Journal of Hydrology 561 (2018) 427–443

Contents lists available at ScienceDirect

Journal of Hydrology

journal homepage: www.elsevier.com/locate/jhydrol

Research papers

Modelling solute dispersion in periodic heterogeneous porous media: Model benchmarking against intermediate scale experiments

Samer Majdalani [a,*], Vincent Guinot [a,b], Carole Delenne [a,b], Hicham Gebran [c]

[a] HSM, CNRS, IRD, Univ Montpellier, Montpellier, France
[b] LEMON, Inria, Univ Montpellier, Montpellier, France
[c] Mathematics Department, Lebanese University, Fanar, Lebanon

ARTICLE INFO

Article history:
Received 12 December 2017
Received in revised form 19 February 2018
Accepted 9 March 2018
Available online 28 March 2018
This manuscript was handled by C. Corradini, Editor-in-Chief, with the assistance of Rao S. Govindaraju, Associate Editor

Keywords:
Solute transport
Heterogeneous porous media
Intermediate scale
Fickian behaviour
Dispersion modelling

ABSTRACT

This paper is devoted to theoretical and experimental investigations of solute dispersion in heterogeneous porous media. Dispersion in heterogenous porous media has been reported to be scale-dependent, a likely indication that the proposed dispersion models are incompletely formulated. A high quality experimental data set of breakthrough curves in periodic model heterogeneous porous media is presented. In contrast with most previously published experiments, the present experiments involve numerous replicates. This allows the statistical variability of experimental data to be accounted for. Several models are benchmarked against the data set: the Fickian-based advection-dispersion, mobile-immobile, multirate, multiple region advection dispersion models, and a newly proposed transport model based on pure advection. A salient property of the latter model is that its solutions exhibit a ballistic behaviour for small times, while tending to the Fickian behaviour for large time scales. Model performance is assessed using a novel objective function accounting for the statistical variability of the experimental data set, while putting equal emphasis on both small and large time scale behaviours. Besides being as accurate as the other models, the new purely advective model has the advantages that (i) it does not exhibit the undesirable effects associated with the usual Fickian operator (namely the infinite solute front propagation speed), and (ii) it allows dispersive transport to be simulated on every heterogeneity scale using scale-independent parameters.

1. Introduction

In many circumstances, the classical Fickian operator fails to account correctly for the behaviour of solutes in heterogeneous porous media. The Advection-Dispersion (AD) model exhibits poor performance. Attempting to calibrate this model against field or laboratory data has been seen to lead to contradictory conclusions. Field scale dispersion data have been reported to yield a growing trend for the dispersion coefficient D with the scale of the experiment (Gelhar et al., 1992). A number of laboratory experiments, in contrast, indicate that no clear trend can be identified for the variations in D with experiment scale. For instance, Silliman and Simpson (1987) report an increasing trend for the dispersion coefficient. In Irwin et al. (1996), an increasing trend is found for $D(x)$, but the authors notice that this conclusion may be biased by experimental noise. In Sternberg et al. (1996), identifying a trend for the variations of D with distance is found very difficult if not impossible. In Danquigny et al. (2004), no scaling trend is identified for the dispersion coefficient, even over short distances. More recently, laboratory experiments carried out on an artificial, periodic porous medium (Majdalani et al., 2015) show that contradictory trends in $D(x)$ can easily be inferred if the breakthrough curves are not sampled with sufficient accuracy and the tracer experiments are not replicated a sufficient number of times. Several models with scale-dependent dispersion have been proposed in the literature (Aral and Liao, 1996; Jayawardena and Lui, 1984; Pickens and Grisak, 1981a,b; Yates, 1990, 1992; Zhang et al., 1994; Zhou and Selim, 2002). All these models have shown a good ability to reproduce field- or laboratory-obtained experimental breakthrough curves via a proper parameter tuning. This makes a benchmarking of their respective predictive capabilities very difficult (Gao et al., 2009). The following models have been used extensively for benchmarking against experimental data sets.

The Fractional Advection-Dispersion (FAD) model builds up on the Continuous Time Random Walk (CTRW) formalism (Klafter et al., 1987; Metzler and Klafter, 2000; Montroll and Weiss, 1965). FAD occurs when the motion of the solute molecules is

* Corresponding author.
E-mail address: samer.majdalani@umontpellier.fr (S. Majdalani).

https://doi.org/10.1016/j.jhydrol.2018.03.024
0022-1694/© 2018 Elsevier B.V. All rights reserved.

non-Brownian. Different behaviours may be obtained depending on the assumptions made on the characteristic times and lengths of molecule jumps (Klafter et al., 1987; Kumar et al., 2010; Kavvas et al., 2015, 2017). In the presence of trapping effects, an inverse power law asymptotic behaviour may be observed for the probability density function of solute residence time in the porous media. This results in subdiffusive dispersion processes, with a variance of molecule positions growing slower than time. Another type of non-Fickian behaviour is that of Levy motion, whereby the characteristic time for particle motion is finite, but the characteristic length of the jumps in molecule positions is infinite (Benson et al., 2000a,b). The resulting behaviour is called superdiffusion, with a variance of molecule positions growing faster than time. All these models share the common feature that the governing equations incorporate fractional derivatives with respect to time and/or space, hence the term "fractional". FAD models have been tested against experimental data sets obtained from laboratory experiments (Berkowitz et al., 2000; Cortis and Berkowitz, 2004; Huang et al., 2006; Lévy and Berkowitz, 2003). In Huang et al. (2006) the best fit was obtained by making the dispersion parameters scale-dependent. In Sun et al. (2014), a FAD model was tested against in situ data obtained from experiments at the scale of 1 m to 1 km. Comparisons with data observed at the metric scale (Berkowitz et al., 2008) showed that time-varying fractional orders of differentiation were essential in reconstructing the heavy tailing in the observed breakthrough curves.

The Mobile-Immobile (MI) model (Gaudet et al., 1977; Van Genuchten and Wierenga, 1977) is based on the assumption of a mobile region (where the solute obeys a standard AD model) exchanging with an immobile region. The MI formalism has been used to describe different physical settings. The simple structure of this model allows analytical solutions to be obtained for a number of configurations (De Smedt and Wierenga, 1979; Goltz and Roberts, 1988; Parker and Valocchi, 1986; Van Genuchten et al., 1984). Several versions of the MI model with a scale-dependent dispersion coefficient have been explored in Gao et al. (2010). The best fit against the experimental laboratory results obtained in Huang et al. (1995) was achieved for a dispersion coefficient varying exponentially with the travelled distance.

The Multiple Rate (MR) model (Haggerty and Gorelick, 1995) is a generalization of the MI model. Several immobile regions exchange with the mobile region according to different exchange rates. Increasing the number of regions and varying the exchange kinetics allows for anomalous diffusion processes to be reproduced via a proper distribution of the exchange rates between the mobile and immobile fractions (Dentz and Berkowitz, 2003).

Multiple Region Advection-Dispersion (MRAD) models have been proposed to account for the dispersion of solutes in heterogeneous soils in the presence of macropores, high- or low-permeability inclusions or several spatial scales of hydraulic heterogeneity. Note that the term MRAD is not the name given to these models by their authors but a term proposed by the authors of the present paper for the sake of terminology convenience. In these models, several different mobile regions, each having its own velocity fields and dispersion coefficient, exchange mass. Several closure models have been investigated for the exchange between the two regions. Although most applications include two mobile regions (Ahmadi et al., 1998; Cherblanc et al., 2003, 2007; Gerke and Van Genuchten, 1993a,b, 1996; Gwo et al., 1998; Skopp et al., 1981), applications with three mobile regions have been reported (Gwo et al., 1996). Two region models have been tested against numerical experiments (Cherblanc et al., 2003, 2007; Davit et al., 2010) and laboratory experiments (Golfier et al., 2007, 2011). They are shown to become equivalent to a single region model with a Fickian behaviour (that is the AD model) in the limit of long times and travel distances (Ahmadi et al., 1998; Davit et al., 2010; Golfier et al., 2011). Conversely, they are deemed more accurate than the AD model for small times and highly contrasted hydraulic properties (Golfier et al., 2011).

All these models have shown a good ability to reproduce field- or laboratory-obtained experimental breakthrough curves via a proper parameter tuning. This makes a benchmarking of their respective predictive capabilities very difficult. As shown in Golfier et al. (2011), tracer tests involving a strong heterogeneity allow for a better model discrimination than tests involving weakly variable porous media. Moreover, pulse tracer tests are also deemed more discriminatory in terms of model response than step injection tests, especially for long time and/or travel distances (Golfier et al., 2011). However, most experiments report either step tracer tests (Huang et al., 1995; Irwin et al., 1996; Li et al., 1994; Niehren and Kinzelbach, 1998; Silliman and Simpson, 1987; Sternberg et al., 1996) or very long pulses that may be interpreted as a succession of two steps (Saiers et al., 1994; Silliman and Simpson, 1987; Golfier et al., 2011). A few exceptions are reported in Golfier et al. (2011), Greiner et al. (1997), Tran Ngoc et al. (2011).

As shown in a previous publication (Majdalani et al., 2015), the AD, FAD and MI models with scale-independent parameters fail to account for the behaviour of experimental breakthrough curves at small space and time scales when the porous medium is strongly heterogeneous and periodic. Two main reasons were identified for this. Firstly, the size of the Representative Elementary Volume (REV) (Bear, 1972) is at least one order of magnitude larger than the spatial period of the Model Heterogeneous Porous Medium (MHPM). Dispersion models are not valid at spatial scales smaller than the REV size. Secondly, a Laplace analysis of the theoretical AD, FAD and MI modelled breakthrough curves (Majdalani et al., 2015) shows that these models yield infinite signal propagation speeds. An infinite concentration wave speed is clearly physically unrealistic. Besides, the finite propagation speed of the concentration signal exerts a strong influence on the behaviour of the experimental breakthrough curves for small times and distances (Majdalani et al., 2015), which explains that the above three models are more inaccurate for small times and short distances than for long time and distances. That Fickian-based dispersion models only seem to become more accurate as the spatial scale increases is only due to the fact that the Peclet number increases with distance (therefore, dispersion, albeit modelled wrongly, has a decreasing importance in the modelled signal) (Majdalani et al., 2015). These conclusions are to be extended to the FAD model with superdiffusive behaviour. Indeed, this model is obtained under the assumption of heavy-tailed PDFs for the particle jump length (Metzler and Klafter, 2000), thus allowing for infinite particle velocities. A conclusion of the study (Majdalani et al., 2015) is therefore that models where advective processes play a predominant role should be expected to give better results than AD- and FAD-based models at small scales.

The experimental results in Majdalani et al. (2015) also indicate that previously identified scale dependency of the dispersion coefficient may easily be explained by the variability between the replicates of a same experiment.

The objectives of the present paper are the following.

(i) Build a high-quality experimental database for Intermediate Scale Experiments (ISE) of dispersion of tracers in heterogeneous porous media. In Majdalani et al. (2015) it was chosen to build a periodic heterogeneous porous medium made of a series of 15 cm long columns enclosing high permeability conduits surrounded by single-sized glass beads. However, for a single period and two periods, the results were biased by the influence of the inlet and outlet boundary conditions. Consequently, experiments were meaningful for a minimum of three successive periods. In the present work,