Transport Upscaling Using Multi-Rate Mass Transfer in Three-Dimensional Highly Heterogeneous Porous Media

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Abstract

A methodology for transport upscaling of three-dimensional highly heterogeneous formations is developed and demonstrated. The overall approach requires a prior hydraulic conductivity upscaling using an interblock-centered full-tensor Laplacian-with-skin method followed by transport upscaling. The coarse scale transport equation includes a multi-rate mass transfer term to compensate for the loss of heterogeneity inherent to all upscaling processes. The upscaling procedures for flow and transport are described in detail and then applied to a three-dimensional highly heterogeneous synthetic example. The proposed approach not only reproduces flow and transport at the coarse scale, but it also reproduces the uncertainty associated with the predictions as measured by the ensemble variability of the breakthrough curves.

Keywords: upscaling, heterogeneity, solute transport, mass transfer

1. Introduction

Upscaling flow and transport has been disregarded by some on the basis that it is not needed because our computers are capable of handling larger and larger numerical models. However, we know by experience that there will always be a discrepancy between the scale at which we can characterize the medium, and the scale at which we can run our numerical codes. This discrepancy renders upscaling necessary in order to transfer the information collected at the measurement scale into a coarser scale better suited for numerical modeling.

In the last decades, many reviews have been published dealing with upscaling but mostly focusing on hydraulic conductivity upscaling [e.g., 57, 41, 48]. In comparison with the effort devoted to the upscaling of hydraulic conductivity, less attention has been paid to upscaling for solute transport modeling. For

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example, Dagan [11] noted that hydraulic conductivity upscaling induces a loss of information and advised to compensate for this loss by splitting the solute plume into subplumes with effective dispersivities derived from stochastic theory. Rubin et al. [44] developed an upscaling method to derive effective block-scale dispersivities using a perturbation method, which accounts for the loss of subgrid variability in the upscaled numerical model. These two approaches are based on analytical techniques, which have a limited range of application because of their underlying assumptions. Numerical methods, on the contrary, are more general, since they are not restricted by the geometry of the domain, the type of boundary conditions, or the degree of heterogeneity. Scheibe and Yabusaki [49] examined the impact of hydraulic conductivity upscaling using the power-averaging method with different exponents [33]. They found that although flows and heads can be preserved after upscaling the hydraulic conductivities, the discrepancy on transport predictions is substantial. Cassiraga et al. [7] applied the simple-Laplacian technique [57] to upscale hydraulic conductivity and evaluated the impact of upscaling on solute transport for various degrees of heterogeneous media in two dimensions. They concluded that the prediction of solute transport at the coarser scale will provide reasonably good estimates of the early particle arrival times but will largely underestimate the late travel times; the explanation for this behavior was the existence and connectedness of extreme-valued hydraulic conductivities at the fine scale, which are lost after upscaling. To overcome this inability, Fernàndez-Garcia and Gómez-Hernández [16] extended this study and introduced an enhanced block dispersion tensor to compensate for the loss of information. They found that, with this approach the median travel time could be reproduced but that the tails of the breakthrough curves were largely underestimated. They suggested that a mass transfer process should be introduced at the coarse scale to make up for the information at the small scale that cannot be resolved by the upscaled model in heterogenous media. Fernàndez-Garcia et al. [18] examined the use of a mass transfer process with different memory functions as part of the constitutive transport equation at the coarse scale, in conjunction with hydraulic conductivity upscaling with the simple-Laplacian technique in 2D. The results showed that considering a double-rate or a truncated power-law mass transfer model at the coarse scale was enough to properly describe the ensemble average behavior of the main features associated with the breakthrough curves. However, the uncertainty associated with the predictions is underestimated after upscaling due to the lack of memory in space during the upscaling process.

It is important to note that the use of a mass transfer process as part of the constitutive equation for transport at the coarse scale model has also been proposed by Guswa and Freyberg [26], Zinn and Harvey [64], Willmann et al. [59] and Frippiat and Holeyman [20]. However, these studies mainly focus on upscaling up to a completely homogeneous aquifer. Guswa and Freyberg [26] conclude that a mass exchange term is needed only if the equivalent hydraulic conductivity is larger than the geometric mean of the underlying conductivity field, Zinn and Harvey [64] suggest that a mass exchange is necessary and conclude that the multi-rate model should better compensate for the loss of resolution than the single-rate model, and later Fernàndez-Garcia et al. [18] demonstrated that, indeed, the double-rate model and the power-law mass transfer model outperform the single-rate model for upscaling purposes.

In the current work, we extend to 3D the study by Fernàndez-Garcia et al. [18], who proposed a transport upscaling method using a multi-rate mass transfer. We also introduce an elaborated interblock Laplacian-with-skin hydraulic conductivity upscaling approach, for optimal reproduction of the flows at the coarse scale. Although the extension of the methodology to three-dimensions may appear as conceptually straightforward, we have found that it is necessary to make some adjustments to efficiently reproduce the breakthrough curves. Additionally, unlike most studies that focused primarily on a single realization analysis, the present study analyzes the upscaling at the ensemble level in order to analyze also how prediction uncertainty upscales.

The outline of this paper is as follows. We first introduce the flow and transport governing equations at two different support scales. Next, the importance of using an interblock Laplacian-with-skin hydraulic conductivity upscaling is illustrated, with emphasis on the numerical implementation in three-dimensions. We then describe the transport upscaling with mass transfer in two dimensions and discuss the modifications of the method for its application in three-dimensions. Finally, numerical tests demonstrate the accuracy and efficiency of the method. We end with a discussion on the weaknesses and strengths of the proposed approach, with an indication of avenues for improvement.

2. Methodology

2.1. Background

2.1.1. Fine scale equations

At the fine scale, denoted herein by the superscript f, under steady-state flow conditions and in the absence of sinks and sources, the flow equation of an incompressible fluid in saturated porous media in a Cartesian coordinate system can be obtained by combining the continuity equation and Darcy's law [1]:

$$\nabla \cdot \left[\mathbf{K}^f(\mathbf{x}^f) \nabla h^f(\mathbf{x}^f) \right] = 0 \tag{1}$$

where $h^f[L]$ is the piezometric head; $\mathbf{K}^f[LT^{-1}]$ is a symmetric positive-definite rank-two tensor; $\mathbf{x}^f$ represents the fine scale coordinates.

Similarly, using the solute mass conservation equation and assuming that Fick's law is appropriate at the local scale, the three-dimensional advective-dispersive equation (ADE) for solute transport is often written as [19]:

$$\phi^f \frac{\partial C^f(\mathbf{x}^f,t)}{\partial t} = -\nabla\cdot\left[\mathbf{q}^f(\mathbf{x}^f)C^f(\mathbf{x}^f,t)\right] + \nabla\cdot\left[\phi^f \mathbf{D}^f \nabla C^f(\mathbf{x}^f,t)\right] \tag{2}$$

where $C^f\,[ML^{-3}]$ is the dissolved concentration of solute in the liquid phase; ϕ^f [dimensionless] is the porosity; $\mathbf{q}^f\,[LT^{-1}]$ is the Darcy velocity given by $\mathbf{q}^f(\mathbf{x}) = -\mathbf{K}^f(\mathbf{x})\nabla h^f(\mathbf{x})$; $\mathbf{D}^f\,[L^2T^{-1}]$ is the local hydrodynamic dispersion coefficient tensor with eigenvalues (associated with the principal axes, which are parallel and perpendicular to the direction of flow) given by [5]:

$$D_i^f = D_m + \alpha_i \frac{|\mathbf{q}^f|}{\phi^f} \tag{3}$$

where α_i are the local dispersivity coefficients, more specifically, α_L, α_T^H and α_T^V are, respectively, the longitudinal dispersivity coefficient and the transverse dispersivity coefficient in the directions parallel and orthogonal to flow, and D_m is the effective molecular diffusion coefficient.

The fine scale transport equation (2) is only valid if the Fickian assumption is satisfied at the small scale. Here, we assume that the ADE is capable of reproducing the tracer spreading at the fine scale. Salamon et al. [47] at the MADE site and Riva et al. [42] at the Lauswiesen site have shown that for cases in which, apparently, the transport spreading does not look Fickian at the macroscopic scale, the ADE equation is applicable if the small-scale variability of hydraulic conductivity is properly modeled at the smallest scale possible.

2.1.2. Coarse scale equations

There are two main approaches to get the coarse scale equations. On one hand, those who work analytically from the fine scale equations and apply regularization techniques to derive the equations that would express the state of the system on a larger scale. Examples of these works can be found in [9, 39, 32, 25]. On the other hand, those who empirically postulate the coarse scale expression (after the fine scale one) and then try to determine the parameter values of the postulated coarse scale expressions. Examples of these works can be found in [43, 24, 23, 26]. In the first approach, the authors generally obtain equations which are nonlocal, that is, the parameters associated to a given block at the coarse scale depend not only on the fine scale parameters values within the block, but also on the values outside the block. This fact is recognized by some authors using the second approach when the coarse block parameters are computed on local flow and/or transport models which extend beyond size of the block being upscaled, so that the influence of the nearby cells is captured [57]. We have opted, in this paper, for the second approach.

At the coarse scale, denoted herein by the superscript c, the flow equation is taken to have the same expression as the fine scale equation, but with $\mathbf{K}^f$ replaced by an upscaled hydraulic conductivity tensor $\mathbf{K}^c$:

$$\nabla\cdot\left[\mathbf{K}^c(\mathbf{x^c})\nabla h^c(\mathbf{x^c})\right] = 0 \tag{4}$$

where $h^c[L]$ designates the coarse scale piezometric head, and $\mathbf{x}^c$ refers to the coarse scale coordinates.

In earlier studies of transport at the coarse scale, only upscaling of the flow controlling parameters was performed [e.g., 49, 7, 37]. That is, upscaled $\mathbf{K}^c$ values were derived, and the same advection dispersion equation was used both at the fine and coarse scales. However, recent findings have demonstrated that the transport equation to be used at the coarse scale should include an enhanced dispersion tensor and a fictitious mass exchange process as a proxy to represent the mass transfer processes taking place within the coarse block and largely associated with the within-block heterogeneity [e.g., 64, 16, 59, 18].

We have chosen the multi-rate mass transfer model (MRMT) [27, 6] as the mass exchange expression to be used at the coarse scale. Alternative models such as the continuous time random walk [3] or fractional derivatives [2] could be used as well. Fernàndez-Garcia et al. [18] discussed the use of the MRMT for upscaling purposes in 2D, and its versatility to treat complex heterogeneities; furthermore, it was successfully applied at the MADE aquifer by Feehley et al. [15] and at the Lauswiesen site by Riva et al. [42]. Many transport codes based on the MRMT model [e.g., 61, 6, 45, 50] indicate the great potential of this approach.

The upscaled transport equation, including the MRMT model, can be described by the following governing equation [27, 6]:

$$\phi_m^c \frac{\partial C_m^c(\mathbf{x}^c,t)}{\partial t} = -\nabla\cdot\left[\mathbf{q}^c(\mathbf{x}^c)C_m^c(\mathbf{x}^c,t)\right] + \nabla\cdot\left[\phi_m^c\mathbf{D}^c\nabla C_m^c(\mathbf{x}^c,t)\right] - \phi_m^c\Gamma(\mathbf{x}^c,t) \tag{5}$$

where ϕ_m^c [dimensionless] defines the pore volume fraction of the mobile domain; $C_m^c[ML^{-3}]$ is the solute concentration in the mobile region of the coarse block; $\mathbf{q}^c[LT^{-1}]$ is the Darcy velocity derived from the upscaled hydraulic conductivity; $\mathbf{D}^c[L^2T^{-1}]$ is an enhanced block dispersion tensor, which includes the fine scale local hydrodynamic dispersion (α_i) and a macrodispersivity term (A_i) [16, 18]:

$$D_i^c = D_m + (\alpha_i + A_i)\frac{|\mathbf{q}^c|}{\phi_m^c} \tag{6}$$

where D_i^c are the eigenvalues of $\mathbf{D}^c$ associated with the principal axes, which are parallel and perpendicular to the flow direction. The additional mass exchange term $\Gamma(\mathbf{x}^c, t)$ $[ML^{-3}T^{-1}]$ can be expressed in terms of mobile concentrations by using a convolution product with a memory function $g(\mathbf{x}^c, t)$ $[T^{-1}]$ [6, 28]:

$$\begin{aligned} \Gamma(\mathbf{x}^c, t) &= \beta(\mathbf{x}^c) \int_0^t g(\mathbf{x}^c, \tau) \frac{\partial C_m^c(\mathbf{x}^c, t-\tau)}{\partial \tau} d\tau \\ g(\mathbf{x}^c, t) &= \int_0^\infty \alpha f(\mathbf{x}^c, \alpha) e^{-\alpha t} d\alpha \end{aligned} \tag{7}$$

where $\beta(\mathbf{x}^c)$ [dimensionless] is the so-called capacity ratio; α $[T^{-1}]$ is a continuous positive variable representing the multiple mass transfer rate coefficients, and $f(\mathbf{x}^c, \alpha)$ $[T]$ denotes the probability density function of the mass transfer rate coefficients. Therefore, once $f(\mathbf{x}^c, \alpha)$ is given, the MRMT model equation (5) can be numerically solved. It is worth emphasizing that, the macrodispersivity term A_i and the mass transfer model are introduced as fictitious processes to make up for the presence of low and high conductivity zones which are smeared out after upscaling, and for the diffusive-like process occurring within the coarse block due to the heterogeneity. In this respect, it is consistent with Zinn and Harvey [64], Willmann et al. [59], and Riva et al. [42] who used the MRMT model to account for the subgrid heterogeneity in the upscaled transport model.

2.2. Hydraulic conductivity upscaling using the Laplacian-with-skin method

In contrast with the previous studies by Fernàndez-Garcia and Gómez-Hernández [16] and Fernàndez-Garcia et al. [18] that used the simple-Laplacian scheme to compute the block equivalent conductivities in two dimensions, here, we use a more sophisticated interblock Laplacian-with-skin three-dimensional full-tensor hydraulic conductivity upscaling technique, which is an extension of an earlier two-dimensional approach [21]. In essence, the Laplacian-with-skin upscaling scheme is an improved version of the simple-Laplacian approach. With regard to the simple-Laplacian method, Li et al. [37] have already demonstrated that it fails to reproduce interblock flow at the coarse scale and further underestimates contaminant spread at the MADE site. The major disadvantage of the simple-Laplacian approach is the assumption that the upscaled conductivity tensor is diagonal. For the details on the different upscaling processes, the reader is referred to the work by Wen and Gómez-Hernández [57], or more recently by Li et al. [37].

Gómez-Hernández [21] presented the Laplacian-with-skin approach recognizing the nonlocal nature of the upscaled conductivity tensor. The skin (a ring of cells surrounding the block) is used to approximate the actual boundary conditions around the block being upscaled without having to solve the flow problem for the entire aquifer (as previous authors had done, i.e., White and Horne [58]). For each block being upscaled, the algorithm consists of three steps: (a) isolate the block, plus a surrounding ring of cells (referred to as the skin), and solve a local flow problem numerically for a set of boundary conditions inducing fluxes in different directions across the block; (b) for each boundary condition the spatially-averaged flow and gradient within the block are calculated; (c) and then, the components of the upscaled hydraulic conductivity tensor are determined by solving the following overdetermined system of linear equations by a standard least squares procedure:

$$\begin{bmatrix} \langle \partial h/\partial x\rangle_1 & \langle \partial h/\partial y\rangle_1 & \langle \partial h/\partial z\rangle_1 & 0 & 0 & 0 \\ 0 & \langle \partial h/\partial x\rangle_1 & 0 & \langle \partial h/\partial y\rangle_1 & \langle \partial h/\partial z\rangle_1 & 0 \\ 0 & 0 & \langle \partial h/\partial x\rangle_1 & 0 & \langle \partial h/\partial y\rangle_1 & \langle \partial h/\partial z\rangle_1 \\ \langle \partial h/\partial x\rangle_2 & \langle \partial h/\partial y\rangle_2 & \langle \partial h/\partial z\rangle_2 & 0 & 0 & 0 \\ 0 & \langle \partial h/\partial x\rangle_2 & 0 & \langle \partial h/\partial y\rangle_2 & \langle \partial h/\partial z\rangle_2 & 0 \\ 0 & 0 & \langle \partial h/\partial x\rangle_2 & 0 & \langle \partial h/\partial y\rangle_2 & \langle \partial h/\partial z\rangle_2 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ \langle \partial h/\partial x\rangle_n & \langle \partial h/\partial y\rangle_n & \langle \partial h/\partial z\rangle_n & 0 & 0 & 0 \\ 0 & \langle \partial h/\partial x\rangle_n & 0 & \langle \partial h/\partial y\rangle_n & \langle \partial h/\partial z\rangle_n & 0 \\ 0 & 0 & \langle \partial h/\partial x\rangle_n & 0 & \langle \partial h/\partial y\rangle_n & \langle \partial h/\partial z\rangle_n \end{bmatrix} \cdot \begin{bmatrix} K^c_{xx} \\ K^c_{xy} \\ K^c_{xz} \\ K^c_{yy} \\ K^c_{yz} \\ K^c_{zz} \end{bmatrix} = - \begin{bmatrix} \langle q_x\rangle_1 \\ \langle q_y\rangle_1 \\ \langle q_z\rangle_1 \\ \langle q_x\rangle_2 \\ \langle q_y\rangle_2 \\ \langle q_z\rangle_2 \\ \cdots \\ \langle q_x\rangle_n \\ \langle q_y\rangle_n \\ \langle q_z\rangle_n \end{bmatrix} \quad (8)$$

where q_x q_y q_z are the components of the Darcy flux $\mathbf{q}$ obtained from the local solution of the flow equation; angle brackets indicate spatial averaging within the block; subscript n denotes an index referring to the different boundary conditions; $K^c_{xx} \cdots K^c_{zz}$ are the components of the upscaled equivalent conductivity $\mathbf{K}^c$. Note that the requirement of symmetry is enforced implicitly [62] in this system of equations.

Although we are aware of the works by Zijl and Stam [63] and Bierkens and Gaast [4] in which they argue that the upscaled conductivity tensor may be non-symmetric, we prefer to maintain symmetry at the block level to preserve its physical meaning: that opposite gradient vectors should induce opposite specific discharge vectors. Likewise, we enforce positive definiteness, since it is non-physical that the scalar product of the gradient vector and the specific discharge be positive (flow never goes upgradient). However, the approach would be equally applicable without imposing symmetry on the upscaled conductivity tensor.

Since we plan to solve the flow equation by finite differences, a further improvement in the hydraulic conductivity upscaling consists in computing the upscaled hydraulic conductivity tensors at the block interfaces rather than at block centers. This is done by isolating an aquifer volume centered at the interface, plus a skin, prior to solving the local flow problem [62]. In fact, this suggestion of an upscaled hydraulic conductivity based on the interface agrees with the works of Chen et al. [8], Wen et al. [55], and He and Durlofsky [31], who already pointed out that the upscaling of transmissibility (the equivalent to interblock conductivity in petroleum engineering) provided a more accurate coarse scale result than permeability upscaling.

2.3. Transport upscaling using mass transfer

Both in petroleum engineering and in subsurface hydrogeology, many studies have demonstrated that hydraulic conductivity upscaling is not enough to reproduce transport at the coarse scale [e.g., 49, 8, 16]. We have adopted the method proposed by Fernàndez-Garcia et al. [18] to address this problem, whereby the coarse scale transport equation includes a mass transfer term to compensate for the loss of information at the coarse scale. The problem we face is replacing a heterogeneous block within which the heterogeneity induces solute dispersion by a homogeneous block with enhanced dispersion and an associated multi-rate mass transfer process, the parameters of which have to be determined to induce the same solute dispersion induced by the within block heterogeneity. To this extent mass transport is solved at the fine scale using a particle tracking random walk approach and the residence times of the particles within the block are computed resulting in a cumulative distribution of residence times $F_\tau(\tau)$. The objective of transport upscaling is to determine the multi-rate mass transfer parameter resulting in the same residence time distribution. This is accomplished by a curve fitting process making use of an approximate solution for the residence time distribution of the multi-rate mass transfer transport equation in 1D, $F^*_\tau(\tau)$. The Laplacian transform of $F^*_\tau(\tau)$ is given by [29, 18]:

$$\widetilde{F}^*_\tau(p) \approx \frac{1}{p}\exp\left[L_b\left(\frac{v_m}{2D^c_\ell} - \sqrt{\frac{v_m^2}{4{D^c_\ell}^2} + \frac{\widetilde{\psi}(p)}{D^c_\ell}}\right)\right] \tag{9}$$

where $L_b[L]$ is the mean travel displacement of solute mass particles; the mobile velocity $v_m[LT^{-1}]$ is defined by:

$$v_m = \frac{\|\mathbf{q}^c\|}{\phi^c_m} \qquad \phi^c_m = \frac{\phi^c_e}{1+\beta} \tag{10}$$

and $\widetilde{\psi}(p)$ is defined by:

$$\widetilde{\psi}(p) = p + \beta\int_0^\infty f(\alpha)\frac{p\alpha}{p+\alpha}d\alpha \tag{11}$$

p is the Laplace transform variable; $f(\alpha)$ is the density function given in terms of the mass transfer coefficients, the expression of which depends on the multi-rate process considered. For instance, for the case of the double-rate mass transfer process it is:

$$f(\alpha) = \frac{\beta_1}{\beta}\delta(\alpha-\alpha_1) + \frac{\beta_2}{\beta}\delta(\alpha-\alpha_2) \tag{12}$$

with

$$\beta_1 + \beta_2 = \beta \tag{13}$$

where β_1 and β_2 [dimensionless] are the capacities of each immobile phase; α_1 and α_2 $[T^{-1}]$ are the transfer rates in each immobile phase and $\delta(\cdot)$ is the Dirac delta.

We also found that, in order to preserve the mean travel time of the plume to each control plane, it was not enough to match the particle residence time distributions for each block but that it was necessary to make a local upscaling of the effective porosity. For our purpose it was sufficient to define a coarse scale effective porosity ϕ_e^c [dimensionless] piecewise in between each pair of control planes as follows:

$$\phi_{e,i}^c = \frac{\bar{\tau}_{cp,i}^f - \bar{\tau}_{cp,i-1}^f}{\bar{\tau}_{cp,i}^c - \bar{\tau}_{cp,i-1}^c} \qquad i = 1, 2 \cdots n_{cp} \tag{14}$$

where $\bar{\tau}_{cp,i}^f$ is the mean travel time at the i control plane computed at the fine scale with porosity ϕ_f; $\bar{\tau}_{cp,i}^c$ is the average travel time computed with unit porosity at the coarse scale at the i^{th} control plane, and n_{cp} is the number of control planes. This estimated effective porosity is an artificial numerical value which also compensates for the loss of information in the upscaling process. This need of upscaling the porosity to preserve the mean travel times is also reported by Zhang [60] and Fernàndez-Garcia et al. [18].

For each block, once the particle residence time distribution has been obtained numerically, the model-independent nonlinear parameter estimation program, PEST [14], is used to determine the best set of mass transfer parameters in equation (9) that matches the distribution $F_\tau(\tau)$. For this purpose, a penalty function is established as follows:

$$P(\mathbf{\Theta}) = \xi_1[\bar{\tau} - \bar{\tau}^*(\mathbf{\Theta})]^2 + \xi_2[\sigma_\tau^2 - \sigma_\tau^*(\mathbf{\Theta})^2]^2 + \sum_{i=1}^{n_q} \omega_i \left[F_\tau(\tau_i) - F_\tau^*(\tau_i, \mathbf{\Theta})\right]^2 \tag{15}$$

where $\mathbf{\Theta}$ represents a vector with the transport parameters being estimated (we have noted explicitly the dependence of the distribution function on $\mathbf{\Theta}$), $\bar{\tau}$ is the average residence time computed from the particle distribution, $\bar{\tau}^*(\mathbf{\Theta})$ is the average residence time of $F_\tau^*(\tau)$, which can be derived from equation (9) as [36, 18]:

$$\bar{\tau}^*(\mathbf{\Theta}) = \frac{L_b}{v_m}(1 + \beta) \tag{16}$$

${\sigma_\tau}^2$ is the variance of residence time computed from the particle distribution, $\sigma_\tau^*(\mathbf{\Theta})^2$ is the variance of the distribution, which can be derived from equation (9) as:

$$\sigma_\tau^*(\boldsymbol{\Theta})^2 = \frac{2D_\ell^c}{v_m^3}(1+\beta)^2 L_b + \frac{2L_b}{v_m}\beta \int_0^\infty \frac{f(\alpha)}{\alpha} d\alpha \tag{17}$$

n_q is the number of particles that travel through the block, and ξ_1, ξ_2 and ω_i are weight coefficients, which in this case are all set to 1.

The PEST code has to evaluate multiple times expression (9) for different sets of the mass transfer coefficients being determined; for this purpose we have used the code STAMMT-L [29].

3. Numerical Evaluation

3.1. Model Configuration

Consider a synthetic three-dimensional confined aquifer under a uniform, natural-gradient flow condition, as shown in Figure 1, it will be the reference. A set of 30 hydraulic conductivity fields was generated using the code GCOSIM3D [22]. The field is parallelepipedic with dimensions of $x = 200$ m, $y = 140$ m, and $z = 70$ m and a discretization of $\Delta x = \Delta y = \Delta z = 1$ m. Only the inner domain consisting of $180 \times 120 \times 60$ cells will be uniformly upscaled to $18 \times 12 \times 12$ blocks, resulting in an overall scale-up factor of 500. The following standardized exponential semivariogram was used for the simulation of the isotropic hydraulic conductivity field:

$$\frac{\gamma_x(r)}{\sigma_x^2} = 1 - \exp\left[-\frac{r}{\lambda_x}\right] \tag{18}$$

where λ_x $[L]$ is the range with a value of 12 m in all the directions and r $[L]$ is the directional lag distance. The variance σ_x^2 of the natural logarithm of hydraulic conductivity is 4.0 (similar to the one found, for instance, at the MADE site [40]), to represent highly heterogeneous media. The aquifer was modeled with constant head boundaries at $x = 0$ m and $x = 180$ m and with no-flow boundaries at the remaining model faces. The average hydraulic gradient induced by the constant head boundaries is 0.01. The porosity is assumed constant and equal to 0.3.

At the fine scale, the five-point block-centered finite-difference groundwater flow model MODFLOW 2000 [30] was employed to solve the flow equation (1). The interface velocities were calculated, and then utilized in the random walk particle tracking code RW3D [17, 46], which was used to solve the fine scale transport equation (2). In this approach, the evolution in time of each particle is comprised of a deterministic component, which depends only on the local velocity field, and a superposed Brownian motion responsible for dispersion. A hybrid scheme is used for the velocity interpolation which provides local as well as global divergence-free velocity fields within the solution domain. Meanwhile, a continuous dispersion tensor field provides good mass balance at grid interfaces of adjacent cells with contrasting hydraulic conductivities [35, 46]. Furthermore, in contrast to the common constant-time scheme used in random walk modeling, a constant-displacement scheme [56], which modifies automatically the time step size for each particle according to the local velocity, is employed in order to decrease computational effort.

At the coarse scale, the nineteen-point block-centered finite-difference groundwater model FLOWXYZ [38] was employed to solve the flow equation (4). The most remarkable characteristic of this forward flow simulator is the capacity to deal with full conductivity tensors defined at block interfaces. Hydraulic conductivity tensors are defined at the block interfaces eliminating the need to average conductivity tensors at adjacent blocks to approximate their values at the interfaces. This scheme has been shown to perform better than the MODFLOW LVDA package [38], and has been successfully applied in other studies [e.g., 62, 37]. Again, the RW3D was used to solve the coarse scale multi-rate transport equation (5) based on the methodology presented by Salamon et al. [45]. Mass transfer processes are efficiently incorporated into the particle tracking algorithm by switching the state of the particle between mobile/immobile states according to appropriate transition probabilities.

For the sake of simplicity, we neglect dispersion, and only consider advection, at the fine scale, i.e., $D_m = 0$ and $\alpha_i = 0$. A total of 20000 particles (a number that we have tested yields stable transport predictions for this specific case) randomly distributed in a rectangular-shaped area of 60 m width and 30 m height located orthogonal to the principal flow direction in the plane at $x = 20$ m were released at time $t = 0$. The variable time step was computed on the basis of a grid Courant number of 0.01. A unit mass was assigned to each particle. Control planes are located within the aquifer to measure the mass arrival at 10 m intervals (see Figure 2).

Figure 1: A realization of reference lnK field (σ^2_{lnK}=4.0) overlaid with the discretization of the numerical model at the coarse scale.

3.2. Flow upscaling results

Prior to transport upscaling we wish to demonstrate the effectiveness and robustness of the interblock Laplacian-with-skin approach to flow upscaling as compared with other methods, such as the block-centered

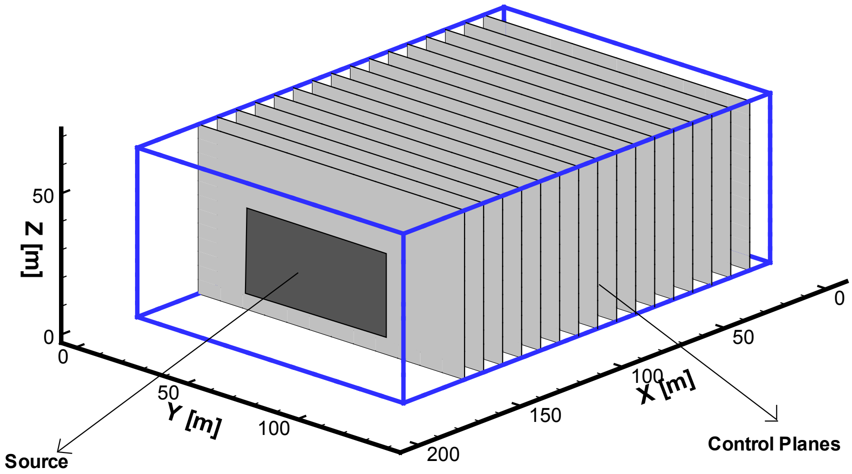

Figure 2: Sketch of transport simulations. The shaded rectangle located in the upstream zone delineates the initial particle injection zone. Control planes are also shown for measuring the mass fluxes.

simple-Laplacian approach, the interblock-centered simple-Laplacian approach, and the Landau-Lifshitz-Matheron conjecture for 3D isotropic media [41]. For this purpose a single realization is analyzed. Our goal, as that of any upscaling exercise, is to generate a heterogeneous coarse model which predicts the interblock flows as close as possible to those derived from a fine scale simulation. We will focus on interblock flow reproduction and disregard the analysis of piezometric heads, since the errors in piezometric head reproduction are always much smaller.

We compare the coarse scale flows obtained after solving the flow equation with the upscaled conductivities, with the reference flows obtained from the solution of the flow equation at the fine scale. The mismatch between these two values is measured by a Relative Bias defined as:

$$RB = \left(\frac{1}{N} \sum_{N} \frac{\left| q_x^f - q_x^c \right|}{q_x^f} \right) \cdot 100, \tag{19}$$

where N is the number of interblocks used to compute the relative bias; q_x^f is the specific discharge computed on the fine scale solution, and q_x^c represents the specific discharge from the coarse scale simulation. Because the x flow direction plays an important role in this case, the flow comparisons mainly focus on this direction. Similar results (not displayed) are obtained for the orthogonal directions. Also, as noted by Vermeulen et al. [54], the boundary conditions have an impact on the performance of upscaling for the nearby blocks, for this reason, and in order to filter out this impact in the comparison of the different methods, only the inner $14 \times 8 \times 10$ blocks are used to calculate the relative bias.

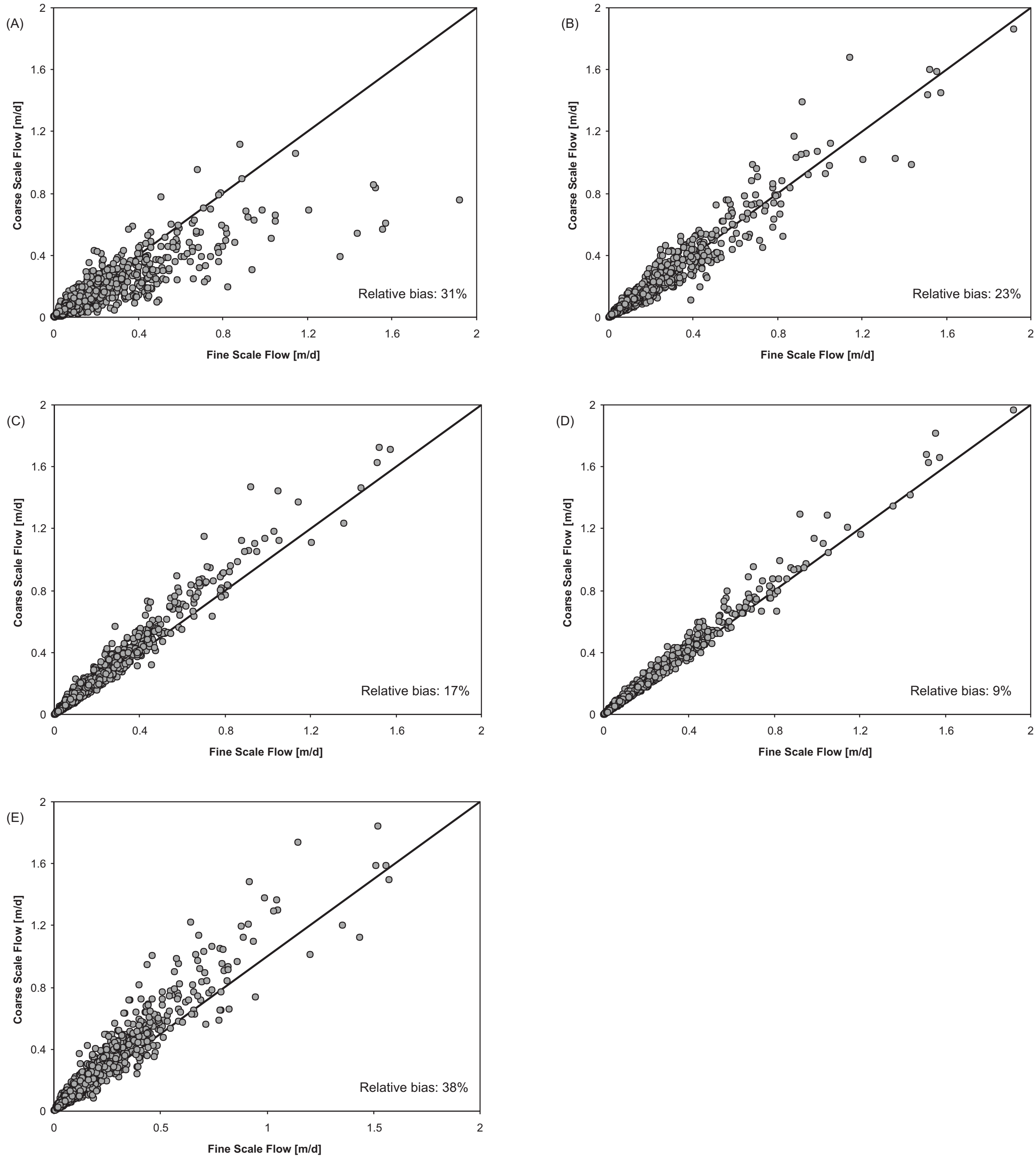

Figure 3: Flow comparisons at the fine and coarse scales on a single realization. (A) the block-centered simple-Laplacian method; (B) the interblock-centered simple-Laplacian method; (C) the interblock-centered full-tensor Laplacian-with-skin (skin size 3 m); (D) the interblock Laplacian-with-skin (skin size: 10 m along rows, 10 m along columns and 5 m along layers); (E) Landau-Lifshitz-Matheron conjecture for 3D isotropic media.

Figure 3 shows the cross-plots between the flows computed on the fine scale (reference values) and the ones computed on the coarse scale for several upscaling approaches. Results indicate: (1) interblock upscaling is better than block-centered upscaling, since it avoids the additional averaging process within the coarse flow simulator needed to approximate the interblock values (31% relative bias using block-centered simple-Laplacian to 23% relative bias using interblock-centered simple-Laplacian, see Figures 3A and 3B). This result agrees with previous finding [38]. (2) Compared with the simple-Laplacian method, the Laplacian-with-skin significantly improves the coarse scale results (23% relative bias using interblock simple-Laplacian to 9% relative bias using interblock Laplacian-with-skin, see Figure 3B and 3D); the main reasons for these results are the use of a full hydraulic conductivity tensor to represent the interblock property and the use of a skin to approximate the "real" boundary conditions around the interblock, in contrast with the simple-Laplacian approach which seeks a diagonal hydraulic conductivity tensor with boundary conditions directly at the block sides. (3) The significance of the skin size is evident as it was already pointed out by Zhou et al. [62] (17% relative bias using interblock Laplacian with a skin size of 3 m, down to 9% relative bias using interblock Laplacian with a skin of 10 m in the x and y directions and 5 m in the z direction, see Figures 3C and 3D). The high variance of hydraulic conductivity, as is the case in this example with σ^2_{lnk}=4.0, can result in local flows departing significantly from the average flow direction (along the x axis in this case), in which case the use of a full tensor and the skin size is more important. (4) For a mild isotropic heterogeneous field, the Landau-Lifshitz-Matheron conjecture (a close expression that gives the upscaled conductivity as a p-norm of the fine scale conductivities within the block, in which p only depends on the dimensionality of the problem) performs well [12]. However, when the global variance increases, the conjecture loses its accuracy and it is better to resort to the numerical flow experiments as is the case here, i.e., the Laplacian-with-skin method (38% relative bias using conjecture to 9% relative bias using interblock Laplacian-with-skin of 10 m along rows, 10 m along columns and 5 m along layers), see Figure 3D and 3E).

In short, the best reproduction of the fine scale flows is given by the interblock-centered Laplacian-with-skin approach. This scheme is retained for the subsequent transport upscaling.

3.3. Transport upscaling results

We examined two transport upscaling approaches using the same set of upscaled hydraulic conductivities obtained in section 3.2; in the first one, we only model advection using the velocities from the coarse scale flow simulation, and in the second one, we include the multi-rate term in the transport equation at the coarse scale and perform transport upscaling to determine enhanced macrodispersion coefficients, upscaled effective porosities and the parameters of the multi-rate transfer model. The multi-rate model estimates the mass transfer parameters as described in section 2.3. It should be noted that we do not make the comparison with an intermediate model including only enhanced macrodispersion coefficients, since it has already been shown [e.g., 64, 16, 20, 59, 18] that upscaled macrodispersion coefficients are not sufficient to reproduce the transport behavior for highly heterogeneous media.

As mentioned previously, the synthetic studies of Fernàndez-Garcia et al. [18] have shown that the double-rate mass transfer model is better than the singe-rate model in 2D mass transport upscaling. Herein, we only consider the double-rate mass transfer model to represent the mass transfer process, i.e., in each coarse block, the solute transport is assumed to happen in three zones: transport in the mobile zone is mainly by advection, while transport in the other two immobile zones is by diffusion-like processes.

With regard to the double-rate mass transfer model, the mass transfer rate density function $f(\alpha)$ and the memory function $g(t)$ are:

$$\begin{aligned} f(\alpha) &= \frac{\beta_1}{\beta}\delta(\alpha-\alpha_1) + \frac{\beta_2}{\beta}\delta(\alpha-\alpha_2) \\ g(t) &= \alpha_1\frac{\beta_1}{\beta}e^{-\alpha_1}t + \alpha_2\frac{\beta_2}{\beta}e^{-\alpha_2}t \end{aligned} \tag{20}$$

Accordingly, the parameters being estimated, are collected as a vector in $\mathbf{\Theta} = [\alpha_1, \alpha_2, \beta_1, \beta_2, A_l]$. Notice that the parameters are spatially variable since they are estimated for each upscaled block independently.

We compare the effectiveness of the transport upscaling by analyzing the breakthrough curves at different control planes in one specific realization and by looking at the ensemble results. For the ensemble results we will look at the early (5^{th} percentile of the BTC), median (50^{th} percentile) and late (95^{th} percentile) travel times.

Results using the advective-only model and the double-rate transport model are shown (see Figure 4 for the reproduction of BTCs in one realization and Figure 5 for the ensemble behavior of early, median and late travel times). From these results, we see that: (1) In contrast to the advective-only model, the double-rate mass transfer upscale model displays a higher accuracy to reproduce the fine scale breakthrough curves, in particular, the late travel times. Therefore, it is important to include the fictitious mass transfer process for solute transport predictions after upscaling. (2) In agreement with the study of Fernàndez-Garcia and Gómez-Hernández [16], it is shown that the advective-only model even when using a sophisticated hydraulic conductivity upscaling (interblock Laplacian-with-skin here) can result in overestimating the early travel times and underestimating the late travel times in very heterogeneous media. (3) The small deviations in the reproduction of the BTCs by the mass transfer model may be due to fact that the upscaled mass transfer parameters are derived from a one-dimensional analytical solution of the double-rate transport model (see

Equation (9)).

3.4. Propagation of Uncertainty

Due to the inherent lack of information in groundwater modeling, an uncertainty assessment is commonly requested in solute transport simulations [e.g., 47, 42]. Quantifying the uncertainty associated with flow and transport modeling should be important for the decision maker to assess the degree of confidence of his decisions. Here, we have analyzed how the uncertainty is estimated from the ensemble of realizations at the fine scale and after flow and transport upscaling. We will analyze the propagation of uncertainty through the upscaling process, along the same line as Fernàndez-Garcia and Gómez-Hernández [16].

The use of 30 realizations may seem a small number to perform an uncertainty evaluation in such a heterogeneous aquifer. However, our purpose is not so much to analyze the number of realizations needed to obtain a good estimation of model uncertainty, but rather to compare the uncertainty derived from 30 realizations, before and after upscaling. If uncertainty upscales well for 30 realizations, it should do so for a larger number of realizations.

We evaluate uncertainty by calculating the spread in the ensemble of cumulative breakthrough curves at all the control planes. More precisely, we quantified uncertainty by the 95% confidence interval related with the early, median, and late arrival time of particles to each control plane. The early arrival time reflects the fastest pathways between source and control plane, which is for example of importance for the safety assessment of nuclear waste repositories. The late arrival time constitutes important information for the calculation, for example, of clean-up times in contaminated aquifer remediation.

The evolution of uncertainty with the travel distance is shown in Figure 6. We can see that: (1) For the early arrival time, the advection-only model and double-rate mass transfer model show a slight overestimation of the uncertainty. (2) For the median arrival time, the double-rate mass transfer model is better in reproducing the uncertainty estimated at the fine scale than the advective-only model. (3) For the late time, it is evident that the use of double-rate mass transfer model clearly outperforms the advective-only for distances larger than 60 m, and less clearly (because of the scale the results are plotted) for the shorter distances.

In highly heterogeneous formulations, hydraulic conductivity upscaling is not sufficient to preserve the uncertainty. Transport upscaling, through the use of a mass transfer process at the coarse scale is needed for proper upscaling of the uncertainty associated with solute transport predictions.

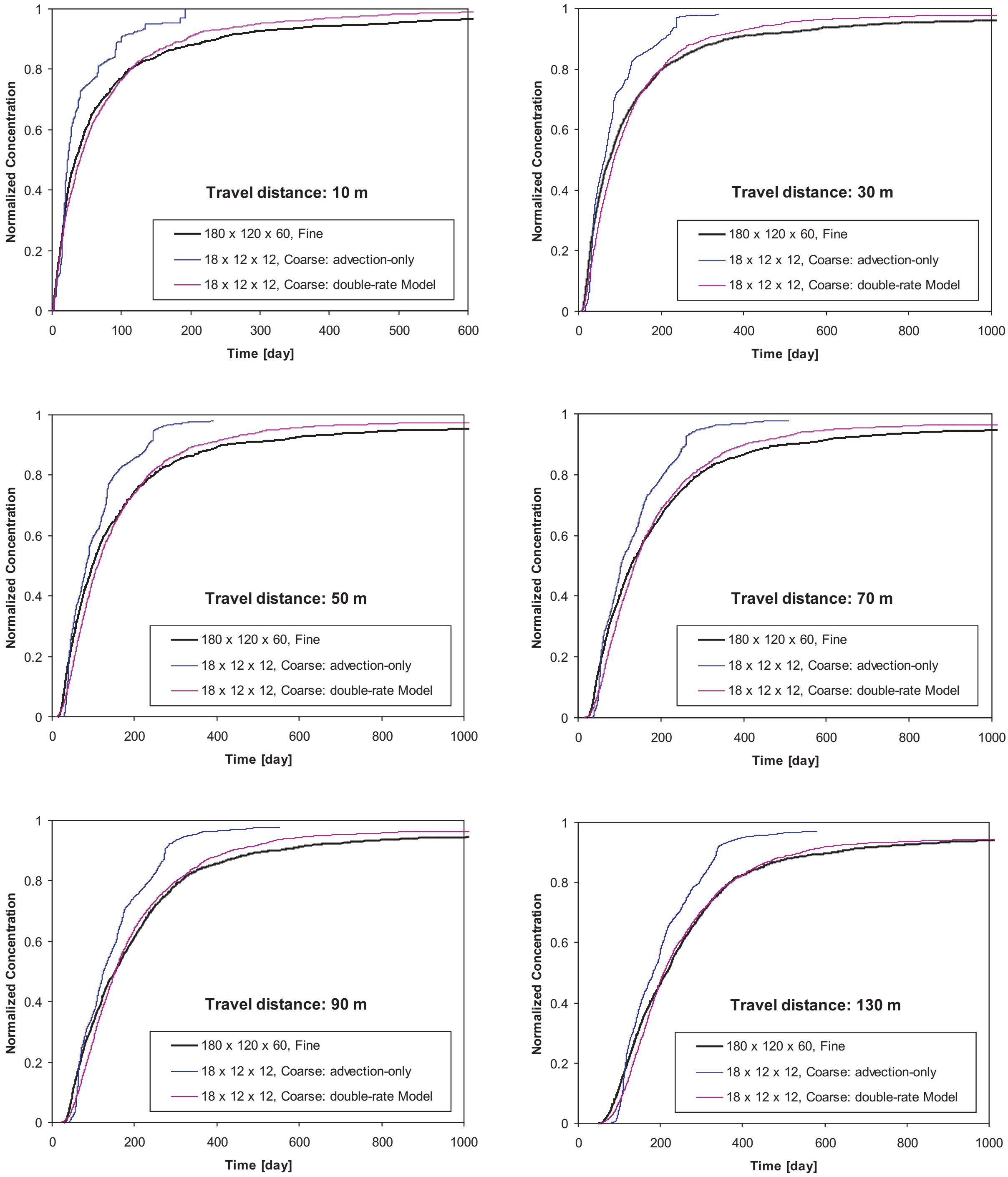

Figure 4: Comparison of fine scale cumulative breakthrough curves with those obtained by the upscaled transport models at six different control planes.

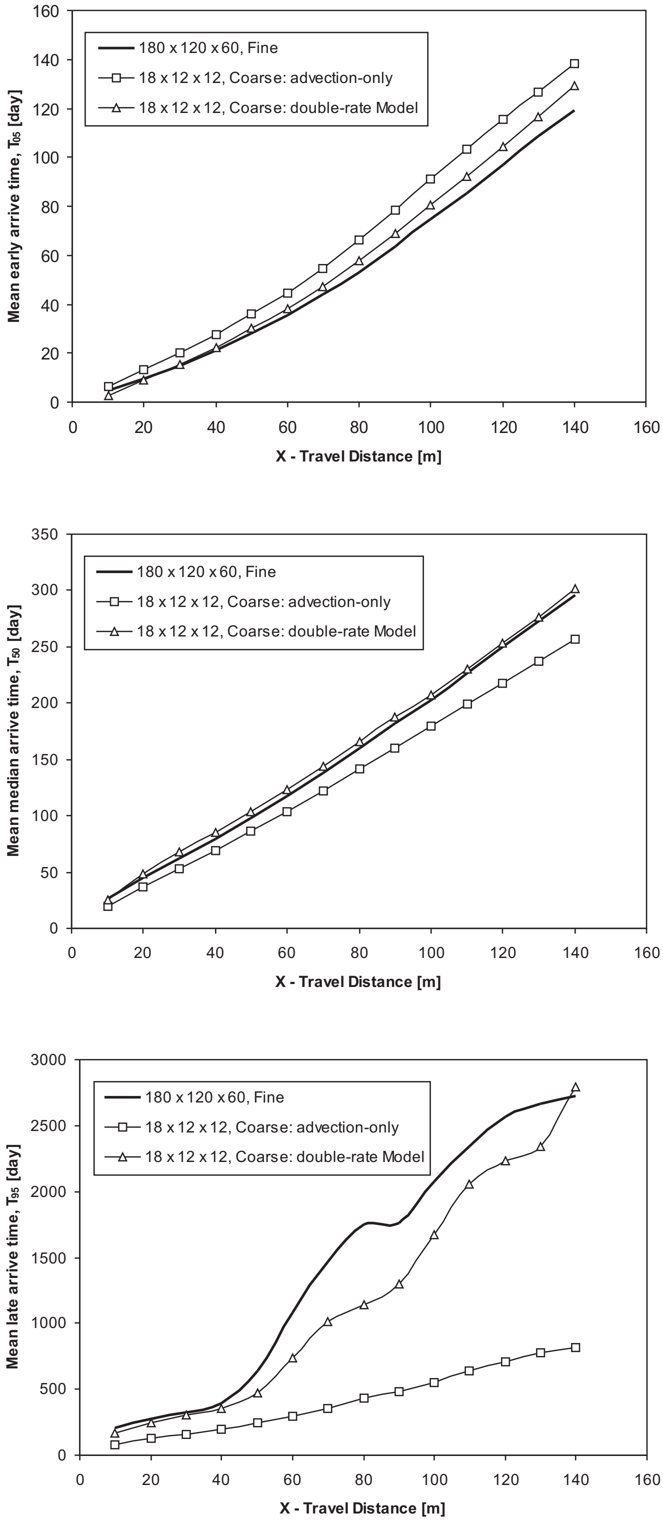

Figure 5: Ensemble travel times (early, median, and late travel times) as a function of travel distance, and comparison of the fine scale simulations to the upscaled simulations.

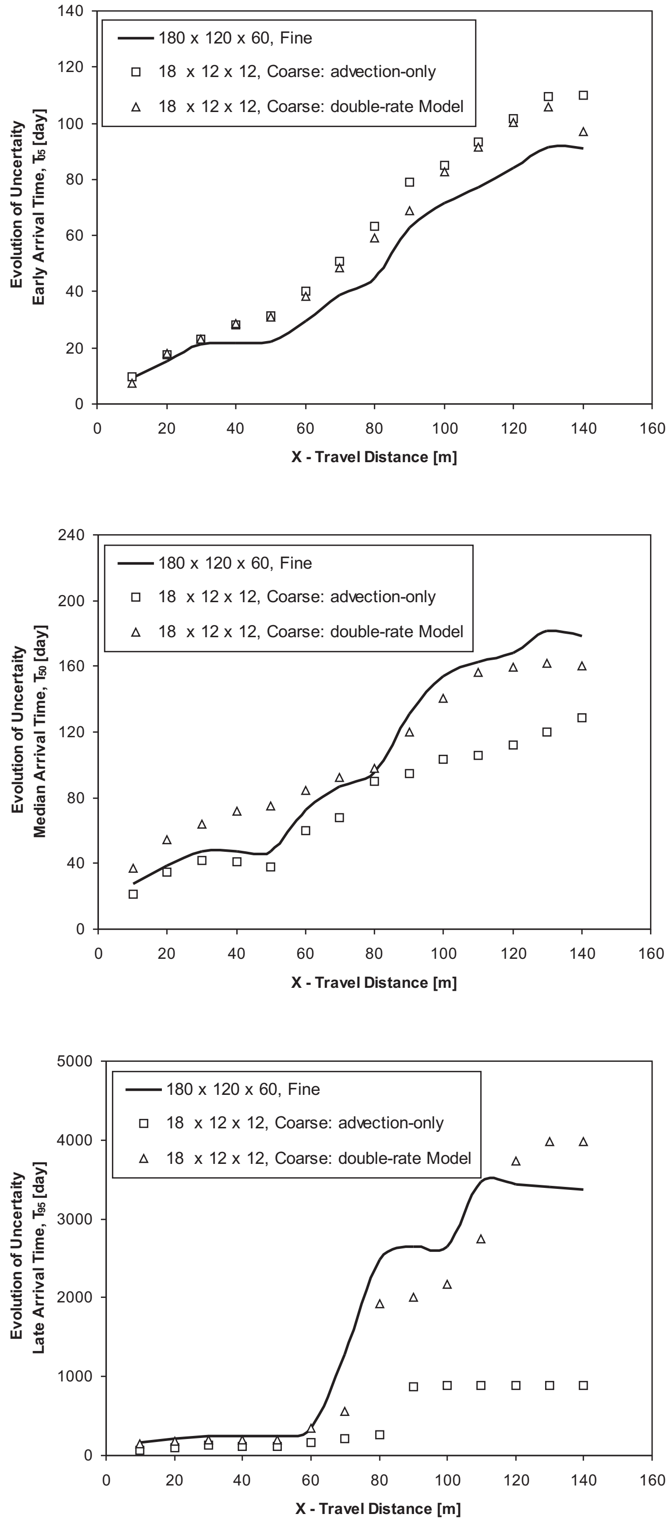

Figure 6: Evolution of uncertainty as a function of travel distance for the early, median, and late travel times, as measured by the width of the 95% confidence interval derived from an ensemble of 30 realizations. Calculations were performed at the fine scale, and at the coarse scale for two different upscaling approaches.

4. Discussion

We have presented and demonstrated an algorithm for transport upscaling to reduce the computational burden of transport predictions in three-dimensional highly heterogeneous media. But, how general is the algorithm? Will it work for different case studies? Will it work for different transport experiments? Although we recognize that the results obtained are specific to the case study under consideration, we believe that the upscaling procedure is general and it should work for other settings, as discussed below.

We present the results for statistically isotropic fine scale conductivities. What if the fine scale conductivities had been statistically anisotropic, with a much larger correlation length in the horizontal plane than in the vertical direction? What if the fine scale conductivities display curvilinear features, such as those associated with channels? In the case of statistical anisotropy, it would be necessary to adjust the size of the coarse blocks proportionally to the correlation lengths in each direction, in order to reduce the amount of smoothing in the directions of shortest continuity. In the case of curvilinear features, the proposed approach will yield upscaled conductivity tensors, the principal directions of which will change from block to block, inducing fluid velocities in the coarse model following those curvilinear features. The proposed approach has no problem in dealing with hydraulic conductivity tensors with arbitrary orientations of their principal directions. The block-by-block upscaling procedure is local, each block is isolated and a local flow exercise is performed in each block; at this local scale, the anisotropic correlation or the curvilinear features should not be clearly distinguishable from the intrinsic heterogeneity of the fine scale conductivities within the block; therefore, the upscaling algorithm should perform similarly. The question, remained to be answered, is whether when the blocks are assembled they will capture the global behavior of the statistically anisotropic formation or of the curvilinear features, or some specific corrections have to applied in these cases.

We present an analysis for a confined aquifer under steady-state flow conditions. We have not investigated how the upscaled coarse model would behave under transient conditions. We conjecture that the upscaled model should reproduce the the transient flow response of the the fine scale model with a degree of accuracy similar to the one obtained here for steady-state conditions, since the upscaled block conductivities are determined using different flow configurations applied to the block being upscaled. However, the upscaled transport parameters are based on the particle residence times for a specific velocity field; since, for transient flow conditions, the velocity field changes with time, it should be further investigated how much the upscaled transport parameters change as the velocity field changes, and decide whether these transport parameters should be made time dependent or there is a set of optimal parameters that would work well for the entire transient period. Also, in the case of transient flow, the need to upscale the storage coefficient needs to be addressed. Regarding the application of this approach for an unconfined aquifer, the general upscaling procedure should remain the same, all blocks should be upscaled as if they were fully saturated, and then, special caution should be taken in the numerical simulation model to account for those cells intersected by the phreatic surface at the time of computing the mass balances involving those cells. We have not analyzed this case because we do not have a numerical flow simulator capable of using full conductivity tensors and accounting for a phreatic surface.

The issue of how the upscaled transport parameters will perform under transient conditions (i.e., different velocity fields in time), brings the question of what will happen if the flow geometry changes substantially with respect to that for which the parameters were computed. A priori, we anticipate that the transport parameters would have to be recomputed, since the flow velocity field will change, and so will the residence times in the blocks.

We present a sequential upscaling procedure in which first, we compute the upscaled flow parameters, and then we use these parameters to compute the upscaled transport parameters. However, if the final aim of our analysis were to get the best transport predictions at the coarse scale, even compromising the accuracy of flow reproduction, we could think of performing the flow and transport upscaling jointly, therefore using the particle residence times within the block being upscaled in the computation of the coarse conductivity tensors. This is an interesting avenue of research that has not been investigated in this paper.

We have used a homogeneous porosity throughout the exercise. If porosity had been heterogeneous it would have had to be upscaled, too

There are two main drawbacks in the proposed method: the need to use the particle residence times obtained after a simulation of the flow and transport equations at the fine scale, and the need to correct the porosity at the coarse scale, even though the porosity is homogenous at the fine scale.

The first drawback beats, in principle, the whole purpose of upscaling, which is to avoid having to simulate flow and/or transport at the fine scale. For this reason, this paper loses some of its practicality, and could be justified (from a practical point of view) only if the upscaled model is to be used for a more complex type of modeling (i.e. reactive transport) avoiding the need to run the complex model at the fine scale. In order not to have to obtain the fine scale solution for the transport upscaling, we have tried to follow the same local approach as for the flow upscaling, that is, to isolate the block plus a sufficiently large skin and to solve local transport problems for several boundary conditions, and then derive the upscaled transport parameters; however, we have not succeeded with this approach, which has always resulted in biased transport predictions. There is, therefore, additional research needed in the transport upscaling procedure in order to yield it more practical. Our contribution with this paper is to demonstrate than, in 3D modeling of flow and transport, it is possible to systematically derive flow and transport upscaled parameters as long as we acknowledge that removing the heterogeneity within the block implies turning conductivities into tensors and including an enhanced macrodispersion and a mass transfer process for the solute transport.

The second drawback requires further investigation. We are not the first ones to face the need to make this adjustment for the coarse scale porosity [64, 60, 18]. The need for this correction is due to the accumulation of small biases in the transport modeling for each coarse block. When the transport parameters of each coarse block are computed, they are determined trying to reproduce the particle residence time distribution within the block with emphasis in matching the mean residence time; however, there seems to be a small systematic bias in this determination, which, at the end, forces us to correct the coarse porosities so that the breakthrough curves from the upscaled model are not shifted with respect to those from the fine scale model.

There is a need to know the fine scale parameters over the entire aquifer. Obviously, these parameter values will never be available, they have to be generated on the basis of available data. The issue of scales has been discussed for many years in the literature; it is a very old issue (known as the change of support problem) in mining [34], and a little bit more recent in hydrogeology and petroleum engineering; a good paper on the subject from the hydrogeology literature is the one by [10], in which Dagan talks about measurement and model scales, among other scales. For many years, data were measured, and without further consideration they were used to inform the parameter values of the groundwater flow model elements, until a concern about the so called "missing scale" was risen, mostly in the petroleum literature [52, 53], and the need to account for the disparity of scales between measurements and model cells was recognized [21]. Data are collected at a scale, generally, much smaller than the scale at which models are going to be discretized. Data spatial variability can be characterized at such scale by standard geostatistical methods [13] or by the more powerful and sophisticated multipoint geostatistical approaches [51], and this characterization can be used to generate conditional realizations, at the sampling scale, over discretized grids of multi-million cells. It is not proper to characterize the sampled data and use them directly for the generation of realizations at a larger scale suitable for numerical modeling, since the spatial variability patterns of the "equivalent" properties that should inform the larger blocks are completely different from that of the sampled data. Besides, as we have shown, for the purpose of transport modeling, removing the within-block heterogeneity requires the introduction of additional processes to make up for this loss of variability, which makes virtually impossible to generate the additional parameters directly from a few sampled data. As proposed in this, and many other papers on upscaling, the proper way to account for the disparity of scales is to build fine scale models based on the data, then to upscale them so that the model size is amenable to numerical modeling. It remains open the problem of how to integrate sampled data taken at different scales.

We recognize that the results have been demonstrated in a single case study, but we conjecture that the good performance of the method proposed is not case specific, and we base this conjecture in that the upscaling exercise is performed on a block by block basis at a scale in which the specific features of the different case studies will be less noticeable. We had to upscale several thousands of blocks using a systematic approach, with each block having a different distribution of fine scale conductivities. The upscaled parameters would have been computed similarly had the flow geometry or the conductivity heterogeneity changed. We acknowledge that the final results we present are based on the assembly of these blocks for a specific flow and transport problem, and the performance of this final assembly for a different case study may not work so well as it did in our example.

Further research is needed (i) to avoid the solution of the flow and transport at the fine scale in order to determine the coarse scale transport parameters, (ii) to explain the need for correcting the porosity when moving from the fine to the coarse scale, (iii) to determine how to upscale heterogeneous porosities, (iv) to evaluate the approach for different conditions/scenarios, such as statistical anisotropic conductivities, transient flow conditions or radial flow and (v) to account for data measured at different scales.

5. Summary and Conclusions

We have presented and demonstrated an algorithm for transport upscaling in three-dimensional highly heterogeneous media. This work is an extension of the work by Fernàndez-Garcia et al. [18] in two dimensions. Some of the critical features of this method is that it uses an elaborated Laplacian-with-skin approach to reproduce the flows instead of the simple-Laplacian scheme, the use of a multi-rate mass transfer process at the coarse scale to compensate for the loss of information during upscaling, and the need to perform a piecewise upscaling of effective porosity.

We have used a synthetic example to demonstrate the advantages of the interblock Laplacian-with-skin approach to upscale hydraulic conductivities as compared with other approaches. We found that using interblock centered conductivities and that using a skin to compute them results in a good reproduction of flows at the fine scale.

Moreover, we found that proper transport upscaling is particularly important for the reproduction of the late time behavior of the solute breakthrough curves. We also found that proper transport upscaling is important to not underestimate the breakthrough curve prediction uncertainty.

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