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On using particle tracking methods to simulate transport in single-continuum and dual continua porous media

Ahmed E. Hassan[a,b,*], Mohamed M. Mohamed[c]

[a]*Division of Hydrologic Sciences, Desert Research Institute, 755 E. Flamingo Road, Las Vegas, NV 89119, USA*
[b]*Irrigation and Hydraulics Department, Faculty of Engineering, Cairo University,Giza, Egypt*
[c]*Department of Civil Engineering, University of Florida, Gainesville, FL 32611, USA*

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Abstract

The use of particle tracking methods to predict transport in single continuum and dual continua (mobile–immobile systems and fractured media) is studied. The accuracy of the particle tracking methods with different interpolation and tracking techniques is evaluated, and its transport predictions are compared to analytical solutions, finite element solutions (e.g. SUTRA) and finite difference solution (e.g. MT3D). For a two-dimensional problem with homogeneous conductivity and pulse injection of contaminant, the particle tracking solution matches the analytical solution better than those using standard finite difference and finite element techniques, which suffer from numerical dispersion. Furthermore, the particle tracking method accurately predicts the mean and variance of the stochastic concentration distribution and compares favorably with MT3DMS that employs a total variance diminishing technique for discretizing the advection term. For modeling matrix diffusion in fractured media and mass transfer in dual porosity (mobile–immobile) systems, two approaches are studied and compared. A semi-analytical approach is compared to a particle tracking technique that accounts for matrix diffusion using particle transfer probabilities. An empirical relationship that can be used to map the governing parameter of the semi-analytical approach to the corresponding particle transfer probability is derived. The similarities and differences between these two techniques and their suitability for practical applications are also discussed.
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Keywords: Particle tracking methods; Mobile–immobile systems; Particle transfer proability

1. Introduction

Numerical simulations are being used extensively for analyzing transport in complex geologic media. The characteristics of these geologic media cannot be determined with certainty, which requires the implementation of the numerical simulations within a Monte Carlo framework. Ideally, by processing a large number of fine-scale realizations through groundwater modeling programs, an assessment of aquifer response uncertainty is provided. The issue of the number of realizations needed to achieve convergence for the statistics of concern becomes crucial when the medium heterogeneity increases. This requirement in addition to numerical constraints,

* Corresponding author. Address: Division of Hydrologic Sciences, Desert Research Institute, 755 E. Flamingo Road, Las Vegas, NV 89119, USA. Tel.: +1-702-895-0465; fax: +1-702-895-0427.
E-mail address: hassan@dri.edu (A.E. Hassan).

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such as domain size, grid resolution, time step, and convergence issues increase the computational effort involved in these simulations to the extent that they become prohibitive from both time and cost perspectives, even with today's advanced computing resources. Computationally efficient numerical methods are, therefore, needed to simulate transport processes in highly heterogeneous geologic media, while providing accurate estimates for the expected values and uncertainties of the resulting contaminant distributions.

Standard numerical techniques such as the finite difference and finite element methods, when applied to solve contaminant transport problems, impose some restrictive spatial discretization requirements to avoid numerical dispersion. The random walk particle tracking (RWPT) method provides a robust alternative if the discretization requirements cannot be met. Since the RWPT method is not a direct numerical solution to the governing differential equation, it does not suffer from numerical dispersion (Uffink, 1987). A random walk is generally defined as the movement of a particle (walker) that will undergo a displacement with a magnitude that depends on chance. The irregularity and randomness of the grain skeleton of a porous medium make it impossible to fully describe the solute displacement in a deterministic fashion. As long as the movement of solute particles is unpredictable, it is useful to consider all possible displacements and the probabilities they are realized (Uffink, 1987).

The random walk method is suitable for applications where other methods are, at comparable computational effort, plagued by numerical dispersion. Since particles in the random walk method are not lost nor destroyed, the method conserves mass exactly. Strictly speaking, no grid is needed for the RWPT method. However, in many applications a grid may be employed for the definition of the velocity and dispersion characteristics in the modelled domain. In addition, interpretation of the particle distribution as a concentration field may require a grid or some other means for converting the spatial distribution of particles to concentration values. A further advantage of the RWPT method is the ease with which it can be implanted over any flow model. By switching local dispersion off, the pathlines for the average flow field may be obtained (Kinzelbach, 1988). It is thus easy to model purely advective transport with infinite Peclet number.

The main problem with the RWPT method is the random fluctuations of computed concentrations. The relative size of these fluctuations can be diminished by increasing the number of particles used. As, however, the statistical fluctuations are proportional to the square root of the number of particles in a cell, the increase in the total number of particles does not show equivalent reduction in the random fluctuations of computed concentrations (Kinzelbach, 1988). However, when the random walk method is used in the context of Monte Carlo simulation to get ensemble properties, the effects of these random fluctuations may diminish as the number of realizations increases. In other words, a transport simulation on a single heterogeneous velocity realization may show significant random fluctuations in the computed concentrations, but when the results over many Monte Carlo realizations are averaged, these fluctuations are minimized.

Significant effort has been devoted to developing different RWPT-based approaches to model non-reactive (e.g. Uffink, 1987, 1990; Tompson and Gelhar, 1990; Kinzelbach and Uffink, 1991; LaBolle et al., 1996; Hassan et al., 1998) as well as reactive (e.g. Kinzelbach, 1988; Valocchi and Quinodoz, 1989; Andricevic and Foufoula-Georgiou, 1991) contaminant transport in single-continuum porous media, and little attention has been paid to extending the RWPT method to dual-continuum models (e.g. dual-porosity models, dual-permeability models, and fractured systems). Recently, Liu et al. (2000) developed a particle transfer probability expressions for RWPT methods to account for mass transfer processes associated with fractured porous media.

The objectives of this study are to evaluate the robustness of the RWPT method in predicting transport statistics in single-continuum heterogeneous porous media, and to evaluate two different approaches (relying on RWPT) for predicting transport in fractured systems. Following this introduction we present an overview of the RWPT method in single-continuum and compare the performance of RWPT to other numerical as well as analytical solutions for both homogeneous and heterogeneous media. We also present in

Section 2 a sensitivity analysis showing the effect of different factors on the performance of the RWPT method. Section 3 focuses on the approaches that are used to predict transport statistics in fractured porous media using RWPT. We compare two approaches and develop an empirical relationship between the governing parameters of these approaches. The results of the numerical simulations are discussed in Section 4 and their implications for large-scale numerical studies are highlighted. Section 5 summarizes the work and presents the main conclusions that can be drawn from this study.

2. Particle tracking method in single-continuum porous media

The RWPT method is a method from statistical physics, which has been used in the analysis of diffusion and dispersion processes in porous media for a long time (e.g. Scheidegger, 1954; De Josselin de Jong, 1958). The idea of applying the RWPT method to solute transport problems is based on an analogy between the random walk equation and the Fokker–Planck equation for diffusion. An extensive quantity (such as the mass of some solute tracer) is represented by a large number of particles to which the properties of that quantity are assigned. The particles are displaced in space over discrete time steps by the action of some driving mechanism such as a chemical potential field or a velocity field. The solution of the differential equation governing the movement of the extensive quantity can be approximated by tracking the particles movement in time and space. This tracking in two dimensions is based on the equation (Kinzelbach, 1988; Tompson and Gelhar, 1990; LaBolle et al., 1996)

$$\begin{aligned} x_{t+\Delta t} &= x_t + \left[V_x(x_t, y_t, t) + \left(\frac{\partial D_{xx}}{\partial x} + \frac{\partial D_{xy}}{\partial y} \right) \right] \Delta t \\ &\quad + \sqrt{2D_{xx}\Delta t}\, Z_1 + \sqrt{2D_{xy}\Delta t}\, Z_2, \\ y_{t+\Delta t} &= y_t + \left[V_y(x_t, y_t, t) + \left(\frac{\partial D_{yx}}{\partial x} + \frac{\partial D_{yy}}{\partial y} \right) \right] \Delta t \\ &\quad + \sqrt{2D_{yx}\Delta t}\, Z_1 + \sqrt{2D_{yy}\Delta t}\, Z_2 \end{aligned} \tag{1}$$

where x and y are the coordinates of the particle location, V is the velocity, D_{ij} is the ij component of the dispersion tensor, Δt is the time step, and Z is a normally distributed random number with zero mean and unit variance. The second term that is multiplied by Δt on the right hand side of Eq. (1) is an effective velocity that combines the local velocity at location (x_t, y_t) and time t plus the gradient of the dispersion tensor at location (x_t, y_t), and the last two terms account for the local-scale dispersion and Brownian diffusion. Implementation details of this method can be found in Tompson and Gelhar (1990), LaBolle et al. (1996) and Hassan et al. (1997, 1998), just to name a few.

A number of factors affect the accuracy and robustness of the RWPT method. The number of particles used to represent the solute mass, the size of the time step, the velocity interpolation scheme, and the projection function that converts particle distribution to concentration values over a domain grid are usually the key issues when using RWPT methods for heterogeneous media. Same factors affect the solution for homogeneous cases except the velocity interpolation scheme, which is not needed in this case. In the following analysis, we start with a homogeneous two-dimensional transport problem, for which an analytical solution exists. We compare the prediction of a number of numerical codes and that of the RWPT to the analytical solution. The objective is to first evaluate the performance of RWPT method compared to other approaches and to select another numerical code to be used for evaluating the RWPT method in predicting stochastic concentration mean and variance in the heterogeneous case.

2.1. Homogeneous case

A two-dimensional $x-y$ homogeneous aquifer is assumed with dimensions 48 m × 16 m (Fig. 1) and a constant velocity in x-direction (V_x) of 0.5 m/d. The longitudinal and transverse dispersivities are chosen to be the same with a value of 0.1 m. A total mass of 4 kg is released as a small areal source of contamination with dimensions 0.2 m × 0.2 m. The analytical solution for such case is given as (e.g. Ogata and

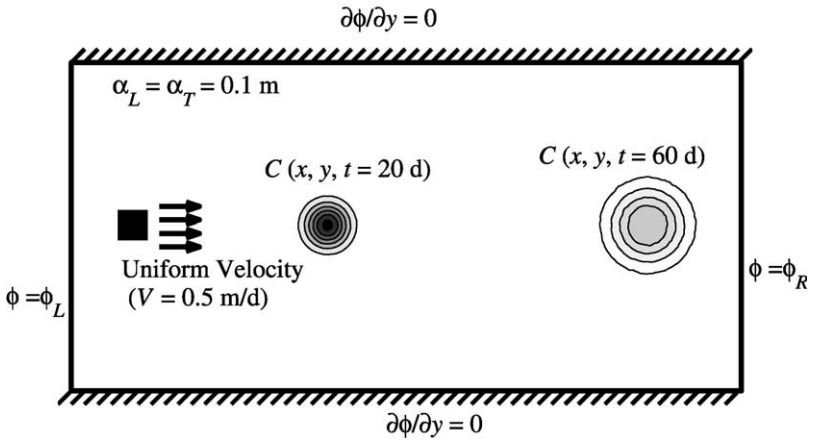

Fig. 1. Schematic diagram of the two-dimensional simulation domain and boundary conditions.

Banks, 1961)

$$C(x,y,t) = \frac{C_0 A}{4\pi t\sqrt{D_{xx}D_{yy}}} \times \exp\left[-\frac{(x-x_0)^2}{4D_{xx}t} - \frac{(y-y_0)^2}{4D_{yy}t}\right] \quad (2)$$

where C_0 is the initial concentration, A is the source area, and (x_0, y_0) is the center of the source area. The two-dimensional concentration distribution is obtained at two times, $t = 20$ days and 60 days after release. The analytical solution given in Eq. (2) is valid for a point source instantaneous release. The analytical solution for an instantaneous release of a rectangular source, with length L in the direction of flow and width W transverse to this direction, can be approximated by Eq. (2) if $L^2/48D_{xx}t$ and $W^2/48D_{yy}t$ are both small (Charbeneau, 2000). For the problem studied here, the two values are equal and after 20 and 60 days they are 0.000833 and 0.000278, respectively. These values are small enough to allow the use of Eq. (2) for this problem.

Several software packages are used to solve this transport problem and to compare to the RWPT method as well as to the analytical solutions. Two finite element codes: SUTRA (Voss, 1984), and FEFLOW (Diersch, 1998) and a finite difference code: MT3D (Zheng, 1990) and MT3DMS (Zheng and Wang, 1999) are used for this purpose. The latter code has new features and solvers that are of particular importance for the comparisons presented in the following sections.

SUTRA is a well-known finite element model that simulates flow and solute transport in the subsurface. The simulation domain shown in Fig. 1 is discretized into grid cells of size 0.2m × 0.2 m. Comparisons between SUTRA solution and the analytical solution are given in Fig. 2A. It can be seen from the figure that SUTRA solution has a significant longitudinal 'numerical' dispersion and does not match the analytical solution. A finer mesh and/or higher dispersivity values are usually used to avoid such inaccuracy of the finite element solution. We, however, used a grid of size 0.1 m × 0.1 m and increased the longitudinal and transverse dispersivities to 0.2 m, but with no improvement in the performance of SUTRA relative to the corresponding analytical solution. Similarly, FEFLOW solution entails some inaccuracy due to numerical dispersion (Fig. 2B). In fact, FEFLOW and SUTRA solutions are in very close agreement.

MT3D is another well-known model that uses the finite difference method. We used MT3D/MT3DMS and selected two solvers that differ in the finite difference discretization of the advection term. The first solver (denoted in Fig. 2 as MT3D) relies on a standard central differencing discretization for the concentration gradient governing the advection term. The second solver (denoted as MT3D-TVD) relies on a third-order total variance diminishing approach with

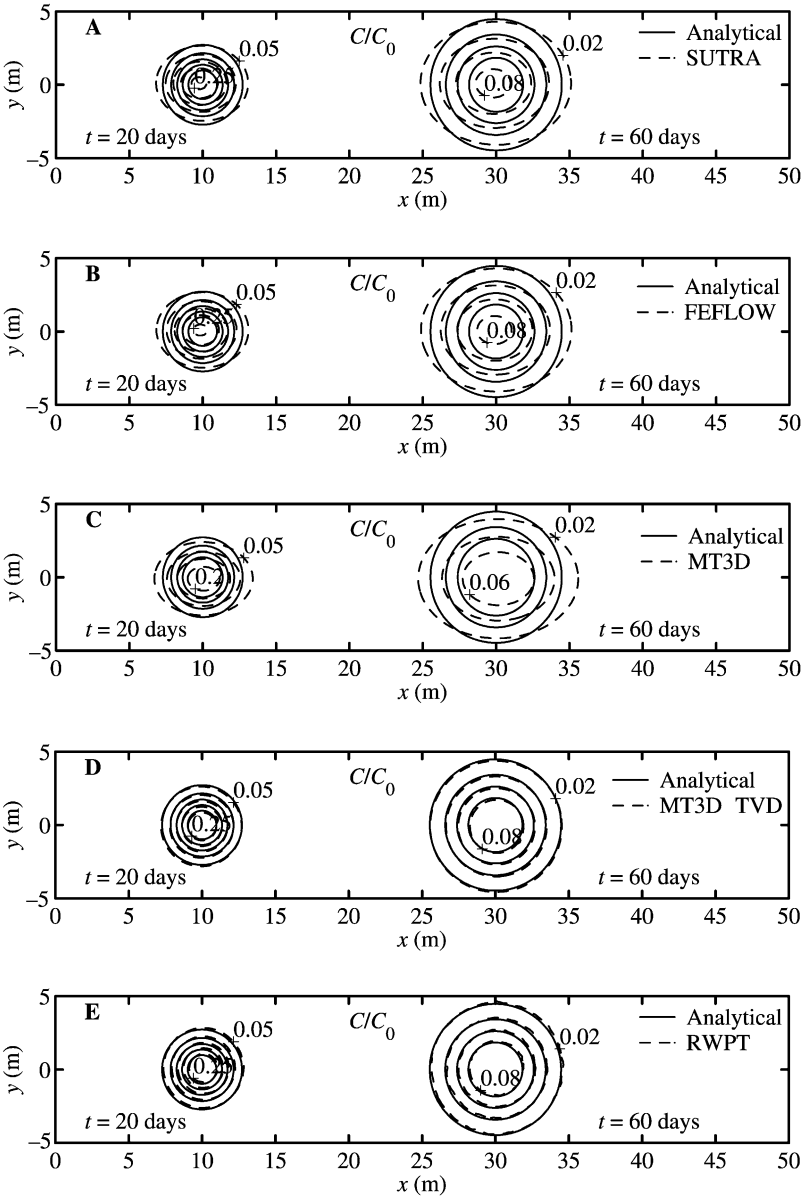

Fig. 2. Comparison between analytical solution (Ogata and Banks, 1961) and different numerical codes for transport in a two-dimensional heterogeneous domain.

a universal flux limiter (Zheng and Wang, 1999) for discretizing the advection term.

Fig. 2C shows that MT3D with the standard central differencing induces more numerical dispersion than SUTRA and FEFLOW. The results of MT3D-TVD, shown in Fig. 2D, show an excellent agreement with the analytical solution and completely eliminated numerical dispersion. The third-order TVD solver with the universal flux limiter significantly minimizes numerical dispersion leading to much better results

compared to the standard central differencing. However, the computational burden associated with the TVD solver is much larger than the standard solver (Zheng and Wang, 1999).

Finally, the performance of the RWPT method for this homogeneous case is displayed in Fig. 2E. Similar to the MT3D-TVD solution, the RWPT solution is in excellent agreement with the analytical solution. However, a very large number of particles ($\sim$2.5 millions) was necessary to achieve the smoothness of the solution, otherwise oscillations exist around the circular contours. These oscillations are the result of the discrete nature of the particles and the random component of their movement. It will be seen later that in the heterogeneous case, such large number of particles is not necessary when implementing the Monte Carlo simulations as the ensemble statistics automatically average out these oscillations.

In the homogeneous case, RWPT is not very sensitive to time step size as long as overshoot problems are avoided, which is achieved by keeping $V_x\ \Delta t < \Delta x$. The oscillations in the obtained concentration distribution can be reduced by increasing the number of particles and/or increasing the size of the projection area used to project particles distribution to concentration values. Nine simulation cases are considered where the time step size, Δt, the number of particles, NP, and the projection area, Ω, are varied to study the sensitivity of the resulting concentration to these variations. Using the same problem configuration and parameters as in Fig. 2, these three parameters are varied around their base-case values: $\Delta t = 0.1$ days, NP = 102,400, and $\Omega = 2\Delta x \times 2\Delta y$. Fig. 3 shows the sensitivity of the RWPT solution to these factors. The figure displays the concentration values along the longitudinal centerline of the simulation domain. As mentioned earlier, velocity interpolation effect comes into play only in the heterogeneous case. It can be seen from the figure that only Ω (the projection area) has some noticeable effect on the obtained concentration. The time step and the number of particles, as long as properly selected, do not have any effect when they are further refined (time step is decreased and number of particles is increased).

2.2. Heterogeneous case

Since the MT3D-TVD solution is shown to match the analytical solution of the homogeneous case, we use it here to evaluate the performance of the RWPT method in predicting transport in heterogeneous domains. In this case, we deal with the concentration distribution in a statistical sense. That is, we evaluate, using Monte Carlo procedure, the ensemble mean and variance of the stochastic concentration distribution in two dimensions. An isotropic, heterogeneous, spatially correlated conductivity field, $K(x, y)$, with a lognormal distribution, a unit variance, and an exponential covariance structure is used to represent medium heterogeneity. A rectangular domain of size $52I \times 26I$ is used for the heterogeneous case, where I is the correlation length of the $\log K$ field. The discretization grid for this domain is assumed to consist of uniform squares of size $0.2I \times 0.2I$ with five grid cells per conductivity correlation length. The boundary conditions and the macroscopic mean velocity in the x-direction are the same as used for the homogeneous case, Fig. 1.

The comparison between MT3D-TVD and the RWPT method is shown in Fig. 4, where the normalized mean concentration, $\langle C\rangle/C_0$, and the concentration variance, σ_C^2/C_0^2, are plotted. These moments are obtained by averaging over 1000 realizations of the transport solution through the expressions $\langle C\rangle = (1/1000)\sum_{i=1}^{1000} C_i$ and $\sigma_C^2 = (1/1000)\sum_{i=1}^{1000} C_i^2 - \langle C\rangle^2$, where C_i is the two-dimensional spatial distribution of the concentration in realization i. Fig. 4 shows that the results for the mean concentration are very close in terms of both the plume shape and the concentration values. The variance of the stochastic concentration, usually used to express the prediction uncertainty, is also matching closely between the two approaches. It is apparent that the MT3D-TVD results for the variance are not as smooth as the RWPT results, which is attributed to the smaller number of realizations employed in the former model due to the long computational times involved. In terms of CPU time, the RWPT method was at least three times faster than the MT3D-TVD method.

These results indicate that in addition to being computationally efficient, the RWPT method is also robust and accurate in predicting concentration

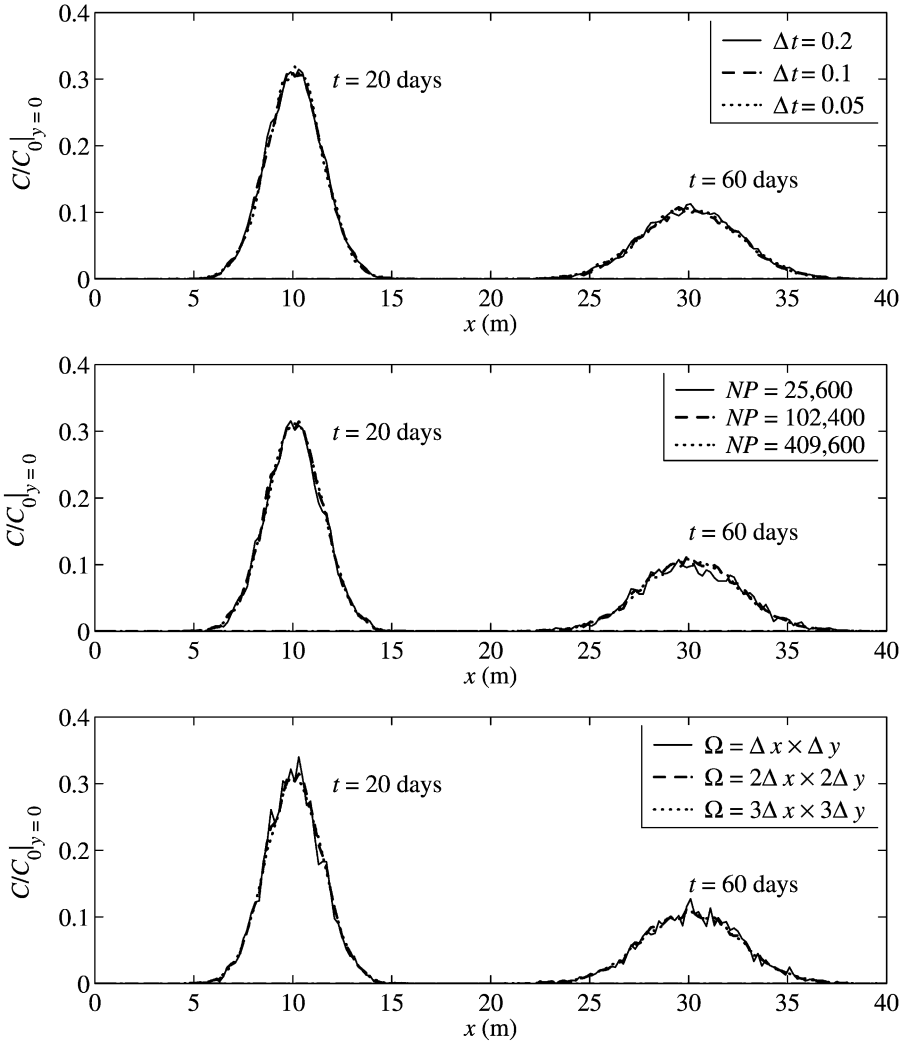

Fig. 3. Sensitivity of the RWPT solution for the concentration in the homogeneous case to the time step, the number of particles and the projection area.

moments under heterogeneous conditions, which is usually the case for field problems. The oscillations resulting from the discrete nature of the particles do not persist in a Monte Carlo simulation as these oscillations are random and they cancel each other when the ensemble statistics are computed. The factors affecting the results of the RWPT method are similar to the homogeneous case, but with the addition of the velocity interpolation scheme and the method of incorporating the effects of sharp contrasts in medium properties (e.g. dispersion gradient term). We discuss in Section 2.3 the results of a number of simulations assessing the sensitivity of the RWPT results to these parameters.

2.3. Sensitivity analysis in the heterogeneous case

Fig. 5 shows the sensitivity of the RWPT solution of the mean concentration to the size of the time step, the number of particles, and the size of the projection

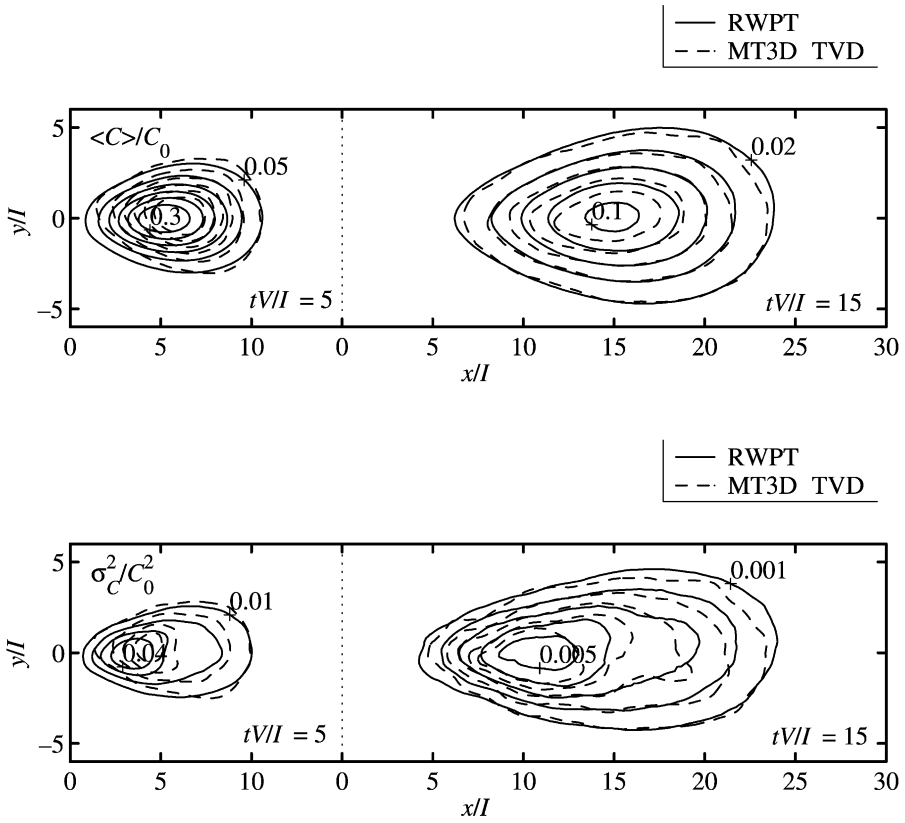

Fig. 4. Comparison between the RWPT and the MT3D-TVD solutions for the concentration mean and variance in the heterogeneous case.

area, Ω. As can be seen, the largest effect on the mean concentration is obtained when increasing the number of particles, NP, from 51,200 to 102,400. Any further increase in the number of particles does not affect the mean concentration. Fig. 6 is similar to Fig. 5, but for the concentration standard deviation. Here both the number of particles and the size of the projection area have some effects on σ_C, although not significant.

The heterogeneous conductivity distribution leads to spatially varying velocity field that is only known at grid block interfaces. Particle velocity needs to be interpolated from the surrounding known velocity values. A number of studies dealt with the velocity interpolation and presented different alternatives for computing particle velocity. These include, but are not limited to, Pollock (1988), Goode (1990), Schafer-Perini and Wilson (1991) and LaBolle et al. (1996). The common result of these approaches is that for heterogeneity such as the one considered here, the linear and bilinear interpolation yield similar results (e.g. Goode, 1990, (Fig. 17) and LaBolle et al., 1996, (Figs. 12–14)). To check the sensitivity of the results of the RWPT method to the velocity interpolation scheme, we employed a bilinear and an inverse distance interpolation scheme.

Discontinuities in effective subsurface transport properties that may arise in discrete velocity fields of numerical groundwater flow models violate the smoothness assumption upon which the standard RWPT techniques are based (LaBolle et al., 2000). These authors developed generalized stochastic differential equations applicable to the case of discontinuous coefficients (e.g. dispersion coefficients) and developed a new random walk method that numerically integrates these equations. That method is applicable for cases of abrupt changes in transport parameters and velocity values. The new random-walk equations proposed by LaBolle

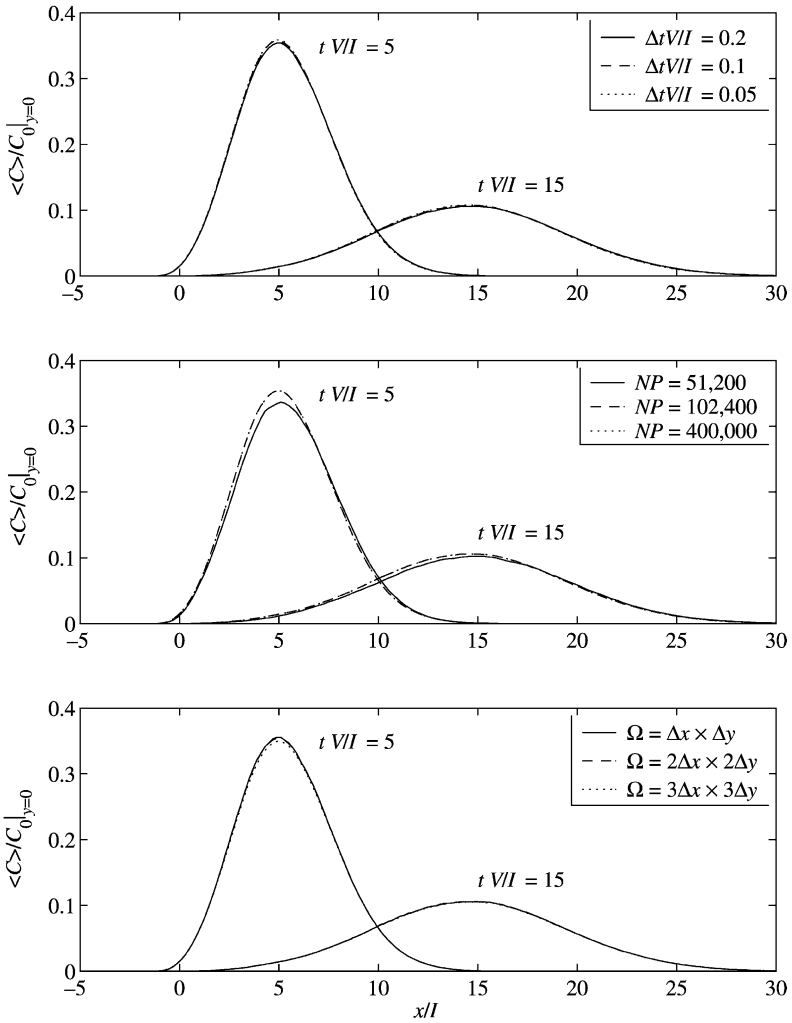

Fig. 5. Sensitivity of the RWPT solution for the mean concentration to the time step, the number of particles and the projection area.

et al. (2000) can be written as

$$
\begin{aligned}
x_{t+\Delta t} &= x_t + V_x(x_t, y_t, t)\Delta t \\
&\quad + \sqrt{2D_{xx}(x_t + \delta x, y_t + \delta y, t)\Delta t}\, Z_1 \\
&\quad + \sqrt{2D_{xy}(x_t + \delta x, y_t + \delta y, t)\Delta t}\, Z_2, \\
y_{t+\Delta t} &= y_t + V_y(x_t, y_t, t)\Delta t \\
&\quad + \sqrt{2D_{yx}(x_t + \delta x, y_t + \delta y, t)\Delta t}\, Z_1 \\
&\quad + \sqrt{2D_{yy}(x_t + \delta x, y_t + \delta y, t)\Delta t}\, Z_2
\end{aligned}
\tag{3}
$$

where δx and δy are defined as

$$
\begin{aligned}
\delta x &= \sqrt{2D_{xx}(x_t, y_t, t)\Delta t}\, Z_1 + \sqrt{2D_{xy}(x_t, y_t, t)\Delta t}\, Z_2, \\
\delta y &= \sqrt{2D_{yx}(x_t, y_t, t)\Delta t}\, Z_1 + \sqrt{2D_{yy}(x_t, y_t, t)\,\Delta t}\, Z_2
\end{aligned}
\tag{4}
$$

The idea in this approach is to evaluate the advective step of the particle using the velocity at the current position of the particle, (x_t, y_t), and at time t. The dispersive step is performed using dispersion coefficients evaluated at an intermediate location, $(x_t + \delta x, y_t + \delta y)$, where the increments δx and δy represent dispersive steps from the current

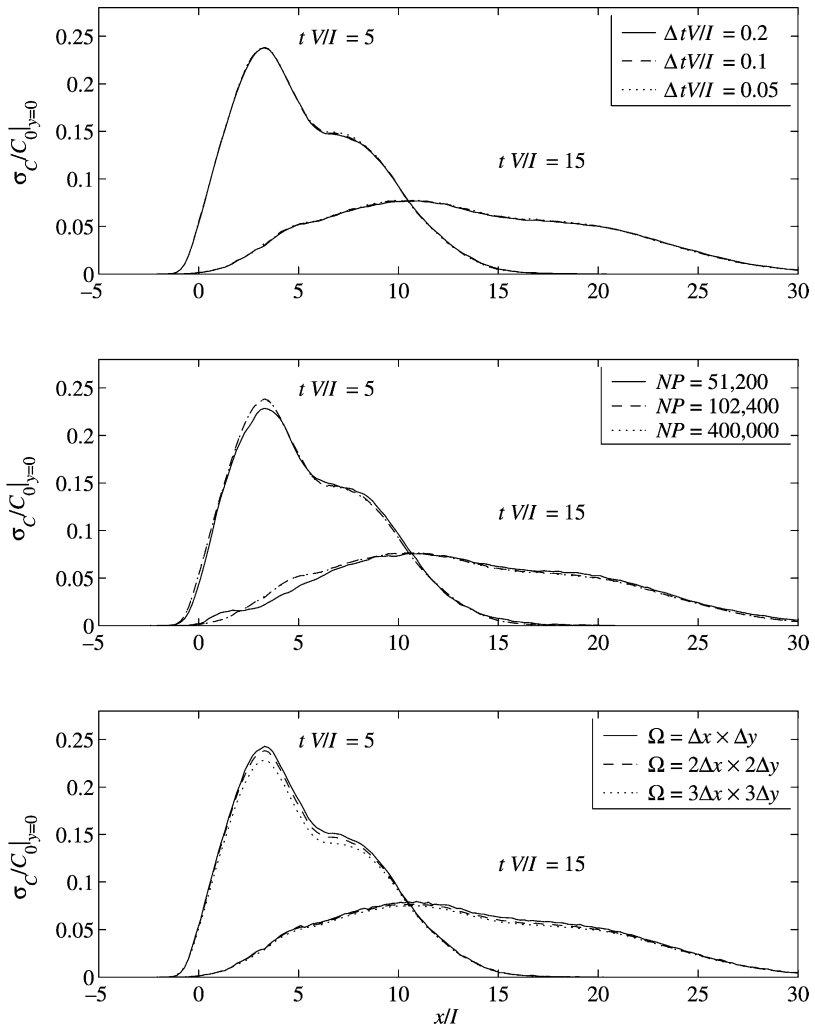

Fig. 6. Sensitivity of the RWPT solution for the concentration standard deviation to the time step, the number of particles and the projection area.

location, (x_t, y_t), to the intermediate location, $(x_t + \delta x, y_t + \delta y)$. Comparing Eqs. (3) and (4) to Eq. (1), one can see that the dispersion gradients (which are not defined at interfaces between domain blocks) are not needed in LaBolle et al.'s (2000) approach.

It is of interest to compare the performance of this modified approach with the traditional RWPT method. We plot in Fig. 7 the mean and standard deviation of the stochastic concentration along the longitudinal centerline of the simulation domain. The figure compares the traditional RWPT method using bilinear and inverse square-distance interpolation schemes to the modified RWPT approach of LaBolle et al. (2000) with bilinear interpolation for the velocity. It is apparent that the modified approach is not producing any different results than the traditional one for the type of heterogeneity used here. Also, the change of velocity interpolation scheme has a minor effect on the concentration statistics.

3. Particle tracking methods in dual continua

As mentioned earlier, no progress has been made in developing the RWPT method for dual continua porous media (dual porosity, dual permeability and

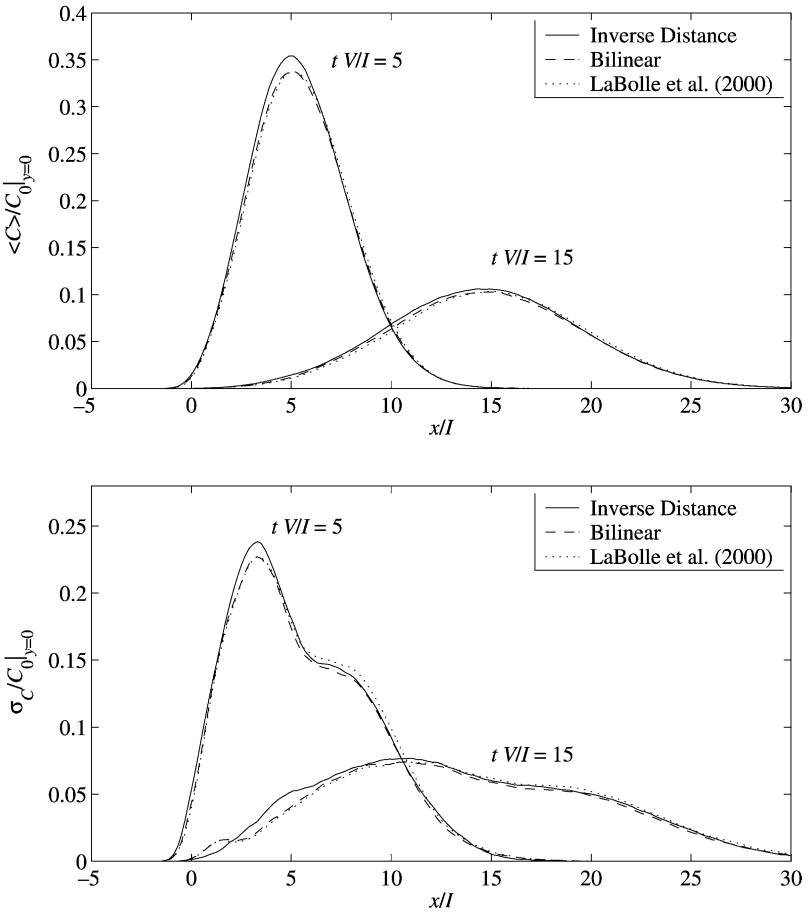

Fig. 7. Sensitivity of the RWPT solution for the concentration mean and variance to different velocity interpolation and implementation schemes.

fractured media). We adapt and compare two approaches to account for matrix diffusion in fractured systems, where mass diffusion occurs from the high-velocity fracture flowpaths into the surrounding porous blocks. The purpose is to compare the two approaches, study and evaluate their underlying assumptions, identify the sets of parameters needed for each approach, and explore the possibility of relating the two sets of parameters. By doing so one can have the option to select any of the two approaches for modeling purposes once a single set of parameters is obtained.

3.1. Retention function approach

For the analysis of transport in a fractured system, matrix diffusion can be accounted for using the retention function semi-analytical solution presented by Cvetkovic et al. (1999). The mass flux or the concentration breakthrough curves can be first obtained using a RWPT method (or any other transport solution) without accounting for matrix diffusion. That is, these breakthrough curves are obtained as if transport occurs in the fractures with no fracture-matrix interaction. The matrix diffusion effect can be accounted for using a retention function that depends on the matrix properties. The mass flux or concentration breakthrough with matrix diffusion effect can thus be evaluated from the expression

$$Q_{\mathrm{md}}(t) = \int_0^{\infty} \gamma(t,\tau) Q(\tau) d\tau \tag{5}$$

where $Q(\tau)$ is the mass flux breakthrough at time τ with no fracture-matrix interaction, $Q_{md}(t)$ is the mass flux after accounting for matrix diffusion, and $\gamma(t,\tau)$ is the retention function that incorporates the effect of mass transfer between the fracture and the rock matrix. This retention function is given as (Cvetkovic and Dagan, 1994; Cvetkovic et al., 1999)

$$\gamma(t,\tau) = H(t-\tau)\frac{\kappa\tau}{2\sqrt{\pi}(t-\tau)^{3/2}}\exp\left(-\frac{(\kappa\tau)^2}{4(t-\tau)}\right) \quad (6)$$

where H is the dimensionless Heaviside function ($H(t-\tau)=1$ for $(t-\tau)>0$ and 0 for $(t-\tau)\leq 0$), τ is the particle travel time (days), t is the time at which the flux is obtained, and κ is the matrix diffusion parameter ($d^{-1/2}$) defined as

$$\kappa = \frac{\theta_m\sqrt{D_m^* R_m}}{b} \quad (7)$$

where θ_m is the matrix porosity, b is the effective fracture half-aperture (m), D_m^* is the effective diffusion coefficient in the rock matrix (m^2/d) and R_m is the dimensionless retardation coefficient in the rock matrix. The main assumptions underlying the derivation of the above analytical retention function are a constant aperture along the streamtube, diffusion only perpendicular to the fracture plane, well-mixed conditions over the cross-sectional area of the fracture, and homogeneous rock matrix with no advection and with infinite capacity such that diffusion fronts do not intersect. These assumptions may not be satisfied in the field unless matrix blocks are sufficiently large for the diffusion fronts not to intersect. It should be mentioned here that Eq. (5) conserves mass as the original $Q(\tau)$ is mass conservative and the retention function, γ, has the effect of just delaying the arrival of the mass to the control plane location.

We apply this approach to the same two-dimensional heterogeneous domain used in the single-continuum case, but assuming that particles travel in connected fractures from the source to a compliance boundary or a control plane placed normal to the mean flow direction. The series of connected fractures and the surrounding matrix blocks are assumed to have the same characteristics everywhere in the domain such that the strength of matrix diffusion is spatially invariant. An initial contaminant source is then released and assumed to exist only in fractures. Particles representing this source are tracked in the simulation domain over discrete time steps until they reach and cross the control plane. Once they cross, the mass flux breakthrough is obtained as a function of time, $Q(t)$, and is then convoluted with the retention function γ to obtain the breakthrough with fracture-matrix interaction, $Q_{md}(t)$.

3.2. Particle transfer probability (Liu et al., 2000)

Liu et al. (2000) introduced the first RWPT approach applicable to fractured porous media, where they developed the particle transfer probability expressions necessary for diffusing the particles into the matrix and back to the fractures. The basic idea is then to track the particles in space over discrete time steps and use the transfer probabilities to move the particles back and forth between the two continua. The approach assumes that each grid block in the simulation domain contains two overlapping continua; one represents the fractures containing the rapid migration of water and solutes, and the other represents the surrounding matrix blocks that have a larger quantity of water, which is immobile under the assumption of no advection in the matrix. Assuming no water flux between fractures and matrix blocks, the particle transfer probabilities of Liu et al. (2000) can be written as forward transfer probability, $P_{fm} = F_{fm}\Delta t/(V_f C_f)$, and a backward transfer probability, $P_{mf} = F_{mf}\Delta t/(V_m \bar{C}_m)$. In these expressions, the net solute transport rate between the fractures and the matrix within a grid cell is expressed as a transport rate from fractures to the matrix (F_{fm}) minus another transport rate from the matrix to fractures (F_{mf}). The particle transfer probabilities are expressed in terms of these transport rates, the concentration, C_f, and water volume, V_f, in the fractures, and the 'block-averaged' concentration, $\bar{C}_m$, and water volume, V_m, in the matrix. Liu et al. (2000) then derived expressions for flux rates in terms of measurable quantities and arrived finally at the expressions

$$P_{fm} = \frac{2D_m^* A}{V_f(1-\lambda)(B-b)}\Delta t \quad (8)$$

$$P_{mf} = \frac{2D_m^* A}{V_m(1-\lambda)(B-b)}\Delta t \quad (9)$$

where P_{fm} is the probability that a particle in the fracture will diffuse into the matrix, P_{mf} is the probability that a particle in the matrix will diffuse back to the fracture,D_m^* is the diffusion coefficient in the matrix, A is the fracture surface area, B is half the spacing between the fractures, b is the fracture half-aperture, λ is a shape factor expressing how the matrix volume increases as one approaches the center of the matrix block (e.g. $\lambda = 1/3$ for parallel fractures), V_m is the volume of water in the matrix block, V_f is the volume of water in the fractures, and Δt is the time step. According to P_{fm}, particles can diffuse into the matrix and are thus subject to the matrix advection (if any) and dispersion/diffusion. On the other hand, when particles are found to diffuse back from the matrix to the fractures according to P_{mf}, they start moving with the underlying velocity of the fractures.

This approach is implemented using the two-dimensional heterogeneous settings as in the retention function approach described above. Once the particles that are in the fractures reach the control plane of interest, the mass flux breakthrough, $Q_{md}(t)$, can be obtained and compared to that of the retention function approach. It can be seen from Eqs. (7)–(9) that the common governing parameters between the two approaches are D_m^*, b, and θ_m (since V_m in Eq. (8) is a function of θ_m). The main difference between the two approaches is that the first approach assumes infinite matrix extent and thus infinite capacity for mass diffusion, whereas the second approach accounts for the finite extent of matrix blocks through the parameter B in Eqs. (8) and (9).

3.3. Modified particle transfer probability

In the derivation of the transfer probabilities, Liu et al. (2000) relate the spatially varying contaminant concentration in the matrix, C_m, to the average concentration over the matrix volume, $\bar{C}_m$. This results in expressing the backward probability, P_{mf}, in terms of the averaged concentration leading to a very slow release of particles even if they are associated with high concentration gradient between the matrix and the fractures. This implies that complete mixing of contaminant mass associated with each particle occurs immediately once the particle is in the matrix or in the fracture, which if realistic for fractures, is not so for the matrix. We propose a simple modification to computing the backward probability and implementing the fracture-matrix interaction into the RWPT method. Consistent with the distribution of contaminant concentration in the matrix as hypothesized by Liu et al. (2000), the modified probability can be written as

$$P_{mf}^* = P_{mf} + \left(1 - \frac{s}{S}\right)^2 (P_{fm} - P_{mf}) \quad (10)$$

where s is a coordinate axis normal to the fracture-matrix interface, $S = B - b$ is the distance from that interface to the center of the matrix block, and P_{fm} and P_{mf} are as expressed by Eqs. (8) and (9). Eq. (10) yields a backward probability that is different for different particles according to their locations within the matrix. However, since the implementation of the approach does not account for spatial locations of particles within the matrix block, s/S can be hypothetically computed according to the number of particles that exist in the matrix for each grid cell. That is, in each grid cell, the number of particles that exist in the matrix can be computed and the particles can be hypothetically distributed such that they span a certain range of residence times from zero to the value that is associated with complete mixing within the block. This mean that the mass associated with one particle is assumed to have experienced complete mixing conditions (thus $P_{mf}^* = P_{mf}$), and that another particle has just diffused into the matrix (thus $P_{mf}^* = P_{fm}$). The remaining particles are assumed to have backward transfer probabilities between these two extremes as expressed by Eq. (10).

3.4. Results and comparisons

The retention function approach is compared to the particle transfer approach using both Liu et al.'s (2000) probabilities and the modified backward probability in Eq 10. Four test cases are presented where the governing parameter of the retention function approach, κ, is assigned the values 0.01, 0.03, 0.1, and 0.3, respectively. For the RWPT approach with the particle transfer probability, it is assumed that the average length of fractures within

Table 1
Input values of the parameters used in the two-dimensional heterogeneous cases

Flow and transport parameters		Parameters for the retention function approach and the particle transfer probabilities	
Parameter	Value	Parameter	Value
Log-K covariance	$\sigma_f^2 e^{-r/I}$	B	0.1 m
Log-K variance, σ_f^2	1.0 and 4.0	b	10 μm
Log-K correlation length, I	1.0 m	$(1-\lambda)$	0.001, 0.0023, 0.0083, 0.0214
Domain size	$51.2\,I \times 25.6\,I$	θ_m	0.1
Grid size	$0.2\,I \times 0.2\,I$	D_m^*	1×10^{-12}, 9×10^{-12}, 1×10^{-10}, 9×10^{-10} m^2/d
Mean velocity in x, V	0.5 m/d	Δt	0.05 d
Source size	$4.0\,I \times 2.0\,I$	P_{fm}	0.0001, 0004, 0012, 0042
α_L	0.1 m	P_{fm}/P_{mf}	400
α_T	0.1 m	κ	0.01, 0.03, 0.1, 0.3 d$^{-1/2}$

each grid cell, L, is 0.5 m, and as such, one can express the fracture surface area and water volume in fractures and in the matrix as

$$A = 2L \qquad V_f = 2bL$$
$$V_m = (\Delta x \times \Delta y - 2bL)\theta_m \tag{11}$$

Table 1 shows the rest of the parameters used for these test cases, where all parameters are fixed, except for D_m^* and $(1-\lambda)$, which are varied according to the variation of κ to yield the corresponding values of P_{fm} and P_{mf}. Since the objective was to obtain the values of P_{fm} and P_{mf} that provide the closest match with the retention function results, we considered the parameter λ to be a flexible one that can be changed to yield the identified values of P_{fm} and P_{mf} for each value of κ.

Fig. 8 shows the results for the first two test cases. In each case, we show the total mass flux breakthrough without matrix diffusion effect, Q (thick solid line), and the breakthrough with matrix diffusion effect, Q_{md}, obtained using the retention function of Eq. (5) (thick dashed line), the RWPT employing Eqs. (8) and (9) (dotted line), and the RWPT employing Eq. (10) for the backward transfer probability (thin solid line). In addition, the breakthrough curves are obtained at two sections located at distances $x/I = 10$ and 20 from the initial source location. When the retention function parameter $\kappa = 0.01$ d$^{-1/2}$, there is very minimal diffusion in the matrix and as such Q and Q_{md} are very close. The retention function and the RWPT results are essentially indistinguishable. Increasing κ to 0.03 d$^{-1/2}$ increases the matrix diffusion effect and leads to a long tailing behavior for Q_{md}. The results of the retention function approach show a slight delay in the arrival of the mass (especially the peak mass flux) to the breakthrough planes as compared to the particle transfer approach. However, the magnitude of the peak is close between the two approaches. It should be mentioned that for each case, we obtain Q_{md} using the retention function first and then use trial-and-error experiments to obtain the closest match with the particle transfer probability. The two cases shown in Fig. 8 show good agreement between the two approaches, which is essentially due to the fact that the matrix diffusion is not significant, and thus the different assumptions underlying both approaches do not have a strong influence yet. In addition, the original transfer probabilities of Liu et al. (2000) and the modified probability of Eq. (10) yield the same result.

Fig. 9 shows the remaining two cases where κ values are increased an order of magnitude compared to the first two cases. The original Q with no matrix diffusion is not shown in this figure, but it is similar to Fig. 8. As can be seen in the figure, when $\kappa = 0.1$ d$^{-1/2}$, the difference in the peak arrival time between the particle transfer approaches and the retention function approach increases, but the values of the peak flux are still comparable. On the other hand, Liu et al.'s (2000) probabilities yield a quick drop after the peak with no significant tailing behavior. The modified probabilities perform better in terms of producing

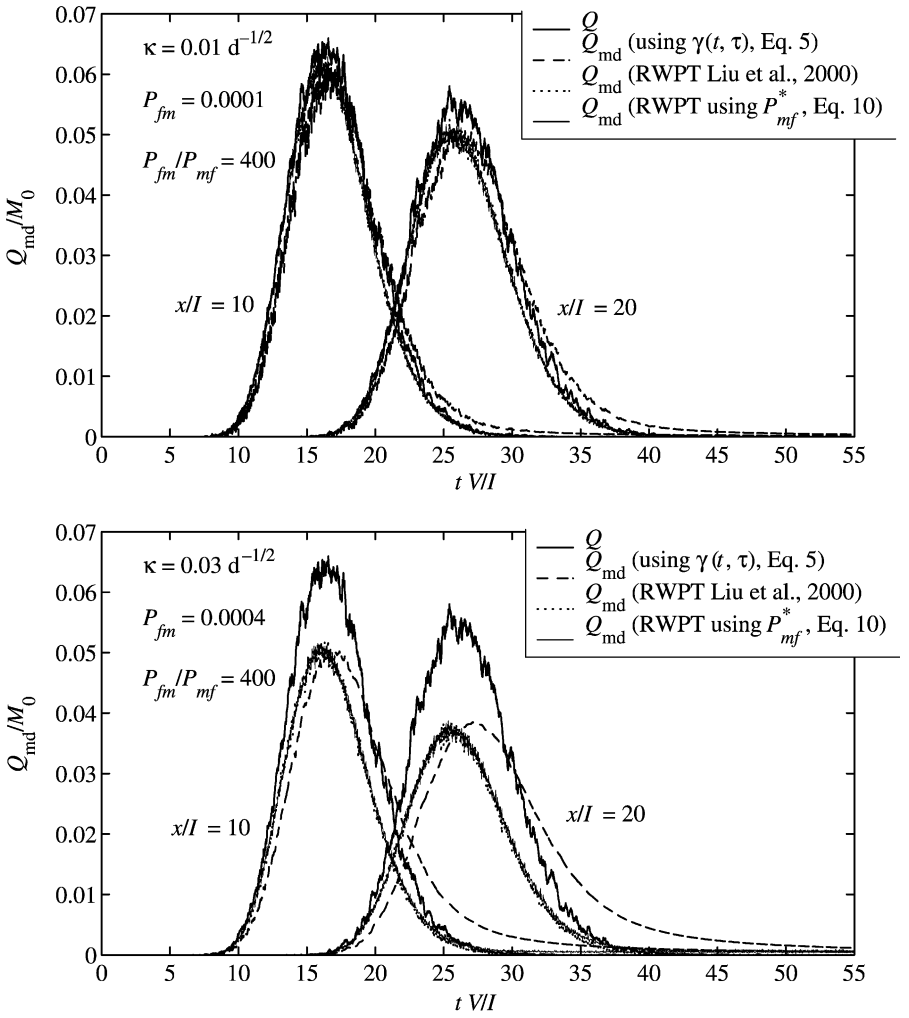

Fig. 8. Solute mass flux in a fractured system obtained using the retention function approach (with $\kappa = 0.01$ and $0.03\ d^{-1/2}$) and the RWPT method with particle transfer probability.

a long tailing behavior close to the one predicted by the semi-analytical (retention function) approach. This fact is even more apparent for $\kappa = 0.3\ d^{-1/2}$, where the modified probability of Eq. (10) leads to the typical matrix diffusion features of a delayed peak and a long tailing behavior. These results compare to the semi-analytical solution better than do those based on Liu et al.'s (2000) original probability as can be seen in Fig. 9. The original particle transfer probability leads to a reduction of the mass flux values but no delay in the peak arrival and no tailing behavior. This is due to the fact that P_{mf} is very small and once the particles diffuse into the matrix, they are released to the fracture very slowly over time and with small quantities that do not constitute a significant tailing. Using P^*_{mf} as given in Eq. (10) leads to a faster release of large numbers of particles from the matrix to the fractures, which facilitates their arrival to the breakthrough boundary and produces the tailing behavior. Fig. (9) clearly shows that the effects of the assumption of infinite matrix availability underlying the derivation of the retention function appear for $\kappa = 0.3\ d^{-1/2}$, where a larger portion of the contaminant mass has diffused into the matrix and will require more time

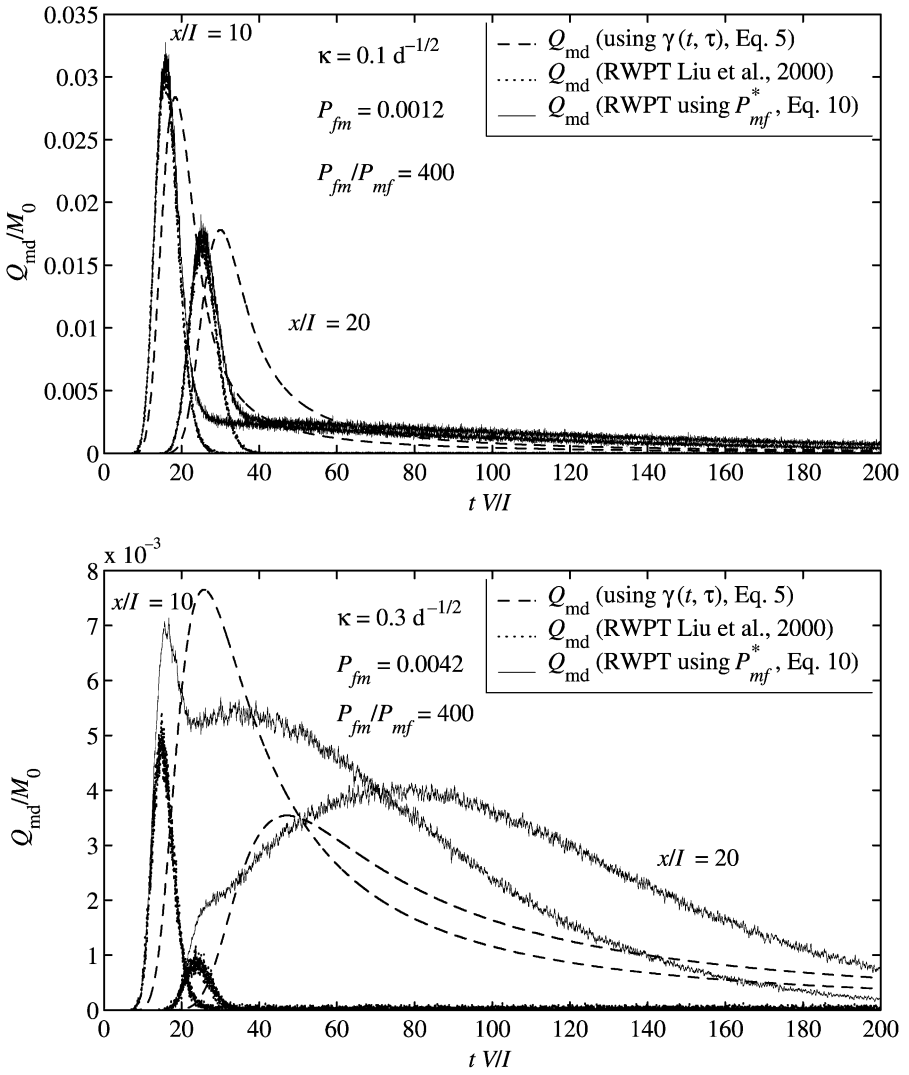

Fig. 9. Solute mass flux in a fractured system obtained using the retention function approach (with $\kappa = 0.1$ and 0.3 $d^{-1/2}$) and the RWPT method with particle transfer probability.

to reach the breakthrough planes as compared to the RWPT transfer probability.

In addition to the four test cases discussed above, we also compared the results using other values for κ in an attempt to find a relationship between κ and the particle transfer probability, P_{mf}. Such a relation would be very helpful if only one parameter can be estimated from the available data of a certain field application. For each value of κ, Q_{md} is obtained and a trial-and-error experiment is performed until the value of P_{mf} that provides the closest result to Q_{md} is identified. The pairs of κ and P_{mf} are then analyzed for the existence of a relationship. Fig. 10 displays the data points and the relationship between κ and $P_{mf}/\Delta t$, where a best fit line with a correlation coefficient R of about 0.98 is obtained. This relation enables estimating the two parameters from limited field data and thus provides flexibility in using one or both approaches to model transport in fractures and matrix diffusion.

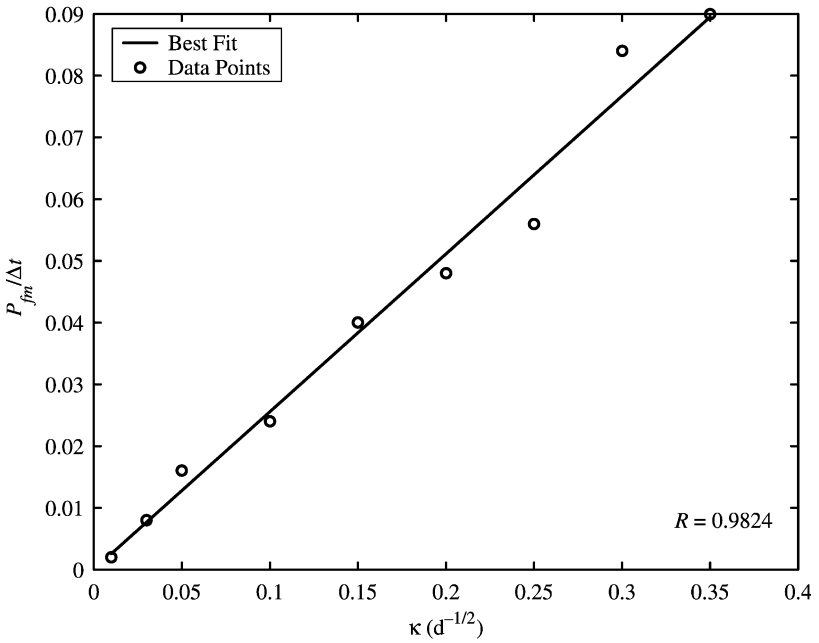

Fig. 10. Empirical relation between the retention function parameter, κ, and the forward transfer probability divided by the time step, $P_{mf}/\Delta t$.

4. Discussion

The application of the particle tracking technique to transport in porous media is very broad in today's practice. The approach has some advantages as well as some disadvantages as discussed in Section 1. There are other approaches for simulating transport in heterogeneous media that may be more common and well established as compared to the RWPT method. However, the computational burden encountered when applying these approaches to large numbers of realizations prohibits its use in stochastic modeling of field scale problems. This warrants devoting some efforts to studying and enhancing the capabilities of the RWPT methods and extending them to dual domain models that have gained popularity in recent years and fractured systems that are very common in the real world. This is particularly important considering the fact that these random walk methods have been and will remain widely used for modeling transport in large-scale numerical models.

As an example of this aspect, we show in Section 2 the large numerical dispersion associated with the standard finite difference technique as implemented in MT3D. This numerical dispersion is minimized by using the total variance diminishing technique (Zheng and Wang, 1999), but this dramatically increases the computational time making this solution algorithm impractical for large-scale Monte Carlo simulations. The computational efficiency of the RWPT technique makes it advantageous in these types of simulation. Furthermore, the oscillation problem associated with the RWPT method is usually overcome when analyzing the ensemble of a large number of transport simulations.

The computational burden associated with standard finite difference and finite element techniques becomes even worse when it comes to modeling transport in fractured and dual domain systems. Since many of the real world aquifers are fractured and highly heterogeneous, a large number of realizations is usually required to obtain accurate results, and time considerations may hinder the investigation of an adequate number of scenarios, which may be essential for a given problem. These reasons make a computationally efficient approach such as the random walk approach the preferred tool for performing these modeling tasks under the imposed restrictions of limited budgets and time frames. Developing and studying RWPT methods for simulating transport in

dual domain and fractured systems is thus important from both scientific and practical perspectives. The results presented in this study shed some lights on different RWPT-based techniques for handling matrix diffusion in fractured systems.

The retention function approach is limited to cases where transport of contaminants occurs in a single fractured unit. In this case one relies on the assumption that particles migrate in a series of connected fractures with the same characteristics such that the retention function approximation is valid. In addition, the realism of the approach with its underlying assumptions depends on the fracture intensity and the average spacing between them. For widely spaced fractures, the approach may be realistic since the matrix availability can be considered unlimited. On the other hand, the RWPT approach with the particle transfer probability may be more advantageous in composite aquifers where transport occurs in multiple units with different fracture characteristics that may be combined with non-fractured units. In this case, it is relatively easy to switch the particle transfer between fractures and matrix blocks on and off depending on the characteristics of the geologic unit in which particles migrate.

5. Summary and conclusions

The RWPT technique for simulating transport in single continuum and dual continua porous media is studied and evaluated using different test cases and implementation approaches. For single continuum cases, both homogeneous and heterogeneous domains are used and results of the RWPT method are compared to analytical and numerical methods for two dimensions. For a simple homogeneous case, RWPT performs better than traditional finite difference (e.g. MT3D) and finite element (e.g. SUTRA and FEFLOW) methods, which introduce numerical dispersion. MT3D with total variance diminishing (TVD) solver significantly reduces numerical dispersion. For heterogeneous media, the RWPT method compares favorably with MT3D-TVD in terms of mean concentrations and uncertainty.

The RWPT method in homogeneous domains shows some sensitivity to the size of the support volume or projection area that is used to convert particle distribution to concentration values, but it shows a lesser sensitivity to time step and number of particles. For heterogeneous simulations, the concentration mean of the RWPT method is slightly sensitive to the number of particles used, whereas the variance shows minor sensitivity to the number of particles and the projection area. However, all these sensitivity results are minimal and of no significant effect on the resulting solution. Furthermore, velocity interpolation techniques do not have any noticeable effect on the concentration statistics for the test cases considered here where medium heterogeneity is assumed.

Two approaches for simulating matrix diffusion in fractured systems using RWPT methods are evaluated. A semi-analytical approach is compared to a RWPT technique, which accounts for matrix diffusion using particle transfer probabilities (Liu et al., 2000). We derived an empirical relationship that can be used to map the governing parameter of the semi-analytical approach to the corresponding particle transfer probability. The performance of the RWPT method with Liu et al.'s (2000) particle transfer probability is found to be inferior to that relying on a modified transfer probability proposed in Eq.(10). The latter leads to a delayed and reduced peak of mass flux and a long tailing behavior of the breakthrough curves, which are typical features of matrix diffusion effect. The original transfer probability of Liu et al. (2000) only reduces the peak of the mass flux and does not cause a delay in the arrival of the peak nor does it produce a noticeable tailing behavior.

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