mathrm{T}} - \alpha_{\mathrm{L}})v_i v_j/|\mathbf{v}|, \qquad (18)$$

where α_{T} and α_{L} (L) are transverse and longitudinal dispersivities, respectively, and D^* is effective molecular diffusivity $(L^2 T^{-1})$. Velocity direction is oriented at an angle ϕ with the x axis as illustrated in Figure 6. We compare predictions from approximation (17a) with finite difference numerical solutions to (1). Again, the finite difference algorithm uses an explicit updating scheme. Choosing $\Delta x = \Delta y = 0.005$ and $\Delta t = 0.0005$ satisfies the stability criteria referenced in section 3.2.1 and ensures a grid Peclet number $Pe \approx \Delta x/\alpha_{\mathrm{L}}$ less than one. Particle simulations use 10^6 particles. As in the previous simulations, the initial distribution in each simulation is $c(\mathbf{x}, 0) = c_0$ specified within a square region of $\Delta x = \Delta y = 0.01$, as shown in Figures 9a–9c, and $c(\mathbf{x}, 0) = 0$ outside of this region.

Coarse particle and finite difference solution contours of $c(\mathbf{x}, t)/c_0$ at $t = 2.5$, 5.0, and 5.0 plotted in Figures 9a, 9b, and 9c, respectively, for parameters given in Table 2 compare well. These and the previous results show that diffusions de-

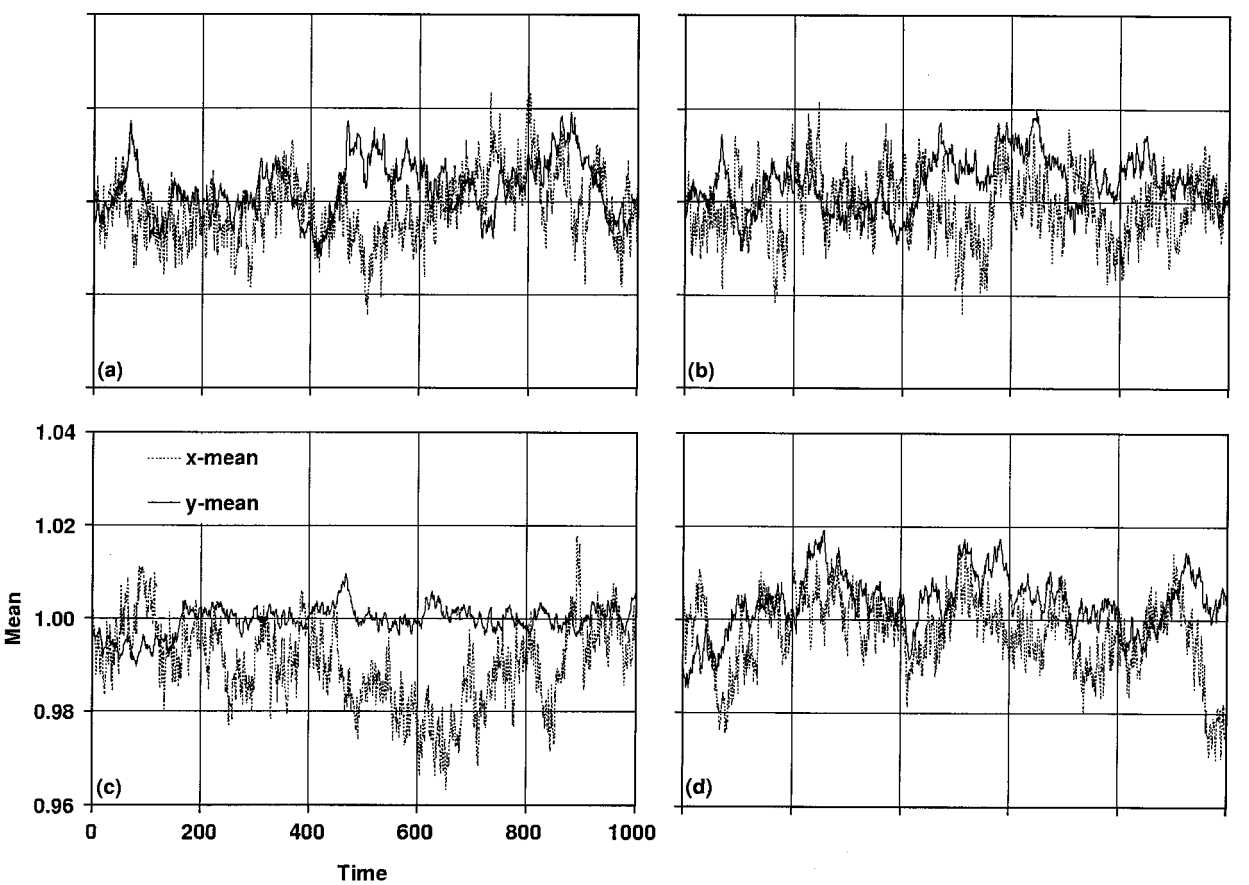

Figure 7. Simulated first moments in the x and y directions as a function of time for the system illustrated in Figure 6 with periodic boundaries in the x direction and reflective, no-flux boundaries at $y = 0$ and $y = 2$, and initially uniform number density. (a–d) Parameters given in Table 1.

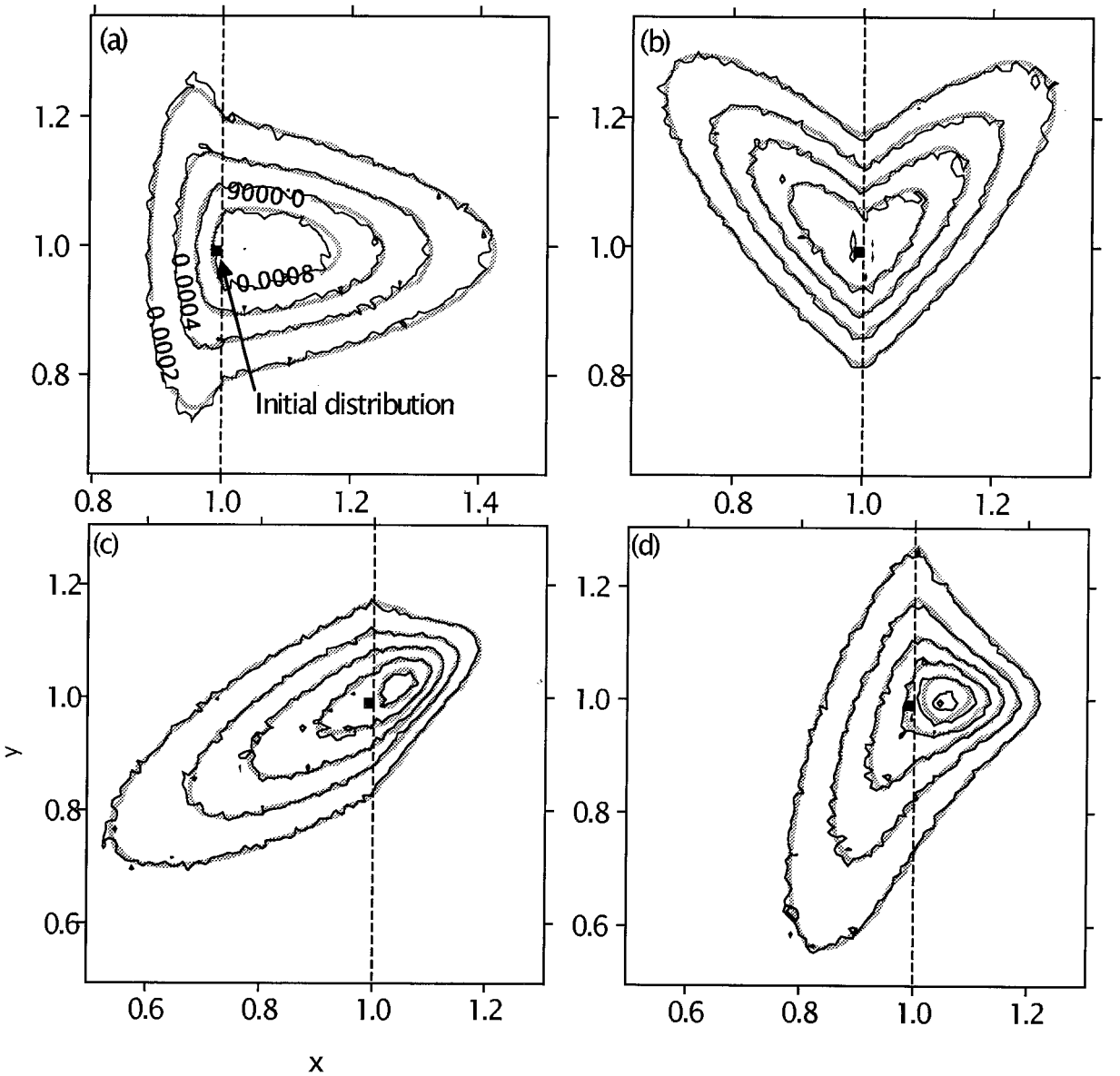

Figure 8. (a–d) Contours of $c(\mathbf{x}, t)/c_0$ from simulations of anisotropic diffusion in the composite media shown in Figure 6 with parameters given Table 1. Figure 8a shows contour intervals used in Figures 8a–8d.

scribed by (15a) correspond with (1) and that approximation (17a) simulates (1) in composite media with surprising accuracy.

4. Discussion and Conclusions

Standard diffusion theory only applies when effective transport properties are sufficiently smooth, yet discontinuities in transport properties arise naturally in porous media at abrupt contacts between geologic materials with contrasting transport properties. Limitations of standard diffusion theory have precluded development of diffusion processes that obey spatially averaged transport equations in composite media. In this paper we have (1) generalized SDEs to the case of discontinuous coefficients and (2) developed random-walk methods for numerically integrating these equations. The new results apply to problems found in many scientific disciplines and offer a unique contribution to diffusion theory and the theory of SDEs. Examples demonstrated convergence of the new methods to ADEs in composite media and applications to subsurface-transport problems including (1) one-dimensional diffusion in composite porous media with constant coefficients in subdomains, (2) isotropic two-dimensional diffusion in a system of circular cylinders packed in regular square arrays within a matrix of contrasting material, (3) two-dimensional anisotropic diffusion in a composite system with contrasting diffusion tensors, and (4) transport by advection and dispersion in composite porous media.

Standard theory shows that diffusions described by the new generalized SDEs obey ADEs when coefficients are sufficiently smooth. Further, we have demonstrated that these diffusions obey ADEs in composite media. Thus, in cases where coefficients are smooth, we conclude that the new methods may often be more robust approximations than standard Itô-Euler techniques.

The new simulation techniques possess the computational advantages of standard random-walk methods, including the

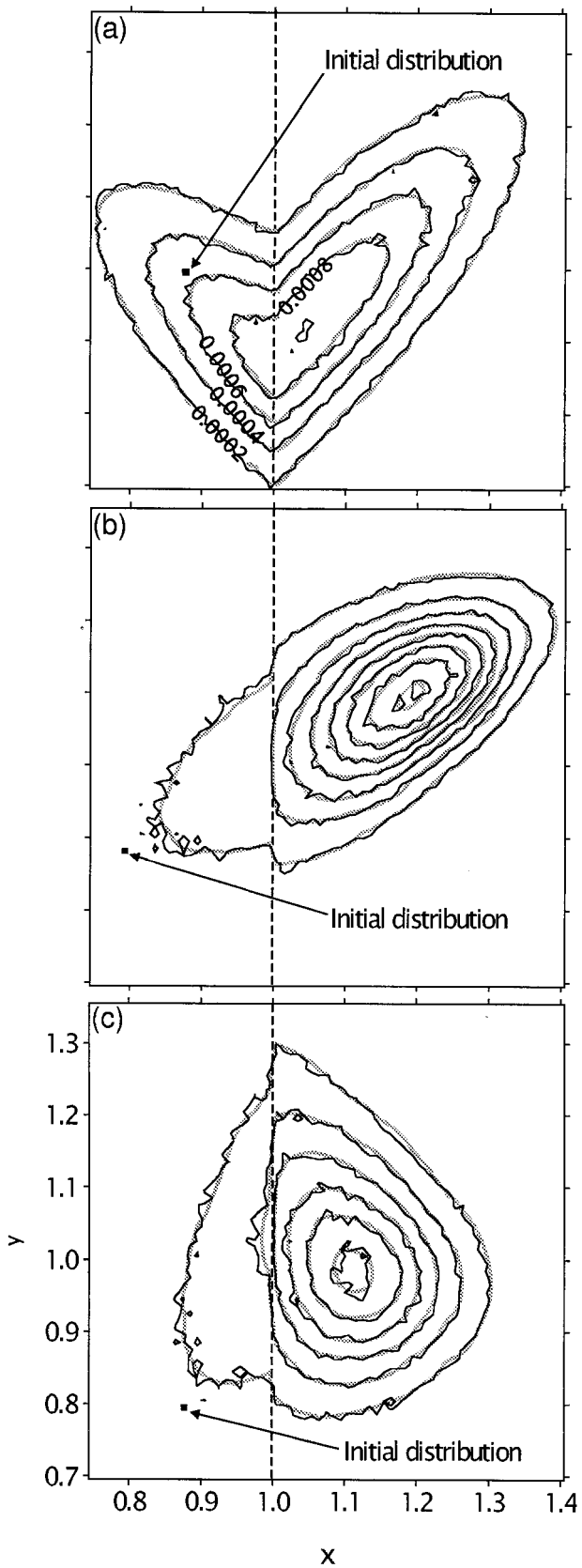

Figure 9. (a–c) Contours of $c(\mathbf{x}, t)/c_0$ from simulations of advective-dispersive transport in the composite media shown in Figure 6 with parameters given Table 2. Figure 9a shows contour intervals used in Figures 9a–9c.

ability to efficiently simulate solute-mass distributions and arrival times while suppressing errors such as numerical dispersion, common to finite difference methods when Peclet numbers are large. As such, the new methods are appropriate for problems characterized by immense computational grids, such as those now commonly produced through the use of geostatistical simulation techniques for subsurface characterization. Finally, unlike alternatives relying on Itô-Euler integration and spatial interpolation to ensure smooth coefficients, the new methods will converge exactly in the limit, without the need to simultaneously refine the interpolation scheme and time step.

Appendix A: Necessary Conditions for Weak Convergence

LaBolle et al. [1998] use a graphical technique to show that approximations with symmetric transition-probability density satisfy necessary conditions for weak convergence developed therein. Since transition-probability densities specified by (11a) and similar approximations are asymmetric, however, the simple graphical techniques used by *LaBolle et al.* [1998] are not applicable here. Instead, here we will show mathematically that (11a) approximates the diffusion equation of interest.

Functions $p(x, t)$ satisfy the one-dimensional diffusion equation

$$\int f(x, t)\,\frac{\partial p(x, t)}{\partial t}\,\mathrm{d}x = \int [D'(x, t) f'(x, t) + D(x, t) f''(x, t)] p(x, s)\,\mathrm{d}x \quad \text{(A1)}$$

for all smooth and bounded test functions f and are weak solutions of (1) with $v = 0$ [*LaBolle et al.*, 1998], where the f' and f'' denote the first and second partial derivatives of f with respect to x. For the problem at hand it is convenient to consider the case in which $D(x, t)$ has constant values within subdomains and a single jump of size $|D_2 - D_1|$ at x_0, $D = D_1$ for $x < x_0$, and $v = 0$. Here $D' = 0$ except at x_0, where it is given as

$$D' = (D_2 - D_1)\delta(x - x_0). \quad \text{(A2)}$$

Substituting this result into (A1) yields

$$\begin{aligned}\int f\,\frac{\partial p}{\partial t}\,\mathrm{d}x &= \int (D'f' + Df'')p\,\mathrm{d}x \\ &= \int [(D_2 - D_1)\delta(x - x_0) f' + Df'']p\,\mathrm{d}x \\ &= \int Df''p\,\mathrm{d}x + (D_2 - D_1) f' p|_{x_0}. \quad \text{(A3)}\end{aligned}$$

We will show that the density $p_\varepsilon(x, t)$ generated by (11a) satisfies (A3).

To facilitate computations, we will consider the following variation of approximation (11a):

$$\Delta X(t) = \tfrac{1}{2}\{B[X + B(X)Z] + 2B(X) - B[X - B(X)Z]\}Z, \quad \text{(A4)}$$

where Z has distribution $\Phi(Z)$ with mean zero and variance $\Delta t = \varepsilon^2$. We could work directly with (11a), but use of (A4) simplifies the math that follows. The density p_ε generated by the Markov-chain approximation (A4) satisfies the equations [*LaBolle et al.*, 1998]

Table 2. Parameters Corresponding to Simulation Results Presented in Figures 9a–9c

Figure	Θ_1	$\Theta_1\lvert\mathbf{v}\rvert$	α_L	α_T	ϕ, deg	D^*
			Region 1			
9a	1/3	$\Theta_1\,(10^{-1})$	10^{-1}	10^{-2}	−45	10^{-5}
9b	1/3	$\Theta_1\,(10^{-1})$	10^{-1}	10^{-2}	30	10^{-5}
9c	1/3	$\Theta_1\,(10^{-1})$	10^{-1}	10^{-2}	60	10^{-5}
			Region 2			
9a	1/3	$\Theta_1\,(10^{-1})$	10^{-1}	10^{-2}	45	10^{-5}
9b	1/2	$\Theta_1\,(10^{-1})$	$\Theta_2\,(10^{-2})/\Theta_1$	1.5×10^{-3}	30	10^{-5}
9c	1/2	$\Theta_1\,(5\times10^{-2})$	$\Theta_2\,(2\times10^{-2})/\Theta_1$	3.0×10^{-3}	0	10^{-5}

$$\int_0^t \int f(x, s)\,\frac{\partial p_\varepsilon(x, s)}{\partial t}\,\mathrm{d}x\,\mathrm{d}s = \int_0^t \int L_\varepsilon f(x, s) p_\varepsilon(x, s)\,\mathrm{d}x\,\mathrm{d}s + o(\varepsilon^2) \tag{A5a}$$

$$L_\varepsilon f(x, t) = \frac{1}{\varepsilon^2}\int [f(z, t) - f(x, t)] p_\varepsilon(z, t + \varepsilon^2|x, t)\,\mathrm{d}z, \tag{A5b}$$

where L_ε is commonly referred to as the generator (or infinitesimal operator) of the Markov chain [*Arnold*, 1992]. Expanding f in a Taylor series yields

$$\begin{aligned}\varepsilon^2 \int_0^t \int f(x, s)\,\frac{\partial p_\varepsilon(x, s)}{\partial t}\,\mathrm{d}x\,\mathrm{d}s &= \int_0^t \int \int [f(z, s) - f(x, s)] \\ &\quad \cdot p_\varepsilon(z, s + \varepsilon^2|x, s) p_\varepsilon(x, s)\,\mathrm{d}x\,\mathrm{d}z\,\mathrm{d}s + o(\varepsilon^2) \\ &= \int_0^t \int \int [(z - x) f'(x, s) - \tfrac{1}{2}(z - x)^2 f''(x, s)] \\ &\quad \cdot p_\varepsilon(z, s + \varepsilon^2|x, s) p_\varepsilon(x, s)\,\mathrm{d}x\,\mathrm{d}z\,\mathrm{d}s + o(\varepsilon^2).\end{aligned} \tag{A6}$$

Noting that $(z - x)$ is a realization of $\Delta X(t)$, we have, retaining terms to order ε^2,

$$\begin{aligned}\int (z - x) p(z, t + \varepsilon^2|x, t)\,\mathrm{d}z &= \frac{1}{2}\int \{B[x + B(x)Z] + 2B(x) \\ &\quad - B[x - B(x)Z]\} Z\Phi(Z)\,\mathrm{d}Z \\ &= \frac{1}{2}\int \{B[x + B(x)Z] - B[x - B(x)Z]\} Z\Phi(Z)\,\mathrm{d}Z\end{aligned} \tag{A7a}$$

$$\begin{aligned}\frac{1}{2}\int (z - x)^2 p(z, t + \varepsilon^2|x, t)\,\mathrm{d}z &= \frac{1}{8}\int \{B[x + B(x)Z] + 2B(x) - B[x - B(x)Z]\}^2 Z^2\Phi(Z)\,\mathrm{d}Z \\ &= \varepsilon^2\bar{D}(x) + o(\varepsilon^2),\end{aligned} \tag{A7b}$$

where $\bar{D} = (D_1 + D_2)/2$ at x_0 and $\bar{D} = B^2/2$ elsewhere. Substituting these relationships into (A6), we have

$$\begin{aligned}\varepsilon^2 \int_0^t \int f\,\frac{\partial p_\varepsilon}{\partial s}\,\mathrm{d}x\,\mathrm{d}s &= \int_0^t \int \int [(z - x) f' - \tfrac{1}{2}(z - x)^2 f''] p_\varepsilon(z, s + \varepsilon^2|x, s) p_\varepsilon\,\mathrm{d}x\,\mathrm{d}z\,\mathrm{d}s \\ &= \frac{1}{2}\int_0^t \int \int [B(x + BZ) - B(x - BZ)] Z f' p_\varepsilon \Phi(Z)\,\mathrm{d}x\,\mathrm{d}Z\,\mathrm{d}s \\ &\quad + \varepsilon^2 \int_0^t \int \bar{D} f'' p_\varepsilon\,\mathrm{d}x\,\mathrm{d}s + o(\varepsilon^2),\end{aligned} \tag{A8}$$

where we have dropped explicit reference to (x, t). On the interval $[x_0 - B_1 Z, x_0 + B_2 Z]$, B on either side of x_0 will take on the value of B from the remaining side. For all such cases, in the limit,

$$\begin{aligned}\lim_{\varepsilon\to 0} \frac{1}{2\varepsilon^2}\int \int_{x_0 - B_1 Z}^{x_0 + B_2 Z} &\{B[x + B(x)Z] \\ &- B[x - B(x)Z]\} Z f' p_\varepsilon \Phi(Z)\,\mathrm{d}x\,\mathrm{d}Z \\ &= \lim_{\varepsilon\to 0} \frac{1}{2\varepsilon^2}\int \int_{x_0 - B_1 Z}^{x_0 + B_2 Z} (B_2 - B_1) Z f' p_\varepsilon \Phi(Z)\,\mathrm{d}x\,\mathrm{d}Z \\ &= \tfrac{1}{2}(B_2 + B_1)(B_2 - B_1) f' p_\varepsilon|_{x_0} = \tfrac{1}{2}(B_2^2 - B_1^2) f' p_\varepsilon|_{x_0} \\ &= (D_2 - D_1) f' p_\varepsilon|_{x_0}.\end{aligned} \tag{A9}$$

Outside of this interval we have

$$\begin{aligned}\lim_{\varepsilon\to 0} \frac{1}{2\varepsilon^2}\int \int_{x \notin [x_0 - B1Z, x_0 + B2Z]} &[B(x + BZ) - B(x - BZ)] \\ &\cdot Z f' p_\varepsilon \Phi(Z)\,\mathrm{d}x\,\mathrm{d}Z = 0\end{aligned} \tag{A10}$$

because D is constant in this case. From (A9) and (A10) we have, in the limit,

$$\begin{aligned}\lim_{\varepsilon\to 0} \frac{1}{2\varepsilon^2}\int \int [B(x + BZ) - B(x - BZ)] Z f' p_\varepsilon \Phi(Z)\,\mathrm{d}Z\,\mathrm{d}x \\ = (D_2 - D_1) f' p_\varepsilon|_{x_0}.\end{aligned} \tag{A11}$$

Substituting this result into (A8) yields, in the limit,

$$\int_0^t\int f\,\frac{\partial p_\varepsilon}{\partial t}\,\mathrm{d}x\,\mathrm{d}s = \int_0^t\int \bar{D}f''p_\varepsilon\,\mathrm{d}x\,\mathrm{d}s + \int_0^t (D_2 - D_1)f'p_\varepsilon|_{x_0}\,\mathrm{d}s$$

$$= \int_0^t\int Df''p_\varepsilon\,\mathrm{d}x\,\mathrm{d}s + \int_0^t (D_2 - D_1)f'p_\varepsilon|_{x_0}\,\mathrm{d}s, \quad \text{(A12)}$$

where the last equality is justified since singular values of $\bar{D} > 0$ have no effect on the diffusion process. Equation (A12) has the same form as (A3); therefore the density generated by (A4), and version (11a) of this approximation, satisfy (A3) associated with this equation. Similar results can be developed for the algorithm in (17a). In Appendix B we show how this result implies a stochastic calculus with generalized functions.

Appendix B: Stochastic Calculus With Generalized Functions

The forgoing result suggests that one can apply a stochastic calculus with generalized functions to obtain the desired result. For example, in one dimension, expanding (9a) yields

$$\mathrm{d}X = B(X + B\,\mathrm{d}W, t)\,\mathrm{d}W$$

$$= B(X, t)\,\mathrm{d}W + B(X, t)\,\frac{\partial B(X, t)}{\partial x}\,\mathrm{d}W\,\mathrm{d}W$$

$$= B(X, t)\,\mathrm{d}W + \frac{\partial D(X, t)}{\partial x}\,\mathrm{d}t$$

$$= B(X, t)\,\mathrm{d}W + \left.\frac{\partial D(X, t)}{\partial x}\right|_{X\neq x_0}\mathrm{d}t$$

$$+ [D_2(X, t) - D_1(X, t)]\delta(X - x_0)\,\mathrm{d}t, \quad \text{(B1)}$$

which from standard theory corresponds to the diffusion equation

$$\frac{\partial p}{\partial t} = D\,\frac{\partial^2 p}{\partial x^2} + \left[\left.\frac{\partial D}{\partial x}\right|_{x\neq x_0} + (D_2 - D_1)\delta(x - x_0)\right]\frac{\partial p}{\partial x}. \quad \text{(B2)}$$

Equation (B2) is equivalent to (A3) for the problem considered therein.

Appendix C: Taylor Series Expansion of Equation (15a)

When coefficients are smooth, (15a) is a SDE. Here we expand this equation to show that it is, indeed, equivalent to the SDE given in (2a). Expanding the second term in the right-hand side of (15a) in Taylor series for sufficiently smooth D_{ij} and Θ yields:

$$\Theta^{-1/2}(\mathbf{X}, t)\sum_{j,k}\hat{B}_{ijk}\left[X_l + \Theta^{-1/2}(\mathbf{X}, t)\sum_{m,n}\hat{B}_{lmn}(\mathbf{X}, t)\,\mathrm{d}W_m, t\right]\mathrm{d}W_j$$

$$= \Theta^{-1/2}(\mathbf{X}, t)\sum_j\left[\sum_k \hat{B}_{ijk}(\mathbf{X}, t) + \Theta^{-1/2}(\mathbf{X}, t)\right.$$

$$\left.\cdot\sum_{k,l,m,n}\frac{\partial\hat{B}_{ijk}(\mathbf{X}, t)}{\partial x_l}\,\hat{B}_{lmn}(\mathbf{X}, t)\,\mathrm{d}W_m + \cdots\right]\mathrm{d}W_j$$

$$= \Theta^{-1/2}\sum_{j,k}\hat{B}_{ijk}\,\mathrm{d}W_j + \Theta^{-1}\sum_{j,k,l,n}\frac{\partial\hat{B}_{ijk}}{\partial x_l}\,\hat{B}_{ljn}\,\mathrm{d}t + o(\mathrm{d}t)$$

$$= \sum_{j,k}\lambda_k^{1/2}Z_{ijk}\,\mathrm{d}W_j + \Theta^{-1/2}$$

$$\cdot\sum_{j,k,l,n}\frac{\partial}{\partial x_l}(\Theta^{-1/2}\lambda_k^{1/2}Z_{ijk})\lambda_n^{1/2}Z_{ljn}\,\mathrm{d}t + o(\mathrm{d}t)$$

$$= \sum_{j,k}\lambda_k^{1/2}Z_{ijk}\,\mathrm{d}W_j + \Theta^{-1/2}$$

$$\cdot\sum_{j,k,m,n}\frac{\partial}{\partial x_j}(\Theta^{-1/2}\lambda_n^{1/2}Z_{imn})\lambda_k^{1/2}Z_{mjk}\,\mathrm{d}t + o(\mathrm{d}t), \quad \text{(C1)}$$

where we have omitted explicit reference to evaluation of terms at $(\mathbf{X}, t)$ and used the following relationships

$$Z_{ijk} = Z_{jik} \quad \text{(C2a)}$$

$$\sum_k Z_{ijk} = \delta_{ij} \quad \text{(C2b)}$$

$$\sum_m Z_{imn}Z_{mjk} = Z_{ijk}\delta_{nk}, \quad \text{(C2c)}$$

where $\delta_{nk} = 1$ for $n = k$ and 0 otherwise. Expanding the third term on the right-hand side of (15a) in Taylor series yields

$$\Theta^{-1/2}(\mathbf{X}, t)\sum_{j,k,m,n}Z_{imn}(\mathbf{X}, t)\hat{B}_{mjk}[X_l + \lambda_n^{1/2}(\mathbf{X}, t)\,\mathrm{d}W_l, t]\,\mathrm{d}W_j$$

$$= \Theta^{-1/2}(\mathbf{X}, t)\sum_{j,k,m,n}Z_{imn}(\mathbf{X}, t)$$

$$\cdot\left[\hat{B}_{mjk}(\mathbf{X}, t) + \frac{\partial\hat{B}_{mjk}(\mathbf{X}, t)}{\partial x_l}\,\lambda_n^{1/2}(\mathbf{X}, t)\,\mathrm{d}W_l + \cdots\right]\mathrm{d}W_j$$

$$= \Theta^{-1/2}\sum_{j,k,m,n}Z_{imn}\left(\hat{B}_{mjk}\,\mathrm{d}W_j + \frac{\partial\hat{B}_{mjk}}{\partial x_j}\,\lambda_n^{1/2}\,\mathrm{d}t\right) + o(\mathrm{d}t)$$

$$= \sum_{j,k}\lambda_k^{1/2}Z_{ijk}\,\mathrm{d}W_j + \Theta^{-1/2}$$

$$\cdot\sum_{j,k,m,n}\frac{\partial}{\partial x_j}(\Theta^{-1/2}\lambda_k^{1/2}Z_{mjk})\lambda_n^{1/2}Z_{imn}\,\mathrm{d}t + o(\mathrm{d}t), \quad \text{(C3)}$$

where again we have used (C2a)–(C2c) and omitted explicit reference to evaluation of terms at $(\mathbf{X}, t)$. Expressing (15a) in terms of (C1) and (C3), we have

$$\mathrm{d}X_i = v_i\,\mathrm{d}t + \sum_{j,k}\lambda_k^{1/2}Z_{ijk}\,\mathrm{d}W_j$$

$$+ \tfrac{1}{2}\Theta^{-1/2}\sum_{j,k,m,n}\lambda_k^{1/2}Z_{mjk}\,\frac{\partial}{\partial x_j}(\Theta^{1/2}\lambda_n^{1/2}Z_{imn})\,\mathrm{d}t$$

$$+ \tfrac{1}{2}\Theta^{-1/2}\sum_{j,k,m,n}\frac{\partial}{\partial x_j}(\Theta^{1/2}\lambda_k^{1/2}Z_{mjk})\lambda_n^{1/2}Z_{imn}\,\mathrm{d}t + o(\mathrm{d}t), \quad \text{(C4)}$$

which one can show is equivalent to (2a) using the following relationships:

$$\sum_k\lambda_k Z_{mjk} = \sum_{k,m,n}\lambda_n^{1/2}Z_{imn}\lambda_k^{1/2}Z_{mjk} = 2D_{ij} \quad \text{(C5a)}$$

$$\sum_k\lambda_k^{1/2}Z_{ijk} = \sum_{k,m,n}\lambda_k^{1/2}Z_{imn}Z_{mjk} = [(2\mathbf{D})^{1/2}]_{ij} \quad \text{(C5b)}$$

$$\frac{1}{2}\Theta^{-1/2}\left[\sum_{j,k,m,n}\frac{\partial}{\partial x_j}(\Theta^{1/2}\lambda_n^{1/2}Z_{imn})\lambda_k^{1/2}Z_{mjk} + \sum_{j,k,m,n}\lambda_n^{1/2}Z_{imn}\frac{\partial}{\partial x_j}(\Theta^{1/2}\lambda_k^{1/2}Z_{mjk})\right] = \Theta^{-1}\frac{\partial}{\partial x_j}(\Theta D_{ij}) \tag{C5c}$$

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G. E. Fogg and E. M. LaBolle, Hydrologic Sciences, University of California, Davis, One Shields Avenue, Davis, CA 95616. (gefogg@ucdavis.edu; emlabolle@ucdavis.edu)

J. Gravner, Department of Mathematics, University of California, Davis, Davis, CA 95616. (gravner@math.ucdavis.edu)

J. Quastel, Department of Mathematics, University of Toronto, Toronto, Canada M5S 1A1. (quastel@math.ucdavis.edu)

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