Random Walks in Prefractal Porous Media

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Random walks are used to examine dispersion between miscible fluids in one-dimensional, prefractal porous medium. Mixing in such media is anomalous and several nonlocal theories have been proposed to describe this non-Fickian characteristic. This work simulates dispersion via a random walk in a prefractal media by incorporating local and spatially nonlocal effects and compares the results with laboratory experiments. Results suggest that nonlocal models fit experimental data better than local models.

Introduction

The main phenomenon of interest in this work is dispersion in prefractal porous media. Dispersion is used here to describe the mixing of miscible fluids flowing within a porous medium. Prefractal is a term used to describe real objects with fractal characteristics over a limited range of scales (see Feder, 1988 or Mandelbrot, 1983). Recent observations of mixing in natural porous media suggest dispersion is dependent on the time/space scale of observation (Gelhar et al., 1985, 1992; Neuman, 1990). It has been inferred that the scale dependence results from evolving heterogeneity (Sposito et al., 1986). The characteristic of evolving heterogeneity can be defined as a property whose average value changes as the size of the region over which the average is taken changes. Such a property may be described as fractal, although for this work we use a prefractal medium, which is a model that is neither statistically nor actually periodic at the system size. Recent theories of dispersion in such media incorporate nonlocal constitutive relations for the dispersive flux. To our knowledge, only one study has examined the magnitude of the nonlocal effect (Cushman et al., 1994b). In that article it was found for a conservative tracer that nonlocal effects were not important concerning spatial moments through the second. However, for higher spatial moments or for reactive transport nonlocality is critically important.

This work explores the effect that evolving heterogeneity has on mixing in four random walk models. These models differ in the way that dispersive effects are included. The models are labeled as local, distributed, memory and nonlocal. Laboratory experiments are used to compare the results of the random walk models. The simulation goal is to compare different theoretical models for predicting dispersion in prefractal systems. This is accomplished by observing the dispersion profile evolution as the mixing front encounters abrupt changes in the permeability field. The major contribution of this article is to investigate dispersion in media which lacks periodicity, and to verify that nonlocal theories provide a superior modeling tool.

Several reviews (Sahimi, 1993; Haselow et al., 1989; Gelhar et al., 1985) and books (Scheidegger, 1963; Bear, 1972; Greenkorn, 1983) have been written on the subject of transport phenomena in porous media. Many include detailed discussion of the theoretical basis for describing dispersion with random walk models. Each of the above mentioned books includes discussion of dispersion measurement in laboratory experiments. Since there is a great deal of information already available describing the techniques used in this work, this discussion will emphasize what is new and novel about our approach.

Dispersion Background

Dispersion can be described as the amount of mixing between fluids caused by diffusion and flow in a porous medium. Greenkorn (1983) identifies nine mechanisms which cause dispersion: diffusion, tortuosity, autocorrelation in flow paths, recirculation, changes in gross streamlines, hydrodynamic dis-

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persion, turbulence (eddies), dead end pores, and adsorption. It should be noted that the majority of these mechanisms are dependent on the fluid velocities and not on concentration gradients, yet the standard advection-dispersion equation relies only on a concentration gradient as the driving force for mixing.

Measurements of dispersion can be made by observing the rate at which a front between two miscible fluids spread into each other. Dispersion in the laboratory experiments was calculated from the time-concentration data (breakthrough curves). Dispersion in the random walks was determined from the time required for a set number of particles to reach a particular distance from the origin in the model. Dispersion in both cases was calculated by using a form of the true mixing model of Bear (1972)

$$D = \frac{L\upsilon}{2}\left[\frac{(t_{0.841} - t_{0.159})^2}{t_{0.500}^2}\right] \tag{1}$$

where D is the dispersion coefficient (cm^2/s), L is the medium length (cm), υ is the seepage velocity (cm/s), and the ts are the times (s) associated with the dimensionless concentration values (or cumulative fraction of particles) in the subscript. These time values correspond to one standard deviation of a normal distribution, from which the subscripts originate.

The inadequacy of the standard advection-dispersion equation for aperiodic media has prompted much theoretical effort. The development of a nonlocal theory of dispersion begins in the same manner as the standard or classical dispersion theory, with the continuity equation

$$\frac{\partial C}{\partial t} + \nabla \cdot N = 0 \tag{2}$$

where C represents the concentration of a conservative tracer and N represents the flux. Since the dispersion profile resembles a diffusion profile for homogeneous media, Fick's first law with an advective term has been used as the constitutive relationship

$$N = \upsilon C - \rho D \cdot \nabla(C/\rho) \tag{3}$$

where υ is the velocity, ρ is the density of the fluid, and D is the dispersion coefficient. If the fluid is incompressible and D is a constant scaler, this equation may be rearranged to give

$$\frac{\partial C}{\partial t} + \upsilon \cdot \nabla C = D\nabla^2 C \tag{4}$$

which is the standard advection-dispersion equation. This Fickian constitutive theory requires that the process be, in some sense, asymptotic. This result holds for homogeneous media at large distances and long times. However, in heterogeneous media the asymptotic limit may never be reached. This fact has lead to the development of scale-dependent models, some of which use nonlocal constitutive theories.

Several nonlocal models have been proposed (Koch and Brady 1987a,b; Thompson, 1988; Naff, 1990; Neuman, 1993; Cushman and Ginn, 1993a,b, and Cushman et al., 1994a). The concept behind these works is to represent the flux of the tracer as an integral equation over space and time. Cushman and Ginn (1993b) present this relationship as

$$N = \upsilon C(\bar{x},t) - \int_{R^3}\int_0^t D(\bar{y},\tau,t)\cdot\nabla_{\bar{x}-\bar{y}} C(\bar{x}-\bar{y},t-\tau)\overline{dy}\,d\tau \tag{5}$$

This flux can be substituted into the continuity equation and rearranged to form a nonlocal advection-dispersion equation

$$\frac{\partial C}{\partial t} + \upsilon\cdot\nabla C = \nabla\cdot\int_{R^3}\int_0^t D(\bar{y},\tau,t)\cdot\nabla_{\bar{x}-\bar{y}} C(\bar{x}-\bar{y},t-\tau)\overline{dy}\,d\tau \tag{6}$$

Equation 6 is said to be nonlocal because of the presence of integrals and derivatives. This allows it to use information from the entire integrated domain, instead of just the local region around the point of interest. The integration also allows information from different scales of observations to be included. We will use this concept of using information from the entire system to compare results with experimental dispersion measurements in prefractal porous media.

Random Walks: Background and Methods

Random walks have been used since the 1950s (Scheidegger, 1958; De Josselin de Jong, 1958) to model flow of a tracer through a porous medium. The method is based on the analogy between diffusion theory and stochastic processes. It involves studying the movement of many discrete particles through a given flow field. The flow field is determined through knowledge of the permeability field (hydraulic conductivity), the use of the flow equation, and an appropriate set of boundary and initial conditions.

This method has the advantages of being easy to understand, easy to use and modify, and it avoids the numerical problems associated with partial differential equation solvers. Also, it is relatively simple to include nonlocal effects. The key disadvantage is the amount of computer time required. To improve statistical accuracy the number of particles must be increased, yet accuracy is only proportional to the square root of the number of particles.

Many researchers have successfully used random walk methods to model diffusion and dispersion phenomena. The procedure used here is based on a standard random walk method as described in Prickett et al. (1981), (also see Spitzer, 1975; Scheibe and Cole, 1994), in which time is discrete and position is continuous (within the limits of single precision).

The random walk algorithm consists of three parts. First, a tracer particle is placed at the entrance to the model. Then, the particle is moved a deterministic distance, which corresponds to the velocity and time step size. Finally, the particle is moved a random distance whose characteristics are determined by the value of a dispersion multiplier at that point. Equation 7 summarizes the algorithm for particle movement.

$$x_{\mathrm{NEW}} = x_{\mathrm{OLD}} + \upsilon(x_{\mathrm{OLD}})\Delta t + [M(x_{\mathrm{OLD}})\Delta t]^{0.5} RN \tag{7}$$

where x_{NEW} denotes the new time particle position ($t + \Delta t$), and x_{OLD} the old time position (t). υ denotes the velocity

(which can be a function of position, but is not in this work), M denotes the dispersion multiplier, Δt is the time step, and RN is a Gaussian random number of zero mean and unit variance. Random numbers in [0, 1] were obtained using the long period ($> 2 \times 10^{18}$) random number generator of L'Ecuyer with Bays-Durham shuffle, as described in Press et al. (1992). This deviate is then adjusted to provide a Gaussian random number.

Time step size was chosen to be one second. The number of particles was determined to be 1,000. Using additional particles did not improve accuracy or precision. Velocity was assumed to be constant (flow rate, porosity, and cross-sectional area were constant in all systems). Equation 8 shows an empirical relationship (based on laboratory experiments) used to calculate the local value of the dispersion coefficient for any region in the simulation models.

$$D = \alpha v^{1.2} \sqrt{k} \tag{8}$$

Here k is the permeability (cm^2) of the local region, v is the seepage velocity (cm/s), and α is an empirical constant determined by matching the random walk results with experimental data for homogeneous media. α had a single value of 25.7 for all the systems examined in this work.

Experimental variables of the random walk simulations include eight media and four methods of determining the dispersion multiplier, $M(x)$ in Eq 7. Results are shown for only one fluid velocity (0.011 cm/s). Although three velocities were investigated, results for each were very similar. It should be noted that dispersion is calculated in the longitudinal direction only. Transverse dispersion (dispersion in the directions perpendicular to average flow) was examined and found to have no effect on the results; hence, it was not studied. This is appropriate as, in the laboratory experiments, average flow was in a single constant direction, the tracer was added uniformly over the column face, and no radial deviations were detected. In systems with three-dimensional velocity fields, however, transverse effects will be important and must be included.

Figure 1 depicts the porous media models examined. A total of eight different models were examined, of which two are homogeneous and six are heterogeneous combinations of the two homogeneous media. These six models consist of a pattern in which the first and last 25% of the model has the characteristics of one of the homogeneous models, and the middle 50% has characteristics of the other model. This model can be described as a first generation Cantor dust of fractal dimension 0.5. The arrangement was chosen such that the overall system is aperiodic and has abrupt changes within the system. Length was varied in order to examine the observation that dispersion measurements are larger in larger systems (Gelhar et al., 1992; Neuman, 1990). Four methods (local, distributed, memory, and spatially nonlocal) were used to determine the dispersion multiplier.

The local method uses a single, constant value of dispersion.

$$M(x) = D \tag{9}$$

This global value of D the dispersion coefficient was calculated from Eq. 8 using the harmonic mean of the permeability values from each model section. This method describes the system as homogeneous and dispersion as an effective parameter. It is included mainly for comparison, as it is not expected to predict dispersion at smaller or larger scales of measurement, although it should predict the dispersion for the entire system.

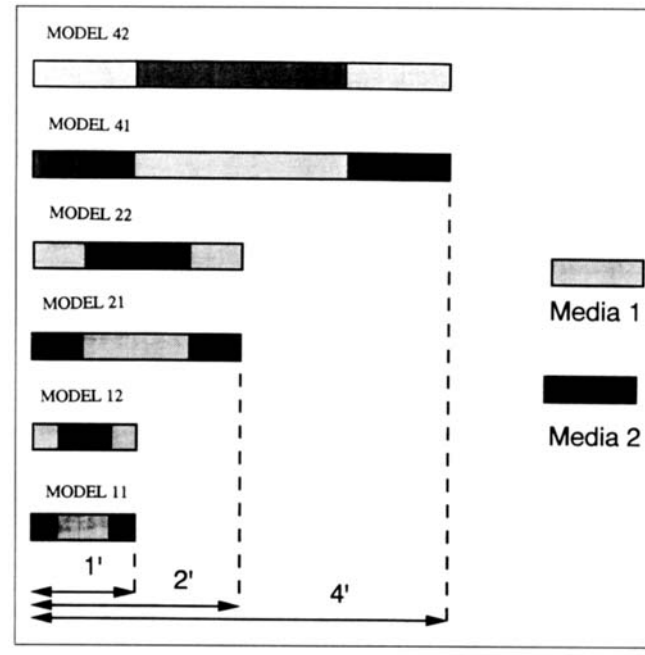

Figure 1. Models of the prefractal porous media.

The distributed method uses a dispersive term which is constant for the section of the model that the particle is in. The multiplier changes only when the medium properties change locally.

$$M(x) = D(x) \tag{10}$$

For example, in model 42, a four ft column, velocity is 0.011 cm/s, and k is 2.27×10^{-6} cm^2 when the particle is between 0.0 and 12.0 in. (305 mm), $k = 0.138 \times 10^{-6}$ cm^2 between 12.0 and 36.0 in. (305 and 914 mm), and $k = 2.27 \times 10^{-6}$ between 36.0 and 48.0 in. (1,219 mm). The dispersion multiplier can then be determined at any point using Eq. 8 and knowledge of these local properties.

The memory method uses a multiplier which is a weighted average of all past and present values of the dispersion term.

$$M(x) = \frac{1}{x} \int_0^x D(\tau) d\tau \tag{11}$$

The dispersion values at any particular point are calculated the same as in the distributed method, but before it is used in Eq. 2, it is averaged with all the previous values of the dispersion multipliers as shown in Eq. 11.

The spatially nonlocal method uses a multiplier which is a weighted average of all the dispersion values in the system.

$$M(x) = \frac{1}{L} \int_0^L w(x) D(x) Dx \tag{12}$$

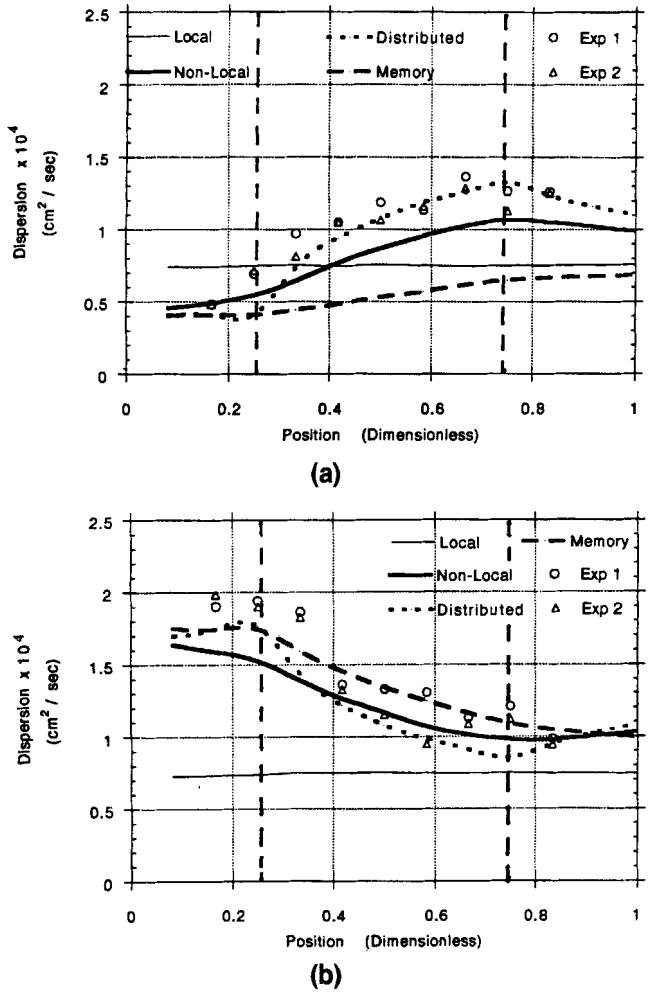

Figure 2. (a) Results from Model 11 (one ft length, 2-1-2 medium); (b) results from Model 12 (one ft length, 1-2-1 medium).

L is the system length and $w(x)$ is an empirical expression derived from the laboratory data. Several different simple forms were examined such as a one-sided ramp function, a two-sided ramp function, a square function, and an exponential function. The closest fit was obtained with an exponential decay curve whose peak is shifted in front of the particle position by 2.5 cm (1 in.). This relationship is given by

$$w(x) = 0.27 \exp\left[-\frac{|x_p - x + 1|}{2}\right] \tag{13}$$

where x_p is the particle's position in inches. Only results from this function are given in this work. The experimental data used to generate this empirical expression are provided on the figures showing the random walk results (Figures 2, 3 and 4). As this is an empirical fit, it provides a good first approximation of the form we can expect such a function to have. Additional experiments and simulations are needed to discover if other parameter dependencies exist (such as porosity, nature of the miscible fluids, and transverse effects).

Laboratory Experiments

The data from the experiments mentioned in this section are used to make comparisons with the random walk methods. A complete description of the laboratory measurements is described in Sternberg (1994). A short description of the method follows.

Laboratory experiments were carried out in 3.2-cm-ID Lucite columns. Three lengths were examined: 30.5, 61.0 and 122.0 cm (12, 24 and 48 in.). The columns were filled with unconsolidated spherical glass beads held in place with fine mesh screens. Two sets of beads were used in the experiments. Beads were packed into the columns in four arrangements: all bead 1, all bead 2, layered in the arrangement of 25% bead 1—50% bead 2—25% bead 1, and the opposite arrangement of 25% bead 2—50% bead 1—25% bead 2. The single bead columns were examined in the 30.5-cm-long columns. The layered arrangements were examined in all three lengths. Figure 1 shows the arrangements and notation for the eight columns.

Table 1 lists the experimental measurements of the properties of porosity, permeability, and dispersion coefficient for each column. Porosity was determined using the liquid satu-

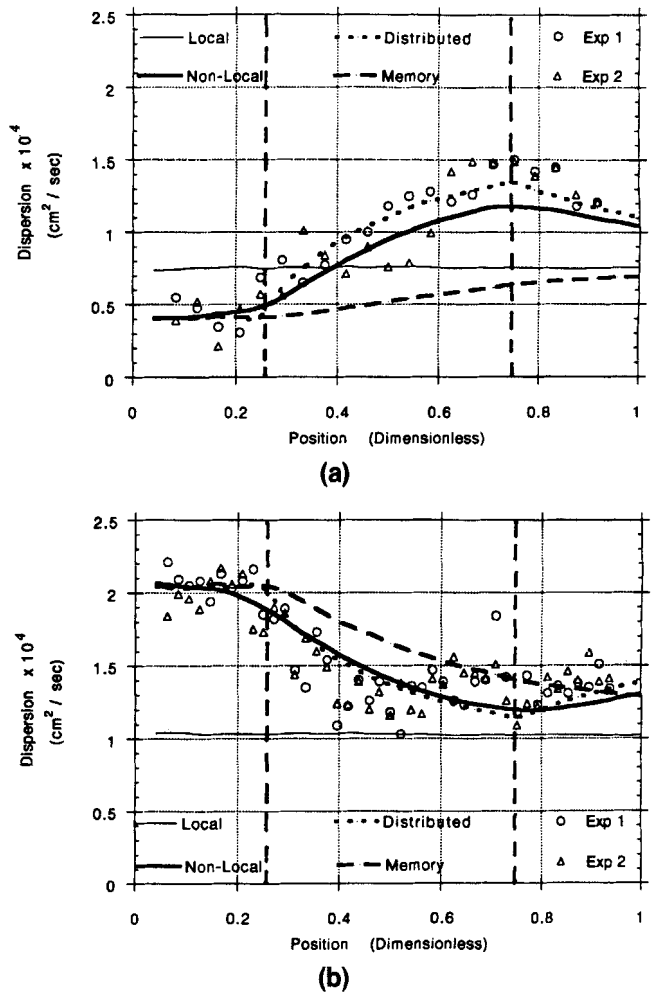

Figure 3. (a) Results from Model 21 (two ft length, 2-1-2 medium); (b) results from Model 22 (two ft length, 1-2-1 medium).

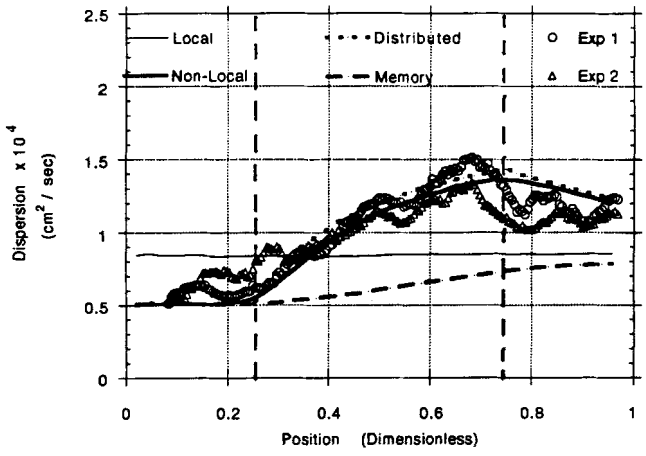

Figure 4. (a) Results from Model 41 (four ft length, 2-1-2 medium); (b) results from Model 42 (four ft length, 1-2-1 medium).

ration method (Scheidegger, 1963). Permeability was determined using Darcy's law at several flow rates (Greenkorn, 1983). Dispersion was measured in each distinct system by monitoring the change in concentration of a chloride tracer as a function of time and position within the columns when a weak concentration solution (0.02-M KCl) was displaced by a strong concentration solution (0.2-M KCl). The more dense solution is introduced at the column bottom and pumped upwards to displace the weaker solution. This direction was chosen to eliminate gravity and viscous fingering effects. The chloride concentrations were measured using a silver/silver chloride electrode and measuring the potential difference obtained with a standard calomel electrode. Measurements were made at many positions along the column length (2.5-cm spacing in the 40.5-cm column, 1.26-cm spacing in the 60.5-column, and 0.63 cm in the 120.0-cm column). Time between measurements at a given electrode was 4 s. The millivolt signal was converted to chloride concentration using a log-linear relationship calibrated for each electrode in each run. Equation 1 shows the relationship used to calculate dispersion from the breakthrough curves.

Table 1. Parameter Values of the Two Layers

Parameter	Layer 1	Layer 2	Units
Porosity	35.0	35.3	%
Permeability	2.27	0.138	$\times 10^{-6}$ cm^2
Dispersion	1.69	0.438	$\times 10^{-4}$ cm^2/s

Results and Discussion

Figures 2, 3 and 4 present the results of this study. Each figure plots dispersion as a function of dimensionless length. Dimensionless length is determined by dividing each position by the final position, and it is used to allow comparison between the three column lengths. Five profiles are shown in each figure, the points represent laboratory data, the lines show the results of the four types of random walk simulations (local, distributed, memory, and nonlocal). Figures 2a–4a provide results for the heterogeneous columns in which the layers are arranged in the 1-2-1 combination. Figures 2b–4b provide results for the 2-1-2 combination. Figures 2a and 2b show results from the one ft columns, Figures 3a and 3b show results for the two ft columns, and Figures 4a and 4b show results for the four ft columns. Boundaries between the layers were at the 25% and 75% of total length positions.

Interestingly, the laboratory results show the dispersion profile changes before it encounters the boundary between layers. This suggests that the velocity profile is altered before exiting the layer, much like the exit effects in flow entering a pipe from a large tank. This type of effect is not included in most theories, and cannot be included by those whose driving force for dispersion is based solely on concentration gradients (Fickian processes). It has been suggested that such effects are nonlocal, because they are created by changes in properties that are not in the local area of the process. This nonlocal effect may be described through the use of a velocity field correlation function. However, the use of such a theory would require a more complete knowledge of the flow field at the pore scale. Comparisons between Figures 2, 3 and 4 show similar responses to changes in the permeability, even though total length in each system is different. This demonstrates that system size has no bearing on this phenomenon.

An additional variable that was examined, but not included in this work, was velocity. Three velocities were used (0.011, 0.016 and 0.026 cm/s). Results for all three values were similar, and since the other velocities added no new information about this system, they were omitted. These results are included in Sternberg (1994).

The local model provides (as expected) a good prediction of the final mixing in each system, but not at intermediate points. This method was adequate for predicting dispersion in the homogeneous media and demonstrates the usefulness of the random walk method.

The distributed model appears to predict the dispersion profiles better; however, it does not display the interesting behavior present at the transitions in permeability. Also, careful examination of these results shows symmetry in the dispersion profile, which suggests that layer order is unimportant. However, as shown in Sternberg and Greenkorn (1994), the order of the layers does indeed change the final value of the dispersion measurement. Hence, while this

method is a distinct improvement over the local model, it does not exhibit the observed behavior.

The memory model provides very poor prediction of the dispersion profiles. This suggests that knowledge of the particles past behavior alone is insufficient to predict its future behavior.

The spatially nonlocal model yields the best fit to the dispersion profiles. This model provides a way for information throughout the medium to affect the mixing process at any given point in the system. This model allows the dispersion profile to react to changes in the medium *before* the changes are encountered. The results are nonsymmetric, which also agrees with the observed experimental results.

Conclusions

The random walks studied show that including nonlocal spatial effects is essential to correctly model dispersion in heterogeneous porous media. Many theories had correctly proposed that nonlocal effects would be important in such media. Our previous experiments have shown that this is true for simple laboratory cases. This work demonstrates that spatially nonlocal random walk models yield more accurate predictions than other tested models.

The laboratory results indicate that the spatially nonlocal effects are most noticeable when flow is from regions of high permeability to regions of low permeability. This suggest that the weight function should account for the relative change of the permeability between layers. No attempt was made to include this complexity in the models.

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