Stochastic Models of Solute Transport in Highly Heterogeneous Geologic Media

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A stochastic model of anomalous diffusion was developed in which transport occurs by random motion of Brownian particles, described by distribution functions of random displacements with heavy (power-law) tails. One variant of an effective algorithm for random function generation with a power-law asymptotic and arbitrary factor of asymmetry is proposed that is based on the Gnedenko–Levy limit theorem and makes it possible to reproduce all known Levy α-stable fractal processes. A two-dimensional stochastic random walk algorithm has been developed that approximates anomalous diffusion with streamline-dependent and space-dependent parameters. The motivation for introducing such a type of dispersion model is the observed fact that tracers in natural aquifers spread at different super-Fickian rates in different directions. For this and other important cases, stochastic random walk models are the only known way to solve the so-called multiscaling fractional order diffusion equation with space-dependent parameters. Some comparisons of model results and field experiments are presented.

Dispersion in naturally fractured and porous aquifers is highly complex due to strongly varying velocity fields. Discussions of different modern approaches to transport in such media can be found in Sahimi (1995), National Research Council (1996, 2001), Faybishenko et al. (2000), and Neuman and Di Federico (2003).

Due to spatial fluctuations of seepage velocity in nonuniform media, solute transport is accompanied by dispersion, giving rise to expansion of a contaminant plume. Under certain conditions, when the seepage velocity correlation length is finite, dispersion has a classical Fickian character, and the spatial scale of the solute plume (R) will increase with time (t) as $R \sim t^{1/2}$. In many experimental studies, however, the tracer plume was found to grow in the direction of flow at a super-Fickian rate as $R \sim t^{1/\alpha}$, with $\alpha < 2$ (Glimm and Sharp, 1991; Uchaikin and Gusarov, 1997; Isichenko, 1992; Bouchard, 1995; Bouchard and Georges, 1990; Klafter et al., 1996; Shlesinger et al., 1982, 1993; Zhang et al., 2006a; Matheron and de Marsily, 1980; Lenormand and Wang, 1995; Bolshov et al., 2008).

Another, more complicated behavior of tracer plumes was analyzed in a review by Neuman and Di Federico (2003). They pointed out that plume behavior is not predicted by the classical Fick's law at comparably early times; both longitudinal and transverse dispersivities increase with travel time (or with travel distance). At later times (or large distance), a quasi-Fickian regime is established.

Two macrodispersion natural gradient tracer tests have been performed in real geologic media with strong heterogeneities, at a site on Cape Cod, Massachusetts (LeBlanc et al., 1991) and the macrodispersion experiment (MADE) at the Columbus Air Force Base in Mississippi (Adams and Gelhar, 1992; Boggs et al., 1993). Plume growth in these experiments in the longitudinal direction was considerably faster than expected from the classical Fickian law $R \sim t^{1/\alpha}$, with $1/\alpha = \gamma \sim 0.8$ for the MADE (Adams and Gelhar, 1992; Boggs et al., 1993) and $\gamma \sim 0.6$ at Cape Cod (LeBlanc et al., 1991). The second macrodispersion experiment, MADE-2 (Boggs et al., 1993), revealed that the concentration profile is very skewed in the direction of the average flow, indicating streamline-dependent anomalous diffusion with heavy tails.

A possible approach to anomalous diffusion modeling is provided by the one-dimensional advection–diffusion equation with so-called fractional derivatives (Benson et al., 2001; Saichev and Zaslavsky, 1997; Montroll and Weiss, 1965; Shlesinger et al., 1982; Schumer et al., 2001; Benson, 1998; Samko et al.

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1987; Goloviznin et al., 2002a,b, 2003, 2005b; Mainardi,1997). A variety of numerical methods have been developed recently for modeling superdiffusion with the one-dimensional fractional-order advection–diffusion equation (FADE) (Liu et al., 2004; Meerschaert and Tadjeran, 2004, 2006; Zhang et al., 2005; Yuste and Acedo, 2003; Lynch et al., 2003; Deng et al., 2004; Oldham and Spanier, 1974). The one-dimensional FADE with fractional-order space derivatives has a fundamental solution that has Levy α-stable density. A governing equation for particles that undergo Levy motion rather than classical Brownian motion readily describes skewed and heavy-tailed solute concentration profiles as observed in macrodispersion experiments such as the MADE. Moreover, the one-dimensional FADE model is compatible with observations of solute behavior in the laboratory and in field tests at Cape Cod (Benson et al., 2000, 2001).

Multidimensional solute dispersion modeling using the fractional diffusion equation with diffusion parameter asymmetry encounters difficulties. Methods for directly solving the multidimensional FADE (Benson et al., 2006; Meerschaert et al., 2001) require the scaling exponent $1/\alpha = \gamma$ to be the same in different directions. There is no physical reason for this restriction, which is just a mathematical artifact of the modeling approach. In fact, the scaling exponent usually varies with direction, as suggested by observations in the MADE field experiment (see below). The rate of solute spreading may be faster in the direction of mean flow and slower in the transverse direction. Moreover, different asymmetric factors and space-dependent parameters should be expected and taken into account.

Difficulties in generalizing FADE to multidimensional transport have stimulated the development of random walk methods in recent years (Goloviznin et al., 2005b; Benson et al., 2006; Zhang et al., 2006a,b). The computational efficiency and flexibility of random walk methods overcomes a number of existing difficulties in the formulation and solution of such problems in the multidimensional case. The purpose of this study was to develop a stochastic random walk model that reduces to the multidimensional FADE model in the special case of isotropic, space-independent parameters. We extended the stochastic model to the multidimensional case with spatial anisotropy, and compared model results with field experiments. The treatment presented here was mainly focused on saturated flow systems; an extension to variably saturated conditions is possible but will not be pursued here.

Approximate Algorithm for Random Variable Generation of α-stable Fractal Levy Distribution

The foundation for developing multidimensional stochastic random walk models is an effective algorithm for random function generation with power-law asymptotics and an arbitrary factor of asymmetry. Generation of stable distributions of random variables with heavy tails is hampered by a lack of analytical expressions for the distribution function and its inverse. Only Gaussian, Cauchy, and Levy distributions constitute exceptions and can be represented analytically (Uchaikin and Zolotarev, 1999; Saichev and Zaslavsky, 1997; Kanter, 1975).

An approach to solving this problem was first proposed in Kanter (1975) for distributions with $\alpha < 1$ and subsequently generalized to any α (Chambers et al., 1976). Generators of such type (Kanter, 1975; Chambers et al., 1976) are termed "exact" algorithms. Despite the existence of "exact" algorithms to simulate stable random variables, other approximate methods are often used in practice because they turn out to be more efficient than exact ones. The so-called "approximation generators" are based on the use of the generalized Gnedenko–Levy limit theorem (Uchaikin and Zolotarev, 1999; Chambers et al., 1976; Mantegna, 1994). Such a generator for symmetric distributions was described in Mantegna (1994). For arbitrary asymmetric distributions, such an approximate generator was developed in Zhang et al. (2006a,b) for random walk approximation of fractional-order multiscaling multidimensional anomalous diffusion. The algorithm is based on the use of the generalized limit theorem (Uchaikin and Zolotarev, 1999; Chambers et al., 1976; Mantegna, 1994) and involves taking the sum of independent, identically distributed, Pareto random variables. The researchers (Zhang et al., 2006a,b) found that the approximate method converged faster to the α-stable variables than an algorithm based on method (Janicki and Weron, 1994), but one parameter (the cutoff of Pareto density from constant to the power-law form) significantly affected the rate of convergence.

We developed a method to generate α-stable random variables on the basis of the generalized limit theorem as well. Our method is more reliable than the Pareto method, however, since it does not require choosing a cutoff parameter.

Consider a one-dimensional distribution function F that may represent a solute concentration distribution as may arise from dispersion in a heterogeneous medium. The distribution of sums of independent random variables X_i is said to belong to the domain of attraction of F if there exist some normalizing constants a_n, b_n such that the distribution of

$$\left(X_1 + X_2 + \ldots + X_n\right)/a_n - b_n \qquad [1]$$

converges to F as $n \to \infty$. It is rather interesting that all so-called stable distributions, and only these, can be obtained as such limits. Distributions of such type are also known as Levy flight distributions or α-stable fractal Levy distributions.

The generalized limit theorem (Feller, 1971) asserts that for any n random variables $X_1, X_2, \ldots, X_n$ with the same power-law dependence for the tails, the domain of attraction is an α-stable fractal Levy distribution with heavy power-law tails if $0 < \alpha < 2$. We propose a new generator for random variables for arbitrary values of the parameter α that is given by

$$X_i = A\left(\frac{1}{y_i^{1/\alpha}} - 1\right)\mathrm{sign}\left(z_i\right) \qquad [2]$$

where y_i and z_i are random variables uniformly distributed in the interval (0,1), $A > 0$ is an arbitrary numerical constant, and

$$\mathrm{sign}\left(z_i\right) = -1 \qquad [3a]$$

if $z > \beta_1$,

$$\mathrm{sign}\left(z_i\right) = 1 \qquad [3b]$$

if $z_i \le \beta_1$, where β_1 is an arbitrary numerical constant in the interval (0,1).

It is not difficult to see that the distribution functions for $0 < \alpha \le 2$ and $x \to \infty$ are

$$F(-x)=\frac{A^{\alpha}}{(x+A)^{\alpha}}(1-\beta_1)\sim(1-\beta_1)A^{\alpha}x^{-\alpha} \quad [4a]$$

$$F(x)=1-\frac{A^{\alpha}}{(x+A)^{\alpha}}\beta_1\sim 1-\beta_1 A^{\alpha}x^{-\alpha} \quad [4b]$$

Probability densities are defined by the relations

$$p(-x)=\frac{A^{\alpha}}{(x+A)^{\alpha+1}}\alpha(1-\beta_1) \quad [5a]$$

$$p(x)=\frac{A^{\alpha}}{(x+A)^{\alpha+1}}\alpha\beta_1 \quad [5b]$$

Thus, the random variables X_i have "heavy" (power-law) tails, and the domain of attraction is an α-stable fractal Levy distribution for $0 < \alpha < 2$. Moreover, for $\beta_1 = 0.5$ we obtain a symmetric distribution, while for $\beta_1 = 0$ or $\beta_1 = 1$ the distributions are fully asymmetric.

In accordance with the generalized limit theorem, normalizing constants are determined by

$$a_n = n^{1/\alpha}$$
$$b_n = \mathrm{E}(X_i)n^{1-1/\alpha} \quad [6]$$

$$\mathrm{E}(X_i)=\frac{A}{1-\alpha}(1-2\beta_1) \quad [7]$$

for $1 < \alpha \le 2$, when the expectation value $\mathrm{E}(X_i)$ exists and centering is required, and as

$$a_n = n^{1/\alpha}$$
$$b_n = 0 \quad [8]$$

for $0 < \alpha < 1$ when $\mathrm{E}(X_i)$ does not exist. The α-stable Levy distributions with heavy tails can be asymmetric relative to zero; the asymmetry factor is given by

$$\beta_1 = \lim_{x\to\infty}\frac{1-F(x)}{1-F(x)+F(-x)} \quad [9]$$

or

$$1-\beta_1 = \lim_{x\to\infty}\frac{F(-x)}{1-F(x)+F(-x)} \quad [10]$$

Equations [1–8] provide a means to construct a generator of stable random variables as the sum of a "sufficiently" large number of X_i. The approach given above is one of several possible algorithms that were explored and tested for generating random variables with heavy tails for the entire range of parameters α and β_1.

Rate of Convergence of Stable Random Variables Generator

The algorithm set forth above was implemented numerically. The rate of convergence of the distribution of random variable sums to stable distributions as a function of n was investigated for various values of the parameters α and β_1, including the cases for which analytical expressions for the probability density exist. Analytical expressions for the probability density of stable distributions are presently known only for two values of the parameter α. For $\alpha = 1$ we have the symmetric Cauchy distribution (Feller, 1971):

$$p(x)=\frac{1}{\pi}\frac{t}{t^2+x^2} \quad [11]$$

and for $\alpha = 0.5$ the asymmetric Levy distribution (Feller, 1971):

$$p(x)=\frac{t}{\sqrt{2\pi}}\frac{1}{x^{3/2}}\exp\left(-\frac{t^2}{2x}\right) \quad [12]$$

The number of X_i or the set length n in Eq. [1] will be denoted as "njump" in the subsequent discussion and illustrative figures. The number of different stable random variables used for the construction of probability densities is denoted as n_j.

It was found that for $0 < \alpha < 1$ the generator converges rapidly for all values of β_1. For greater asymmetry, however, a larger data set is required for convergence. If $0 < \alpha \le 0.5$, then a set length n = njump = 10 is sufficient even for practically fully asymmetric cases ($\beta_1 = 0.1$). Further increase of the set length does not lead to any noticeable variation in probability density for the set (see Fig. 1). At large distances, the distribution has a power-law form (Fig. 2). In the range $0.5 < \alpha < 1$, the required set length increases n = njump = 50 is acceptable for the fully asymmetric case, $\beta_1 = 1$ (Fig. 3). Comparison of numerical results obtained from the approximate stable random variables generator with the

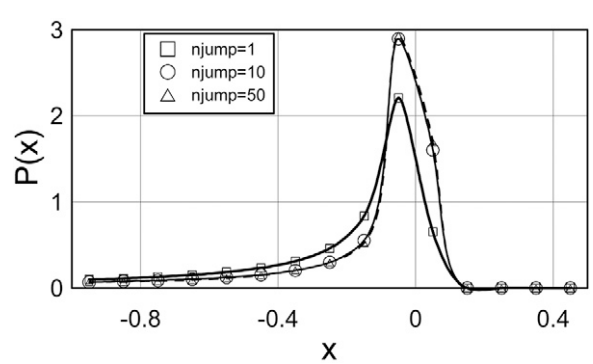

FIG. 1. Probability density $p(x)$ for sums of random numbers generated from Eq. [2], demonstrating convergence to a stable distribution. Parameters are constants $A = 0.5$ and $\beta_1 = 0.1$, and the inverse exponent of time $\alpha = 0.2$. The number of particles is n_j = 200,000; njump is the set length.

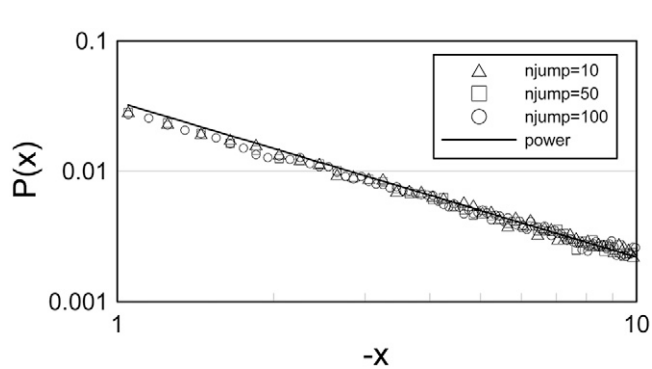

FIG. 2. Probability density $p(x)$ for sums of random numbers generated from Eq. [2] on a log–log scale for $-x > 1$, demonstrating approximate power-law behavior. Parameters are constants $A = 0.5$ and $\beta_1 = 0.1$, and the inverse exponent of time $\alpha = 0.2$. The number of particles is n_j = 200,000; njump is the set length.

Cauchy (Eq. [11]) and Levy (Eq. [12]) analytical distributions shows good agreement (Fig. 4 and 5).

As α increases in the range $1 < \alpha < 2$, the rate of convergence decreases. It was found that, similar to the case $0 < \alpha < 1$, for greater asymmetry a larger set length is required for convergence. For the asymmetrical case with $\alpha = 1.5$ and $\beta_1 = 1$, the required set length is njump = 100, while for the symmetrical case njump = 30 to 50 is sufficient (see Fig. 6). For the $\alpha = 1.9$ asymmetrical case, the acceptable set length was found to be njump = 200. Thus, the new random variable generator proposed here is applicable for the entire range of α and β. The rate of convergence is higher for lower α.

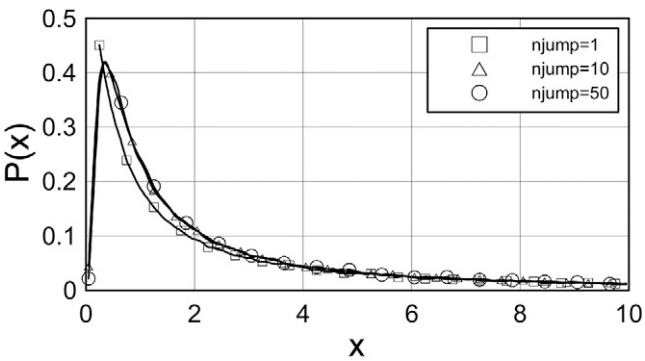

FIG. 3. Probability density $p(x)$ for sums of random numbers generated from Eq. [2], demonstrating convergence to a stable distribution. Parameters are constants $A = 0.7$ and $\beta_1 = 1$, and the inverse exponent of time $\alpha = 0.5$. The number of particles is n_j = 200,000; njump is the set length.

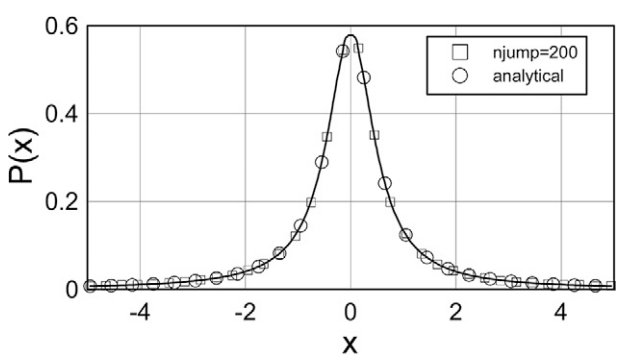

FIG. 4. Comparison of numerical results obtained from Eq. [1–8] with the analytical formula Eq. [11]. Parameters are constants A = 0.5 and $\beta_1 = 0.5$, and the inverse exponent of time $\alpha = 1$ (symmetric Cauchy distribution). The number of particles is n_j = 200,000; the set length is njump = 200.

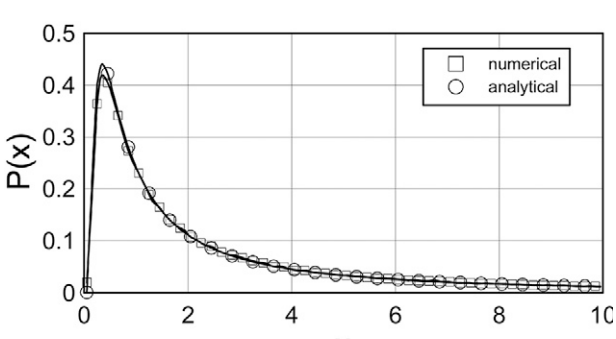

FIG. 5. Comparison of numerical results obtained from Eq. [1–8] with the analytical formula Eq. [12]. Parameters are constants A = 0.7 and $\beta_1 = 1$, and the inverse exponent of time $\alpha = 0.5$ (asymmetric Levy distribution). The number of particles is n_j = 200,000; the set length is njump = 50.

Stochastic One-Dimensional Nonstationary Model of Solute Dispersion with Heavy Tails

We now compute the migration of a large number of solute particles that, at every time step, experience random displacements drawn from a stable Levy distribution. The stochastic equation for particle coordinates in the random walk process is given by

$$x_i^{n+1} = x_i^n + \Delta t^{1/\alpha} \xi_i \qquad [13]$$

where x_i^n is the coordinate of the ith particle at the nth moment of time, ξ_i is a random variable that is obtained from the suggested algorithm Eq. [1–8] for generation of stable random variables, Δt is the time step, and α is the parameter of the power-law distribution.

Equation [13] provides the particle displacement during one time step. In the case of α-stable distributions of ξ_i, the distribution of particle displacements for arbitrary time t does not depend on the time step. It further follows that the spatial scale of the particle distribution changes with time as $x \sim t^{1/\alpha}$. In other words, the motion is self-similar (fractal), and the self-similarity coordinate has the form $x/t^{1/\alpha}$. Figure 7 compares modeling

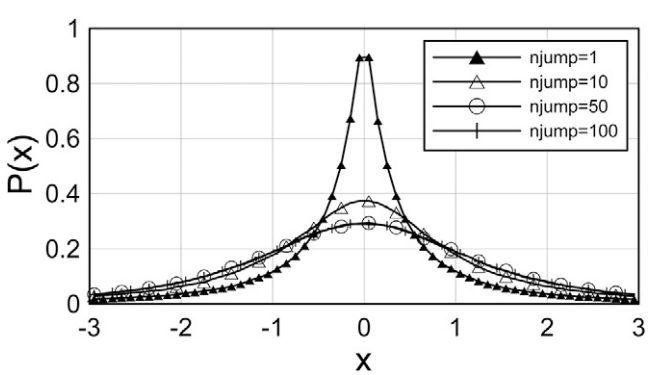

FIG. 6. Convergence of random numbers to stable distribution. Parameters are constants A = 0.5 and $\beta_1 = 0.5$, and the inverse exponent of time $\alpha = 1.5$. The number of particles is n_j = 200,000; njump is the set length.

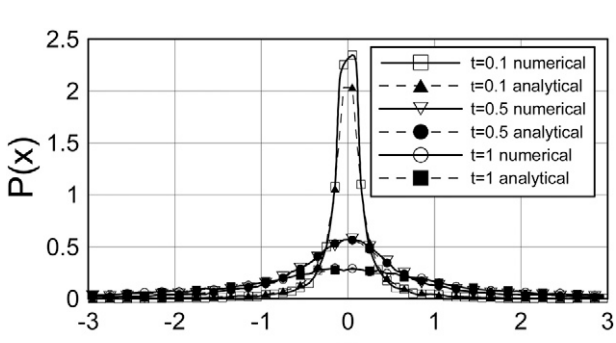

FIG. 7. Solute concentration (probability density [$p(x)$] of particle locations) obtained from a stochastic (numerical) model and from an analytical solution (Cauchy distribution) at different points of time t. The inverse exponent of time $\alpha = 1$ and constant $\beta_1 = 0.5$.

results from Eq. [13] with the known analytical solution for $\alpha = 1$ (symmetrical Cauchy distribution) at different times. The agreement is good.

Figure 8 presents an asymmetrical distribution for parameters $\alpha = 0.5$, $\beta_1 = 0.1$ at time $t = 0.1$, with time step size $\Delta t = 0.001$, and with n = njump = 1, 10, and 30 taken as approximations of stable random variables ξ_i. As was mentioned above, it is not necessary to attain a very good approximation of the stable random variable at each time step. The greater the time step, the more accurate the approximation should be. As the time step is reduced, the sum of displacements will have a distribution that tends to the exact stable Levy distribution. After 100 time steps, results are virtually independent of set length. Hence, for the solution of practical problems, it is not necessary to use the ideal approximation of strongly stable distributions at every time step.

The particle displacement algorithm presented here can be used to model solute transport at field sites. An approach was developed to estimate the parameters A, α, and β from observations (Goloviznin et al., 2005a). The inverse problem of identification of stochastic model parameters from actual measurements can be solved by means of neural networks. Neural networks with different architecture have been considered and recommendations have been developed based on practical measurements and numerical solutions of the forward problem (Goloviznin et al., 2005a).

Two- and Three-Dimensional Stochastic Random Walk Models

Generalization of the one-dimensional anomalous diffusion model presented above to the multidimensional case is straightforward: assuming that particle motions along different directions are independent, we obtain the equation system for the three-dimensional case:

$$x_1^{n+1} = x_i^n + \Delta t^{1/\alpha_1}\xi_i \qquad [14a]$$

$$y_i^{n+1} = y_i^n + \Delta t^{1/\alpha_2}\eta_i \qquad [14b]$$

$$z_i^{n+1} = z_i^n + \Delta t^{1/\alpha_3}\mu_i \qquad [14c]$$

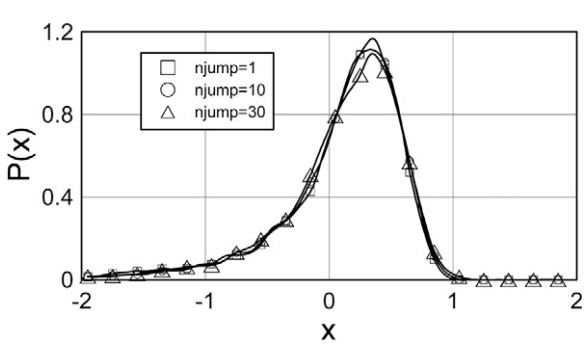

FIG. 8. Asymmetrical distribution of solute concentrations at time $t = 0.1$ for the inverse exponent of time $\alpha = 0.5$, constant $\beta = 0.1$, and time step $\Delta t = 0.001$. Curves are shown for sets with different lengths (njump).

where ξ_i, η_i, and μ_i are α-stable, one-dimensional random variables with heavy tails. These variables will, in general, have different distribution parameters α and β_1.

Two versions of two-dimensional stochastic random walk models were developed. The first version uses Eq. [14a] and [14b], while the second version assumes radial symmetry and is defined by

$$x_i^{n+1} = x_i^n + \Delta t^{1/\alpha}\xi_i \cos\theta_i \qquad [15a]$$

$$y_i^{n+1} = y_i^n + \Delta t^{1/\alpha}\xi_i \sin\theta_i \qquad [15b]$$

where ξ_i are random variables with stable distribution and θ_i is a random variable that is independent of the time step and is uniformly distributed in the interval $[0,\pi]$.

To achieve smooth concentration distributions with a relatively small number of test particles, we interpreted each particle as representing a cubic volume with uniformly distributed density. The contribution of each particle to solute concentration in a given computational grid cell is considered proportional to the share of its cubic volume within the cell. The volume associated with a solute "particle" can vary from one to two grid cells.

Analytical solutions for concentration distributions at different times are shown in Fig. 9 and 10 for the case $\alpha_1 = 0.5$, $\beta_1 = 0$

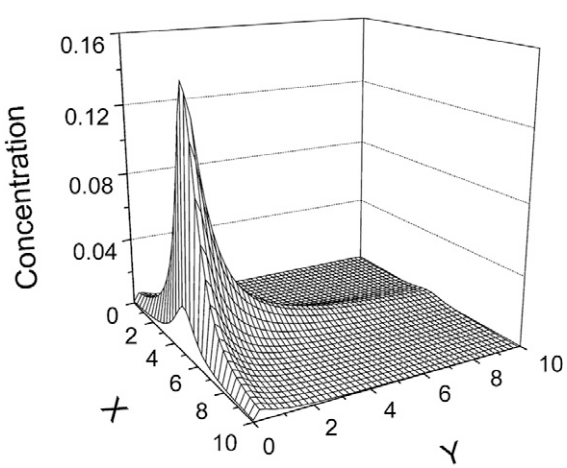

FIG. 9. Analytical solution of solute concentrations at time $t = 1$.

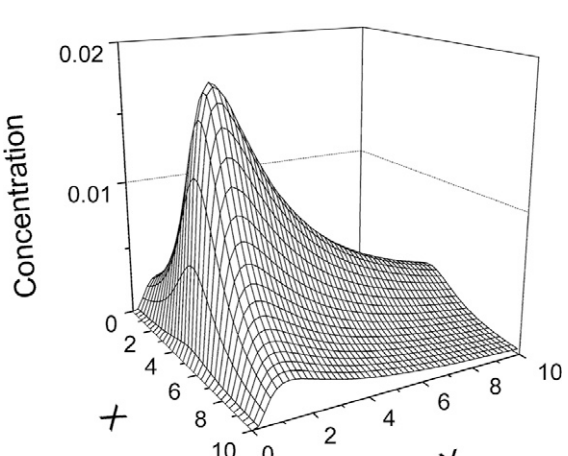

FIG. 10. Analytical solution of solute concentrations at time $t = 2$.

(x axis), and $\alpha_2 = 1$, $\beta_1 = 0.5$ (y axis). Figures 11 and 12 show concentration distributions calculated at different times from the stochastic model (Eq. [14]) with parameters $A = 0.63$, njump = 50, $n_j = 700{,}000$, and $\Delta t = 0.1$. Agreement of the stochastic model with the known analytical Cauchy and Levy solutions is good.

On Statistical Properties of Fractal Levy Flights

It is of interest to consider moments of lower orders for the two- and three-dimensional models developed, including moments of "fractional order" γ that are defined as

$$\left\langle x^{\gamma} \right\rangle = 2\int_0^{\infty} c(x) x^{\gamma} \mathrm{d}x \qquad [16]$$

where $\gamma < \alpha$. It will be seen that certain moments of fractional order exist and can, in principle, be computed.

Let us note that for classical diffusion in one dimension, the mathematical expectation

$$\begin{aligned} \left\langle x^{\gamma} \right\rangle &= 2\int_0^{+\infty} x^{\gamma} \frac{1}{\sqrt{2\pi t}} \exp\left(-\frac{x^2}{2t}\right) \mathrm{d}x \\ &= 2t^{\gamma/2} / \sqrt{\pi} \int_0^{+\infty} 2^{\gamma/2} \left(\frac{x^2}{2t}\right)^{\gamma/2} \exp\left(-\frac{x^2}{2t}\right) \mathrm{d}\frac{x}{\sqrt{2t}} \\ &= t^{\gamma/2} \mathrm{const} \end{aligned} \qquad [17]$$

for any γ.

As already noted, the concentration distribution is self-similar, i.e., it depends only on the similarity variable $x/t^{1/\alpha}$ (in ordinary diffusion, the similarity variable is $x/t^{1/2}$).

In general, there are no analytical expressions for the probability density of Levy flights except for the two cases noted above. Based on the self-similarity of probability density, however, for one-dimensional problems the time dependence of distribution moments can be established. From the normalizing condition

$$C\int_{-\infty}^{+\infty} p\left(x/t^{1/\alpha}\right) \mathrm{d}x = 1 \rightarrow C = t^{-1/\alpha} \qquad [18]$$

it follows that the normalizing constant is proportional to $t^{-1/\alpha}$. Then

$$\begin{aligned} \left\langle x^{\gamma} \right\rangle &= 2t^{-1/\alpha} \int_0^{+\infty} x^{\gamma} p\left(\frac{x}{t^{1/\alpha}}\right) \mathrm{d}x \\ &= 2t^{\gamma/\alpha} \int_0^{+\infty} \left(\frac{x}{t^{1/\alpha}}\right)^{\gamma} p\left(\frac{x}{t^{1/\alpha}}\right) \mathrm{d}\frac{x}{t^{1/\alpha}} \\ &= t^{\gamma/\alpha} \mathrm{const} \end{aligned} \qquad [19]$$

This relation holds only for those values of γ for which the improper integral converges at $\gamma < \alpha$. As already shown, for classical diffusion, $\langle x^{\gamma} \rangle \sim t^{\gamma/2}$, and $\langle x^{\gamma} \rangle^{1/\gamma} \sim t^{1/2}$. For anomalous diffusion with Levy flights,

$$\left\langle x^{\gamma} \right\rangle^{1/\gamma} \sim t^{1/\alpha} \quad (\gamma < \alpha, 0 < \alpha < 2) \qquad [20]$$

For example, if $\alpha > 1$, we obtain $\langle x \rangle \sim t^{1/\alpha}$.

It follows from Eq. [20] that the expectation value $\langle x^{\gamma} \rangle$ for anomalous diffusion increases with time more rapidly than for classical diffusion. Let us consider the results of numerical simulation of two-dimensional anomalous diffusion (model type Eq. [14a–14b]) and try to approximate the expectation value $\langle r^{0.1} \rangle^{10} = [\langle (x^2 + y^2)^{0.05} \rangle]^{10}$ by the function of time t^{pw} const, where pw is an unknown index. Figure 13 presents the results of approximation pw on the basis of the linear regression method for the two-dimensional model given by Eq. [14a–14b] ($\beta_1 = 0.5$, different α). The solid line depicts the theoretical curve pw = $1/\alpha$, while square markers show numerical results. These relations can be readily extended to three dimensions.

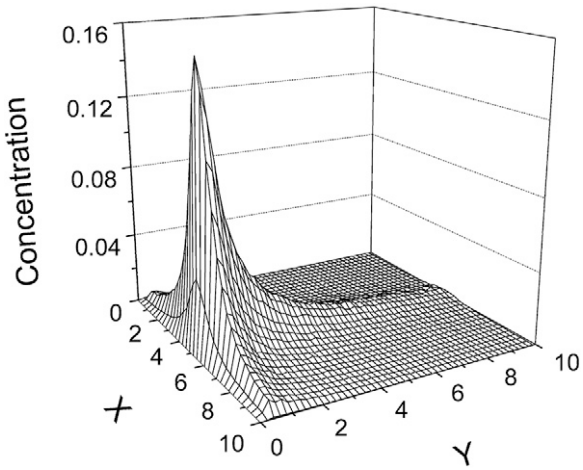

FIG. 11. Stochastic random walk model of solute concentrations at time $t = 1$.

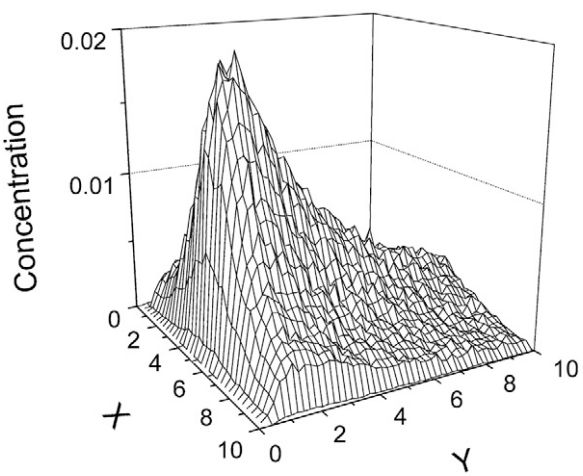

FIG. 12. Stochastic random walk model of solute concentrations at time $t = 2$.

Comparison of Stochastic Model and Fractional Diffusion Model

Results for the one- and two-dimensional stochastic transport problem were compared with calculations made on the basis of the FADE. Specifically, we used a discretized version of the Riemann–Liouville fractional diffusion model with higher order accuracy (see the Appendix) for comparison with the one-dimensional stochastic model. Computations were performed with equal steps in time and space. For the one-dimensional case, concentration profiles from the stochastic model and the fractional derivative model virtually coincide [$\beta_1 = (1 + \beta)/2$]). Even for fully asymmetrical distributions ($\beta_1 = 0$ or $\beta = 1$), at $\alpha = 1.5$ a generator set of length 10 is quite sufficient (Fig. 14).

Figures 15 and 16 show the results for the symmetric FADE in two dimensions, while Fig. 17 and 18 present results for the

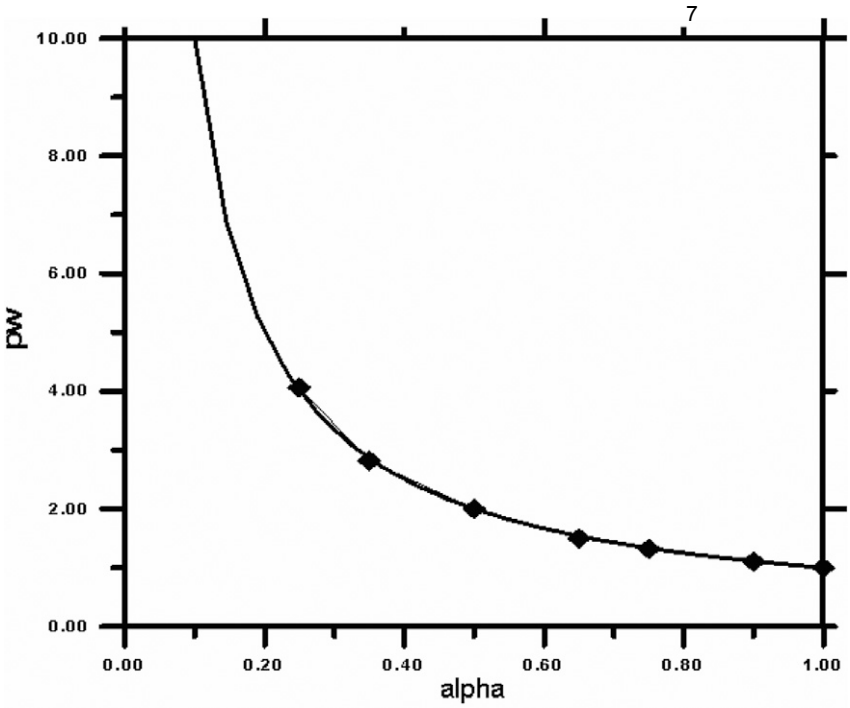

FIG. 13. Dependence of the index pw on the inverse exponent of time α: theoretical (solid line) and computational results for the two-dimensional model, Eq. [14a–14b] (symbols).

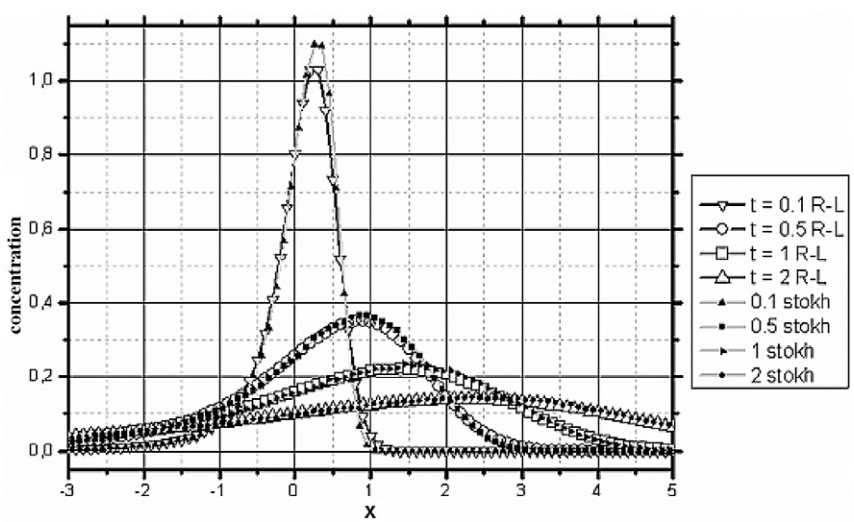

FIG. 14. Comparison of solute concentration profiles obtained with the fractional diffusion model based on Riemann–Liouville (R-L) with results from a stochastic model (stokh) at different times *t*. The length of the set for the stochastic model is njump = 10.

corresponding stochastic model with njump = 50 and n_j = 50,000. Results for the two-dimensional stochastic model (Eq. [14]) practically coincide with the FADE for streamline uniform and space-independent parameters (Goloviznin et al., 2005b).

Comparison of Two-Dimensional Stochastic Model Results and Field Experiment

We present the results of fitting the two-dimensional random walk model to solute concentration data of large-scale field experiments at the MADE site.

Detailed studies were performed for the MADE-1 and MADE-2 experiments to characterize the spatial variability of the aquifer and the spreading of the conservative tracer plume (Adams and Gelhar, 1992; Boggs et al., 1992, 1993). These studies documented the dramatically non-Gaussian behavior and anomalous spreading of the plume (Adams and Gelhar, 1992; Boggs et al., 1992, 1993).

The aquifer at the MADE site resides in a sand–gravel mixture containing clays and alluvial deposits. It is extremely heterogeneous, with a large spread of local permeability values.

The experiments injected about 10 m^3 of water that simultaneously contained different tracers, including conservative tracers such as bromide and tritium (tritiated water). Observations lasted from 15 to 20 months in different experiments. Dispersion of a passive contaminant (bromide and tritium) is in fact two dimensional (in the horizontal direction) with small-scale vertical spreading.

At the MADE site, the separation of the peak and mean position was evident, especially at later times.

The comparison of MADE-2 data and model results for the concentration peak and the center of mass distances from the source is shown in Fig. 19 (the center of mass of the tracer plume position is defined as $X_c(t) = [\int c(x,t)x\mathrm{d}x]/[\int c(x,t)\mathrm{d}x]$, where $c(x,t)$ is the relative concentration and x is the coordinate along the mean flow).

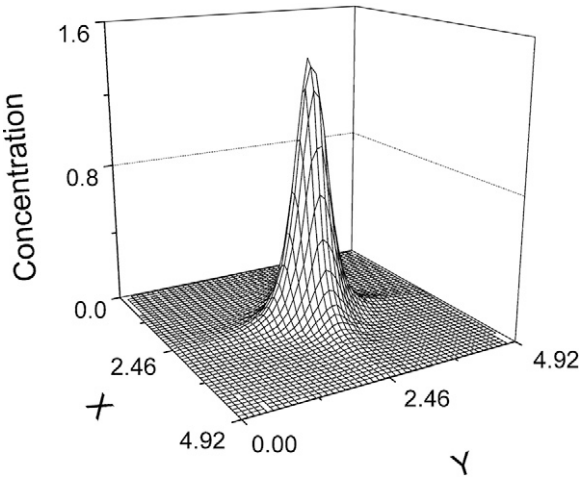

FIG. 15. Concentration distribution from the stochastic model, with the inverse exponent of time α = 1.5 at time t = 0.5.

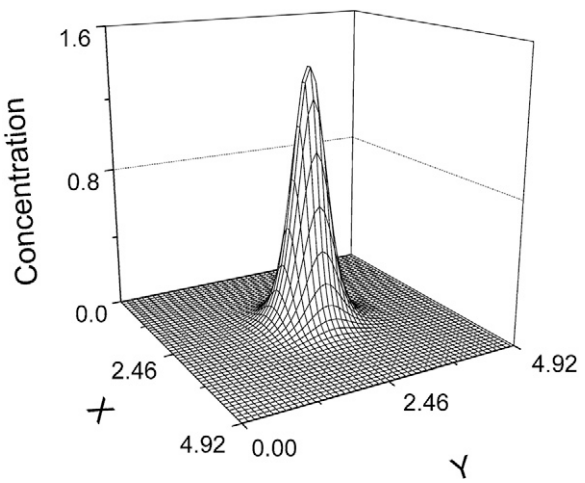

FIG. 16. Concentration distribution from the fractional diffusion model, with the inverse exponent of time α = 1.5 at time t = 0.5.

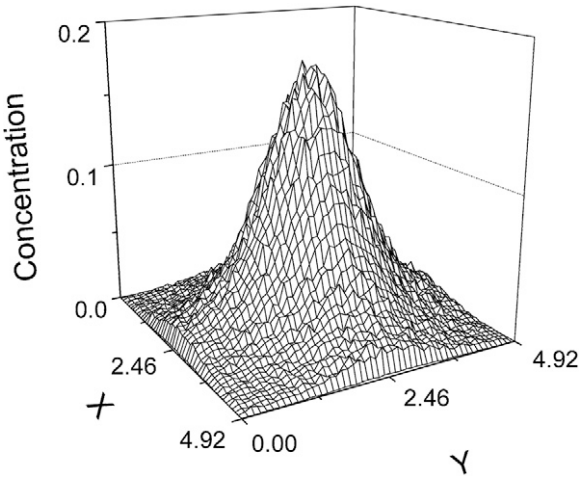

FIG. 17. Concentration distribution from the stochastic model, with the inverse exponent of time α = 1.5 at time t = 0.5.

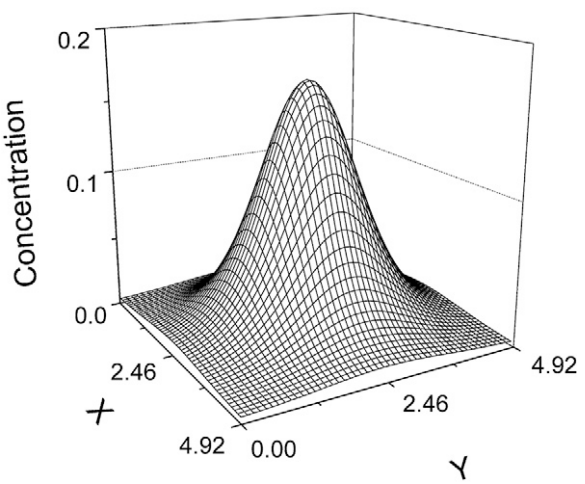

FIG. 18. Concentration distribution from the fractional diffusion model, with the inverse exponent of time α = 1.5 at time t = 0.5.

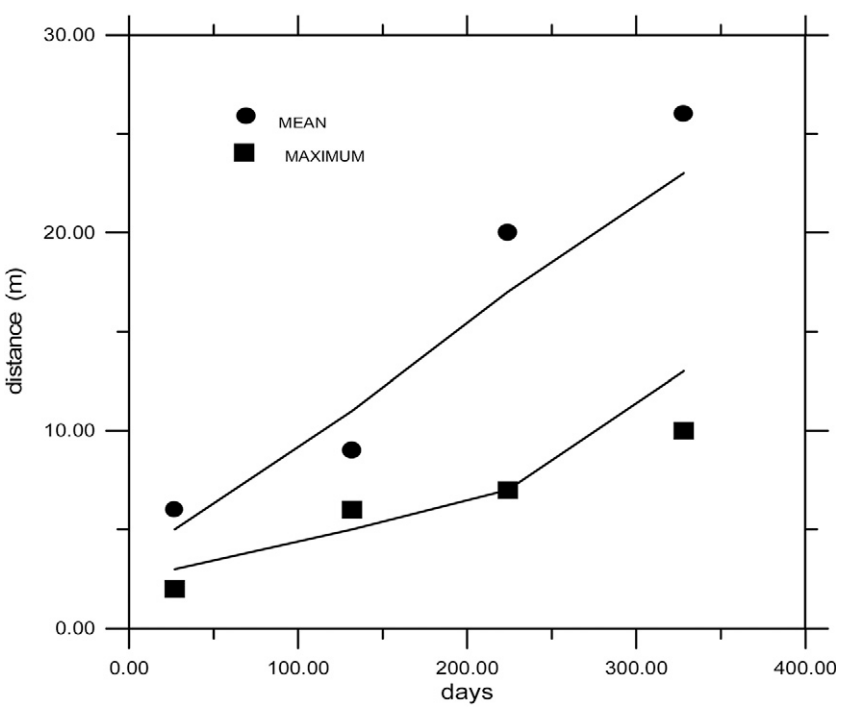

FIG. 19. The second macrodispersion experiment (MADE-2) and two-dimensional stochastic model results. Concentration peak (maximum) and center of mass (mean) positions at different times. Solid lines are two-dimensional stochastic model fit, symbols are the data of observations.

The mean drift velocity was chosen as 0.22 m d^{-1} according to estimations given in Adams and Gelhar (1992) and Boggs et al. (1992, 1993). The index in the power law of time dependence of plume size is $\gamma = 1/\alpha_1 \sim 0.9$ for the longitudinal direction (along the plume) and $\gamma_2 = 1/\alpha_2 \sim 0.6$ for the transverse direction; the asymmetry factor is $\beta_1 = 1$ (strongly asymmetric) for the longitudinal direction and $\beta_1 = 0.5$ (symmetric) for the transverse direction; the diffusivity value is $A = 0.03$ for both directions.

Figure 20 shows the maximal values of concentration in the tracer plume at different times (MADE-1). Open squares correspond to measured concentrations averaged across the vertical direction. Open circles are the observed absolute maximal values of the tracer concentration in the plume. Solid circles are the same, calculated using the two-dimensional random walk model. The best correspondence with experimental data was obtained for $\alpha_1 = 1.1$ and $\gamma_1 = 1/\alpha_1 \sim 0.9$ for the longitudinal direction and $\alpha_2 = 1.8$ and $\gamma_2 = 1/\alpha_2 \sim 0.6$ for the transverse direction.

It can be seen from the figures that the observed and model data corresponding to the vertical averaged concentration are close and have the same slope.

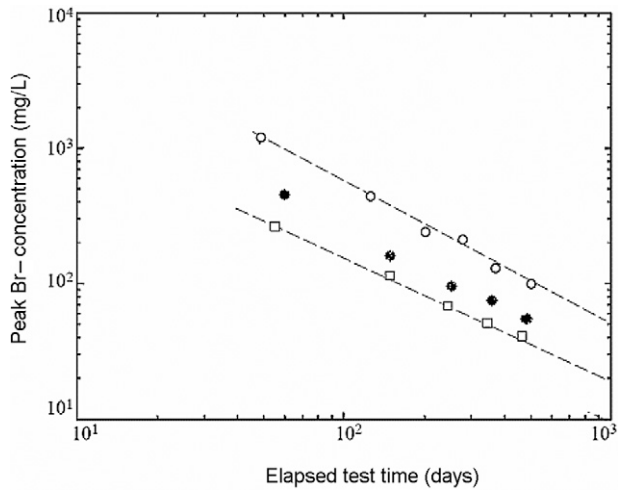

FIG. 20. The first macrodispersion experiment (MADE-1) and two-dimensional stochastic model results. Open squares correspond to measured concentrations averaged across the vertical direction. Open circles are the observed absolute maximal values of tracer concentration in the plume. Solid circles are the same calculated using a two-dimensional random walk model.

Conclusions

A model based on the one-dimensional FADE was compatible with observations of solute plume behavior in macroscale field tests. This model describes skewed and heavy-tailed solute concentration profiles, in general agreement with field and laboratory observations in some highly heterogeneous geologic media. This model, however, is limited to one space dimension.

Multidimensional solute dispersion modeling using FADE with asymmetry and streamline- and space-dependent parameters encounters a number of difficulties. This has motivated the development of random walk methods in recent years as a multidimensional extension of the FADE model. The computational efficiency and flexibility of random walk methods overcomes a number of existing difficulties in the formulation and solution of solute transport problems in the multidimensional case. We developed a novel stochastic random walk model based on an effective algorithm for random numbers with a power-law asymptotic and arbitrary factor of asymmetry. The generator belongs to the class of so-called approximation generators that are based on the Gnedenko–Levy central limit theorem. One-, two-, and three-dimensional stochastic models of solute spreading were developed on the basis of this generator.

Solutions of the one-dimensional and symmetric two-dimensional stochastic problems were compared with calculations made on the basis of FADE models. It was shown that concentrations obtained with the stochastic model agree well with solutions of the fractional diffusion equation (when such solution may be found). Moreover it was shown that the new model is in reasonable agreement with experimental data on solute transport in highly heterogeneous media.

Appendix

Finite Difference Approximation for the Anomalous Diffusion Equation with Riemann–Liouville Fractional Derivatives

The one-dimensional diffusion equation in which the second order of differentiation with respect to space is replaced with the fractional derivative takes the following form (Benson et al., 2001; Benson, 1998; Samko et al., 1987; Goloviznin et al., 2002a,b, 2003):

$$\frac{\partial c(x,t)}{\partial t} = \mathrm{D} D_x^{\alpha} c(x,t) + \frac{\partial uc(x,t)}{\partial x}, \; 1 \le \alpha \le 2$$

$$D_x^{\alpha} = \frac{1}{2}(1+\beta)\frac{\partial^{\alpha}}{\partial x^{\alpha}} + \frac{1}{2}(1-\beta)\frac{\partial^{\alpha}}{\partial(-x)^{\alpha}}, \; -1 \le \beta \le 1 \qquad \text{[A1]}$$

where the function $c(x,t)$ characterizes solute concentration, D_x^{α} is the operator of fractional order α for differentiation with respect to x, $\mathrm{D} > 0$ is a constant of dimension $L^{\alpha}T^{-1}$ (generalized diffusivity), β is a "skewness" coefficient, and x and t are spatial and temporal variables, respectively. For $\alpha \to 2$, the fractional derivative operator approaches the differential operator of the second order that corresponds to ordinary diffusion with exponential decay of solute concentrations at infinity.

There exist several alternative approaches for defining derivatives of fractional order (Benson et al., 2001; Benson, 1998; Samko et al., 1987; Goloviznin et al., 2002a,b, 2003; Liu et al., 2004; Meerschaert and Tadjeran, 2004; Zhang et al., 2005; Yuste and Acedo, 2003; Lynch et al., 2003; Deng et al., 2004). Most commonly used are a generalization of the differentiation operator in the Fourier space, the Grünwald–Letnikov definition, and the Riemann–Liouville definition.

The Fourier method is rather accurate, but is applicable only for periodic boundary conditions. The Grünwald–Letnikov difference scheme does not provide sufficient accuracy for values of the fractional parameter α close to 1 (Goloviznin et al., 2002a,b). An increase in the number of nodes (refinement of grid spacing h) partially solves the problem, but may lead to an unacceptable increase in computational work.

For finite difference solution, we prefer the Riemann–Liouville definition, which includes an integral that can be numerically computed with any specified accuracy. To obtain a second-order accurate spatial approximation, fractional flows referred to the centers of computational grids should be approximated to the third order by the method of trapezoids.

Let us represent the fractional diffusion equation in finite difference form and consider flow ${}^{+\alpha}F_{i+1/2}{}^{n}$. In accordance with the Riemann–Liouville definition, after writing the integral as a sum of integrals on segments bounded by computational grid nodes, we obtain

$$
{}^{+\alpha}F^{n}_{i+1/2} = \frac{1}{\Gamma(2-\alpha)}\frac{\mathrm{d}}{\mathrm{d}x_{i+1/2}}\left[\sum_{k=0}^{i-1}\int_{x_k}^{x_{k+1}}\frac{C(t)\,\mathrm{d}t}{\left(x_{i+1/2}-t\right)^{\alpha-1}} + \int_{x_i}^{x_{i+1/2}}\frac{C(t)\,\mathrm{d}t}{\left(x_{i+1/2}-t\right)^{\alpha-1}}\right], \quad x_0 = a \qquad \text{[A2]}
$$

Flow ${}^{-\alpha}F_{i+1/2}{}^{n}$ can be written in a similar way. To obtain a second-order accurate method of approximation with respect to h, it is sufficient to represent the function $C(x)$ in the form of continuous piecewise-linear functions $\beta_k x + \gamma_k$, where $x \in [x_k, x_{k+1}]$, $\beta_k = (C_{k+1} - C_k)/h$, and $\gamma_k = C_k - \beta_k x_k$. With this, the integration in Eq. [A2] can be performed analytically.

Figure A1 presents concentration profiles obtained with different numerical methods. For small values of the parameter α, a solution obtained with the difference scheme of the first order of spatial approximation (finite-difference method based on Grünwald–Letnikov) shows considerable differences from the more accurate Fourier method. The profile computed from the method of spline approximation to the Riemann–Liouville definition virtually coincides with the Fourier method.

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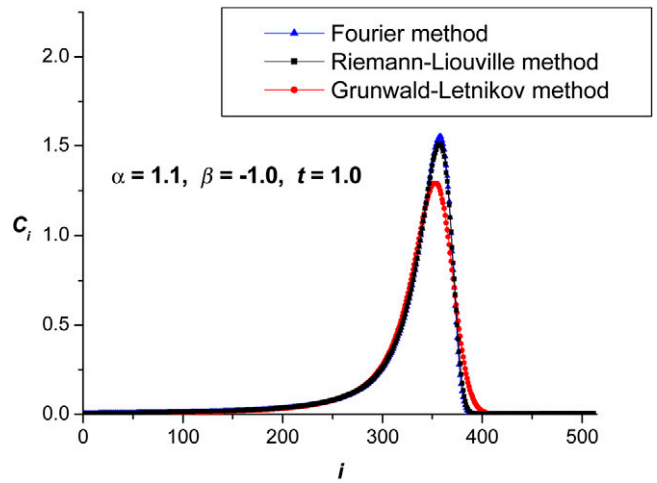

FIG. A1. Profiles of solute concentration (C_i) obtained as solutions to the fractional diffusion equation, with fractional spatial derivatives obtained by the Fourier, modified Riemann–Liouville, and Grünwald–Letnikov methods at the given values of the inverse exponent of time α, constant β, and time t.

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