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Key Points:

- A Random-Walk Particle Tracking (RWPT) model is developed for transport under heterogeneous unsaturated conditions
- RWPT more stable, especially in case of coarse discretization, and more efficient than a series of Eulerian schemes
- RWPT performs well in a range of typical unsaturated soil transport problems, including transient conditions

Supporting Information:

Supporting Information may be found in the online version of this article.

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Unsaturated Transport Modeling: Random-Walk Particle-Tracking as a Numerical-Dispersion Free and Efficient Alternative to Eulerian Methods

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Abstract Lagrangian methods, such as the random-walk particle-tracking (RWPT), are often qualified as a potentially valuable alternative to error-prone Eulerian methods for simulating solute transport in unsaturated porous media. Yet, the RWPT method has not yet been validated against - and compared to - currently used Eulerian solutions for simulating solute transport under a range of typical unsaturated conditions. This paper presents a new implementation of the RWPT approach for advective - dispersive transport problems under variably saturated conditions. We first show that, as previously demonstrated for a heterogeneous dispersion tensor, using an interpolation scheme in the RWPT algorithm performs well for problems with abrupt changes in the water content. The new model is then compared against a simple 1D uniform transport problem, for which an analytical solution exist, and against a variety of 1D and 3D numerical solutions using the different Eulerian schemes implemented in Hydrus software suite. Results show that, while the Eulerian solutions significantly suffer from numerical dispersion in case of a coarse spatial discretization of the simulation domain, the new Lagrangian model provides accurate solutions for all problems. Furthermore, RWPT reproduces accurately solute transport for typical unsaturated flow conditions (infiltration, evaporation). Moreover, the Lagrangian model appears to be orders of magnitude faster than its Eulerian alternative to solve a 3D heterogeneous problem. Thus, RWPT should be seen as an attractive, stable and efficient alternative for simulating solute transport in the vadose zone, especially in case of complex and large problems.

Plain Language Summary Numerical models of the evolution of contaminant in soils is, potentially, a powerful tool to manage the quality of the water resources and the health of associated ecosystems. Yet, most of currently used models generate errors that can significantly deteriorate the quality of predictions. This paper presents a new numerical approach that simulates accurately and efficiently the motion of contaminants in variably saturated porous media by representing the solute as a large number of moving particles. The new model is (a) more accurate to reproduce solute transport in coarsely described and heterogeneous models, and (b) order of magnitude more efficient than equivalent Eulerian models.

1. Introduction

Predicting the evolution of subsurface water quality is a vital task for protecting water resources and the humans and ecosystems exposed to chemicals. This effort remains heavily dependent on numerical models. Providing stable and reliable numerical solutions as well as raising awareness on their limitations is crucial, especially for supporting decision making in case of contamination (Šimůnek & Bradford, 2008). This applies especially to transport models in the vadose zone, which is a key component of the Earth's critical zone and simultaneously represents the main pathway of contaminant toward the groundwater and an important zone of natural remediation (Ashraf et al., 2014; Stenger et al., 2008).

Virtually all vadose zone transport models are solving the advection-dispersion equation (ADE) (Nolan et al., 2005; Zheng & Bennett, 2002). The ADE can be applied to both fully saturated and partially saturated porous media systems using similar numerical schemes and solvers. An array of numerical methods can be used to solve the ADE. Among them, two main approaches can be identified: the Eulerian methods and the Lagrangian methods. Grid-based Eulerian approaches are overwhelmingly used by vadose zone pollution modelers (Šimůnek, 2006; Šimunek et al., 2016). These models have the advantage of being mature, well documented and versatile in terms of simulated processes (Steefel et al., 2015). However, Eulerian schemes are subject to a series

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of numerical error such as numerical instabilities, numerical diffusion and numerical dispersion, especially in case of complex, heterogeneous conditions (Boso et al., 2013). The terms "numerical diffusion" and "numerical dispersion" are often used synonymously (Keunings, 2000). According to Andersson et al. (2011), for a constant liquid flow, if the numerical grid cells are not parallel to the flow, an artificial transport of mass (numerical diffusion) to neighboring grid cells occurs. These errors can occur for all grid based numerical solutions and for advection only problems. Numerical errors of Eulerian solutions may lead to a misrepresentation of dispersion, mixing or dilution.

Langrangian approaches have been highlighted as stable, numerical dispersion free, alternatives to model solute transport under a range of conditions, from complex reactive transport (Dentz et al., 2011; Engdahl et al., 2017) to improved upscaling approach (Guo et al., 2020) and mainly for problems under saturated conditions. During the last two decades, a renewed attention has been focused on the random-walk particle-tracking (RWPT) method (Benson et al., 2017; Delay et al., 2005; Henri & Fernàndez-Garcia, 2015; LaBolle et al., 1996; Salamon et al., 2006). In basic terms, the approach consists in incrementally move a large number of particles to simultaneously simulate their motion by advection and diffusion/dispersion (Ahlstrom et al., 1977; Prickett et al., 1981). RWPT models have demonstrated a great potential to simulate transport under heterogeneous conditions in an efficient and stable manner (e.g., Boso et al., 2013; Ding et al., 2017).

RWPT models also present some challenges in their implementation. For instance, applying RWPT theoretically requires a very large number of particles to avoid sub-sampling induced errors on local concentration or breakthrough curves estimation, which can greatly increase the computational cost. This problem can be tackled by applying post-processed smoothing techniques such as the Kernel Density Estimator to limit the number of particles (Fernàndez-Garcia & Sanchez-Vila, 2011; Pedretti & Fernàndez-Garcia, 2013) or modern parallelization schemes to significantly decrease computation time (Rizzo et al., 2019).

It is also commonly thought that particle tracking techniques do not perform well under transient flow condition, especially when sink or source terms are involved. This specific issue can, in most cases, be addressed by specifying a mass to each particle and extracting or injecting particles according to a specified sink or source mass flux (see an application in Ding et al., 2017). The potential associated sub-sampling effect can be addressed by readjusting local density of particle in any under or over particle populated volume of the simulated domain (LaBolle, 2006; Winston et al., 2018). A few studies applied the RWPT technique to simulate transient transport in soils. For instance, Bechtold, Haber-Pohlmeier et al. (2011) reproduced transport experiments to better understand solute mass dynamic during evaporation. Later, Cremer et al. (2016) investigated the combined impact of dynamic boundary conditions and soil heterogeneity. Yet, the performance and accuracy of the used RWPT algorithm under such transient and heterogeneous conditions is not assessed, especially in comparison with equivalent Eulerian models.

Moreover, RWPT algorithms may produce errors when the applied dispersion tensor presents discontinuities due to the strong spatial variability in the velocity field, which is often solved over a grid-based flow model (Delay et al., 2005; LaBolle et al., 1996; Salamon et al., 2006). This has been shown for saturated heterogeneous porous media but remains true when applied in unsaturated conditions. Moreover, the vadose zone can also present strong spatial variability in water contents (e.g., layering), which has the potential to create similar error in the particle displacement. Corrective algorithms such as the interpolation method (LaBolle et al., 1996), the generalized stochastic differential equations method (LaBolle et al., 2000) and the reflecting barrier method (Bechtold, Vanderborght, et al., 2011; Lim, 2006) can be implemented in order to reduce errors due to discontinuities, but their efficiency on dealing with discontinuities in the water content remains to be shown.

Since decades, a wide array of Eulerian (e.g., Leonard, 1991; Lin & Falconer, 1997), Lagrangian (e.g., Benson et al., 2017; Boso et al., 2013) or mixed Eulerian-Lagrangian (e.g., Neuman, 1984; Sorek, 1988; Yeh, 1990) schemes have been compared to and validated against more mature - and often limited or unstable - approaches. During its initial development in the late 70's and 80's, the RWPT approach has been tested against contemporary Eulerian approaches (Ahlstrom et al., 1977) or a series of theoretical solutions (Prickett et al., 1981). Since then, particle tracking and grid-based techniques have radically evolved and improved to solve more complex and "realistic" contamination scenarios and the performance of RWPT has not been tested for typical unsaturated modeling setting, that is, infiltration of solute in different soil types, more complex flow conditions such as

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transient infiltration/evaporation systems or soils with spatially variable hydraulic properties and water content described in three dimensions.

Originally developed to solve transport under both saturated and unsaturated conditions (Ahlstrom et al., 1977), the application of RWPT in unsaturated media remains virtually nonexistent despite the potential high benefits that it could represent it term of efficiency and reliability. The fact that the community lacks of (a) an implementation scheme specific to unsaturated condition, that is, accounting for spatially variable water contents, (b) an illustration of the performance (numerical stability and efficiency) of modern RWPT compared to currently used Eulerian models, and (c) its application to soil-specific conditions (e.g., heterogeneous water content, transient condition such as during infiltration/evaporation period) can explain the marginal use of the Lagrangian method to model transport in soils. In this context, we present a new implementation scheme that accounts for discontinuities in both the dispersion tensor and the water content. To illustrate the performance and applicability of the method, the new scheme is then tested against commonly used Eulerian schemes solving conservative transport under different space and time discretization, dimensions, heterogeneity and soil-specific flow conditions.

2. Method

2.1. Random-Walk Particle-Tracking Algorithm for Unsaturated Conditions

The RWPT method has been widely used to simulate physical processes involving diffusion (Ibe, 2013). Delay et al. (2005) or Salamon et al. (2006) provide extensive reviews of the method. Here, we recall briefly its background, highlight the specificity of its application to unsaturated media, and propose a new implementation scheme.

2.1.1. Fundamental Equations

Three dimensional transport in the unsaturated zone for a conservative solute is described by the ADE:

$$\frac{\partial(\theta c)}{\partial t} = -\nabla \cdot (\theta \mathbf{u} c) + \nabla \cdot (\theta \mathbf{D}^{w} \cdot \nabla c), \tag{1}$$

where c [M L^{-3}] is the solute concentration, θ [L^3 L^{-3}] is the water content, $\mathbf{u}$ [L T^{-1}] is the average velocity vector in the porous medium and $\mathbf{D^w}$ [L^2 T^{-1}] is the hydrodynamic dispersion tensor in the water phase given by (Bear, 1972):

$$\mathbf{D^w} = (\alpha_T |\mathbf{u}| + D_w)\,\delta + (\alpha_L - \alpha_T)\,\frac{\mathbf{u}\mathbf{u}^T}{|\mathbf{u}|}, \tag{2}$$

where α_L [L] and α_T [L] is the longitudinal and transverse dispersivities, respectively, D_w [L^2 T^{-1}] is the molecular diffusion and δ is the Kronecker delta function.

Fokker-Plank-Kolmogorov Equation (FPKE) and Itô-Taylor integration scheme Traditionally, RWPT is used to solve stochastic differential equations or FPKE under the form:

$$\frac{\partial P(\mathbf{x}_p, t)}{\partial t} = -\nabla \cdot (\mathbf{A}(\mathbf{x}_p)\, P(\mathbf{x}_p, t)) + \nabla\nabla : (\mathbf{B}(\mathbf{x}_p)\, P(\mathbf{x}_p, t)), \tag{3}$$

where $P(\mathbf{x}_p, t)$ is the probability density for a particle to be at the location $\mathbf{x}_p$ at time t, $\mathbf{A}(\mathbf{x}_p)$ [L T^{-1}] is the mean of the jump velocity and $\mathbf{B}(\mathbf{x}_p)$ [L^2 T^{-1}] is the displacement matrix that can be seen as the statistical dispersion of the velocity around its mean (Delay et al., 2005).

A FPKE can be iteratively solved by applying, in the limit of a large particle number and an infinitesimally small time step, the Itô-Taylor scheme (Ito, 1951):

$$\mathbf{x}_p(t + \Delta t) = \mathbf{x}_p(t) + \mathbf{A}(\mathbf{x}_p, t)\,\Delta t + \mathbf{B}(\mathbf{x}_p, t) \cdot \xi(t)\sqrt{\Delta t}, \tag{4}$$

where $\mathbf{x}_p$ is the particle location, Δt is the time step of the particles jump and ξ is a vector of independent, normally distributed random variables with zero mean and unit variance.

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This scheme, which correspond to the RWPT method, basically consists in moving particles by successive displacement characterized by a mean of size $\mathbf{A}(\mathbf{x}_p)\Delta t$ and a covariance $\mathbf{B}(\mathbf{x}_p)\Delta t$.

Equivalence with the Advection-Dispersion Equation The ADE Equation (1) and the FPKE Equation (3) are similar in their forms, being both composed of a drift term that can be assimilated to advection, and a diffusive/dispersive term. A full equivalence is obtained by adapting the drift term such that

$$\mathbf{A} = \mathbf{u}(\mathbf{x}_p) + \nabla \cdot \mathbf{D}(\mathbf{x}_p) + \frac{1}{\theta(\mathbf{x}_p)}\mathbf{D}(\mathbf{x}_p) \cdot \nabla\theta(\mathbf{x}_p). \tag{5}$$

The displacement matrix relates to the dispersion tensor as:

$$2\mathbf{D} = \mathbf{B} \cdot \mathbf{B}^T. \tag{6}$$

Using these expressions of **A** Equation (5) and **B** Equation (6), the ADE can be solved using the RWPT algorithm Equation (4).

Note that the FPKE Equation (3) relies on a stationary assumption implying temporally invariable velocity, dispersion tensor and water content. However, this stationary assumption can be relaxed when the time of a particle jump is smaller than the time discretization of any transport parameters, allowing the application of the RWPT algorithm to transient problems (Delay et al., 2005).

Issues with discontinuities Natural subsurface systems are highly heterogeneous and often characterized by abrupt changes in soil and geological materials (e.g., Hartemink et al., 2020). Physical properties of grid-based flow solvers used under heterogeneous conditions are parameterized at a series of discrete points, which generates discontinuities in output parameters (velocities, porosity/water contents) that are subsequently used to solve transport. Yet, the stochastic differential Equation (3) and the related RWPT algorithm Equation (4) require smooth transitions in the water front within flow fields in order to preserve local solute mass conservation, that is, $\nabla \cdot \mathrm{D}$ and $\nabla \cdot \theta$ has to be small (Tompson & Gelhar, 1990). Techniques such as the interpolation method (LaBolle et al., 1996) and the reflection method (Bechtold, Vanderborght, et al., 2011) have been successfully tested to improve local solute mass conservation and reduce errors due to discontinuities in the dispersion tensor under heterogeneous conditions. Salamon et al. (2006) provide a clear description of those methods and discuss their advantages and limitations.

Interestingly, Bechtold, Vanderborght, et al. (2011) improved the reflection method to resolve the main limitations described by Salamon et al. (2006) and highlighted the sensitivity of the interpolation method to the grid resolution. The sensitivity of the solution to the discretization depends most likely on a set of parameters controlling the size of the particle jumps (e.g., advective flux, diffusion, dispersivity, characteristics of the heterogeneity) and is therefore complex to systematically test. Yet, in practice, geostatistical models used to describe spatial variability in aquifer materials employ resolutions fine enough to make both smoothing methods valuable to improve local mass conservation. Under unsaturated conditions, similar discontinuity related issues are likely to arise from the water content, which can also significantly vary over a short distance in the vadose zone, producing sharp transitions if solved on a grid (e.g., in case of soil layering). The relevance of accounting for water content discontinuities in RWPT algorithms has not yet been illustrated.

2.1.2. Implementation

For this study, we adapt the code RW3D (Fernàndez-Garcia et al., 2005; Henri & Fernàndez-Garcia, 2014, 2015) to the simulation of conservative transport in heterogeneous unsaturated porous media. RW3D is a Fortran 90, object-oriented code that has been previously developed to solve complex ADE-based transport problem in saturated and heterogeneous porous media (e.g., Henri et al., 2016). RW3D accounts for spatial and temporal variability in all parameters. Flow parameters (Darcy flux, water content), obtained from an external flow solver or not, can be described at each node or cell edge of a pre-defined rectilinear grid. The main modification required to adapt the code for unsaturated conditions is the implementation of transient, spatially variable water content and resolve discontinuities in θ, which is done by using the interpolation method.

Interpolation of water contents RW3D already uses interpolation schemes for treating discontinuities in the dispersion tensor (Salamon et al., 2006). This interpolation method approximates a continuous gradient term in

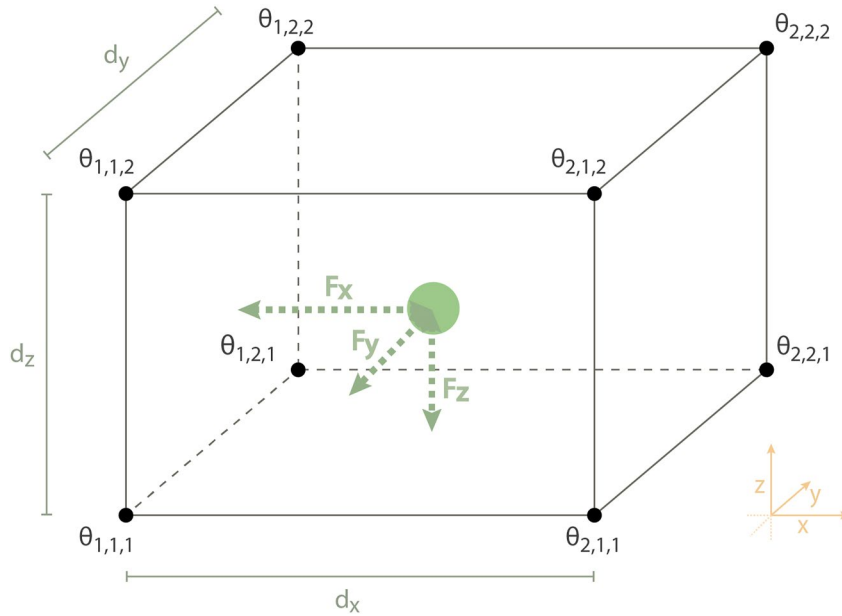

Figure 1. Illustration of the trilinear interpolation scheme.

the drift vector at every interface, resulting in a smooth dispersion tensor. A small time step and a fine discretization are then required to minimize errors due to the applied method (LaBolle et al., 1996).

Similarly, the water content assigned to a particle (θ_p) is estimated using a trilinear interpolation scheme of the water contents estimated at each node of the grid-cell in which the particle is located (Figure 1). The scheme is described as:

$$\begin{aligned}\theta_p = {} & (1-F_x)\times(1-F_y)\times(1-F_z)\times\theta_{1,1,1} + F_x\times(1-F_y)\times(1-F_z)\times\theta_{2,1,1}+\\ & (1-F_x)\times F_y\times(1-F_z)\times\theta_{1,2,1} + F_x\times F_y\times(1-F_z)\times\theta_{2,2,1}+\\ & (1-F_x)\times(1-F_y)\times F_z\times\theta_{1,1,2} + F_x\times(1-F_y)\times F_z\times\theta_{2,1,2}+\\ & (1-F_x)\times F_y\times F_z\times\theta_{1,2,2} + F_x\times F_y\times F_z\times\theta_{2,2,2}.\end{aligned} \tag{7}$$

where F_i is the relative location of the particle with a cell defined as $F_i = (x_{p,i} - x_{c,i})/d_i$, where $x_{p,i}$ is the ith component of the particle location, $x_{c,i}$ is the minimum ith coordinate of the cell in which the particle is located at a given time, and $\theta_{i,j,k}$ is water content values at the node $\{i, j, k\}$ (see Figure 1).

As previously mentioned, RW3D also interpolates Darcy fluxes to smooth the dispersion tensor. This scheme is described by substituting the water content with the flux in any direction in Figure 1 and Equation 7.

2.1.3. Test of the Interpolation Method

The importance of smoothing the water content is tested by analyzing the evolution of the first spatial moment (center of mass) of a plume in a 2D domain, consisting of two parallel layers. Such theoretical systems have been previously used by Salamon et al. (2006) and Bechtold, Vanderborght, et al. (2011) to test the performance of smoothing techniques to improve Lagrangian solutions in case of discontinuities in the dispersion tensor. The simplified setup allows the identification of a theoretical position of the center of mass of a plume and facilitates therefore the identification of a potential degradation of the tested algorithms. This setup is here adapted to test the interpolation of water content.

For the two layers we assume similar dispersivities ($\alpha_L = \alpha_T = 0.1$ m), and no diffusion. The horizontal Darcy flux is considered homogeneous (set to the arbitrary values of $q_x = 216$ m/d), so that discontinuities in the dispersion tensor are null, and will therefore not deteriorate transport solutions. The two layers are characterized by different water contents (θ_1 on the top layer, θ_2 on the bottom layer; see Figure 2) and the analysis is done for two ratios of θ_1/θ_2: 10.0 and 100.0. The domain is arbitrary chosen to be 1.5 m wide (L_y) and 1,000 m long (L_x) to ensure

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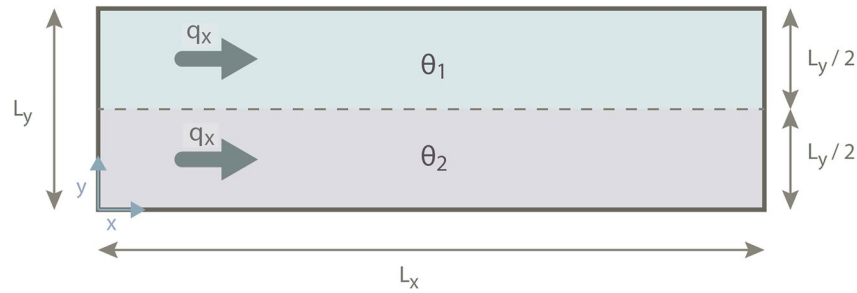

Figure 2. Scheme of the two layers aquifer used to test the importance of water content interpolation.

constant values of the first- and second-order moments before the particles exit the domain. The domain is discretized into two cells vertically ($d_y = 0.75$ m), and 100 cells horizontally ($d_x = 100.0$ m).

5,000 particles are injected at the left edge of the domain and the evolution of the plume's center of mass (first spatial moment) is recorded. The time step between particle jumps is specified in order to preserve the advective displacement, which is done using a grid Courant number (gCu):

$$\Delta t = gCu \times \min\left\{\Delta s/u_x, \Delta s/u_y, \Delta s/u_z\right\}, \tag{8}$$

where Δs is the characteristic size of the grid cell. Different gCu values are used for demonstrating the sensitivity of the random walk solutions to the time step. The same time step defining scheme (Equation 8) is used for the following 1D and 3D examples.

The theoretical position of the center of mass can be estimated as:

$$Y_g^* = \bar{y}_1 \times \left(1 - \frac{\theta_1}{\theta_2 + \theta_1}\right) + \bar{y}_2 \times \left(\frac{\theta_1}{\theta_2 + \theta_1}\right) \tag{9}$$

where $\bar{y}_i$ is the center location of the ith layer.

The solution with water contents estimated in each cell - with values associated to a particle updated only when the particle switches cells - is compared against the solution with values associated to a particle updated at each time step using the interpolation scheme.

2.2. Method for the Lagrangian - Eulerian Comparison

The RWPT algorithm is then tested against a series of 1D and 3D simulations solved using an analytical solution (for simple 1D transport) and a variety of grid-based Eulerian models. The objective here is to evaluate the applicability of the RWPT method to typical unsaturated modeling settings and to directly compare its performance under a variety of conditions. The different scenarios are then based on an increasing degree of complexity of the problem in order to cover the range of flow and transport conditions frequently represented in soil models - from 1D steady state homogeneous cases to dynamic infiltration/evaporation scenarios and 3D heterogeneous conditions.

2.2.1. Eulerian Approach

A large array of Eulerian schemes, each performing differently under specific modeling setting, have been developed over the last decades to solve the ADE (Equation 1). We use the widely used Hydrus software suite (Šimunek et al., 2016) as an example of this variety of Eulerian approach. Solutions from these Eulerian schemes are then used for assessing performance of the RWPT method. We do not aim to compare the performance of the different Eulerian schemes, for which an extensive literature exists. Hydrus 1D is used for the solution of the one-dimensional problems (Jacques et al., 2008) and Hydrus 3D for the three-dimensional system (Šimunek et al., 2018). Hydrus 3D is a popular model in the field of vadose zone and has many applications in the field of solute transport.

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For the estimation of the advective fluxes required in Equation 1, water flow should be simulated first. Water flow in unsaturated soils is described by the Richards-Richardson equation (Richards, 1931; Richardson, 1922), which, in one dimension and assuming no sink-terms, is given by:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z}\left(K\left(\frac{\partial h}{\partial z} + 1\right)\right), \tag{10}$$

where θ [L^3 L^{-3}] is the volumetric water content, t [T] is time, z [L] is the vertical spatial coordinate, (considered positive upwards), h [L] is the pressure head, K [LT^{-1}] is the hydraulic conductivity. For potential limitations of Equation 10 please refer to Diamantopoulos and Durner (2012) and Diamantopoulos et al. (2012). The solution of Equation 10 requires the definition of soil hydraulic properties (SHPs), that is, the water retention curve $\theta(h)$ (SWRC) and the unsaturated hydraulic conductivity curve $K(h)$ (HCC). The so-called Mualem-van Genuchten (MVG) model (van Genuchten, 1980) is the most popular model in vadose zone hydrology, despite its shortcomings (Weber et al., 2019). For the MVG model, SWRC and HCC are described by:

$$\theta(h) = \begin{cases} \theta_r + (\theta_s - \theta_r) \times (1 + |\alpha h|^n)^{-m} & \text{if h} < 0 \\ \theta_s & \text{if h} \geq 0 \end{cases} \tag{11}$$

$$S_e = \frac{\theta(h) - \theta_r}{\theta_s - \theta_r} \tag{12}$$

$$K(S_e) = K_s \times S_e^l \times \left[1 - \left(1 - S_e^{1/m}\right)^m\right]^2 \tag{13}$$

where θ_s and θ_r [L^3 L^{-3}] are the saturated and residual water contents, respectively, α [L^{-1}], n [-], m [-], and l [-] are shape parameters, m = 1-1/n, $n > 1$, and S_e [-] is the effective saturation.

For the numerical solution of Equation (1), a series of space derivatives and time derivative solvers are used, which has the advantage of illustrating the diversity (even though not exhaustive) of Eulerian solutions for transport problems. The program offers two different options for calculating space derivatives:

- the Galerkin finite element method (GFE), which is a linear approach known to be subject to numerical oscillations when steep concentration fronts are being simulated (Šimunek et al., 2013)
- the Upstream Weighted Finite Element (UWFE), which applies non-linear basis functions that ensure that more weight is placed on flow velocities of nodes located at the upstream side of an element. This removes oscillations in the concentration front, however, as stated earlier, this is expected to add numerical dispersion to the solution.

A finite different scheme is used for the approximations of the time derivative in Equation (1). Hydrus proposes three options:

- an explicit scheme (EX), which is often considered unstable;
- a Crack-Nicholson (CN) implicit scheme, which is the standard solution;
- a fully implicit solution (IS), which may result in additional numerical dispersion.

All combinations of space and time derivatives solvers are here tested. For each problem we provide then six Eulerian solutions in total (two space weighting schemes, three times derivatives schemes). We refer to Šimunek et al. (2013) for more theoretical details about all schemes.

Hydrus is also used to obtain the flow and water content fields. The resulting values of the Darcian nodal fluxes were used for the advection component in RW3D. The same applies for the water content fields.

2.2.2. Homogeneous 1D Porous Medium

The first group of simulations includes a steady state water flow in a 1D vertical profile of length L = 100 cm (Figure 3). Simulations are conducted for two macroscopic (Darcy) fluxes (q_z) equal to 0.1 and 10 cm $days^{-1}$ with a constant water content of 0.3 cm^3 cm^{-3} throughout the column. We used a Neumann type boundary condition at the top and a unit gradient condition (free drainage) at the bottom of the column. We assume a longitudinal

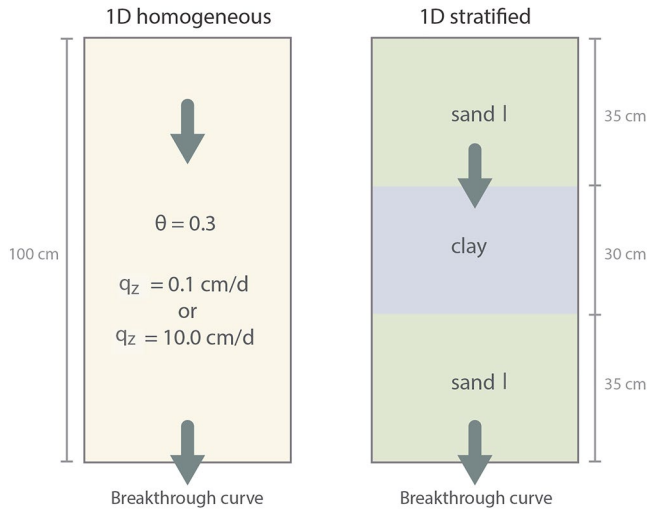

Figure 3. Scheme of the 1-dimensional soil conceptualized as a homogeneous column (left) and a stratified system (right).

dispersivity equal to 1 cm. Solute mass is applied at the top boundary ($L = 0$ cm) for 1.3 and 32 days (t_{inj}) for the scenario with $q_z = 0.1$ cm days^{-1} and $q_z = 10.0$ cm days^{-1}, respectively. We solve both problems for two different spatial discretizations of the simulation domain (1.0 and 10.0 cm).

For the transport simulations with Hydrus, the effect of the size of time steps on the solution is recalled by specifying three different values of Courant number: 0.1, 0.5 and 1.0. The total number of Hydrus simulations conducted for the first group was then: 2 fluxes × 2 discretizations × 3 Courant numbers × 6 solutions resulting in 72 different breakthrough curves at the bottom ($L = 100$ cm) of the soil profile.

For the RW3D simulations, 2 grid-Courant number (gCu) controlling the time step between particle jumps are tested: 0.2 and 1.0 (see Equation 8). Mass release is simulated by injecting a total of 10^5 particles at the top of the domain. Breakthrough curves of exiting particles at the bottom of the domain (i.e., outlet) are built using a Kernel density estimator method in order to reduce the potential impact of subsampling (Pedretti & Fernàndez-Garcia, 2013). This will be the case for all RW3D simulations presented in this study.

The accuracy of all numerical solutions is compared against BTC curves from an analytical solution, assuming a solute pulse input and a Dirac delta initial distribution (solute free columns, Toride et al. (1995)).

2.2.3. Layered 1D Porous Medium

The simulations for the high Darcy flux (10 cm days^{-1}) presented in the previous paragraph are repeated for a one dimensional layered system. We assume a vertical material distribution of sand I (0–35.0 cm) over clay (35.0–65.0 cm) over sand I (65.0–100.0 cm) (Figure 3). As for all the following scenarios, the soil hydraulic parameters used to simulate the flow field are shown in Table S1 in Supporting Information S1). After achievement of steady state conditions in the layered profile, the water content was approximately distributed as follow: Water content from 0 to 35 cm was equal to 0.43 cm^3 cm^{-3}, 0.38 cm^3 cm^{-3} from 35 to 65 cm and finally, 0.2 cm^3 cm^{-3} from 65 to 100 cm. A pulse of solute was applied for one day and the longitudinal dispersivity was equal to 1 m. For the layered profile and for the following solutions, an adaptive time step is used, where the product of Courant and Peclet number should always be lower than two (Perrochet & Bérod, 1993). This ensures the convergence of all solutions and allow an assessment of the performance using an optimized time step estimation.

In addition to BTCs at the column outlet, concentration spatial distributions will also be analyzed for each scenarios by tracking the number of particles in each grid cell. To avoid potential errors due to subsampling, the number of injected particles in increased to 10^6.

2.2.4. Homogeneous 1D Infiltration/Evaporation

Lagrangian approaches can face challenges in their implementation in case of more complex, dynamic conditions typically encountered in soils, such as periodic infiltration and evaporation events. To test this, we performed a dynamic simulation for a 2.0 m soil column assuming a very permeable material for 10,000 days. The material parameters were identical to the coarse sand parameters in Table S1 in Supporting Information S1, except of the saturated hydraulic conductivity parameter which was fixed equal to 10,000 cm/day. At the top, We applied a mass flux of 5.0 mg/cm2/d (5 cm/d × 1 mg/cm3) for one day, followed by a constant flux of solute free water of 0.5 cm/day for 9 days. After the wetting period of 10 days, we assumed an evaporation period with an evaporation flux of 0.5 cm/day until the end of the simulation period. At the bottom, we assumed a constant water table at 2 m, sustaining the upward flux during the evaporation period. For $t = 0$, we assumed a solute free column and a hydrostatic pressure head distribution with $h = 0$ cm at $z = 2$ m and $h = -200$ cm at z The simulation domain consists of 100 nodes (dz = 2.0 cm) and dispersivity was assumed equal to 1.0 and 10.0 cm. For this exercise, the main objective is less to compare the performance of the Eulerian schemes and RWPT, but more to validate the application of the Lagrangian approach under complex flow conditions. For this reason, only one Hydrus scheme (CN-GFE) is used.

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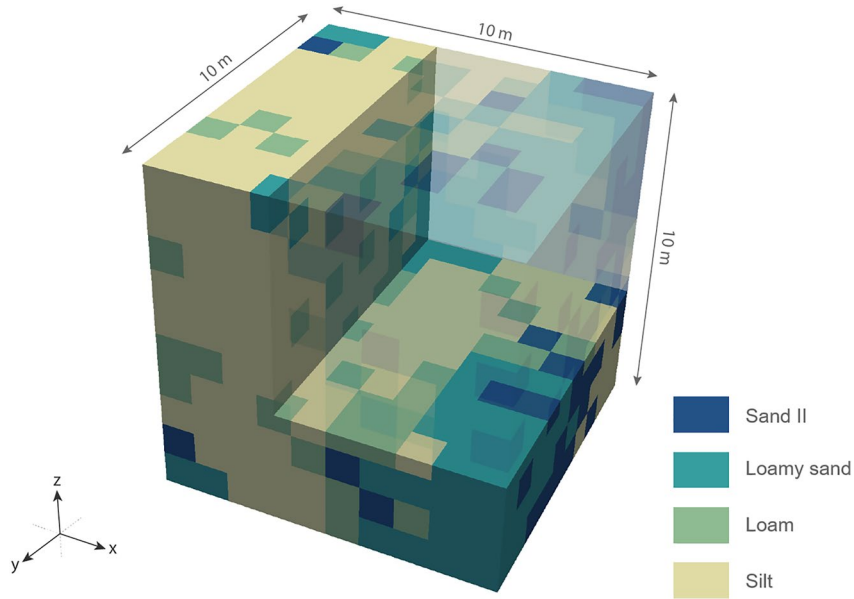

Figure 4. Scheme of the facies spatial distribution resulting from the TPROGS model used in the 3D simulations.

2.2.5. Heterogeneous 3D Porous Medium

In the third scenario, we assume a 3D heterogeneous cube with dimensions 100.0 × 100.0 × 100.0 cm. The cube is divided in 10 × 10 × 10 sub cubes, each one having different SHPs (either sand II, loamy sand, loam or silt, see Figure 4). The material sand II was assumed identical with the material sand I but we a lower saturated water content. In that way, we increased the water content difference at the interface between the least permeable materials (loamy sand, loam and silt) with the sandy II material. The spatial variability of the four hydrofacies is described using TPROGS, a geostatistical approach based on the probability/Markov chain method Carle (1999). Parameters are not representing a specific site and are chosen for illustration only. The proportions of each facies are fixed to 10% of sand, 35% of loamy and, 25% of loan and 30% of silt. The remaining geostatistical parameters are specified in the Table S2 in Supporting Information S1.

At the top surface, we apply a constant water flux of 0.08 cm hr^{-1}, and a unit gradient condition (free drainage) at the lower surface. Once we achieve steady state conditions throughout the profile, we apply solute mass at a constant rate for 5 hr. Solution are achieved for two discretizations (5.0 and 10.0 cm). To simplify our analysis of the performance of the different numerical solutions, both longitudinal and transverse dispersivities are assumed equal to 1.0 cm. Solute transport is monitored for 500 hr and BTCs at the lower surface are calculated for both Hydrus and RW3D simulations. For the Hydrus solution of the 3D transport problem, we use only the CN-GFE method (see 1D problems) due to the very high calculation time of problem (10's of hours) and due to non-convergence issues for fixed Peclet-Courant numbers. The time step for the solution is calculated following Perrochet and Bérod (1993), similarly to the layered problem. For this scenario, computational times required for both Hydrus and RW3D to solve the transport problem are recorded (PC: Intel Core i7-7700 @ 3.6 GHz, 32 Gb of RAM).

3. Results

3.1. Interpolation of Water Content

For a relatively mild water content interface ($\theta_1/\theta_2 = 10$), simulation outputs show a good performance for both schemes, with or without interpolation (Figure 5). Yet, using water content interpolation generates an exact center of mass location, after convergence, while omitting it leads to a minimal error. This result is independent of the time step between particle jumps.

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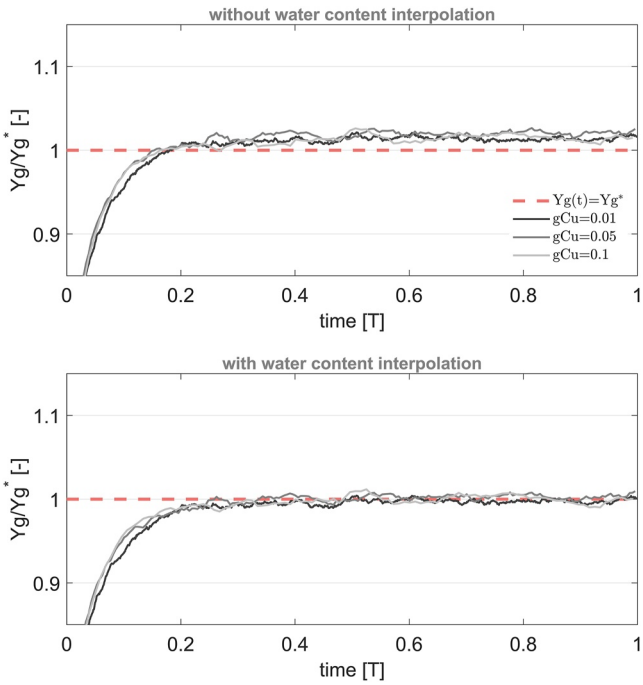

Figure 5. Center of mass location for $\theta_1/\theta_2 = 10$ applying linear interpolation between the water content at the particle location at time t and t + Δt (bottom) and without applying the interpolation scheme (top) for different temporal discretization criteria (expressed in terms of grid Courant number).

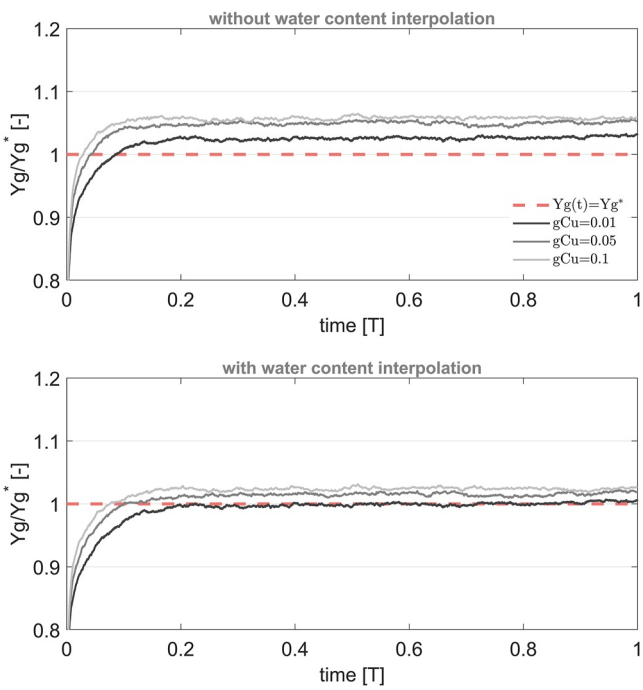

Figure 6. Center of mass location for $\theta_1/\theta_2 = 100$ applying linear interpolation between the water content at the particle location at time t and t+Δt (bottom) and without applying the interpolation scheme (top) for different temporal discretization criteria (expressed in terms of grid Courant number).

In case of sharper water content interface ($\theta_1/\theta_2 = 100$), the solution without interpolation is degraded, especially for larger time steps (Figure 6). When the water content assigned to a particle is interpolated, the estimation of the location of the plume center of mass is improved. The solution converges toward to the true solution when the time step is reduced. Yet, larger time steps (Cu = 0.1) produce only minimal error. The good performance of the interpolation method under our modeling settings indicates then that the resolution is fine enough to obtain a satisfactorily solution.

These results highlight that, as previously shown for the dispersion (Salamon et al., 2006), the interpolation of water content can improve the reproduction of mass distribution in systems with sharp water content interfaces. Therefore, this new scheme was implemented into RW3D and applied to all comparative scenario presented later on in this paper in order to ensure a better accuracy and stability of the solutions.

3.2. Homogeneous 1D Conditions

First, we recall the main limitations in the application of a series Eulerian methods in order to confront them to the new RWPT method. All tested Eulerian solutions appear to produce accurate outputs in case of fine spatial ($dz = 1$ cm) and temporal (i.e., low Courant number) discretization (Figure 7) and agree with the analytical solution. By increasing the cell size ($dz = 10$ cm), all Eulerian solutions generate significant errors on the concentration signal. Space weighting options mostly affect the magnitude of the error. GFE solutions produce slightly late arrival times and an increased dispersion. On the other hand, UWFE schemes produce early arrival times and an even greater overestimation of dispersion. The choice in the time derivative scheme influences less the solution performance. The EX performs the best, slightly limiting the overestimation of dispersion, followed by the Crack-Nicholson implicit scheme (CN) and finally the (IS, which produces the greater error (Figures 7b and 7d). The advective velocity does not significantly impact model performance. As shown in Supplementary Information, Figure S1 in Supporting Information S1, by increasing the Courant number (from 0.1 to 1.0), thus increasing the time discretization, all Eulerian solutions see their performance being degraded.

The analytical solution used to assess the performance of each scheme assumes an infinite 1D column. Yet, our modeling setting presents a finite column. Boundary conditions used in the Eulerian simulations (zero gradient at the outlet) could then generate undesired errors. Figure S2 in Supporting Information S1 shows that observing BTCs in quasi-infinite setting (the domain downstream of the observation plane is being made longer) leads to similar concentration signal in case of a fine discretization ($dz = 1$ cm) and a moderately later arrival of mass in case of a coarser discretization ($dz = 10$ cm). Nevertheless, increasing dz alone produces much larger error on the solution than the boundary condition: solving the problem on a coarse grid leads to significant numerical dispersion even in case of a semi-infinite column.

Choosing the spatial and temporal discretization as well as the spatial and temporal derivative schemes is then an important and sensitive step in the modeling of transport using Eulerian methods. The performance of each solution is highly dependent to the problem defining couple Peclet - Courant numbers. A more detailed analysis of the solution sensitivity to Peclet and Courant number is provided in Figure S3 in Supporting Information S1.

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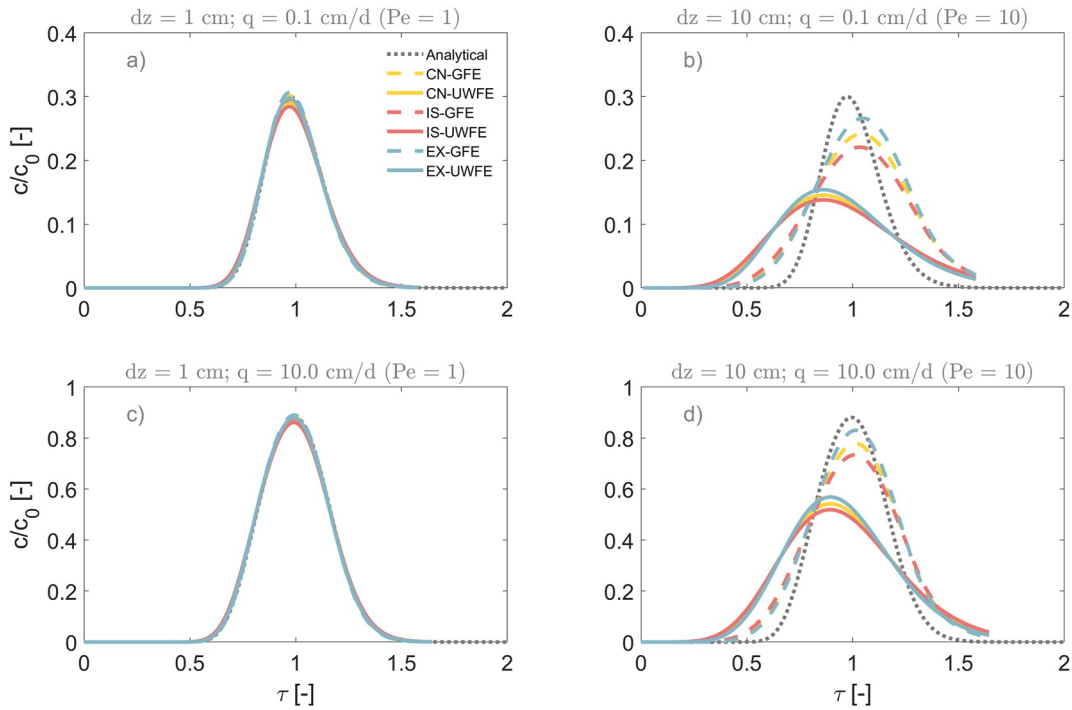

Figure 7. Breakthrough curve resulting from 1D homogeneous simulations using Hydrus 1D with different schemes, for a spatial discretization of 1 cm (left hand frames) and 10 cm (right hand frames) and a flux in the 1D column fixed to 0.1 cm/d (top frames) and 10.0 cm/d (bottom frames). The temporal discretization criteria (Courant number) is fixed to 0.1. The analytical solution is given in dashed line. Time is normalized as: $\tau = t/t_c$ where $t_c = L/u + t_{inj}/2$.

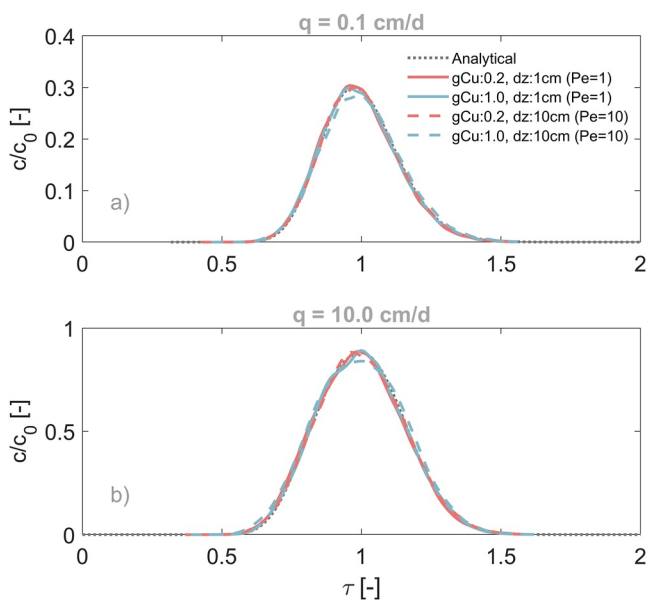

Figure 8. Breakthrough curve resulting from 1D homogeneous simulations using a Lagrangian scheme implemented into RW3D for two different spatial ($dz = 1$ and 10 cm) and temporal ($gCu = 0.2$ and 1.0) discretizations and for two Darcy velocity (top: $q = 0.1$; bottom: $q = 10.0$ cm/d). The analytical solution is given in dashed line. Time is normalized as: $\tau = t/t_c$ where $t_c = L/u + t_{inj}/2$.

Summarizing, and as previously shown, Eulerian schemes should be applied only when the Peclet and Courant number can be maintained low. In vadoze zone hydrology, a criterion that is commonly used for adapting the time step is that the product of Pe × Cu should always be lower than two (Perrochet & Bérod, 1993). However, even when the time step can be limited as a function of velocity conditions, a fine spatial discretization is still required, which can significantly limits the applicability of Eulerian methods to larger scale problem due to large calculation times.

Using the same flow fields than the Eulerian schemes, the previously presented Lagrangian scheme produces an accurate reproduction of the concentration signal in any spatial (dz = 1 and 10 cm) and temporal (gCu = 0.2 and 1.0) discretization scenario (Figure 8). Potential errors of the proposed implementation of the RWPT method due to boundary conditions at the bottom of the domain (e.g., due to dispersive particle jumps out of the domain) are also tested by extending the simulated column to 200 cm and observing the arrival of mass at a control plane located at $z = 100$ cm. Results shows no significant boundary effect on the solutions (Figure S4 in Supporting Information S1), even if dispersivity is increased (Figure S5 in Supporting Information S1).

Thus, for simulations with a coarse discretization of the domain, it appears that transport cannot be solved accurately with any of the tested Eulerian schemes due to a significant numerical dispersion. On the other hand, the RWPT method remains stable and allows therefore a coarser flow field to be considered.

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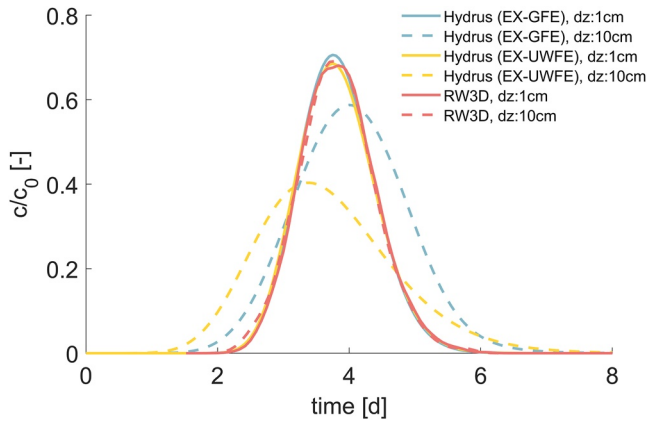

Figure 9. Breakthrough curve resulting from layered 1D simulations using the best performing Hydrus schemes (for each options for calculating space derivatives) and using a Lagrangian scheme implemented into RW3D for two different spatial (dz = 1 and 10 cm). For the random-walk particle-tracking method, the grid-Courant number is fixed to 0.5. For the Eulerian schemes, the time step is constrained by automatically maintaining the product of Courant and Peclet number below two.

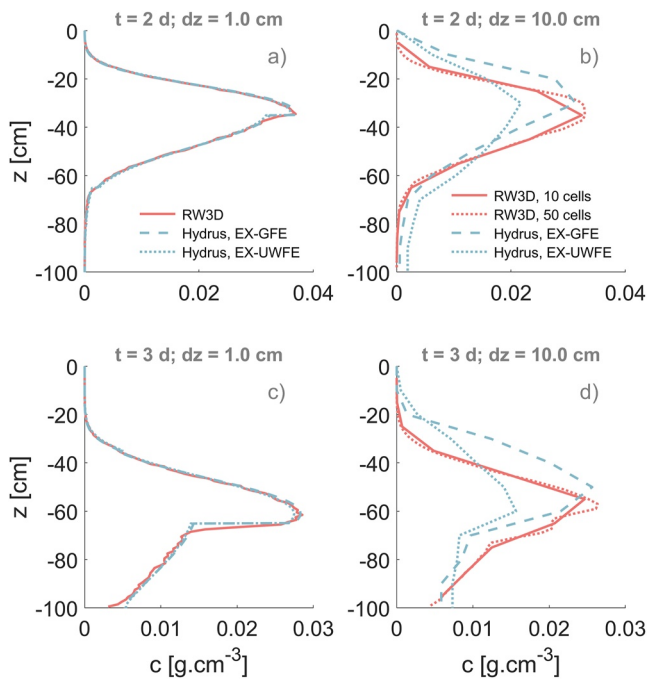

Figure 10. Concentration (z-) profile for two different times (2 and 3 days; top and bottom frames, respectively) and two different domain discretization (1.0 and 10.0 cm; left and right frames, respectively). Solutions using RW3D are shown in red. Solution using Hydrus are shown in blue. For the Lagrangian approach, concentration profiles rebuilt using two arbitrary grid (10 cells and 50 cells) are shown.

3.3. Layered Porous Media

Breakthrough curves. In case of a 1D layered system, similar limitations of Eulerian methods can be observed. For a fine spatial discretization (dz = 1.0 cm), all schemes converge to a common solution (Figure 9, only the best performing GFE and UWFE scheme is shown; see Figure S6 in Supporting Information S1 for all schemes). For a coarser spatial discretization (dz = 10.0 cm), all solutions appears to overestimate macrodispersion, with breakthrough curves characterized by a greater spread than for dz = 1.0 cm.

As in case of homogeneous conditions, the GFE space weighting schemes is less impacted - yet not exempt of artificial dispersion - by the spatial discretization than the UWFE schemes (Figure 9). This results show that even by maintaining the product of Courant and Peclet number below two (Perrochet & Bérod, 1993), the Eulerian solutions produce significant error in case of large dz.

On the other hand, the Lagrangian scheme remains insensitive to the spatial discretization (Figure 9). Here again, only the Lagrangian approach appears to ensure a satisfactorily solution in case of coarse spatial discretization.

Concentration profiles. Concentrations profiles produced by Hydrus schemes and by RW3D lead to similar conclusions (Figure 10, left frames). For a fine discretization of the domain (dz = 1.0 cm), both the Eulerian and Lagrangian methods produce similar spatial concentration distributions. A slight difference appears near the interfaces (depth of 35 and 65 cm): the RWPT approach generates a smoother solution, the Eulerian schemes producing a step-like concentration evolution. Yet differences appears to be minimal and localized.

Differences in term of performance between the two approaches are more dramatic in case of a coarser grid (dz = 10.0 cm; Figure 10, right frames). The Eulerian schemes produce artificial dispersion, leading to a lower peak of concentration and larger spread of the mass within the domain (compared to the finely resolved solution, i.e., using a dz = 1.0 cm). The solution also appears to be highly sensitive to the chosen scheme, the UWFE approach producing more dispersed solutions than the GFE one, which is consistent with observations in BTCs (Figure 9).

On the other hand, the RWPT method leads to concentration profiles closer to the ones produced with a fine discretization: peak concentrations and solute spread are well reproduced. Yet, coarser flow model still generates differences in concentrations, especially at material interfaces and at later times (Figure 10, right lower frame).

Interestingly, the Lagrangian transport solver being grid-less, concentration fields can be estimated using a domain discretization different than the flow solver. Smoother solutions can then be produced by increasing the number of cell in which concentration are estimated (Figure 10).

3.4. Homogeneous 1D Dynamic Conditions

The two-steps dynamic of the infiltration/evaporation system is well represented by both the Eulerian and the RWPT schemes. Both the downward motion of the plume due to infiltration and the evaporation-induced upward motion are identically described. For the case of Hydrus, a no-flux concentration assumed at the soil surface, resulting in mass accumulation near the soil surface during the evaporation period. Such accumulation of mass is accurately reproduced by the RWPT method. Interestingly, no concentration

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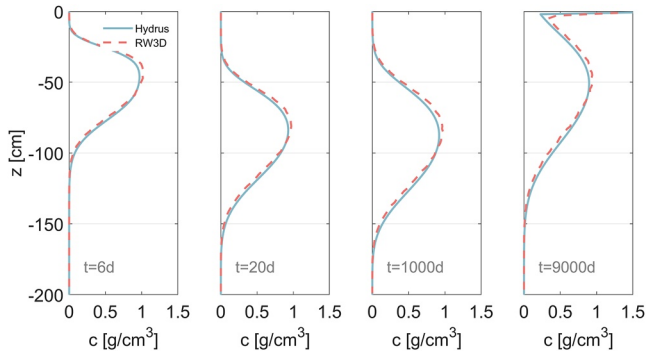

Figure 11. Concentration (z-) profile at different times as simulated by Hydrus (blue line) and RW3D (red dashes).

gradient is constrained in RW3D, but a local increase of concentration is still observed by not letting particles exit the top border of the domain, where they are instead bouncing and eventually accumulating. This phenomenon is often observed in natural system impacted by salt accumulation (e.g., Nachshon et al., 2011).

We observe minor differences on the upper concentration front during the evaporation period, with a slight decrease of the upper flux reproduced by Hydrus compared to RW3D leading to a minor decrease in the mass accumulated right below the soil surface. Increasing the dispersivity coefficient leads to similar results (Figure 11) (Figure S7 in Supporting Information S1).

Peters et al. (2019) recently raised awareness on potential discrepancies between expected and simulated dispersive fluxes when the advection dispersion model is used to reproduce unsaturated systems with sink or source terms. The minor excess in downward fluxes observed with Hydrus could be related to this issue, but more investigation would be required to validate this assumption.

Through this example, the RWPT method displays an aptitude in modeling more complex, dynamic systems that are characteristic of unsaturated soils.

3.5. 3D Heterogeneous Media

BTCs. Results from 3D simulations display a good agreement between Hydrus and RW3D in case of fine spatial discretization (dz = 5.0 cm; Figure 12, top frame). Yet, the breakthrough curve generated by Hydrus shows slightly earlier arrivals and less late mass arrival (i.e., tailing), compared with RW3D. These observations (higher early peak of mass and less tailing) are very pronounced in case of coarser spatial discretization (dz = 10.0 cm; Figure 12, bottom frame). RW3D produces identical outputs for simulations with as few as 10,000 particles and large time steps (gCu = 0.8) and with a larger number of particles (50,000) and smaller time steps (gCU = 0.2).

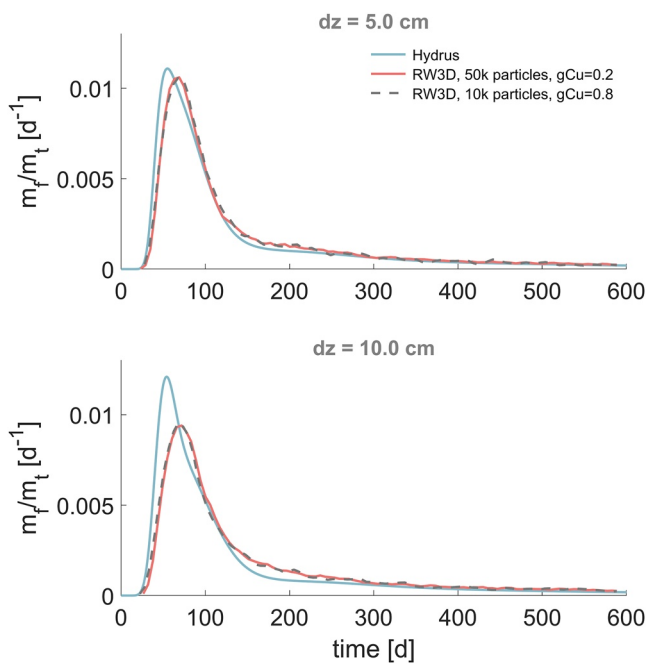

Figure 12. Breakthrough curve resulting from 3D heterogeneous simulations using Hydrus 3D and the Lagrangian scheme implemented into RW3D for two different spatial (top: dz = 5.0 cm; bottom: dz = 10 cm). The mass fluxe recorded at the bottom of the domain (m_f [g.d^{-1}]) is normalized by the total mass injected (m_t [g]).

There is no clear and obvious explanation of the differences between the Eulerian and Lagrangian schemes. We identify here two potential phenomena. First, we can rationaly state that such differences partially originates from numerical dispersion produced by Hydrus. Previously analyzed 1D scenarios have highlighted Hydrus as being affected by numerical dispersion in case of coarse spatial discretization. In this 3D example, a coarser discretization (dz = 10.0 cm) leads to a higher peak of mass and a slightly less intense tailing effect. This observation indicates that in heterogeneous 3D conditions, numerical dispersion can lead to mass leaving artificially low velocity zones, which prevents local accumulation of mass (Guo et al., 2019) and potentially produces earlier first arrivals and an underestimation of tailing.

Another issues explaining (at least in part) the different solution generated by RW3D and Hydrus is potential errors in the flow solution obtained using the GFE solver. Indeed, solution convergence issues typically affecting such method can lead to locally non-zero divergence in the flow field (i.e., $\nabla.\mathbf{u} \neq 0$), and eventually to the trapping of mass in some zones of the domain if particle-based solvers are used. The Figure S8 in Supporting Information S1 displays indeed that Hydrus3D does not produce a divergence-free flow field in our case. This could then explain the later arrival of mass and the increased spread produced by RW3D. Overall, current limitations in solving 3D heterogeneous flow make it challenging to clearly evaluate transport solutions.

Efficiency. In terms of computational efficiency, the Lagrangian approach appears to be orders of magnitude faster than the Eulerian scheme (Figure 13)

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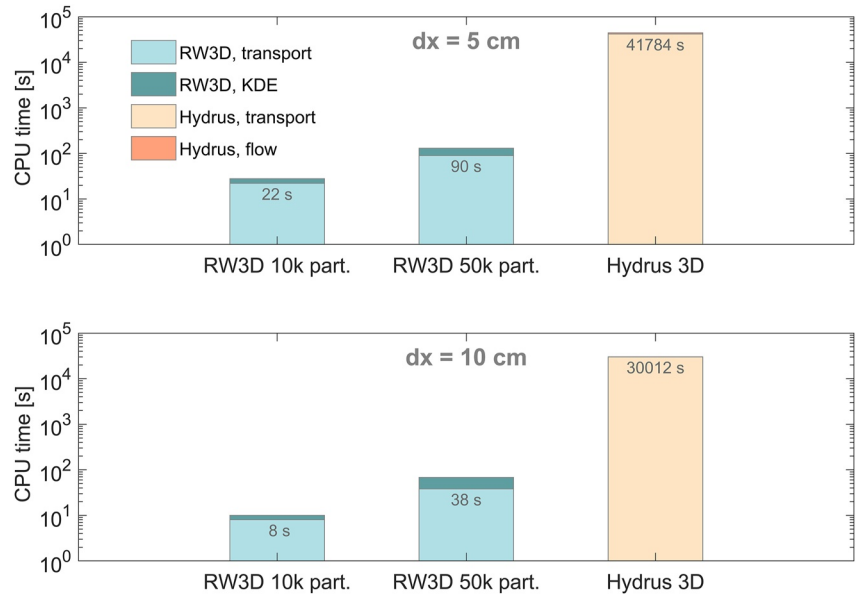

Figure 13. Computational time used by RW3D and Hydrus 3D to solve the 3D heterogeneous scenario. Since Hydrus 3D solves iteratively the flow and the transport problem at each time step, the CPU time required to solve flow only is first estimated in order to approximate Hydrus's CPU time for transport only. The time required to solve the flow is shown to be insignificant in comparison with the time required to solve the transport. RW3D uses a Kernel Density Estimator to improve the computation of breakthrough curves. The corresponding CPU time is also shown.

to reproduce the results displayed in Figure 12. Hydrus took about 3×10^4 s and 4×10^4 s to solve the 3D transport problem with a spatial discretization of 10.0 and 5.0 cm, respectively. Again, a coarse discretization appeared to be prone to numerical errors, so a discretization of 5.0 cm or less should be considered. The largest CPU time is therefore a best case scenario.

On the other hand, RW3D produced consistent outputs regardless of the spatial and temporal discretizations. Efficiency is then analyzed for a gCU = 0.8, and solutions with a $dz = 5.0$ and 10.0 cm can both be considered valid. The Lagrangian method required 8 and 22 s to solve the problem with 10,000 particles for a $dx = 10.0$ and 5.0 cm, respectively; 38 and 90 s were required with 50,000 particles. Kernel Density Estimator performs well to produce BTCs with a limited amount of particles, which can significant improve efficiency.

When estimating BTCs is the objective, RW3D represents a very attractive alternative to Eulerian methods, both in terms of accuracy and efficiency. A significantly larger number of particles is often required to build a concentration map from local particle density. Yet, even in case of a million of injected particles, CPU times remains significantly lower than for Hydrus 3D.

4. Conclusion

This paper presents a new Lagrangian, RWPT method as a valuable alternative to widely used Eulerian methods for simulating transport in unsaturated conditions. The new algorithm uses the interpolation method to improve the accuracy of the solution in case of sharp interfaces in both the dispersivity tensor and the water content field. While the relevance of using the interpolation method has been previously demonstrated in case of heterogeneity in the velocity field, this study explicitly show that such corrective algorithms are also important in case of sharp interface in the water content.

The resulting Lagrangian approach performs well against results of an analytical solution (1D homogeneous), independently of the spatial and temporal discretization. Outputs from 1D and 3D solute transport models are compared with and validated against the series of Eulerian solver proposed in the widely used software suite Hydrus. In contrast to the new RWPT method, Eulerian solutions suffer from artificial dispersion, especially in case of coarse spatial discretization, and even while maintaining a low product of Courant and Peclet numbers,

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as often advised. We also demonstrate the capability of the RWPT method to accurately simulate transport under complex flow conditions typical of unsaturated soils, that is, with periodic infiltration and evaporation periods.

Performances of the RWPT method are also analyzed in terms of computational time. The particle tracking approach appears to be order of magnitude faster than the more unstable Hydrus solver to model a heterogeneous 3D transport problem. RWPT is, therefore, an attractive, efficient and stable alternative for studying transport of contaminants in the unsaturated, especially for large scale problems requiring relatively coarse spatial discretization.

Outlook will be focused on extending the unsaturated-RW3D algorithm for more complex transport problems, including non-linear sorption, first order and higher order chemical reactions. We expect that the advantages of the extended RW3D against the Eulerian solutions presented in this article will hold.

Data Availability Statement

The online resources located here: https://doi.org/10.5281/zenodo.6607599 include the source codes of the proposed implementation of the random walk particle tracking method as well as the compiled executable and input files necessary to reproduce the results.

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