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Solute transport in periodical heterogeneous porous media: Importance of observation scale and experimental sampling

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SUMMARY

This paper focuses on the effects of the observation scale and sampling on the dispersion of tracers in periodical heterogeneous porous media. A Model Heterogeneous Porous Medium (MHPM) with a high degree of heterogeneity was built. It consists of a preferential flow path surrounded by glass beads. 44 tracer experiments were carried out on several series of periodic MHPM to investigate the effect of the observation scale on solute dispersion. Each series was replicated several times, allowing for a statistical description of the unit transfer function of the MHPM. No significant trend was found for the dispersion coefficient as a function of the size of the MHPM. However, given the variability of the breakthrough curves from one experiment replicate to another, under-sampling might easily lead to conclude that the dispersion coefficient is variable with distance. Depending on the samples used, it would be as easy to (wrongly) detect an increasing trend as to detect a decreasing one. A confidence interval analysis of the experimental breakthrough curves in the Laplace space shows that (i) there exists a model with scale independent parameters that can describe the experimental breakthrough curves within the limits of experimental uncertainty, (ii) this model is not the advection–dispersion (AD) model, (iii) the modelling error of the AD model decreases with the number of periods, (iv) the size of the Reference Elementary Volume for the dispersion coefficient is between 10 and 20 periods. The effects of sampling prove to override those of scaling. This, with the invalidity of the AD model, leads to question attempts to calibrate and/or identify trends in the dispersion coefficient at intermediate scales from a limited number of experiment replicates.

1. Introduction

Understanding solute transport in porous media is important to predict the fate of contaminants in natural soils. Natural soils are highly heterogeneous and implicate various transport mechanisms. Tracer experiments on laboratory soil columns made a big contribution to identify these mechanisms. The most widespread transport model for inert solute is the advection–dispersion (AD) model. Fitting this model against real-world tracer tests has been reported to induce an increase in the dispersion coefficient with the travelled distance (see e.g. Dagan, 1989; Gelhar et al., 1992; Zhou and Selim, 2003). The variability of the dispersion coefficient with distance has motivated a strong interest for alternative models. Recently, laboratory tracer experiments have been used as a benchmarking basis to test the respective accuracy of a variety of normal and anomalous transport models (see e.g. the review in Gao et al., 2009). It has also been suggested by some authors that the dispersion coefficient in Mobile–Immobile models should be made exponentially dependent on distance in order to accurately reproduce experimental breakthrough curves in laboratory column experiments (Gao et al., 2010).

A number of laboratory tracer experiments in porous media are available from the literature (Silliman et al., 1998). Laboratory experiments on heterogeneous porous media are very few in comparison to those done on homogeneous porous media. One cause might be the practical difficulty to conceive and construct heterogeneous porous media.

Silliman and Simpson (1987) used fine sand inclusions (small cubes) with coarse sand surrounding. They found a change in the shape of the breakthrough curve at each of five measurement sections, and inferred a continuous increase in dispersivity with distance. Saiers et al. (1994) used a coarse sand inclusion (tubule) with finer sand surrounding. The inclusion constituted a

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preferential flow path and contributed to more than 60% of the mass balance. The breakthrough curves exhibited multiple inflection points that cannot be accounted for by the classical advection dispersion (AD) model. In comparison to other studies, the work of Saiers et al. (1994) is distinguished by the high degree of heterogeneity of the porous media. This fact is reflected in the shape of the breakthrough curves. Much more than a mere tailing effect, the breakthrough curves exhibited a clear two-step shape. Li et al. (1994) used coarse inclusions (polyethylene porous cylinders) with finer sand surrounding. The resulting breakthrough curves exhibited asymmetry. Huang et al. (1995) used a 12.5 m long column with a wide range of soil materials. Their breakthrough curves exhibited irregular and nonsigmoidal distributions and were in most cases poorly described by the AD model.

Sternberg et al. (1996), Irwin et al. (1996) were the first to use a periodical heterogeneous porous media, where the heterogeneity consisted of a succession of several glass bead sets tested in several serial orders. The breakthrough curves were used to infer the variation of the dispersion coefficient with distance. Unfortunately, the breakthrough curves are not shown in the publications and no estimate is provided for the goodness-of-fit of the AD model. It is thus impossible to determine whether fitting the coefficients of the AD model was meaningful. However, both studies present the results of dispersion estimations. Sternberg et al. (1996) found that dispersion does not necessarily increase with scale, but that it can increase and decrease depending on the particular arrangement of properties in the medium. They deduced that the system length was unimportant in observing the scale effect. Conversely, in the study of Irwin et al. (1996), dispersion appeared to be scale dependent up to a certain travel distance. But, quoting the authors' own words, "because of the scatter in the data, it could be argued that the apparent increase in longitudinal dispersion is simply a result of the variation in the data". This raises the issue of sampling effects.

Greiner et al. (1997) used aggregates of lower permeability in a surrounding of higher permeability. Niehren and Kinzelbach (1998) used cylindrical cellpore filters (less permeable) in a quartz-sand surrounding (more permeable). Tran Ngoc et al. (2011) used spherical clay inclusions (micro porosity) surrounded by sand (macro porosity). Danquigny et al. (2004) performed tracer tests on two kinds of heterogeneous porous material: a channel structured medium (5.6 m × 1 m × 1 m), with channels crossing the whole tank, and a statistically correlated random structure. They found that the breakthrough curves of the statistically correlated random structure could be fitted with the AD model (at least for long distances), but that the fit is quite poor for channel structured model due to the non-Gaussian distribution of the concentration.

Zin et al. (2004) designed an artificial porous medium with several conductivity contrasts, using low conductivity inclusions (small beads) surrounded by a high conductivity matrix (large beads). They found that the breakthrough curves of the low-contrast medium showed no tailing, while those of high-contrast medium showed tailing. Golfier et al. (2011) used fine sand inclusions (lenses) with coarse sand surrounding. Their breakthrough curves showed asymmetry. The low degree of heterogeneity in the studies of Sternberg et al. (1996), Irwin et al. (1996) might have led to some contradictory conclusions concerning the dispersion scale dependence. As to the study of Saiers et al. (1994), the authors were not mainly interested in the problematic of dispersion scale dependence, but rather in colloidal mobilization and transport in a structured porous media.

In the light of the abovementioned studies, it appears that studying the effect of heterogeneity on solute transport requires two conditions: (i) a high degree of heterogeneity of the porous media (as the experimental model of Saiers et al. (1994)), and (ii) a periodical heterogeneous porous media (as the experimental protocol of Sternberg et al. (1996), Irwin et al. (1996)). The first condition guarantees that the effects of material heterogeneity are clearly visible, while the second allows the influence of scale effects to be assessed. This also allows for a better quality of the experimental data, with easily replicable experiments yielding reliable statistics.

In this study, we conceived a Model Heterogeneous Porous Medium (MHPM) with a high degree of heterogeneity. The purpose is to (i) enrich the experimental database of laboratory tracer tests with high quality data, (ii) answer the following six questions.

- (Q1) Can a scaling trend be observed for the dispersion coefficient in this highly heterogeneous MHPM?
- (Q2) If this is not the case, can the variations of the dispersion with the observation scale be attributed to experimental artefacts such as sampling effects?
- (Q3) Does there exist a model with uniform coefficients (thus scale-independent) to describe adequately the present MHPM response?
- (Q4) Can the AD model be considered valid at the scale of this MHPM?
- (Q5) If it cannot, does a scale exist above which the AD model can be deemed a satisfactory approximation of the experimental response of the MHPM?
- (Q6) Can more advanced models such as the Mobile–Immobile (MI) or Continuous Time Random Walk (CTRW) models be considered as better candidates than the AD model to simulate small-distance and short-time MHPM behaviour?

The detailed characteristics of the experimental setup is described in Section 2 of the paper. The results of experimental breakthrough curves are shown in Section 3. Such experiments are usually termed Intermediate Scale Experiments (ISE) in that they involve a scale larger than the typical size of the heterogeneity but they do not achieve the size of the Reference Elementary Volume (REV) above which the continuous medium formalism should be expected to be valid (Bear, 1972).

Section 3 is also devoted to an analysis of the transfer function of the MHPM. Section 4 is devoted to discussion. A point-by-point answer to the six questions above is given throughout Section 3 and 4.

2. Materials and methods

2.1. Model heterogeneous porous media

The MHPM consists of a set of PVC columns (10 cm in diameter, 15 cm in length each) containing 1 mm glass spheres (40% porosity) surrounding a cylindrical cavity (2.5 cm in diameter, 10 cm in length) placed in the centre of the column (Fig. 1). The cylindrical cavity has 100% porosity and thus acts as a wide pore that induces preferential water and solute transfer. The pore volume V_0 of one MHPM period is about 461 ml.

The water and solute flow enters and leaves the column by 4 inlets and outlets (4 mm in diameter). A 500 μm sieve prevents the glass spheres from clogging the inlets and outlets, possibly blocking the flow. Fig. 2 shows real captures of the MHPM. The incoming discharge (imposed by a peristaltic pump) is equally distributed between the 4 inlets. It is expected that the flow of the central inlet will mainly transfer through the central cavity, while the flow of the three remaining inlets will mainly transfer through the surrounding glass beads.

After carrying out the series of experiments, four randomly chosen columns were disassembled. In each of them, the position of the sieve was compared to what it was before the experiment. No detectable displacement was noted.