

# On the Nonlocality of Reversible-Time Particle Tracking Methods

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This article presents a novel methodology for time reversal in groundwater transport simulations. A variance minimization procedure leads to a stochastic method, analogous to the Random Walk Particle Tracking Method (RWPTM), capable of tracking groundwater solute concentration profiles over time. This method, called the Reversible-Time Particle Tracking Method (RTPTM), can backtrack in time a certain solute plume, of arbitrary shape, and identify the most probable spill location. The correspondence to the “classical” RWPTM is established for forward-time simulations, and the method is applied to one-dimensional transport of both conservative and nonconservative solutes in spatially invariant and variant groundwater flow fields with great success.

Keywords: particle methods, reverse-time simulations, heterogeneity, pollution source identification

## Introduction

As the world’s population continues to grow, the demand for freshwater will continue to increase. From the 1% of the total freshwater available, 96% is in the form of groundwater, which in the case of the U.S. accounts for about half of the population’s source of drinking water. However, quantity is not the only problem; the quality of the drinking water is also a concern since this vital resource is vulnerable to contamination. With the advances of technology, more and more human activities are polluting the groundwater systems. Water pollutants may originate from point or nonpoint sources. A point source is characterized by the presence of identifiable, small-scale sources, while the non-point problem refers to larger-scale, more diffuse/spread contamination originating from many smaller sources. In the U.S. EPA 305(b) Report (U.S. EPA, 1998a), 37 states have reported that they found potential sources of groundwater contamination. Major sources of point-source pollution are leaking underground storage tanks, landfills, septic systems, and hazardous waste sites.

Once a contaminant is detected, an effort should be and is being made to clean up the groundwater system. This procedure is followed by the recovery of the cost from the identified responsible private parties. In case a private party is not identified, the money comes from federal or state sources (e.g., Superfund), which essentially is taxpayers’ money. The legislation has not been an unqualified success in assigning the liability and the cost

of cleanup is staggering. This can be seen by the fact that in 1994 the National Academy of Sciences estimated that over a trillion dollars, or approximately \$4,000 per person in the United States, would be spent in the next thirty years on cleanup of contaminated soil and groundwater (U.S. EPA, 1999). In many cases, it is hard to find out which companies or parties are responsible for the contamination due to lack of tools to identify the pollution source. Therefore, there exists a need for tools that can reconstruct the plume’s spatial and temporal history (National Research Council, 1990).

It is common practice for hydrogeologists to use computer simulation models in order to assess and study potential sources of contamination. Each and every one of the probable locations is considered as an a priori known feasible solution. Extensive simulations running forward in time provide a good understanding of how close to the present contaminant spatial configuration this potential source leads. This process, repeated as many times as the number of potential sources, ultimately pinpoints the most probable solute source. The disadvantage associated with this methodology is the extensive computational burden imposed. Backward or reversed time simulations employing conventional modeling approaches have been unsuccessful for solute source identification for the following reasons:

- Finite difference and finite element methods constitute numerical mass transport models of unstable behavior when used for backtracking of present contaminant configurations. The stability criteria are, for all practical purposes, restrictive enough to make their use nonfeasible. Atmadja and Bagtzoglou (2001a) have demonstrated this unstable behavior for a finite difference transport model.
- Particle tracking methods (PTMs), though effective in handling time reversal for purely advective contaminant transport,

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are incapable of treating time reversal for dispersive and/or reactive transport.

Even though direct approaches that perform time inversion in the context of contaminant source identification have enjoyed success and visibility lately, their widespread use is still limited due to the sophistication and nonconventional nature of the methods involved. Morrison (2000a, 2000b, 2000c), Atmadja (2001), Atmadja and Bagtzoglou (2001a, 2001b), and Bagtzoglou and Atmadja (2003) present thorough reviews of the hydrogeological applications of this field. The purpose of this article is to present a new methodology for time reversal in groundwater transport simulations. Based on a particle approach, a stochastic method is developed that can backtrack in time a certain solute plume, of arbitrary shape, and identify the most probable spill location. The Reversible-Time Particle Tracking Method (RTPTM), presented herein, is similar to the Random Walk Particle Tracking Method (RWPTM) in that it is fundamentally a particle method that involves an operator splitting algorithm, but differs in the treatment of the diffusive part of the process. It was considered, therefore, essential to present first a review of the “classic” RWPTM, followed by the development of the RTPTM. Then, the RTPTM and the RWPTM are compared and their interrelationship and equivalence, for forward time simulations, is established. Finally, the method is applied to one-dimensional test cases involving conservative and nonconservative solute transport in spatially invariant and variant flow fields.

### The Random Walk Particle Tracking Method

Contaminant transport in groundwater is represented by the advection-dispersion-reaction equation in its three-dimensional (3D) form

$$R \frac{\partial C}{\partial t} = \nabla \cdot [D \cdot \nabla C] - \nabla \cdot [v C] + r(C, t) \quad (1)$$

where  $\nabla$  is the gradient operator,  $C$  is the groundwater solute concentration,  $R$  is the retardation coefficient,  $D$  is the dispersion coefficient tensor,  $v$  is the transport velocity vector,  $t$  is time, and  $r(C, t)$  is a species reaction rate.

“Particle models,” according to Hockney and Eastwood (1981), is a generic term for the class of simulation models where the discrete representation of physical phenomena involves the use of interacting particles. Each particle may have a set of attributes such as mass, charge, position, momentum, heat, or vorticity. The basic concept behind this method is to represent the distribution of an extensive quantity—in the case of Eq. (1) the mass of the solute species—as a large collection of particles. Each particle is translated under the influence of diffusive-dispersive and/or advective driving forces over discrete time steps. The mass that each one of these particles carries can also be altered depending on the reaction terms involved. Since Ahlstrom et al. (1977) first applied this technique to numerical investigations of groundwater contaminant hydrology, many other investigators (Schwartz et al., 1983; Bagtzoglou

et al., 1992; Zimmerman et al., 2001) have used PTMs with success.

Neuman (1981) and Ewing (1988), among many others, reported the successful application of operator-splitting techniques to advective-diffusive transport problems in porous media. Equation (1), although involving a reaction term, can be treated using a time-splitting procedure according to which the change in concentration at a particular point in space, over a time period  $\Delta t = t_{n+1} - t_n$ , is defined as the sum of the changes in concentration due to diffusion, advection, and reaction. This can be expressed as

$$\Delta C = \Delta C_{AD} + \Delta C_R \quad (2)$$

where  $\Delta C$  is the total concentration change, and  $\Delta C_{AD}$ ,  $\Delta C_R$  are the changes in concentration due to advective-diffusive and reactive processes, respectively. Expanding the individual terms of the RHS of Eq. (2) we get

$$\begin{aligned} \Delta C_{AD} \\ = C^* - C^n \approx - \int_{t_n}^{t_{n+1}} \{ \nabla \cdot [v C] - \nabla \cdot [D \cdot \nabla C] \} dt \end{aligned} \quad (3)$$

and

$$\Delta C_R = C^{n+1} - C^* \approx \int_{t_n}^{t_{n+1}} \{ r(C^*, t_n) \} dt \quad (4)$$

where  $C^n$  and  $C^{n+1}$  are the concentrations at time  $t_n$  and  $t_{n+1}$ , respectively, and  $C^*$  is the concentration at the end of the advective-diffusive part of the time-splitting procedure. Then, Equation (3) can once again be expressed in a partial differential equation form as

$$R \frac{\partial C^*}{\partial t} - \nabla \cdot [D \cdot \nabla C^*] + \nabla \cdot [v C^*] = 0 \quad (5)$$

which is the well known Advection-Dispersion Equation (ADE). Similarly, Eq. (4) takes the Ordinary Differential Equation (ODE) form

$$\frac{dC^{n+1}}{dt} = r(C^*, t_n) \quad (6)$$

Following the time-splitting approach these equations are treated separately but are coupled via the concentration values after the advective-diffusive step. Wheeler and Dawson (1988) proved convergence of such an operator splitting algorithm.

We now concentrate on Eq. (5) and its numerical representation. The Fokker-Planck equation (FPE) reads (Haken, 1983; Gardiner, 1985)

$$\frac{\partial f}{\partial t} + \nabla \cdot (A f) - \nabla \cdot \left[ \nabla \cdot \left( \frac{1}{2} B \cdot B^T f \right) \right] = 0 \quad (7)$$

where  $f$  is some extensive quantity;  $A$  is a deterministic forcing vector; and  $B$  is a deterministic scaling (square) matrix. In order to make Eq. (7) analogous to Eq. (5), the following assignments have to be made:

$$\begin{aligned} f &= f(x, t) \Leftrightarrow C^* \\ A &= A(x, t) \Leftrightarrow \frac{1}{R}(v + \nabla \cdot D) \\ B \cdot B^T &= B(x, t) \cdot B^T(x, t) \Leftrightarrow 2D \end{aligned} \quad (8)$$

The particle locations are updated according to the step equation

$$X^{n+1} = X^n + A(X^n, t_n)\Delta t + B(X^n, t_n)W(t_n) \quad (9)$$

where the index  $n$  indicates time  $t_n$ ;  $X$  is the particle position vector;  $A$  and  $B$  have been previously described; and  $W(t_n)$  is a random forcing vector given by  $W(t_n) = Z_n \sqrt{\Delta t}$ , where  $Z_n$  is a normalized random number, at time  $t_n$ , with mean of zero and unit variance.

Equation (6) models the reaction mechanisms in terms of concentration (mass density) rather than mass itself. This usually leads to the development of a grid-based approach, which accommodates spatially nonhomogeneous reaction terms much easier than the particle-based algorithm. The reaction operator is applied on a grid basis and Eq. (6) is integrated by any appropriate ODE solver, such as the Euler or fourth-order Runge-Kutta method. This approach requires knowledge of the concentration field after the advection-diffusion operator has been applied. In addition, the changes in concentration due to reaction must be reflected in the particle masses and locations. Thus, a critical stage in applying the particle-grid methods to reactive problems is the accurate, or consistent, transformation of particle spatial locations and masses to a smooth concentration field used in the reaction modeling and vice versa. For first-order kinetics, however, the particle-based or particle-particle approach can give quite satisfactory results, as reported by Valocchi and Quinodoz (1989). It can also resolve the problem of consistent interpolation, which, especially in cases where high order interpolation or projection functions are used, requires an extensive additional computational effort (Bagtzoglou et al., 1992).

### The Reversible-Time Particle Tracking Method

Equation (9) is incapable of reversing the diffusion effects on the particle spatial spreading. This weakness stems from the fact that the diffusive part of the step equation involves the square root of time. It is therefore apparent that even if one were to reverse the time in the advective part of Eq. (9), the diffusive process would be still forcing the particles to spread around a receding center of mass. The RTPTM modifies the step Eq. (9) by introducing a variance or second moment minimization approach.

Diffusion or dispersion can be expressed in terms of the spatial variance of the particle cloud as

$$2D = \frac{d}{dt}(\sigma^2) \quad (10)$$

The spatial variance or normalized second-order moment matrix, corrected for the center of mass, reads

$$\begin{aligned} \sigma^2 &= \frac{1}{M} \int_{\Omega} C(x - \bar{x})(x - \bar{x})^T d\Omega \\ &= \sum_{i=1}^{N_p} \tilde{m}_i (X_i - \bar{X})(X_i - \bar{X})^T \end{aligned} \quad (11)$$

where  $N_p$  is the number of particles in the system;  $M = \sum_{i=1}^{N_p} m_i$  is the total solute mass within domain  $\Omega$ ;  $\bar{x}$  or  $\bar{X}$  and  $x_i$  or  $X_i$  are the position vectors for the plume center of mass and each individual particle, respectively, as expressed in their continuous or discrete form;  $\tilde{m}_i = \frac{m_i}{M}$  is the normalized particle mass; and the superscript  $T$  stands for the transposed vector.

When the solute transport process advances forward in time,  $D$  is a definite positive matrix, implying that the general 3D spread of solute about the center of mass  $\bar{x}$  is increasing. Similarly, negative diffusion and time reversal can be easily implemented by decreasing the spatial spread of the particle cloud around its center of mass.

The step equation for the RTPTM consists of the following set of equations:

$$\begin{aligned} X^n &= \bar{X}^n + F(\bar{X}^n, t_n)Q(t_n) \\ \bar{X}^n &= \bar{X}^{n+1} - A(\bar{X}^{n+1}, t_{n+1})\Delta t \\ F(\bar{X}^n, t_n) \cdot F^T(\bar{X}^n, t_n) &= \sigma_n^2 \\ \sigma_n^2 &= \sigma_{n+1}^2 - 2\Delta t D \end{aligned} \quad (12)$$

where vector  $Q(t_n) = Q^n = R^n S^n$ , vector  $R^n$  is a normalized random vector with mean of zero and unit variance and vector  $S^n$  is a random sign ( $\pm$ ) vector.

Equation set (12) constitutes the backbone of the RTPTM, being easily reduced to its two- or one-dimensional (2D or 1D) form. The method can be also applied to forward time simulations, by rearranging these equations, yielding results equivalent to the classical RWPTM. Consider a particle  $p$ , which at time  $t_n$  has a position vector given by  $X^n$ . Assume further that this particle is displaced, according to Eqs. (9) and (12), to  $X_*^{n+1}$  and  $X_{**}^{n+1}$ , respectively. If the two methods were to be equivalent the two positions should be equal, thus yielding

$$\begin{aligned} X^n + A(X^n, t_n)\Delta t + B(X^n, t_n)W(t_n) \\ = \bar{X}^n + A(\bar{X}^{n+1}, t_{n+1})\Delta t + F(\bar{X}^{n+1}, t_{n+1})Q(t_{n+1}) \end{aligned} \quad (13)$$

After some algebraic manipulations and assuming that the time step  $\Delta t$  is sufficiently small, the following equivalence is obtained:

$$\begin{aligned} B(X^n, t_n) &= \Phi(\varepsilon^n, t_n) + \Psi(\bar{X}^{n+1}, t_{n+1}) \\ \Phi(\varepsilon^n, t_n) &= \frac{\varepsilon^n}{Z^n \sqrt{\Delta t}} + \frac{A^*(\varepsilon^n, t_n) \sqrt{\Delta t}}{Z^n} \\ \Psi(\bar{X}^{n+1}, t_{n+1}) &= F(\bar{X}^{n+1}, t_{n+1}) \frac{R^{n+1} S^{n+1}}{Z^n \sqrt{\Delta t}} \end{aligned} \quad (14)$$

where an eccentricity vector

$$\varepsilon^n = \bar{X}^n - X^n \quad (15)$$

and a differential flow or differential drift velocity vector

$$A^*(\varepsilon^n, t_n) = A(\bar{X}^n, t_n) - A(X^n, t_n) \quad (16)$$

are introduced.  $\Phi$  and  $\Psi$  are nonlocal matrices, depending on the eccentricity of a specific particle and the center of mass of the particle cloud, respectively, which are both functions of time. Equation set (14) proves the existence of matrices  $B$ ,  $\Phi$ , and  $\Psi$ , which would ensure the equivalence of results in the

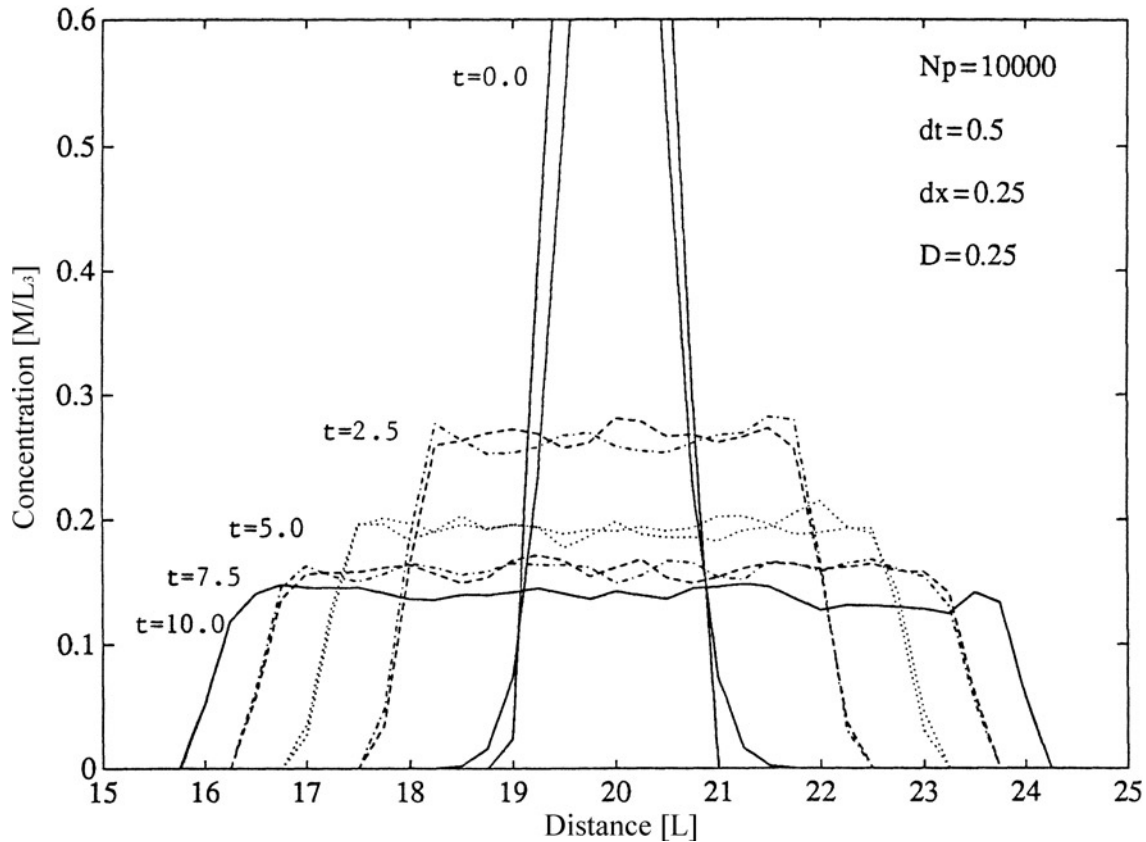
classical RWPTM and the forward-time RTPTM approach. The equivalence established by Eq. (13) is probabilistic in the sense that, on the average, the particle position as derived by the two methods is equal. This does not mean that the solutions obtained by RWPTM and RTPTM are identical. From the perspective of source identification, however, which is the interest here, the two solutions are indeed identical, as the first three moments are equal for the two methods.

## Test Cases

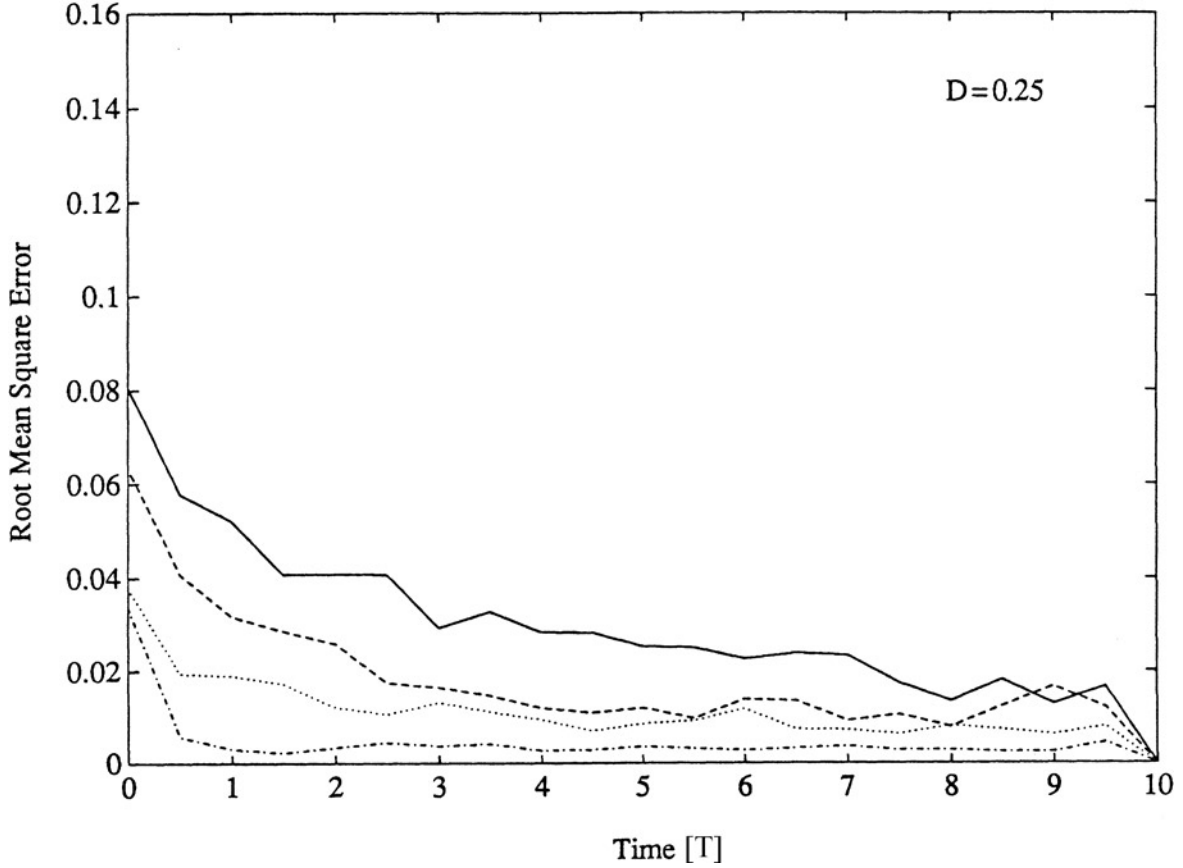
### Pure Diffusion

Consider a purely diffusive process, where  $v = 0$  and  $D_o = D_{\text{dif}} = 0.25$  [L<sup>2</sup>/T]. In the midpoint of a spatially invariant 1D domain  $x = [0, 40]$ , a Gaussian concentration profile is centered. The plume is diffused for a time  $t = 10$  [T] and then backtracked or diffused in reversed time to its original position. Figure 1 shows the concentration profile as it diffuses, forward and backward, in time. The numerical experiment involved  $N_p = 10,000$  particles, a grid spacing  $\Delta x = 0.25$  [L], and a time step  $\Delta t = 0.5$  [T].

There are nine curves, depicted in Figure 1, corresponding to time  $t = 0.0, 2.5, 5.0, 7.5, 10.0$ , and  $7.5, 5.0, 2.5, 0.0$  [T]. It is worthwhile noting that once time  $t = 10.0$  has been reached,



**Figure 1.** Concentration profiles at various times for a purely diffusive transport process. Overlapping curves correspond to forward and backward time simulations, respectively. Note that the graph is shown up to a concentration of 0.6 only for ease of comparison purposes.



**Figure 2.**  $E_{\text{rms}}$  as a function of time and number of particles. Solid,  $N_p = 500$ ; dashed,  $N_p = 1,000$ ; dotted,  $N_p = 2,000$ ; dashed-dotted,  $N_p = 10,000$ .

the time reversal starts yielding to only one  $t = 10.0$  curve. The results are clearly supportive of the claim that RTPTM is a time-reversible, accurate algorithm. Minimal discrepancies between curves, corresponding to equal forward and backward times, are attributed to the stochastic nature of the method. It should be noted that the total mass of the system is conserved to 0.1%.

Figures 2 and 3 present results from a parametric analysis, aimed at identifying the method's sensitivity. The 1D, purely diffusive process, described above, is simulated via the RTPTM using various values for the diffusion coefficient ( $D_o$ ) and the number of particles involved ( $N_p$ ). The root mean square error ( $E_{\text{rms}}$ ) is calculated as a measure of the discrepancy observed between the forward and backward simulated concentration profiles, at a specific time, as follows:

$$E_{\text{rms}}(t) = \frac{1}{N_g} \sqrt{\sum_i^{N_g} [c_i(t) - \hat{c}_i(t)]^2} \quad (17)$$

where  $N_g$  is the number of grid points; and  $c_i(t)$ ,  $\hat{c}_i(t)$  are the concentration values at node  $i$  at time  $t$  running forward and backward in time, respectively.

Figure 2 depicts the  $E_{\text{rms}}$  dependence on the number of particles and time for a constant diffusion coefficient  $D_o = 0.25$

$[L^2/T]$ . All four curves show an exponential-like decrease of the error with time. Figure 2 also shows the high sensitivity of the method to the number of particles, leading to minimal  $E_{\text{rms}}$  values when more than 2,000 particles are used. It also identifies early times, or the solute source proximity, as the area where most of the errors are likely to occur. The word “minimal” is used here in the relative sense when one compares the reduction attained in the  $E_{\text{rms}}$  values (less than 0.01 to 0.004) with the associated fivefold computational expense increase that using 10,000 versus 2,000 particles entails.

Figure 3 identifies the dependence of the  $E_{\text{rms}}$  on the value of the diffusion coefficient. These simulations involved a constant number of particles ( $N_p = 1,000$ ). The number of 1,000 particles was chosen in order to amplify the diffusion coefficient effect on the error. It can be seen that a decrease in  $D_o$ , from 1 to 1/16, has negligible effect on the  $E_{\text{rms}}$  for times  $t \geq 1$  [T]. During the first two time steps, however, the discrepancies are small, yet extant. It is important to realize that the above observations suggest that, counterintuitively, there exists a need to use more particles near the solute source in order to minimize the uncertainty involved. This would necessitate the implementation of adaptive approaches, whereby the number of particles involved in the simulation is increased as the plume is further tracked backwards in time.

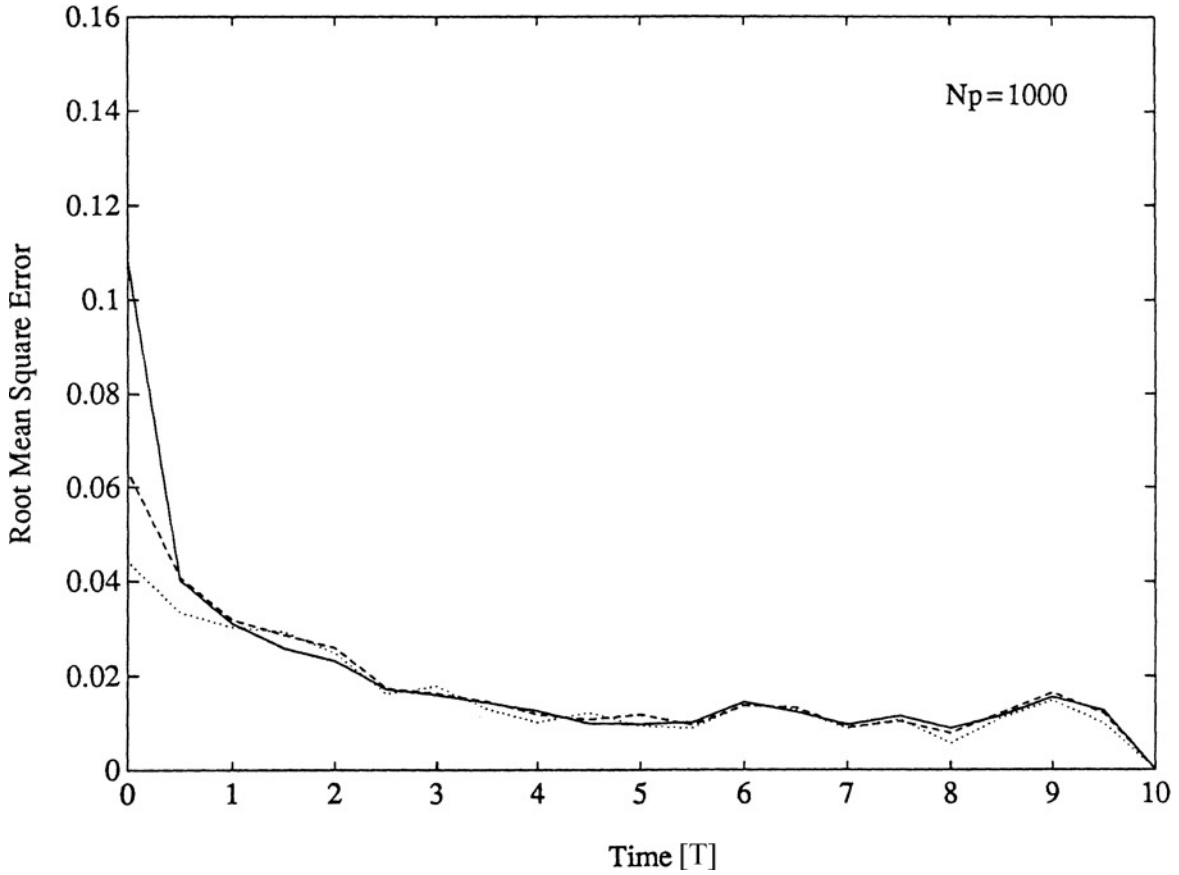


Figure 3.  $E_{rms}$  as a function of time and diffusion coefficient. Solid,  $D_o = 1$ ; dashed,  $D_o = 1/4$ ; dotted,  $D_o = 1/16$ .

#### Advection-Dispersion in Spatially Invariant and Variant Flow Fields

First, we consider an advective-dispersive process in a 1D domain  $x = [0, 100]$  with a spatially invariant, steady-state flow field  $v = 2.0$  [L/T] and a dispersivity  $\alpha = 0.5$  [L]. The molecular diffusion is  $D_o = D_{dif} = 0.0$  [L<sup>2</sup>/T], resulting in a dispersion coefficient  $D = \alpha v + D_{dif} = 1.0$  [L<sup>2</sup>/T]. Adsorption is assumed negligible, implying that the retardation coefficient  $R = 1.0$ . An instantaneous injection (spill) of solute mass  $\Delta M = 500$  [M] at location  $x = 14.0$  occurs at time  $t = 0.0$ . This type of initial condition can be expressed in terms of the Dirac delta function, as

$$C(x, 0) = \Delta M \cdot \delta(x) \quad (18)$$

where  $\Delta M$  is the total amount of solute contained in the domain and the Dirac function is defined as

$$\begin{aligned} \delta(x) &= 0 \\ \forall x &\neq 14.0 \\ \int_{-\infty}^{+\infty} \delta(x) dx &= 1 \end{aligned} \quad (19)$$

Finally, the solution has to be bounded, leading to  $C(\pm\infty, t) = 0$ . The solute plume is translated under the influence of advection, dispersion, and reaction until time  $t = 20.0$  [T]. The analytical solution for this problem is the well-known Gauss function (Kinzelbach, 1986)

$$C(x, t) = \frac{\Delta M}{2\sqrt{\pi\alpha vt}} \exp\left[-\frac{(x - vt)^2}{4\alpha vt}\right] \exp(-\lambda t) \quad (20)$$

where  $\lambda$  is the decay constant [T<sup>-1</sup>]. At any time  $t$ , the solution must satisfy the mass conservation criterion

$$\int_{-\infty}^{+\infty} C(x, t) dx = \Delta M \exp(-\lambda t) \quad (21)$$

The analytical solution to the problem described above, for a conservative (nonreacting) solute (i.e.,  $\lambda = 0.0$ ) is first calculated at different times. The solute plume at time  $t = 20.0$  is considered as the “present” field situation. The plume is then approximated via a cell-by-cell uniform particle distribution of equal mass attributes and tracked back in time until a possible solute source is identified. Equation set (12) is applied over discrete time steps until the spatial variance of the particle cloud becomes zero or sufficiently close to zero. The numerical

experiment was conducted with the use of  $N_p = 2,000$  particles, a time step  $\Delta t = 0.1$ , and a grid spacing  $\Delta x = 0.5$ . The algorithm identified with great accuracy both the location and the time of spill occurrence. According to this solute source identification scenario the spill occurred at time  $t = 19.7$  [T], before the present, at a location  $x = 13.8$  [L]. The error involved in the above identification process is less than 1.5% for both location and time of release.

Second, in order to test the RTPTM's ability to identify contamination sources in the presence of heterogeneities in the transport parameters, a test case with spatially variant transport velocity has been designed. We consider an advective-dispersive process in a 1D domain  $x = [0, 100]$  with spatially invariant dispersivity  $\alpha = 0.5$  [L] and molecular diffusion  $D = D_{\text{dif}} = 0.0$  [L<sup>2</sup>/T]. The flow is steady-state with  $v$  [L/T] being a nonstationary random field with mean 30.0 and standard deviation 15.0, and mean 10.0 and standard deviation 5.0 for the first half and second half of the domain, respectively. This results in a variable dispersion coefficient as depicted in Figure 4.

An instantaneous injection (spill) of solute mass  $\Delta M = 57.5$  [M] at location  $x = 47.50$ , occurs at time  $t = 0.0$ . The contaminant is then advected and dispersed until  $t = 0.34$  [T]. The numerical experiment was conducted with the use of  $N_p = 5,750$  particles, a time step  $\Delta t = 0.005$ , and a grid spacing  $\Delta x = 0.5$ . The algorithm identified with great accuracy both the

location and the time of spill occurrence. According to this solute source identification scenario, the spill occurred at time  $t = 0.335$  [T], before the present, at a location  $x = 47.47$  [L]. The error involved in the above identification process is less than 4% for both location and time of release. Figure 5 shows the initial and final concentration distribution through this time-reversal process.

At every time step the first three moments ( $M^0$ : solute mass contained in the plume,  $M^1$ : location of the center of mass of the plume, and  $M^2$ : variance of the plume) of the particle cloud are calculated and stored for postprocessing purposes. Calculating the slope of the linear trend in both Figures 6 and 7, the "stochastically imposed" velocity and dispersion can be determined. In the case of Figure 6, the time derivative of the first moment of the particle cloud (i.e., the velocity with which the plume's center of mass was tracked back to its original position) was found to be 30.43 and 9.52 [L/T], slightly below the ideal  $v = 30.0$  and 10.0 [L/T]. Similarly, from Figure 7 the time derivative of the second moment of the particle cloud (i.e., the dispersion coefficient multiplied by a factor of 2) was found to be 26.96 and 9.98 instead of the ideal 30.0 and 10.0 [L<sup>2</sup>/T]. Again, the errors involved are considered more than satisfactory.

Finally, the solution of Eq. (20), for a decay constant  $\lambda = 0.025$  [T<sup>-1</sup>], at times  $t = 0.0, 5.0, 10.0, 15.0,$  and 20.0 is calculated. Following a procedure similar to the one applied for the

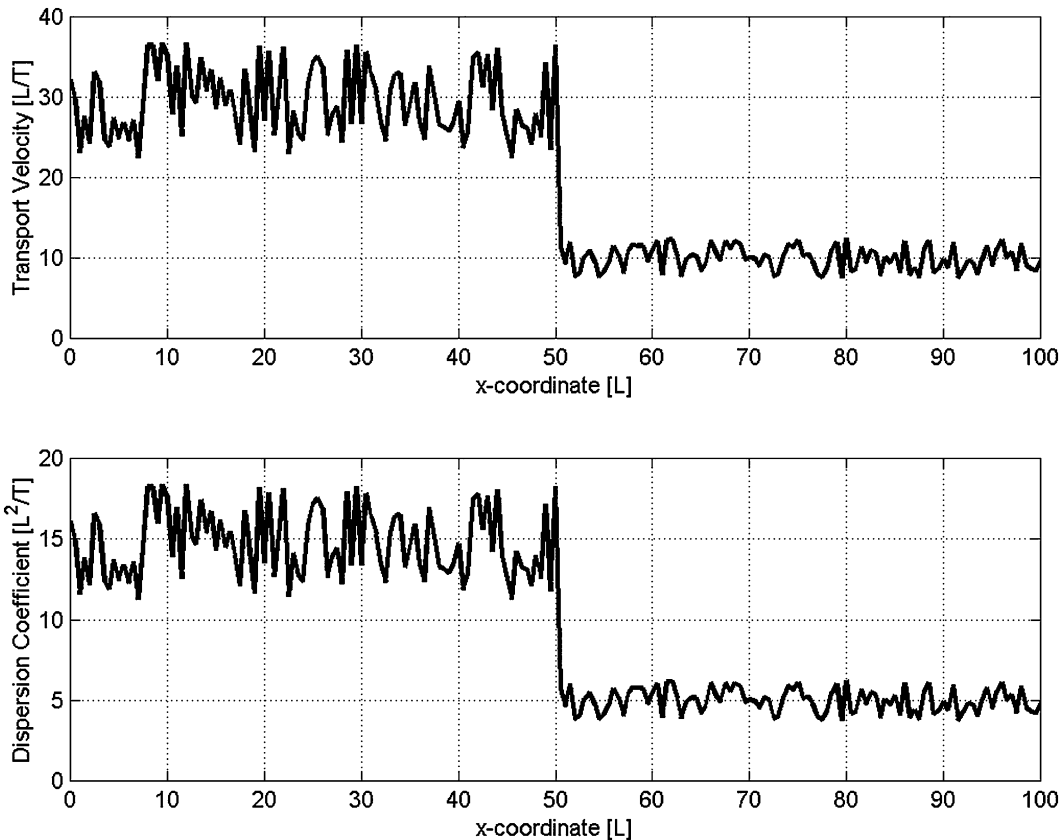
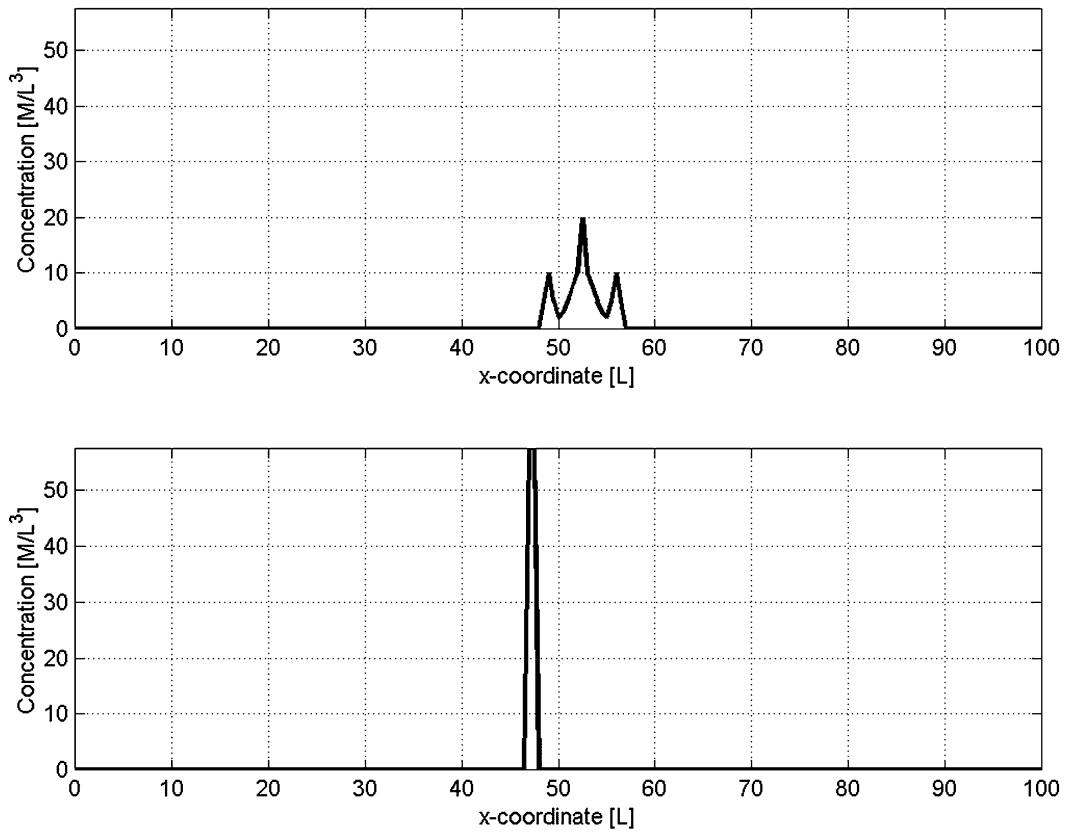
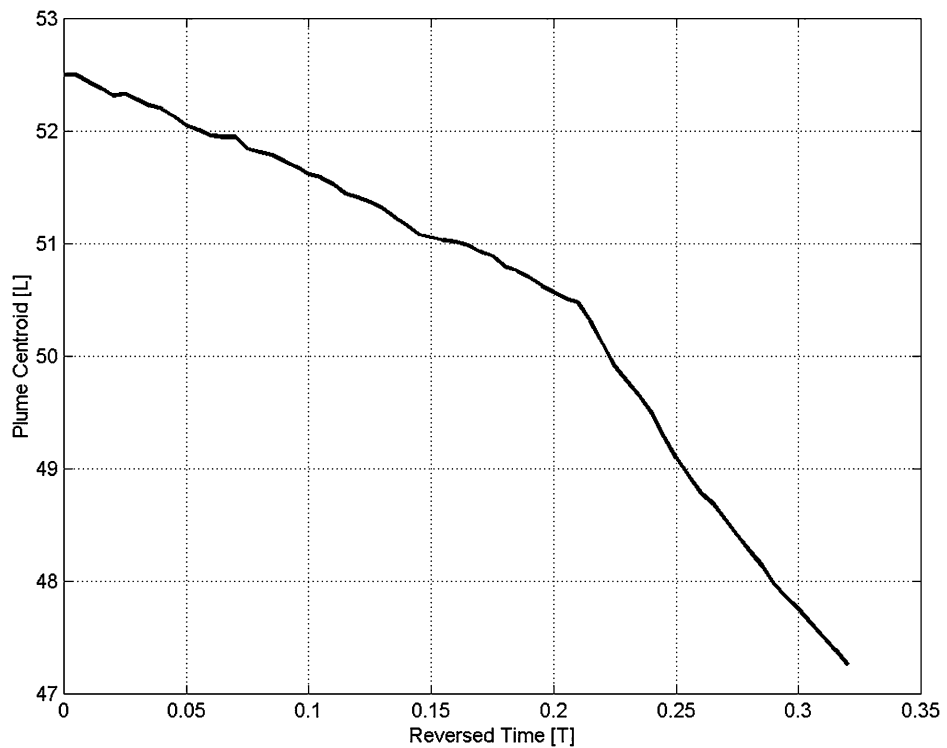


Figure 4. Spatial variation of the transport velocity and dispersion coefficient.



**Figure 5.** Spatial distribution of concentration before and after the time reversal process. Top: present-day spatial configuration. Bottom: spatial configuration after time-reversal analysis.



**Figure 6.** Temporal variation of the plume center of mass location for a conservative (nonreacting) solute.

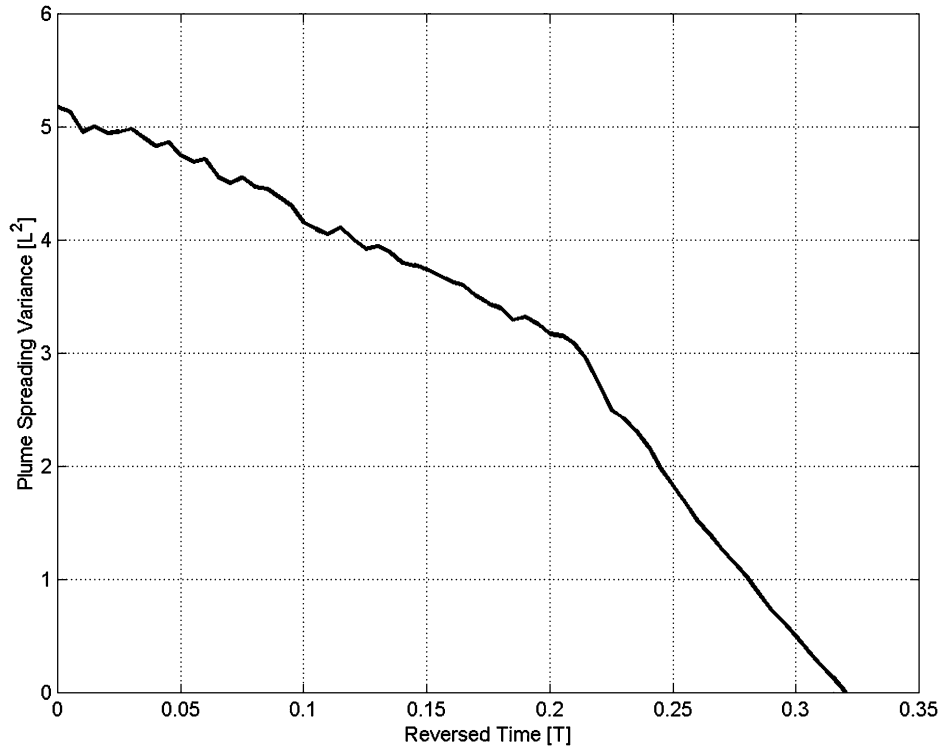


Figure 7. Temporal variation of the plume spatial variance for a conservative (nonreacting) solute.

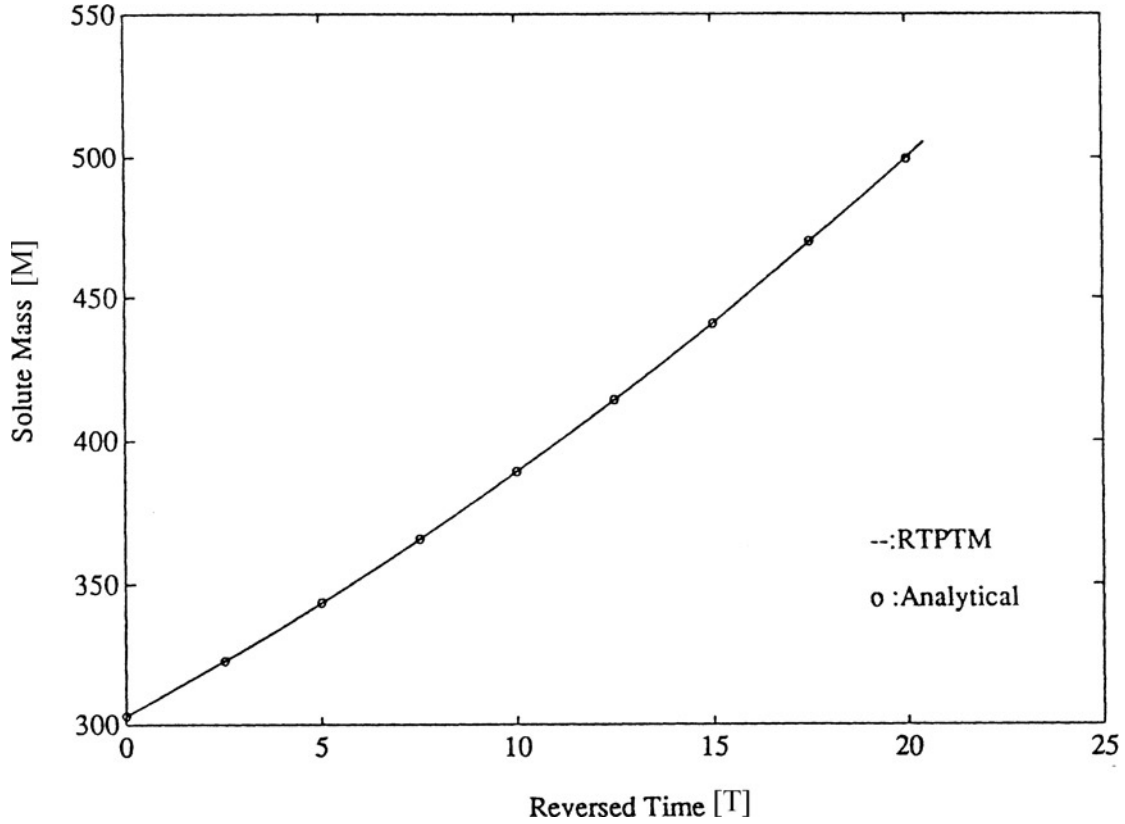


Figure 8. Temporal variation of the total solute mass for the RTPTM and the analytical solution in the case of nonconservative (reactive) transport.

conservative solute case, the analytical solution at  $t = 20.0$  [T] is considered as the present condition. To account for the first-order reaction involved in the process, the mass attribute of each particle is updated. Since the reaction rate is spatially invariant, a particle-based mass updating was followed. This is modeled via the ODE

$$\frac{dm_p}{dt} = \pm \lambda m_p \quad (22)$$

where the + and – signs correspond to backward and forward time simulations, respectively. Since the reaction process amplifies the noise, especially for reversed time simulations, additional computational effort is required in order to maintain the uncertainty at low levels. The results for a reactive solute source identification scenario are depicted in Figure 8. The numerical experiment involved  $N_p = 4,000$  particles, a time step  $\Delta t = 0.05$ , and a grid spacing  $\Delta x = 0.5$ . The total solute mass contained in the plume is in agreement with the analytical solution's conservation criterion of Eq. (21). The error introduced, however, in the source identification was 2.3% and 7% for a spill occurrence of  $t = 20.4$  [T] before present and a spill location of  $x = 13.1$  [L], respectively.

## Conclusions

A novel methodology for time reversing in particle tracking methods was presented. A variance minimization procedure leading to a stochastic method, analogous to the RWPTM, capable of tracking back in time existing groundwater solute concentration profiles, was developed. The method was applied to 1D transport of both conservative (nonreacting) and nonconservative (reacting) solutes in spatially invariant and variant flow fields with great success.

A parametric analysis, aimed at identifying the method's sensitivity, was conducted and the high sensitivity of the method to the number of particles was presented. It was also shown that a decrease in diffusion  $D_o$ , from 1 to 1/16, has negligible effect on the  $E_{\text{rms}}$  for times  $t \geq 1$  [T]. During early time steps, however, the discrepancies are small, yet extant. It is important to realize that the above observations suggest that, counterintuitively, there exists a need to use more particles near the solute source in order to minimize the uncertainty involved. This would necessitate the implementation of adaptive approaches, whereby the number of particles involved in the simulation is increased as the plume is further tracked backward in time.

The ability of the RTPTM to identify groundwater contamination sources in the presence of heterogeneities in the transport parameters was tested with a present-day plume of arbitrary shape tracked in a spatially variant transport velocity that follows a nonstationary random field. The error involved in the above identification process is less than 4% for both spill location and time of release. Finally, the method was tested for systems exhibiting first-order reactions. It was found that since

the reaction process amplifies the noise, especially for reversed time simulations, additional computational effort is required in order to maintain the uncertainty at low levels. The error introduced in the source identification for nonconservative systems was 2.3% and 7% for spill occurrence and location, respectively.

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