Transport in Porous Media **18:** *283-302.1995.*

Experimental Investigation of Solute Transport in Large, Homogeneous and Heterogeneous, Saturated Soil Columns

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(Received: 25 June 1993; in final form: 5 May 1994)

Abstract. Laboratory tracer experiments were conducted to investigate solute transport in 12.5-m long, horizontally placed soil columns during steady saturated water flow. Two columns having cross-sectional areas of 10 x $10\,cm^2$ were used: a uniformly packed homogeneous sandy column and a heterogeneous column containing layered, mixed, and lenticular formations of various shapes and sizes. The heterogeneous soil column gradually changed, on average, from coarse-textured at one end to fine-textured at the other end. NaCl breakthrough curves (BTC's) in the columns were measured with electrical conductivity probes inserted at 50- or 100-cm intervals. Observed BTC's in the homogeneous sandy column were relatively smooth and sigmoidal (S-shaped), while those in the heterogeneous column were very irregular, nonsigmoidal, and exhibited extensive tailing. Effective average pore-water velocities (v_{eff}) and dispersion coefficients (D_{eff}) were estimated simultaneously by tilting an analytical solution of the convection-dispersionequation to the observed BTC's. Velocity variations in the heterogeneousmedium were found to be much larger than those in the homogeneous sand. Values of the dispersivity, $\alpha = D_{eff}/v_{eff}$, for the homogeneous sandy column ranged from 0.1 to 5.0cm, while those for the heterogeneous column were as high as 200cm. The dispcrsivity for transport in both columns increased with travel distance or travel time, thus exhibiting scale-dependency. The heterogeneous soil column also showed the effects of preferential flow. i.e., some locations in the column showed earlier solute breakthrough than several locations closer to the inlet boundary. Spatial fluctuations in the dispersivity could bc explained qualitatively by the particular makeup of the heterogeneities in the column.

Key words: Solute transport experiments, heterogeneous media, dispersion, scale-depcndcncy.

1. Introduction

Solute transport in soil and groundwater is affected by a large number of physical, chemical and microbial processes and media properties. Ignoring microbial processes and assuming equilibrium interactions between solutes in the liquid and solid phases, transport during one-dimensional steady water flow in a uniform medium is generally described with the convection-dispersion equation (CDE) as follows

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x}, \tag{1}$$

where C is the volume-averaged solution concentration (M/L^3), t is time (T), x is distance (L), v = q/ϵ is the average pore-water velocity (L/T) in which q is the Darcian fluid llux density (L/T) and ϵ is the effective porosily (L^3/L^3), R is a retardation factor accounting for linear equilibrium sorption or exchange (dimensionless), and D is the dispersion coefficient (L^2/T).

Assuming negligible diffusion, the ratio of the dispersion coefficient to the pore-water velocity is given by the dispcrsivity, α:

$$\alpha = \frac{D}{v} \tag{2}$$

The dispersivity reflects the scale of mechanical mixing caused by variations in the local fluid velocity around its mean value. The value of α has traditionally been considered a characterisitc of the entire medium (Bear, 1972), usually in the order of 0.1 to 2 cm for repacked homogeneous soil columns (Fried, 1975; Freeze and Cherry, 1979). These results are in contrast to those from field experiments which indicate that α for natural geologic media can be one or several orders of magnilude higher than lhose determined using disturbed laboratory soil columns (Fried, 1975; Anderson, 1979; Pickens and Grisak, 1981a; Molz et al., 1983; Gelhar *et al.*, 1992). Moreover, results from field studies suggest that the dispersivity may increase with travel time, travel distance, and/or measurement scale (Pickens and Grisak, 198 I a; Molz et *al.*, 1986; Huyakorn et *al.*, 1986). This scale-dependency of the dispersivity is considered to be a consequence of the heterogeneous nature of the geological materials (e.g. Pickens and Grisak, 1981 b; Molz et al., 1983; Sudicky, 1986; Moltyaner and Killey, 1988).

The problem of scale-dependent dispersion has been the subject of numerous theoretical and a selected few experimental studies. Theoretical studies by Gelhar et al. (1979), Smith and Schwartz (1980), Dagan (1982, 1984), Poley (1988), and Neuman (1990), among olhcrs, have demonstrated the significant effects of variations in hydraulic properties on dispersive mixing. These studies suggest that the dispersivity of a heterogeneous aquifer at first grows more or less linearly with the solute residence time and/or travel distance as the mixing scale increases, but eventually may approach a constant asymptotic value. The approach lo au asymptotic dispersivity may be very slow and involve relatively long travel times or distances (Dagan, 1984; Sudicky, 1986).

As a complement to the above theoretical studies, several large-scale field experiments have been carried out to investigate solute transport behavior in natural aquifers (Freyberg, 1986; Sudicky, 1986; Moltyaner and Killey, 1988; Garabcdian et *al.*, 1991; Boggs *et al.*, 1992; Jensen et *al.*, 1993). These studies have provided considerable insight into the transport process as affected by both small- and large-scale variations in the hydraulic properties. Unfortunately, large-scale field experiments are very costly, time-consuming, and relatively difficult to execute. As opposed to carefully controlled laboratory tracer studies such experiments also involve a large number of uncertainties which are difficult to control or quantify, including those arising from the natural heterogeneity of the aquifer material, as well as human activities.

An obvious advantage of laboratory tracer studies is that the experimental conditions can be much better controlled and monitored. Many of the early laboratory displacement studies, especially those in soil physics (e.g., Nielsen and Biggar, 1961; Elrick et al., 1966) involved relatively *short* columns (generally less than 50cm) using both disturbed and undisturbed soils. More recently, tracer experiments have been carried out to study transport in much larger columns or lysimeters filled with a variety of soil of geologic materials, including layered or otherwise stratified formations, as well as soil slabs containing lenses of distinctly different permeabilitics, all designed to simulate various heterogeneous conditions at spatial scales of up to about 2 to 3 m (e.g., Sudicky et *al.*, 1985; Refsgaard, 1986, Silliman et *al.*, 1987; Schincariol and Schwartz, 1990). Most of these experiments were used to study convective transport and dispersion parallel to layering as generally occurs in saturated groundwater systems. For example, in flow tank experiments designed to also study density-dependent fl ow, Schincariol and Schwartz (1990) found that more dispersion occured in a lenticular medium than in both homogeneous and layered systems. By contrast, Khan and Jury (1990) studied transport in both repacked and undisturbed vertical soil colutnns of lengths up to 87 cm. They found a significant linear increase in the dispersivity, α, with length of the undisturbed colutnns, but no clear evidence that α related to the measurement scale of the repacked columns. Vertical solute transport in much larger (6-m long) repacked homogeneous and horizontally layered soil columns was studied by Wierenga and van Genuchten (1989) and Porro et *al.* (1993), respectively. Their studies showed that some preferential flow occurred in the homogeneous columns, and that dispersivities were greater in the uniform column (3.5 cm) than in the layered column (1.2cm).

The purpose of this paper is to present experimental results of solute transport throught 12.5-m long homogeneous and heterogeneous soil columns. The length dimensions were chosen such that different heterogeneous formations could be included in the columns, and to allow for a scale-dependent dispersion process to develop as much as possible. Three tracer experiments were conducted during steady saturated water flow: (1) transport of a NaCl solution into a homogeneous sandy column, (2) transport into a heterogeneous column containing a variety of layered, mixed, and lenticular soil formations of various shapes and sizes, and packed such that the particle size changed on average from very coarse at one end to very fine at the other end, and (3) leaching of solute from the same heterogeneous column, but with flow now in the opposite direction, i.e, from the fine-textured to the coarse-textured side. Effective average pore-water velocities and dispersion coefficients were estimated by fitting analytical solutions of equation (1) to the breakthrough curves observed at regular intervals along the horizontally placed columns.

2. Experimental

2.1. COLUMN PREPARATION

The experiments were conducted using 12.5-m long columns constructed with 0.5-cm thick plexiglass plates. The columns had internal dimensions of 10 cm in width and 10cm in height. Two soil columns were prepared, a homogeneous column packed as uniformly as possible with a medium-textured sand, and a heterogeneous column. The heterogeneous column was filled with a wide range of soil materials including clay, fine-, medium- and coarse-textured sands, gravel, and pebbles of 1 to 2cm in diameter, as well as various mixtures of these materials. Several lenses and layers of different shapes and sizes were rather arbitrarily embedded in this heterogeneous column to ensure a relatively high degree of heterogeneity. The soil materials used in both columns were taken from the deposits of a mountain river; they were clean, well-sorted, and contained negligible organic matter. Figure 1 shows a schematic of the experimental setup, including a cross-section of the heterogeneous soil. Notice that, on average, the particle size of the heterogeueous column decreased gradually from very coarse at the inflow end (the M-hand side in Figure 1) to relatively fine at the outlet (right-hand side). The porosity of each material was estimated indirectly from measurements of the particle density and the dry soil bulk density using standard methods (Klute, 1986). The porosity, n, was approximately 0.45 for the clay, and about 0.33 for the other materials. In order to ensure tightly compacted soil cohnnns, the soils were first slightly wetted and carefully mixed before packing. The cohnnns were subsequently packed layer by layer in increments of 1 to 2cm. During packing, piezometers were installed laterally at 50-cm intervals in the center of both columns, while electrodes were installed at 50-cm intervals in the homogeneous column and 100-cm intervals in the heterogeneous column (Figure lc).

After packing, but before closing the columns at the top, the horizontally-placed columns were slowly saturated from the bottom up for three days and then allowed to drain through holes in the bottom plexiglass plate. The wetting and drying cycle promoted further compaction of the soil. Following drainage, a clay paste of about 1 cm thickness was placed on the surface of the packed columns to serve as the upper aquitard (the bottom plexiglass plate served as the lower aquitard). The columns were subsequently covered tightly by a plexiglass plate. To keep close contact between the plexiglass plate and the top clay layer, the inside surface of the top plate was made rough by gluing small pieces of gravel on the surface. A series of braces were installed next to prevent the columns from swelling during the transport experiments. Finally, a high-quality silicon sealant was applied along the plexiglass joints to avoid leakage, while small l-cm long influent and effluent reservoirs were connected to both ends of the soil cohnnns. The reservoirs were separated from the soil columns by plexiglass screens, coarse-textured sand and a nylon mesh. The packed columns thus obtaiucd simulated confined aquifers with thicknesses of approximately 9 cm.

2.2. TRANSPORT EXPERIMENTS

Before starting the transport experiments, the columns were saturated slowly from the bottom using deaired tap water. Entrapped air in the columns was removed by applying a slightly negative pressure at each piezometer. A steady saturated flow field was established in the columns by imposing constant positive pressure heads at the inlet and outlet positions using Mariotte bottles and constant head level apparatus, respectively (Figure 1). Deaircd tap water was continuously applied for about one week to remove as much remaining entrapped air as possible. We assumed that steady-state water flow was reached when fluctuations in the observed drainage rates and piczometer readings became negligible.

Table I summarizes the hydraulic conditions for the three experiments. Although steady flow conditions were established before the transport experiments, the flow rates at the outlets continued to decrease slowly during the tracer studies. For example, the flow rate decreased from 12.09 to 11.25 cm/hr during the first 24 hrs of the transport experiment in the homogeneous sandy column,. The flow rates listed in Table I hence are averages of the measured drainage rates during the experiments. The continued small changes in flow rates were likely due lo slight changes in the soil hydraulic properties because of pore clogging by fine soil particles, and perhaps also because of changes in the concentration and/or ionic composition of the soil solution (Nielsen et al., 1986). The following three tracer experiments were carried out after steady-state flow were established: (1) a tracer injection (transport) experiment in the homogeneous sandy soil column by replacing inflowing tap water with a NaCl solution of concentration C_0 = 6 g/L, (2) a tracer injection experiment in the heterogeneous column by applying the 6 g/L NaCl solution at the coarse end (the left-hand side of Figure 1 b), and (3) a leaching experiment in which the initial saline (6 g/L) fluid introduced during the injection experiment was replaced with deaired tap water applied at the fine-textured end of the heterogeneous column (the right-hand side of Figure lb). The leaching experiment was started after continuously application 6 g/L NaCl solution for 4 days when the entire column reached a more-or-less uniform initial concentration, C_i.

Breakthrough curves (BTC's) were estimated from electrical conductance readings using a calibrated relationship between the specific concentration of a NaCl solution and the corresponding electrical conductance. The electrical conductance was measured be means of electrical conductivity probes inserted laterally into the center of the columns at 50- and lOO-cm intervals in the homogeneous and heterogeneous columns, respectively. The platinum-blackened 5-mm diameter electrodes with cell constants of either 0.9 or 1.1 l/cm were isolated from the surrounding saturated soil by means of screened copper tubes (Figure lc). The clcctrical conductivity of the deaired tap water solution (equivalent to a 0. I54 g/L NaCl solution) was considered negligible in the BTC data analysis.

The transport experiment in the homogeneous column was conducted in March, 1991, at a temperature $17 \pm 2\,^\circ\mathrm{C}$. The injection and leaching experiments in the

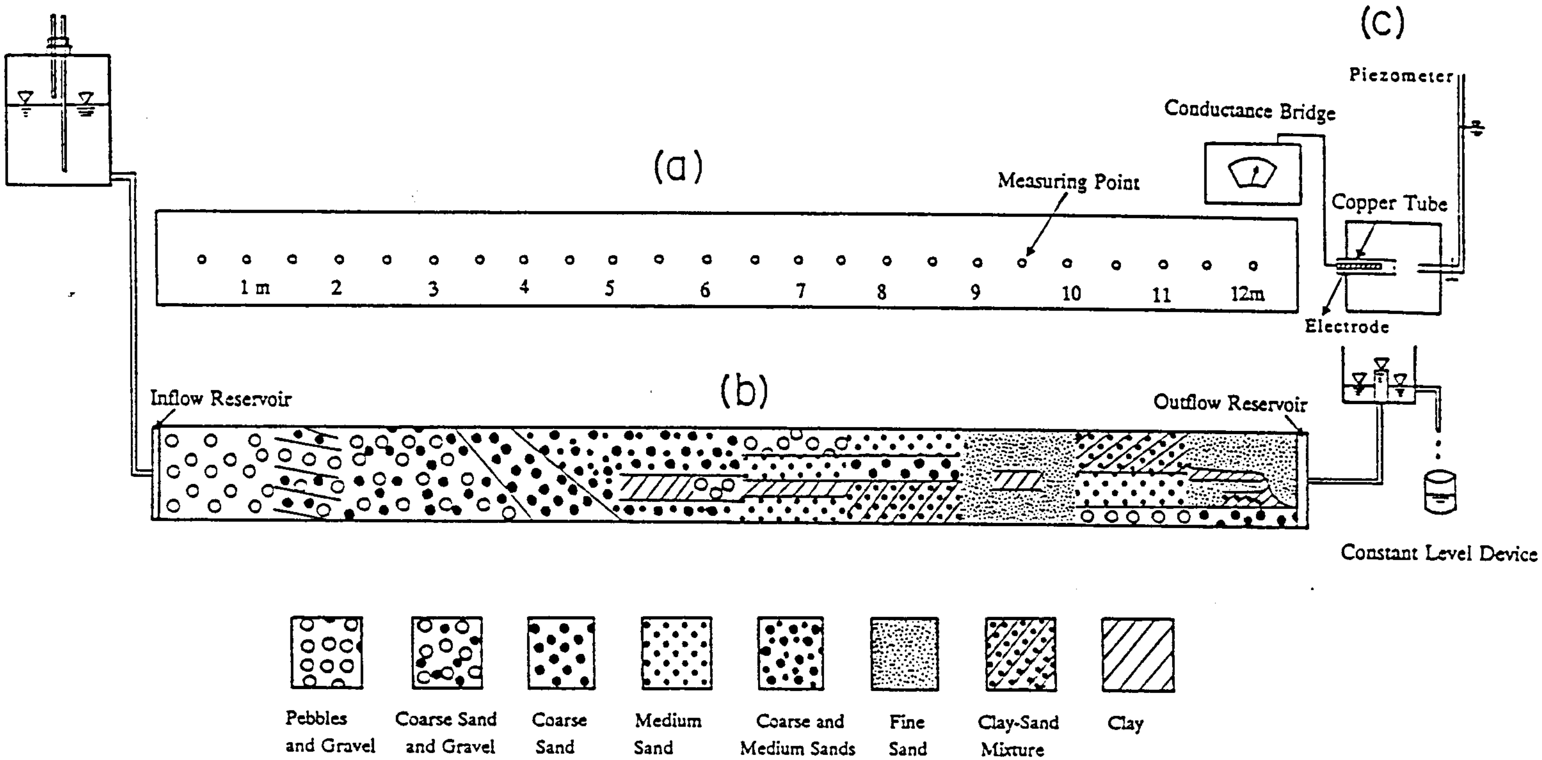


Fig. 1. Schematic of the soil column transport experiments: (a) vertical cross-section of the homogeneous sandy soil showing locations where piezometric heads and electrical conductances were measured, (b) vertical cross-section of the heterogeneous column showing also the inflow and outflow devices, and (c) cross-section of the column with diagrams of the piezometer and electrode.

TABLE I. Summary of hydraulic gradient ($\bar{J}$), flow rate (q), porosity (n), ensemble average of the fitted pore-waler velocity (ii), and effective porosity (ϵ)

Experiment	$\bar{J}$	q $cm\,hr^{-1}$	n –	$\bar{v}$ $cm\,hr^{-1}$	ϵ –
Tracer injection in homogeneous sandy column	0.0539	11.51	0.33	34.7	0.332
Tracer injection in heterogeneous column (coarse → fine)	0.0488	14.33	0.37	61.6	0.233
Leaching from heterogeneous column (fine → coarse)	0.0205	4.93	0.37	20.7	0.238

heterogeneous column were carried out in May 1991, at average temperatures of 22 and 30 °C, respectively. The measured BTC's for the homogeneous columns are shown in Figure 2, while those for the heterogeneous soil are presented in Figures 3 and 4 for the tracer injection and leaching experiments, respectively. The figures show relative concentrations obtained by dividing observed concentrations by the concentration (C_0) of the applied tracer solutions or, in the case of leaching, by the initial concentration (C_i).

3. Data Analysis

Assuming solute transport in a small-scale homogeneous medium, the dispersion coefficient, D, can be readily determined by fitting appropriate analytical solutions of the CDE, equation (l), to the experimental data (e.g., Fried, 1975; Sauty, 1980). More sophisticated approaches, such as the stochastic-analytic approaches of Dagan (1982) and Gelhar and Axness (1983), may be needed to determine the effective flow and transport parameters of heterogeneous media using information about the geostatistical properties of the hydraulic properties. Such information is impractical to obtain for most or all field-scale transport problems (Sudicky and Huyakorn, 1991). In our study we also had no detailed information about the spatial distribution of the hydraulic conductivity and porosity, since only breakthrough curves and steady Darcian flow rates, q, were measured.

In order to evaluate large-scale transport behavior without having detailed information of the hydraulic property distributions inside the columns, we applied, as in the stochastic approach, a macroscopic mean transport equation with effective parameters. The macroscopic convection-dispersion equation with effective

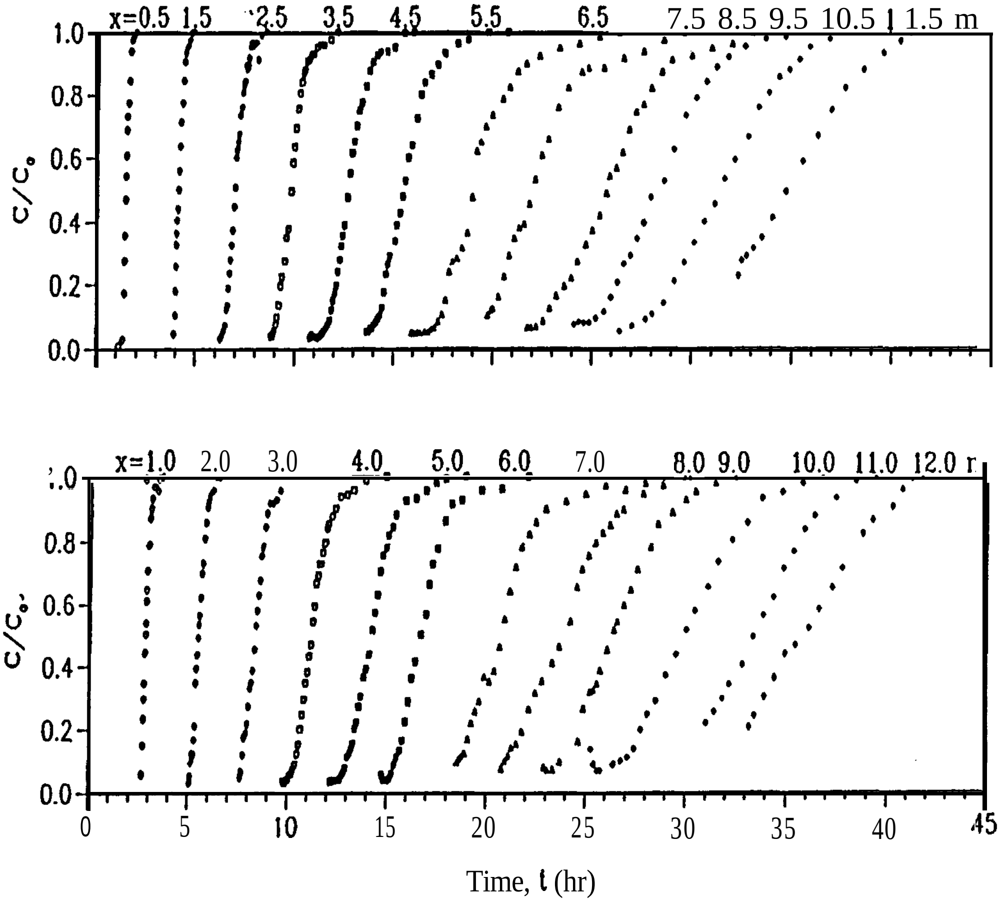
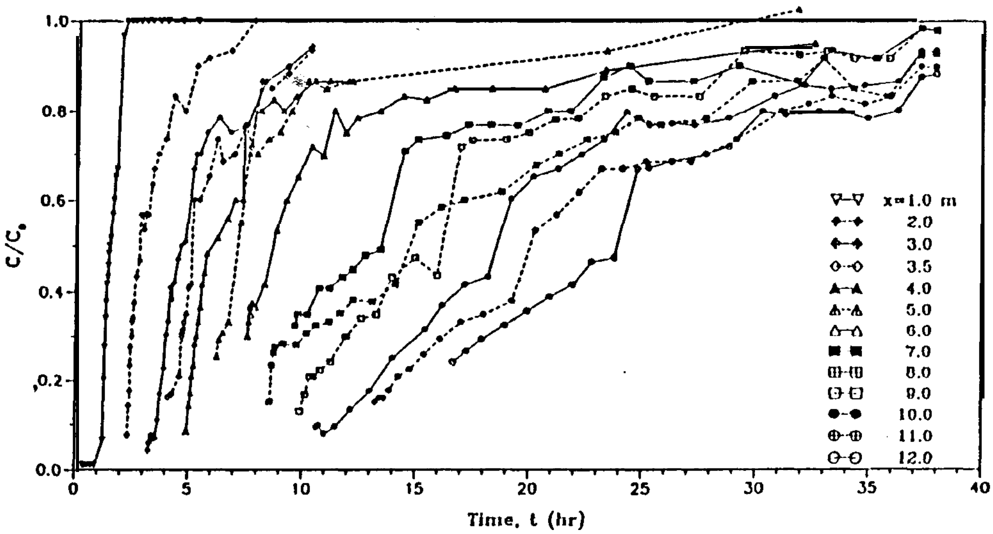


Fig. 2. Observed breakthrough curves for the tracer injection experiment in the homogeneous sandy soil column.

constant parameters for tracer transport in a heterogeneous medium is given by (Moltyaner and Killey, 1988; Jensen et al., 1993).

$$R_{\text{eff}}\frac{\partial\langle C\rangle}{\partial t} = D_{\text{eff}}\frac{\partial^2\langle C\rangle}{\partial x^2} - v_{\text{eff}}\frac{\partial\langle C\rangle}{\partial x}, \tag{3}$$

where $\langle C\rangle$ is the vertical mean concentration, while the subscript eff denotes an effective parameter. Our approach here implies that the measured EC concentrations represent mean values as averaged over the relatively small (0.01 m^2) cross-sectional area of the columns. When the parameters of the mean transport equation, (3), are determined from a breakthrough curve al a certain location, x, they are affected by all of the flow and transport processes between the inlet position and that particular location. Therefore, the estimated effective parameters will represent average values pertaining to the applicable travel distance (between 0 and x) and measurement time period. The spatial distribution of the resulting effective



Fig, 3. Observed breakthrough curves for the tracer injection experiment in the heterogeneous soil **column in which waler flowed from the coarse-textured end to the fine-textured end.**

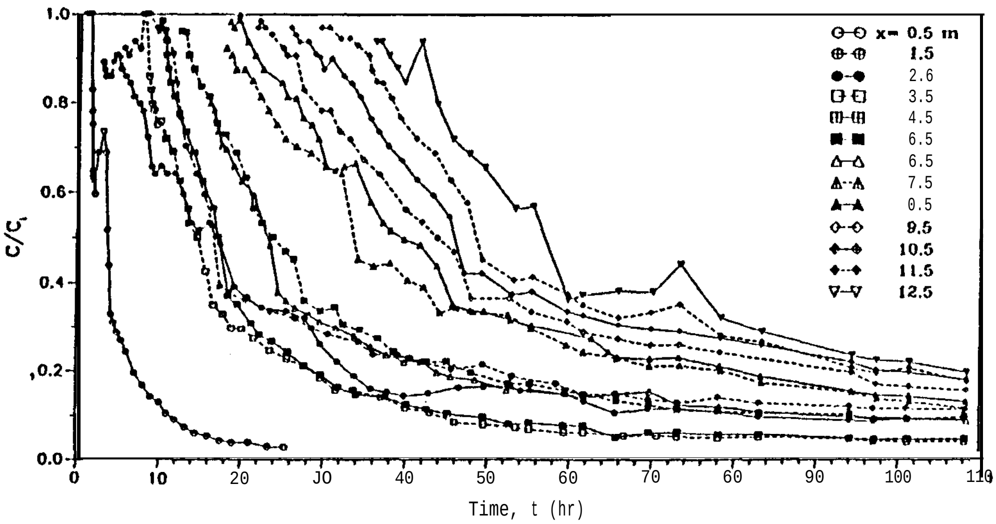


Fig. 4. Obscrvcd breakthrough curves for the leaching experiment in the heterogeneous soil column in which water flowed from the fine-textured end to the coarse-textured end.

parameters should in an approximate manner reflect the macroscopic lransport properties of the heterogeneous medium.

Effective average pore-water velocities, v_{eff}, and dispersion coefficients, D_{eff}, were estimated from the observed BTC's in Figures 2, 3 and 4 using the nonlinear least-squares parameter optimization code CXTFIT of Parker and van Genuchten (1984). This code was used to fit the analytical solution of the macroscopic CDE for a flux-type inlet boundary condition, a uniform initial condition and a uniform semi-infinite system, to the experimental data. The code enables one to fit v_{eff} and

D_{eff}, simultaneously to the measured BTC data. We assumed negligible retardation of the NaCl tracer in the columns, i.e. R = 1 in (1) and R_{eff} = 1 in (3), since the soils involved mostly coarse-&lured materials relatively free of organic matter. Once v_{eff} and D_{eff} were estimated from the BTC at a particular location, the apparent dispersivity, α, was calculated using an equation similar to (2). Table II summarizes the simultaneously fitted pore-water velocities and dispersion coefficients. The coefficient of determination, r^2, in this table reflects the goodness-of-fit (Spiegel, 1992, p. 263) as follows

$$r^2 = 1 - \frac{\sum_1^N (C_{obs} - C_{est})^2}{\sum_1^N (C_{obs} - \bar{C}_{obs})^2}, \tag{4}$$

where the subscripts obs and est denote observed and estimated values, respectively, the bar indicates a mean value, and where N is the number of observed concentration data at a particular observation point.

4. Results and Discussion

The observed solute breakthrough data for the transport experiment in the homogeneous column (Figure 2) show relatively regular, sigmoidal (S-shaped) distributions at most measuring points. These distributions are consistent with CDE type BTC's usually observed for carefully packed, relatively short homogeneous soil columns.

The BTC's for the injection experiment in the heterogeneous column are shown in Figure 3. As compared to those for the homogeneous column, the observed concentrations are much more irregular (less smooth) and exhibit nonsigmoidal distributions. The BTC's in many cases also do not reach the inlet concentration within the experimental time period because of extensive tailing. The BTC's in Figure 4 for the leaching experiment, starting from the opposite (i.e., fine-texlured) side of the heterogeneous column, show very similar features, i.e., irregular distributions with considerable tailing. One notable difference between Figures 3 and 4 is the lack of tailing in the BTC's near the inlet position in Figure 3 (the coarse-textured side), and the more pronounced tailing in the BTC's near the inlet position in Figure 4 (now the fine-textured side). This feature reflects the more heterogeneous nature of the soil near the outlet as depicted in Figure 1, and the relatively homogeneous makeup near the inlet (recall that the flow direction in the leaching experiments is opposite from that in the injection experiment). The BTC's in Figures 3 and 4 exhibit increased tailing and spreading as the transport experiments progressed in time and distance.

Figure 5 shows plots of the fitted effective average pore-water velocity versus distance for each of the three transport experiments. In the figure, $\bar{v}_{eff}$ represents the ensemble average of lhe fitted velocities derived from all measured BTC's during a particular experiment. We emphasize again that the use of an effective pore-water velocities, v_{eff}, assumes that the medium between the inlet and the

TABLE II. Estimated effective average pore-water velocities, v_{eff}, dispersion coefficients, D_{eff}, and coefficients of determination, r^2, for the three transport experiments

Distance, cm	Tracer injection in homogeneous sandy column			Tracer injection in heterogeueous column (coarse → fine)			Leaching from heterogeneous column (fine → coarse)		
	v_{eff} cm hr^{-1}	D_{eff} cm^2 hr^{-1}	r^2 –	v_{eff} cm hr^{-1}	D_{eff} cm^2 hr^{-1}	r^2 –	v_{eff} cm hr^{-1}	D_{eff} cm^2 hr^{-1}	r^2 –
50	34.92	3.1	0.983				16.3	482	0.953
100	35.76	3.3	0.985	67.1	185	0.984			
150	36.54	5.6	0.985				8.2	751	0.927
200	3G.48	8.0	0.992	67.7	867	0.937			
250	35.92	14.1	0.995				19.3	4743	0.980
300	36.08	18.6	0.994	63.2	807	0.950			
350	35.83	20.2	0.996	64.6	1444	0.909	24.1	2348	0.972
400	35.76	27.1	0.996	62.7	1405	0.929			
450	35.42	31.5	0.997				25.5	1676	0.970
500	35.28	39.3	0.998	69.4	1394	0.936			
550	35.56	48.3	0.997				22.3	3338	0.960
600	39.14	52.6	0.997	68.8	5075	0.920			
650	34.24	73.7	0.990				29.0	6793	0.926
700	34.00	77.0	0.993	49.7	11180	0.984			
750	34.15	104.8	0.992				20.7	4683	0.968
800	33.83	122.2	0.997	66.3	10860	0.956			
850	33.1 I	128.8	0.998				20.9	3865	0.970
900	34.19	108.3	0.998	60.1	6084	0.97s			
950	33.70	122.1	0.998				20.9	3852	0.970
1000	33.44	139.3	0.993	54.1	5800	0.980			
1050	33.50	139.8	0.998				20.8	3693	0.950
1100	33.03	120.0	0.99s	53.8	7174	0.983			
1150	33.20	162.5	0.996				20.9	3293	0.943
I200	33.63	165.6	0.992	53.2	6297	0.963			
1250							20.2	3780	0.940

observation point is homogeneous so that the macroscopic transport equation (3) can be invoked. Still, the fitted v_{eff} should be different from one point to another since the nonhomogeneities in the columns change with the transport distance or measurement scale. Notice that v_{eff} for the homogeneous sandy column is almost constant with distance. This is not surprising since the sand used for this column had a relatively uniform particle size, whereas also considerable care was lakerl lo pack lhe column as homogeneously as possible. Table II lists the simultaneously deterrnined values of v_{eff} and D_{eff}. We also estimated the dispersion coefficient for each BTC by assuming a fixed value for the pore-water velocity as calculated from $v = q/n$ in which q is lhe measured Darcian flux (Table I) and n the porosily (0.33)

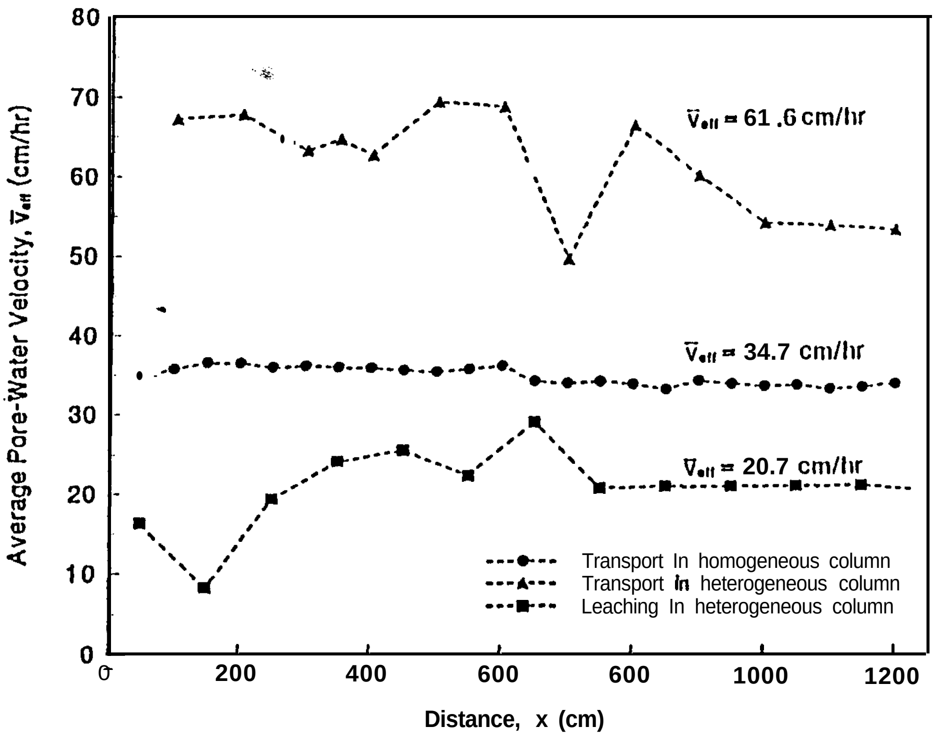


Fig. 5. Fitted effective pore-water velocity versus distance for the three transport experiments. The parameter $\bar{v}_{eff}$ represents the ensemble average velocity.

of the homogeneous column. The fitted dispersion coefficients thus obtained for this column (results not further shown here) were found to be very close to those listed in Table II for the simultaneous fit.

Contrary to the homogeneous column, values of v_{eff} for the tracer injection and leaching experiments in the heterogeneous column varied quite dramatically with distance. The results in Figure 5 show a slowly increasing or decreasing trend with distance, depending on whether flow took place from the coarse-tcxlurcd (left) side to the line-textured (right) side in Figure lb, or vice versa. Although the fitted v_{eff}-values represent average flow velocities from the inlet position lo the measurement location, some dramatic variations are apparent along the column. These variations in v_{eff} can be related directly lo lhe heterogeneities embedded in lhe columns. For example, the significant changes in the flow velocity at approximately x = 1.5 and 6.45 m for the leaching experiment (see Figures 1 b and 5; lhc coordinate in Figure lb runs now from right to left) must have been a consequence of the locally very heterogeneous nature of the medium. The relatively coarse background material in these two parts of the column contained several clay lenses and other mixtures with widely different hydraulic propcrtics. The resulting heterogeneous makeup of the medium causes the flow regime to become two- or three-dimensional, with concomitant variations in v_{eff}.

The effects of heterogeneity on the transport process can also bc demonstrated by considering the effective porosity given by $\epsilon = q/\bar{v}_{eff}$, where q is the imposed water flux, and $\bar{v}_{eff}$ is thc ensemble average of fitted pore-water velocities estimated frotn the BTC's at all tneasuring points (see Table II). Table I summarizes the calculated effective porosities for the three tracer experiments. As mentioned before, the measured porosity n of the individual soils used in the experiments was approxitnately 0.45 for the clay, attd 0.33 for most of the other soil materials, including the sand used for tltc homogeneous column. Given these values and the amounts of each soil type used in the two columns, we estimate the average n to be about 0.33 for the homogeneous sandy soil column, and 0.37 for the heterogeneous soil colutnn. Tlte effective porosity (ϵ = 0.33) was the same as the independently measured value of the porosity, n, for the homogeneous column. In contrast, the calculated effective porosities for the heterogeneous column (ϵ was 0.233 and 0.238 for the injection and leaching experiments, respectively) were much smaller that the estintated porosity (n = 0.37) for the heterogeneous column. The reduced effective porosity for the heterogeneous column was probably caused by mixing of soil materials of significantly different particle sizes during packing, clogging of large pores by relative small particles, and the likely presence of relatively immobile water in low-permeability clay lenses.

Figure 6 contpares typical observed and fitted breakthrough curves for the three transport experiments. The agreement between the observed and fitted curves for the homogeneous column (Figure 6a) was excellent for all BTC's, with most r^2 values exceeding 0.99 as indicated in Table II. The excellent description of the data indicates the general applicability of the CDE equation to transport in the homogeneous column. By comparison, relatively poor description of the observed BTC's were obtained for the heterogeneous column (Figures 6b, c), except in Figure 6b at relatively short travel distances (x <3 m). The descriptions were especially poor at later times when tailing was most pronounced. The discrepancies between the observed and fitted BTC's for the heterogeneous column are an obvious consequence of textural heterogeneities both along and perpendicular to the macroscopic flow direction. These heterogeneities combine to produce preferential flow and related 'physical nonequilibrium effects as discussed by van Genuchten and Cleary (1979) attd Brusseau et al. (1989). among others. Physical nonequilibrium is often modeled using a dual-porosity (or two-region) approaclt in which the medium is partitioned into distinct mobile and immobile (stagnant or non-moving) liquid regions (e.g., Coats and Smith, 1964; van Genuchten and Cleary, 1979). The effects of physical nonequilibrium are cumulative during transport and can lead to considerable tailing as the transport process proceeds in time and space.

As discussed before, most of the breakthrough curves for the homogeneous colutnn ltad signtoidal shapes. However, several BTC's in the homogeneous column also sltowed sonte effects of preferential flow. Figure 7a presents the measured BTC's between x = 7.5 and 9.0m. Notice that the BTC's at x = 8.5 m and 9.0 m are very close and even crossed each otltcr at approximately t = 30 hr. This fact

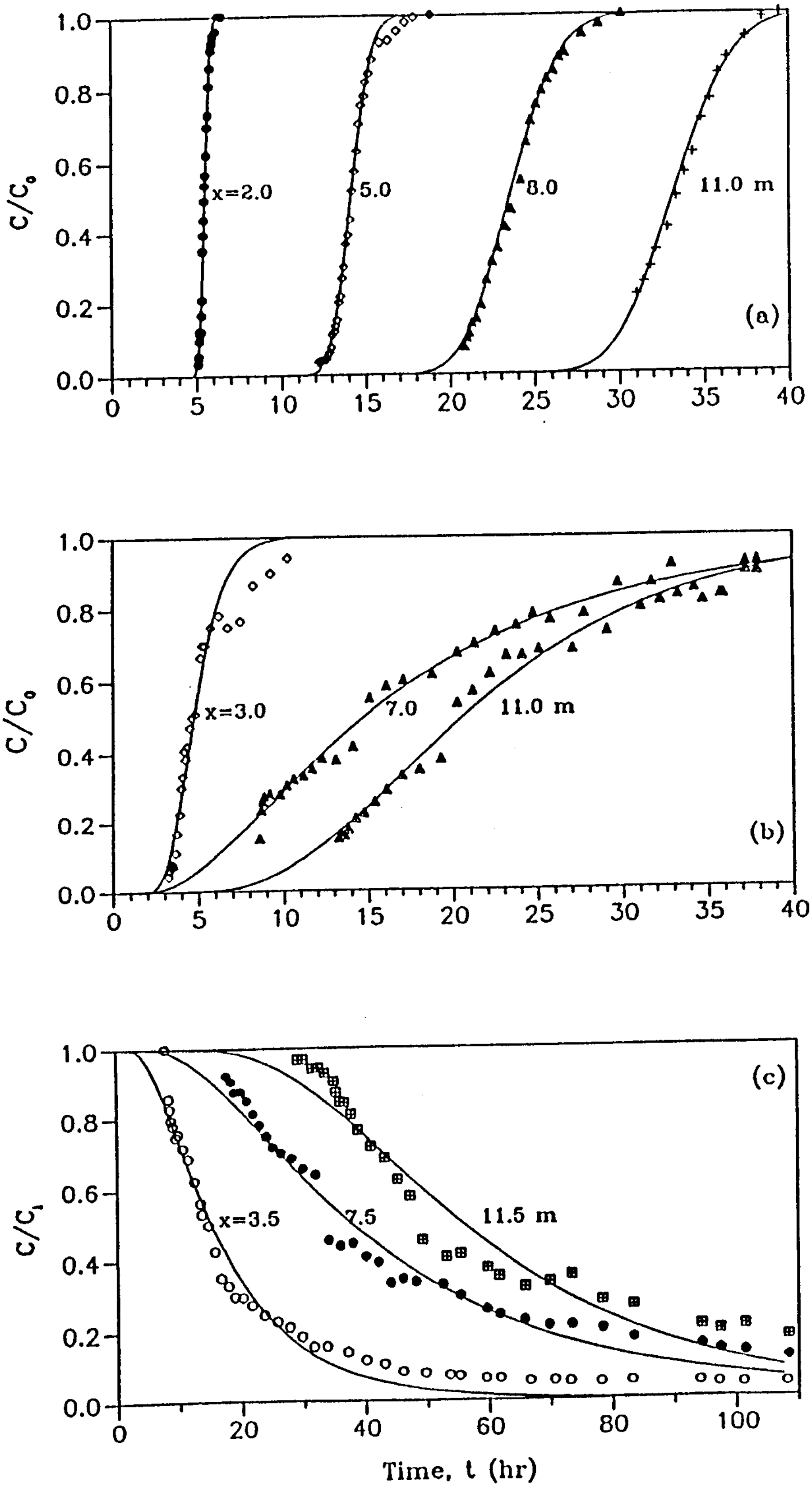


Fig. 6. Observed (symbols) and fitted (solid lines) concentration distributions at several locations in the columns for (a) tracer injection in the homogeneous sandy column, (b) tracer injection in the heterogeneous column, and (c) solute leaching from the heterogeneous soil column. Numbers near the curves indicate distances from the inlet position.

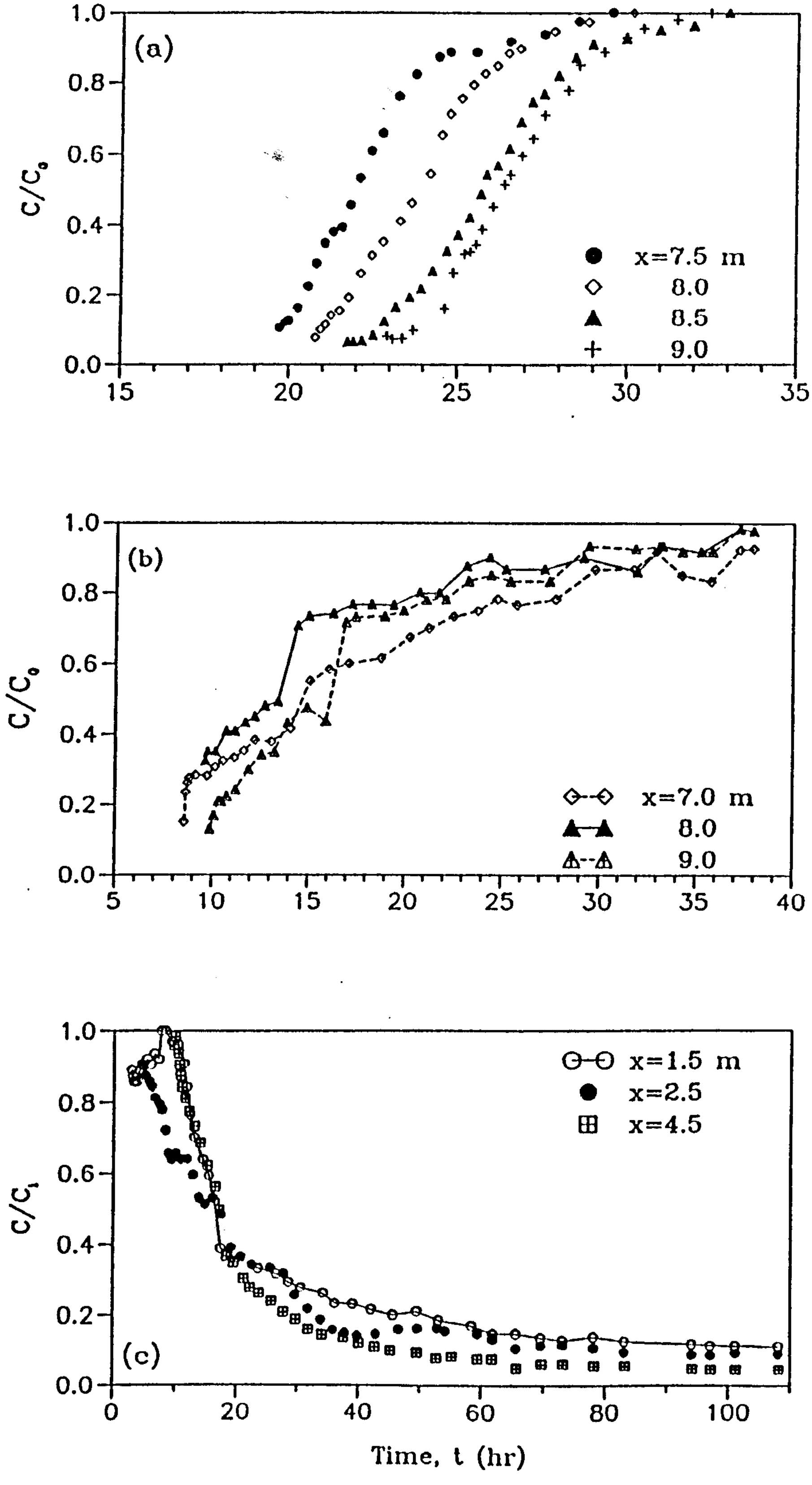


Fig. 7. Selected breakthrough curves showing evidence of preferential flow during (a) tracer injection in the homogeneous sandy column, (b) solute injection in the heterogeneous column, and (c) solute leaching from the heterogeneous soil column. Numbers near the curves indicate distances from the inlet position.

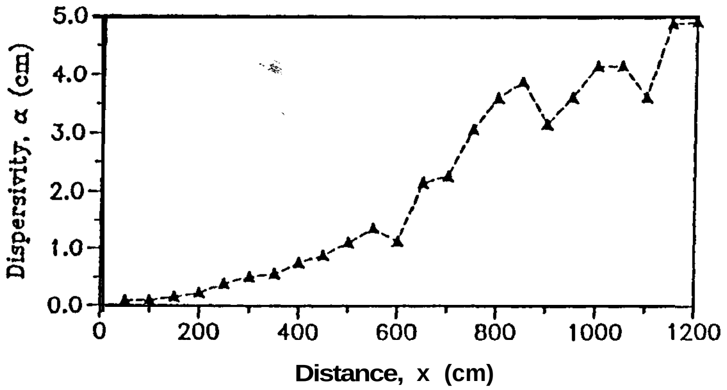


Fig. 8. Calculated dispersivilies as estimated from the filled effective dispersion coefficients and velocities for the homogeneous sandy soil column.

implies the presence of some preferential flow between x = 8.5 and 9.0 m. Similar observations of preferential flow in large uniformly packed soil columns were also made by Porro et al. (1993). in their case for vertical transport. These results show that, in practice, it is very difficult to achieve complete homogeneity when packing relatively large soil columns. For the same reason it is also difficult to imagine the existence of a truly homogeneous aquifer formed in the field by natural forces. Hence, the term 'homogeneous aquifer' as commonly used in hydrogeoiogical studies does not necessarily imply a uniform transport process consistent with such classical theories as the CDE model.

Much more irregular breakthrough curves were obtained for the heterogeneous column. Figures 7b and 7c present several cases where the observed BTC's at different locations cross each other. For example, Figure 7b pertaining to the injection scenario shows that the concentrations at x = 7.0 m were always lower than those at x = 8.0 m, and also lower than those at x = 9.0 m for t > 17 hr. For the leaching experiment (Figure 7c), the concentrations at x = 4.5 m decreased much faster than those at x =2.5 and 1.5 m for t > 15 hr. Note also that the BTC at x = 1.5 m in Figure 7c at first increased before decreasing in response to the imposed leaching process. The above irregularities and tailing phenomena are a result of the nonuniform velocity distribution caused by the heterogeneous nature of the medium, i.e., the presence of clay lenses and various types of sand, silt and clay mixtures in different parts of the column. We also note here that the EC-measured concentrations represent only small sampling volumes in the column; hence, the observed BTC may not accurately reflect average concentrations, (C), over each cross-section as presumed **in the data analysis based on equation (3).** This failure **of** the EC measurements lo represent average concentration undoubtedly contributed to the irregular distributions observed for the heterogeneous column.

Figure 8 shows a plot of the apparent dispersivity, α**, versus distance in** the homogeneous sandy column. Values of α range from 0.1 to 5.0 cm. These values are the same or slightly higher than dispersivities usually observed for transport in

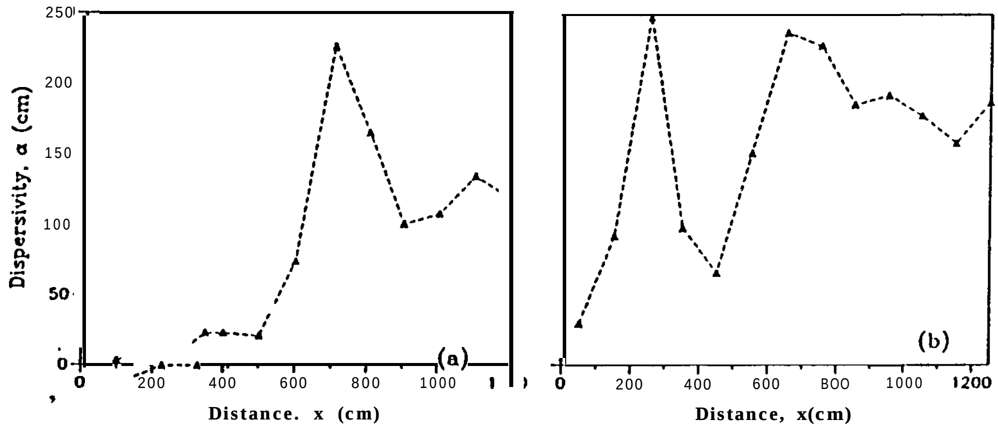


Fig. 9. Calculated dispersivities as estimated from the fitted effective dispersion coefficients and velocities for the injection (a) and leaching (b) experiments in the hclcrogcncous soil column.

relatively small, homogeneous laboratory soil columns. Notice that the dispersivity increased with the measurement scale, especially between x = 6 and 8 m where we previously noted the presence of some preferential flow. The marked increase in α near x = 7 m suggests that preferential flow may play an important part in the development of a scale-dependent dispersion process. An increase in the dispersivity with the measurement scale during transport in a large unsaturated, uniformly packed soil column (6.0 m deep by 0.95 m diameter) was also found by Porro et al. (1993). The dispersivity in their study increased from 2.2 cm at a depth of 82 cm to 7.8 cm at 400 cm.

In marked contrast to the homogeneous column, the dispersivities for the heterogeneous column were much higher and fluctuated greatly, ranging between 3 and 130cm for the injection experiment (Figure 9a), and between 20 and 200cm for the leaching experiment (Figure 9b). The dispersivities for both experiments also exhibit scale-dependency as was the case for the sandy column. The fluctuations in α as shown in Figure 9 appear to be the result of the particular makeup of the heterogeneities in the column. For example, α increases significantly when the heterogeneities appear in the form of nearly horizontal lenses more or less parallel to the flow direction. Solute spreading in those instances is enhanced because of incomplete lateral solute mixing over the entire cross-sectional area of the column (Jury and Fluhler, 1992). On the other hand, the presence of layers perpendicular to the flow direction, or of relatively homogeneous parts in an otherwise heterogeneous column, has a decreasing effect on α (e.g., compare Figures 1 b and 9a at x = 9 m). This last feature is consistent with observations by Parker and Albrecht (1987) and Porro et al. (1993) that layering perpendicular to the transport direction may actually decrease apparent dispersion (or even lead to sharpening of a solute BTC). Finally, we note from Figures 9a, b that, at the same travel distance, the dispersivity for the leaching experiment (starting from fine-textured end of the heterogeneous column) is generally higher than α for the injection experiment (starting from the coarse-textured end). While some effect of the flow direction on the dispersivity is apparent and intuitively logical, the results in Figure 9 do not relate in an obvious manner with the physical makeup of the heterogeneous column (Figure 1b) to permit more definite statements.

5. Conclusions

Tracer experiments were conducted in 12.5-m long homogeneous and heterogeneous saturated soil columns during steady confined flow. Three experiments were carried out: (1) transport of a NaCl solution into a homogeneous sandy column, (2) transport into a hclcrogcneous column containing a variety of materials whose particle size, on average decreased from very coarse at the inlet position lo very fine at the outlet position, and (3) leaching of solute from the same heterogeneous column, but with flow now in the opposite direction. Effective average pore-waler velocities and dispersion coefficients were determined simultaneously by fitting the macroscopic CDE to breakthrough curves observed at regular intervals along the horizontally placed columns.

The observed BTC's for the homogeneous sandy column were relatively smooth and sigmoidal, although a few curves showed some tailing and/or crossing of BTC's, thus indicating the presence of some preferential flow in the homogeneous column. Excellent agreement was obtained between the observed and fitted CDE curves for this column. The dispersivity of the homogeneous column increased with the measurement scale from 0.1 to 0.5 cm, whereas the average pore-water velocity was essentially constant throughout the column. These results suggest that some preferential flow and scale-dependent dispersion are likely to occur in most groundwater systems, even if seemingly homogeneous.

As opposed to the homogeneous column, BTC's for the heterogeneous column were more irregular, nonsigmoidal, and exhibited extensive tailing and crossing of different curves. The observed BTC's for this column were in most cases poorly described with the CDE model. Fitted effective average pore-waler velocities varied rather erratically with distance, while the dispersivities (varying between 3 and about 200 cm) were much higher than those for the homogeneous sandy column. Dispersivities for the tracer injection and leaching experiments in the heterogeneous column also exhibited a scale-dependency, similarly as for transport in the sandy column. Estimated dispersivities for especially the heterogeneous column were found to be much higher than those commonly obtained for relatively short laboratory soil columns. Spatial variations in the dispcrsivity could be explained qualitatively by the particular makeup of the heterogeneities in the column. For example, the dispersivity generally increased with distance in regions where lhe sand layers and clay lenses were situated approximately parallel to the flow direction, but decreased (leading to BTC front sharpening) in regions where the layers were mostly perpendicular to the flow direction.

Acknowledgements

This work was supported in part by the National Science Foundation of China under grant **NSFC-0488012,** 1989. The assistance in the experimental work by Zhen-Guang Huang and Si-Hong Liu of the Department of Geology and Mining Engineering, Fuzhou University, China is sincerely appreciated.

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