Upscaling of solute transport in mildly heterogeneous media: comparison of Fickian and non-Fickian approaches

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Abstract Non-Fickian solute transport models were investigated in the case of mildly heterogeneous porous media using a synthetic two-dimensional ln(K) field, generated randomly. Transport was fully solved using MT3D and exhibited scale-dependence. Monte Carlo simulations were used to quantify uncertainty in the absence of *a priori* data. Then, inverse and direct modelling using the continuous time random walk approach and the fractional advection–dispersion equation were performed in order to assess the upscaling capacities of these models. A sensitive improvement could be observed compared to the classical advection–dispersion model.

Keywords CTRW; FADE; non-Fickian transport; stochastic; synthetic example

INTRODUCTION

The scale-effect in dispersion of solute plumes migrating in aquifers was identified about 20 years ago (see e.g. Pickens & Grisak, 1981) and is still not fully resolved. Stochastic theories have greatly contributed to the understanding of the influence of soil heterogeneity on apparent dispersivities (Gelhar, 1993, and references therein), but their practical application remains painstaking as they require a geostatistical characterization of the aquifer. Recently, new transport models have emerged, some of them being a direct extension of the classical advection–dispersion equation. However, these models make use of unusual mathematical concepts, such as fractional derivatives, which might explain why still only a few practicians exploit them.

This paper shows, using a two-dimensional synthetic example, that even in the case of a weakly heterogeneous medium, the classical Fickian model fails to correctly scale up transport while non-Fickian transport models make an improved deterministic prediction of breakthrough curves (BTC) at the macroscale. Before presenting the reference example to be used, the basics of the continuous time random walk (CTRW) theory are presented, as well as the fractional advection–dispersion equation (FADE).

THEORETICAL BACKGROUND

In a heterogeneous medium, moving particles travel along different paths and at varying velocities. This kind of transport can be represented using a coupled time–space probability density function (PDF) that describes particle transitions in time and in space. In the classical Fickian transport model, this PDF is the Gaussian distribution.

In the CTRW and the FADE frameworks, the PDF is a more general distribution called the *Lévy distribution* (Benson *et al.*, 2000b), characterized by a parameter β ranging from 0 to 2. The importance of the distribution tail increases when β decreases, leading its second-order moment (and even, in the case β < 1, its first-order moment) to become infinite. This allows dissolved particles to travel over a much wider range of velocities and allows the variance of a particle cloud to grow nonlinearly in time, which might be closer to reality in the case of a heterogeneous medium. The Gaussian distribution is a particular Lévy distribution corresponding to β = 2.

Two basic approaches can be followed to describe the random displacement of a particle in a porous media. First, one could assume that the path of the particle can be divided into equidistant transitions in the mean flow direction and determine the distribution $\psi(t)$ of transition durations. Assuming $\psi(t)$ is a Lévy distribution characterized by β_t, Margolin & Berkowitz (2004) have developed a set of analytical expressions to compute concentration distributions in time and in space for various injection conditions. They have also developed inverse modelling tools to infer transport parameters from experimental data; they are used in this study.

In the second approach, the path of a moving particle is divided into transitions of equal duration and the distribution of travel distances $\psi(x)$ is a Lévy distribution (called in this case a *Lévy flight*) characterized by β_x. This approach, however, implicitly assumes that the first moment of the PDF is finite (i.e. that the mean velocity of the particle cloud can be computed) and is valid only in the case $1 < \beta_x < 2$. Starting from this assumption, Benson *et al.* (2000b) ended up with a one-dimensional extended transport equation of the form:

$$\frac{\partial C}{\partial t} = -v\frac{\partial C}{\partial x} + \left(\frac{1+\alpha}{2}\right)\tilde{D}\frac{\partial^{\beta_x} C}{\partial x^{\beta_x}} + \left(\frac{1-\alpha}{2}\right)\tilde{D}\frac{\partial^{\beta_x} C}{\partial (1-x)^{\beta_x}} \tag{1}$$

where C is the concentration, t is the time, x is the longitudinal position, v is the mean migration velocity, α is a skewness parameter and $\tilde{D}$ is comparable to the classical dispersion coefficient but is found to be scale-independent. Equation (1) is usually referred to as the *fractional advection–dispersion equation*, as fractional β_x-order derivatives appear. This model is a direct extension of the classical Fickian model as setting α to 0 and β_x to 2 yields the advection–dispersion equation.

One may wonder which approach is better. Some authors argue that Lévy flight formulations are inadequate in some physical applications (Berkowitz *et al.*, 2002). However, both approaches have already been successfully applied to physical experiments at the laboratory scale as well as in the field (Benson *et al.*, 2000a; Berkowitz *et al.*, 2001).

SYNTHETIC REFERENCE EXAMPLE

The two-dimensional synthetic example used for the purpose of this study is based on an experimental setup consisting of an 80-cm wide and 200-cm long sandbox (Frippiat *et al.*, 2004). A mean gradient of 10% was applied to the confined sample so that a one-dimensional flow in the longitudinal direction was simulated. In practice, fixed-head conditions of 2.2 and 2 m were imposed, respectively, on the upstream and the downstream boundaries. Other boundary conditions are illustrated in Fig. 1.

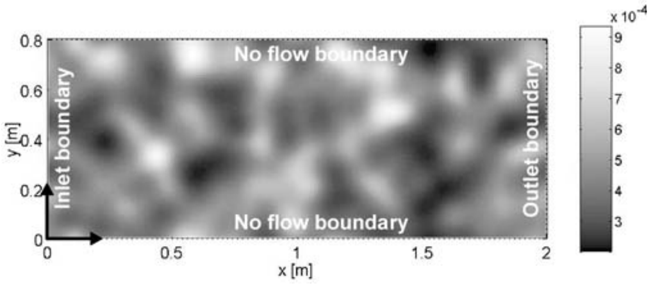

Fig. 1 Two-dimensional K field used as reference example (K in m s^{-1}).

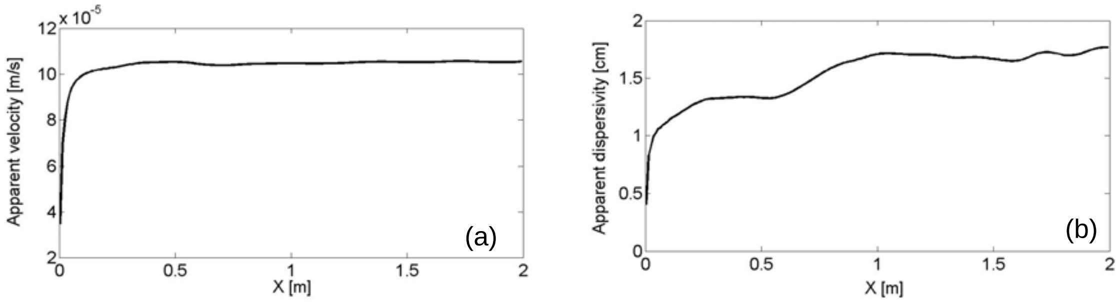

Fig. 2 Apparent transport parameters inferred from transversely averaged BTC.

One realization of a randomly heterogeneous ln(K) field was generated using a Matlab® routine (available at http://matlabdb.mathematik.uni-stuttgart.de) (Fig. 1), assuming a mean of −7.759 (K being expressed in m s^{-1}) and a Gaussian isotropic correlation function characterized by a correlation length of 0.1 m and a variance of 0.076. These values are based on measurements performed on real Brusselean sand samples manually compacted in the experimental tank. The variance is very low, but is nevertheless sufficient for the apparent dispersivity to exhibit a scale effect.

Flow was solved using MODFLOW (Harbaugh *et al.*, 2000) under steady-state conditions. The numerical domain is discretized in 200 × 80 cells of 1 × 1 cm. Assuming a zero initial concentration and a homogeneous step injection through the whole inlet boundary, transport was solved using MT3D (Zheng, 1990) for 30 000 s. Local longitudinal and transversal dispersivities were set equal to the numerical grid (α_L = α_T = 1 cm). Results were stored with a time resolution of 300 s, leading to concentration curves composed of 101 synthetic concentration data. Concentrations were averaged transversely (along the y-direction) and each of the 200 average BTC (for x = 1 to x = 200 cm) was analysed using standard curve-fitting tools to obtain apparent transport parameters (Fig. 2(a) and (b)). Despite a low variance of the ln(K) field, a scale effect clearly appears, leading to macroscale values of dispersivity of about twice the local value.

MONTE CARLO SIMULATIONS

In a second stage of the analysis, 2500 unconditional equiprobable ln(K) fields were randomly generated, similarly to the reference example shown on Fig. 1. Flow and transport were solved for each of these realizations and ensemble-statistics of concentration were computed (ensemble-mean and ensemble-standard deviation of concentration at each location (x,y) of the flow domain). The aim of this procedure is basically to assess the level of uncertainty associated with the reference example used throughout this study, in order to establish whether non-Fickian models contribute to reduce this uncertainty or not. As an example, Fig. 3(a) shows that the BTC simulated at the centre of the model (x = 100 cm, y = 40 cm) for the reference case falls by chance within the one-standard-deviation confidence interval.

As the 2500 equiprobable ln(K) fields where generated unconditionally (i.e. without incorporating any additional information on the aquifer), the one-standard-deviation confidence interval is found to be relatively wide, reflecting a relatively high uncertainty. One could have assessed the benefit of incorporating known local K values in the modelling process (i.e. to generate conditional realizations of the ln(K) field) to reduce uncertainty in BTC prediction, but that was beyond the scope of this paper. It can also be noted in Fig. 3(a) that the ensemble-mean BTC exhibits a larger apparent dispersivity, resulting from the combined uncertainty on the mean front position and on front spreading (Dagan, 1990).

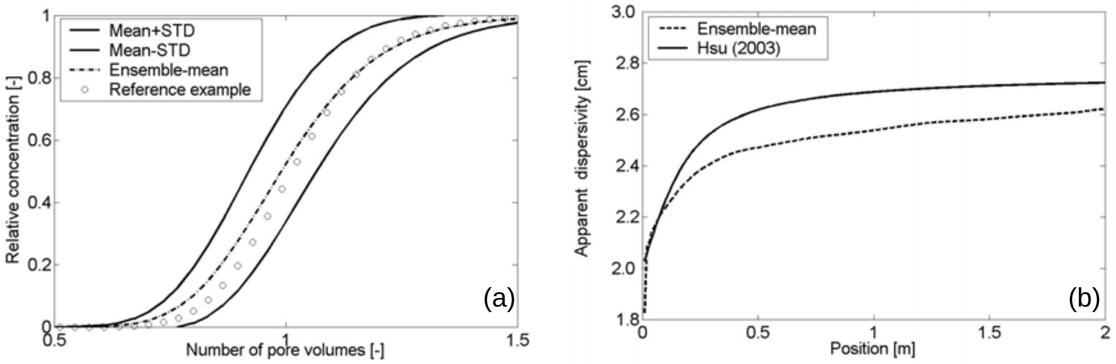

Fig. 3 (a) Confidence interval for the local BTC at x = 1 m and y = 0.4 m, and (b) Spatial development of macrodispersivity.

In addition, it was checked whether one could recover the analytical results of the stochastic theories for macrodispersivity. Figure 3(b) shows the apparent dispersivity of the ensemble-mean concentration distribution compared to the analytical solution proposed by Hsu (2003). The finite size of the flow domain and the curve-fitting method used to obtain transport parameters could account for the discrepancy.

APPLICATION OF THE CTRW FORMALISM

Each of the 200 average BTC of the reference example was re-analysed using Matlab® tools (available at http://www.weizmann.ac.il/ESER/People/Brian/CTRW) in order to obtain non-Fickian transport parameters. Figure 4(a) shows the evolution of β_t with the mean travel distance. It illustrates that β_t (as well as β_x) is a function of the travelled path. Indeed, geological systems can encounter heterogeneities on different hierarchical scales and the size of the largest heterogeneity is likely to influence β the most. As the travel distance increases, the size of this largest heterogeneity must also increase (and so must β), otherwise complete averaging would take place, leading to a Gaussian

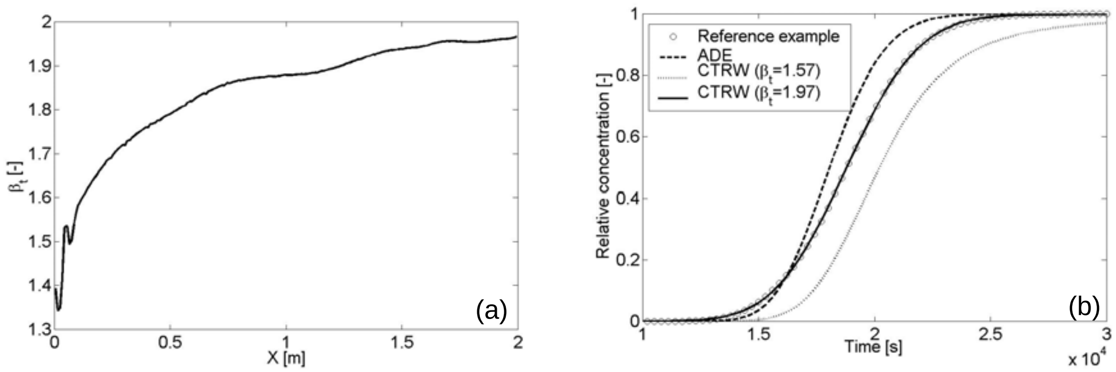

Fig. 4 (a) Evolution of β_t with the mean travel distance and (b) BTC at x = 2 m.

behaviour of the particle cloud (Margolin & Berkowitz, 2000). In this case, this situation was not reached, but β_t had almost converged to its threshold value of 2.

Figure 4(b) shows the average BTC at x = 2 m for the reference example as well as the corresponding curves obtained with the classical model (ADE) and with the CTRW approach, using transport parameters inferred from the analysis of the x = 0.1 m BTC (for the ADE, $v = 9.91 \times 10^{-5}$ m s^{-1} and α_L = 1.12 cm, and for the CTRW, β_t = 1.57). This was done in order to assess whether parameters obtained at a small (arbitrarily chosen) scale could be used to predict concentration distributions at larger scale (i.e. to assess the upscaling capacities of the CTRW approach). The slope of the concentration curve at half relative concentration (i.e. the apparent dispersion coefficient) is better predicted in the CTRW framework but the mean breakthrough time is overestimated. As advective transport is generally well controlled but does not appear explicitly in the CTRW theory, the results in Fig. 4(b) would preach for a hybrid approach, in which only dispersion differs from its classical description (just as in the FADE). For the sake of comparison, the BTC corresponding to the actual β_t value characterizing the BTC at x = 2 m (β_t = 1.97) is also shown in Fig. 4(b).

APPLICATION OF THE FRACTIONAL TRANSPORT EQUATION

According to the methodology proposed by Benson *et al.* (2000a), the transport parameters to be used in equation (1) can also be inferred from BTC analysis. It must be noted that each of the 200 average BTC of the reference example was used in this inverse modelling procedure. Consequently, compared to the CTRW approach which can be used in an actual upscaling process, this methodology requires information at every scale of interest. First, β_x is obtained graphically from the slope m of the apparent dispersivity *versus* mean position plotted on a log–log graph (Fig. 5(a)), according to $\beta_x = 2/(m + 1)$. In this case, numerical points are relatively well aligned and the estimate for β_x is 1.73 ± 0.01 (for the 95% confidence interval). Those values are consistent with the values plotted on Fig. 4(a). Then, the fitting of the standard cumulative distribution to a scaled breakthrough curve yields $\tilde{D} = 4.90 \times 10^{-6}$ m$^{1.73}$ s^{-1}. Figure 5(b) shows the reference BTC at x = 2 m and the solution of the FADE in the corresponding transport conditions. The slope of the curve at half relative concentration is relatively well predicted using the FADE, but significant differences

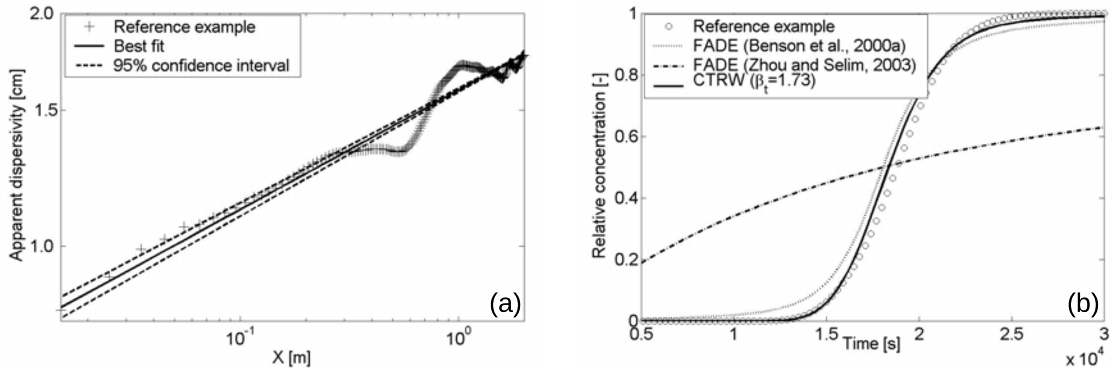

Fig. 5 (a) β_x and the scale effect and (b) BTC at x = 2 m.

remain in the tails of the distribution. Indeed, due to the very low variance of the ln(K) field, the mean concentration distribution appears to be nearly Gaussian, with quickly converging tails. This behaviour cannot be correctly reproduced using the FADE simultaneously to a scale effect in apparent dispersion. The corresponding solution to CTRW, for $\beta_t = \beta_x = 1.73$, is also illustrated in Fig. 5(b).

Zhou & Selim (2003) proposed another method allowing the simultaneous estimation of β_x and $\tilde{D}$ based on the evolution of the spatial variance of the particle cloud in time σ^2_C. They proposed to fit on this plot a nonlinear model of the type $\sigma^2_C = At^B$, where A and B are linked to β_x and $\tilde{D}$. Theoretically, parameters obtained from joint estimation should model more accurately the time dependence of the variance. A value β_x = 1.77 was found. However, the value $\tilde{D} = 3.81 \times 10^{-4}$ m$^{1.77}$ s^{-1} that was also obtained, led to a very poor prediction of BTC at larger scale (Fig. 5(b)). This illustrates the major problem when using nonlinear optimization to fit a given model to experimental data: if the model is too sensitive to one of its parameters, the value of that parameter can be largely influenced by experimental artefacts.

CONCLUSIONS

Flow and transport were simulated numerically in the case of a confined heterogeneous two-dimensional aquifer. Apparent dispersivity exhibited scale-dependence, making the use of the classical advection–dispersion equation with local transport parameters inappropriate. Unconditional stochastic simulations revealed a relatively high level of uncertainty whereas non-Fickian deterministic transport models could relatively safely scale up dispersivity values. It was noted that:

(1) regarding the general CTRW approach, advective transport was poorly predicted as it does not explicitly appear anymore;

(2) this drawback does not exist when using the FADE, but upscaling required a set of macroscale dispersivity values; and

(3) in this example of low heterogeneity level, only a weak tailing behaviour was observed for the BTC, which could not be correctly modelled neither in the CTRW framework nor using the FADE.

Finally, it should be emphasized that the key for successful application of both CTRW and FADE approaches is good estimation of β.

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