

Solute Transport Through Small and Large Unsaturated Soil Columns^a

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ABSTRACT

Unsaturated solute transport experiments were conducted using several small and one large column packed with the same sandy soil material.

Chloride and tritium were used as tracers for water movement. Soil solution samples were collected as a function of time in the small columns and versus time and depth in the large column. The data were compared with solutions of the classical two-parameter convection-dispersion equation and also with solutions of a more elaborate four-parameter transport model that assumes a fraction of the water to be immobile. Fairly symmetrical effluent curves resulted from both the small and large columns, suggesting small transfer coefficients between mobile and immobile regions and small amounts of immobile water. Dispersivity values ranged from 1 cm for the small columns to about 5 cm for the 6-m-long column; retardation factors for chloride were nearly the same (about 0.8) for both column sizes. Thus, while the dispersivity measured in the small columns did not apply to the large columns, the same retardation factors could be applied to the different scale columns.

INTRODUCTION

The movement of surface-applied chemicals in soil and the removal of undesired salts by leaching from soil profiles has been of interest to engineers and soil scientists for many years. In commercial agriculture, large quantities of fertilizer and pesticides are applied on or incorporated into the soil.

With the infiltration of rain or irrigation water, these chemicals move into the root zone for plant uptake. However, when transported too far down into the profile, they cease to be available for plant uptake and become a potential threat to the quality of underlying ground-water systems. The intentional or accidental release of chemical wastes on soils has further stimulated current interest in the movement of chemicals through soils.

The need to know the fate of surface-applied chemicals has, over the years, resulted in the development of various models of solute transport. Many of these models had their origin in chemistry or chemical engineering (Lapidus and Amundson, 1952; Glueckauf, 1949), but were later applied to soils (Day and Forsythe, 1957; Nielsen and Biggar, 1962). One of the most commonly used equations for describing one-dimensional solute transport is the convection-dispersion equation (CDE):

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} \quad (1)$$

where C is the concentration; t is time; x is distance; R is the retardation factor; D is the dispersion coefficient; and v is the average pore-water velocity (defined by q/θ , where q is the Darcian fluid flux density, and θ is the volumetric water content). The dispersion coefficient, D , describes the effects of molecular diffusion and hydrodynamic dispersion on transport.

For relatively homogeneous systems, D is approximately linearly related to the pore-water velocity, v . The correlation coefficient, ϵ , is referred to as dispersivity (or dispersion length). For ground-water systems where flow is largely parallel to soil layers, the value of ϵ has been found to increase with distance from the source (e.g., Gelhar *et al.*, 1985). This increase in dispersivity is thought to be

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a result of variations in the hydraulic conductivity of the aquifer and has been related to the variance in the hydraulic conductivity (Gelhar *et al.*, 1979). In the unsaturated zone, transport is normally across the layering. Instability in the wetting front as a result of soil layering could also cause an apparent increase in dispersivity with distance. However, this has not been documented, largely because the number of studies dealing with solute transport in relatively deep unsaturated geologic profiles is limited. This study was motivated in part by this lack of unsaturated transport studies.

The retardation factor, R , accounts for interactions between the solute and the soil matrix. For linear, equilibrium adsorption, R is given by

$$R = 1 + \rho K/\theta \quad (2)$$

where ρ is the soil's bulk density, and K is a distribution coefficient. If there is no interaction between the solute and the soil, K becomes zero and R reduces to one. In some cases R becomes less than one, indicating that only a fraction of the liquid phase participates in the transport process (van Genuchten and Wierenga, 1986). This may be the case when the chemical is subject to anion exclusion or when relatively immobile water regions are present that do not participate in convective transport. Anion exclusion results from repulsion of anions from negatively charged surfaces associated with clays and ionizable organic matter (Bolt, 1979; James and Rubin, 1986). The concentration of the anion as a function of the distance from the charged surface is often approximated by a step function with zero anions within the exclusion volume, and an anion concentration equal to the bulk solution outside the exclusion volume. It can be shown that the ratio of the exclusion volume, θ_{ex} , to the volumetric water content, θ , is equal to $1-R$. Thus, $1-R$ may be viewed as the relative anion exclusion volume. Anion exclusion has been observed for many soils (Corey *et al.*, 1963; Thomas and Swoboda, 1970; Dyer, 1965; Bond *et al.*, 1982) and was accounted for in models by Krupp *et al.* (1972), Bresler (1973), White *et al.* (1984), and several others.

Analytical and numerical solutions of equation (1) are available to estimate solute concentration as a function of time and soil depth. These solutions have been tested with moderate success in the laboratory, but field studies frequently reveal large discrepancies between measured and computed distributions. Reasons for the discrepancies include: (1) lack of equilibrium in prevailing adsorption/desorption processes (Rao *et al.*, 1979);

(2) extensive temporal and spatial (horizontal and vertical) variability in soil physical properties, fluxes, and water contents (Biggar and Nielsen, 1976; van der Pol *et al.*, 1977; Jury *et al.*, 1982); (3) water and solute movement along preferential pathways and nonuniform mixing with the soil matrix (Quisenberry and Phillips, 1978); and (4) the existence of liquid-filled, dead-end pores or immobile (nonmoving) water (Coats and Smith, 1964).

One way to account for preferential transport and immobile water is to partition the liquid phase of the soil into mobile and immobile regions, to limit convective-dispersive transport to the mobile liquid region, and to assume that diffusion is responsible for the exchange of solute between the mobile and immobile regions. Approximating this diffusional exchange with a first-order rate equation results in (van Genuchten and Wierenga, 1976):

$$\theta_m R_m \frac{\partial C_m}{\partial t} + \theta_{im} R_{im} \frac{\partial C_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 C_m}{\partial x^2} - \theta_m v_m \frac{\partial C_m}{\partial x} \quad (3a)$$

$$\theta_{im} R_{im} \frac{\partial C_{im}}{\partial t} = \alpha (C_m - C_{im}) \quad (3b)$$

where θ_m and θ_{im} are the mobile and immobile water contents ($\theta_m + \theta_{im} = \theta$); C_m and C_{im} are the solute concentrations in the mobile and immobile liquid phases; v_m is the average mobile phase pore-water velocity ($v_m = q/\theta_m$); D_m is the mobile phase dispersion coefficient; and α is a mass transfer coefficient. The retardation factors R_m and R_{im} account for adsorption in the mobile and immobile regions, respectively. The two retardation factors become identical if sorption sites in the soil are distributed between the mobile and immobile regions in the same ratio as the mobile and immobile water contents. In that case (Nkedi-Kizza *et al.*, 1983):

$$R_m = R_{im} = R. \quad (4)$$

The objective of the present study was to determine how well parameter values in equations (1) and (3a, b), determined from short (0.3 m) column experiments, apply to long (6 m) columns filled with the same soil material. Such studies are necessary, because experiments are often performed in the laboratory using short columns. The results of these studies are then applied to field conditions where travel times are much longer. Few studies

have been done to justify such a scaling up from short to very long columns.

A second objective was to investigate the spatial dependence of the dispersivity measured in the long column.

METHODS AND MATERIALS

The general procedure for column studies is to apply water containing a tracer part of the time to vertical, unsaturated soil columns, then measure and analyze the tracer concentration in effluent collected from those columns. By comparing measured tracer concentrations in the column effluent with those computed on the basis of equations (1) or (3a, b), values of the different parameters in the two transport models can be determined. This procedure was followed here, both for the laboratory soil columns and for the larger field lysimeter.

For the smaller laboratory experiments, we used plexiglass columns, 5.1 cm in diameter and 30 cm long (Soil Measurement Systems, 7344 N. Oracle Rd., Tucson, AZ 85704) uniformly filled with Berino loamy fine sand (classified as Typic Haplargid fine loamy, mixed thermic). The soil material was air-dried and passed through a 1-mm sieve. The lower ends of the columns were closed with porous, stainless steel plates that had bubbling pressures of about 140 cm H₂O. The porous plates were held in plexiglass end plates with 0.2-cm clearance between the stainless steel and the plexiglass plates near the center and zero clearance near the edges of the plates. Two tensiometers were mounted on the side of each column, one 5 cm below the soil surface and one 5 cm above the bottom plate. The columns were positioned vertically on top of, and connected to, a vacuum chamber enclosing an automatic fraction collector (ISCO, Lincoln, NE). The suction in the vacuum chamber was adjusted until the pressure heads in the two tensiometers were essentially equal. Suctions were measured with a handheld pressure transducer with digital readout (Marthaler *et al.*, 1983). Equal suction implies unit gradient conditions and uniform unsaturated water contents with depth in the column.

The columns were leached with 0.05 M Ca(NO₃)₂ at a steady flow rate using precision syringe pumps (Soil Measurement Systems, 7344 N. Oracle Rd., Tucson, AZ 85704). When the outflow rate equaled the application rate, the inflow solution was changed to 0.005 M CaCl₂ + 0.2 μCi/ml of ³H₂O. After adding one pore volume of the radio-labeled solution, the input solution was switched

back to the original calcium nitrate solution. Effluent solution was collected in a fraction collector within the vacuum chamber. The chloride concentration in the effluent was determined amperometrically (Cotlove, 1963) with a Buchler chloridometer (Buchler Instruments, Inc., Fort Lee, NJ), while the tritium activity was determined by liquid scintillation counting with appropriate background and quench correction.

Experiments were conducted at one bulk density ($\rho = 1.57 \text{ g/cm}^3$) and four flow velocities (Table 1). Water contents varied slightly between the four experiments. The effective soil-filled column length, L , in each case was 28.7 cm. The multiparameter optimization program of Parker and van Genuchten (1984) was used to determine the parameters D and R in equation (1), and the parameters D_m , R , $\phi (= \theta_m/\theta)$, and α in equations (3a, b).

The large column experiment was conducted in a 6-m-long column constructed from 0.945-m ID galvanized highway culverts. The column was part of a larger facility containing four 0.945-m ID and two 2.44-m ID columns around a central access culvert (Wierenga *et al.*, 1986). The column was filled with the same Berino loamy fine sand used in the laboratory experiments. After settling, the soil density (1.48 g/cm^3) was only slightly less than the 1.57 g/cm^3 measured for the smaller soil columns. Three suction candles 29.2 cm long by 3.5 cm OD (Soil Moisture Equipment Corp., Santa Barbara, CA) were installed 10 cm above the bottom of the column (Figure 1). A constant suction of 60-cm H₂O maintained on the suction candles removed excess drainage water and kept the soil unsaturated. Additional suction samplers were installed at 0.82, 1.25, 2.20, 3.1, 4.0, and 5.0 m below the soil surface. Tensiometers were installed horizontally through the access ports of the column. Pressure heads were determined with a handheld pressure transducer, as in the laboratory study. During filling of the column, a vertical 0.05-m ID aluminum neutron probe access tube was installed in the center of the column. Water contents were measured at 20-cm depth intervals along the length of the column with a neutron probe (CPN Corporation, Martinez, CA). The neutron probe was calibrated in situ by taking gravimetric samples at various times during the experiment. The column was irrigated at a constant rate of 1.91 cm/day with 0.005 M Ca(NO₃)₂ solution using a rotating applicator with 48 syringe needles. The applicator applied water and tracer uniformly over the surface (Wierenga *et al.*, 1986).

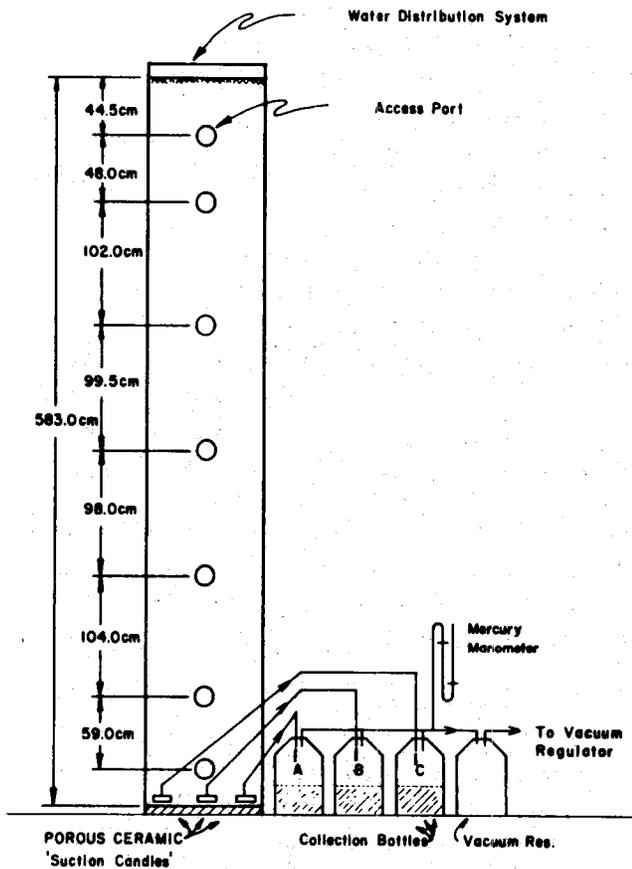


Fig. 1. Schematic of large column with water collection system near the bottom of the lysimeter.

The normal irrigation solution was replaced by a 0.005 M CaCl_2 solution for 10 days. During and after this period, the soil solution was sampled, initially twice a day, and later at one- or two-day intervals.

RESULTS AND DISCUSSION

Small Column Experiments

Table 1 summarizes the experimental parameters and values for D , R , and ϵ of the four leaching experiments conducted in the laboratory.

Table 1. Soil Physical Parameters for the Tritiated Water and Chloride Displacement Experiments Through Small Laboratory Soil Columns

Exp. #	Water content $\theta, \text{cm}^3/\text{cm}^3$	Pore-water velocity $v, \text{cm}/\text{day}$	Pulse time*, T_1	Dispersion coefficient $D, \text{cm}^2/\text{day}$		Dispersivity ϵ, cm		Retardation factor, R	
				^3H	Cl	^3H	Cl	^3H	Cl
1	0.197	4.21	1.126	2.8	3.9	0.66	0.94	0.89	0.66
2	0.209	10.76	1.165	6.9	8.3	0.64	0.77	0.90	0.76
3	0.211	28.4	0.848	26.9	25.3	0.95	0.89	0.97	0.83
4	0.235	53.3	1.084	50.2	46.2	0.94	0.87	0.99	0.88
Mean	—	—	—	—	—	0.80	0.87	0.94	0.78

* In pore volumes ($T_1 = vt_1/L$, where T_1 is the applied pulse length).

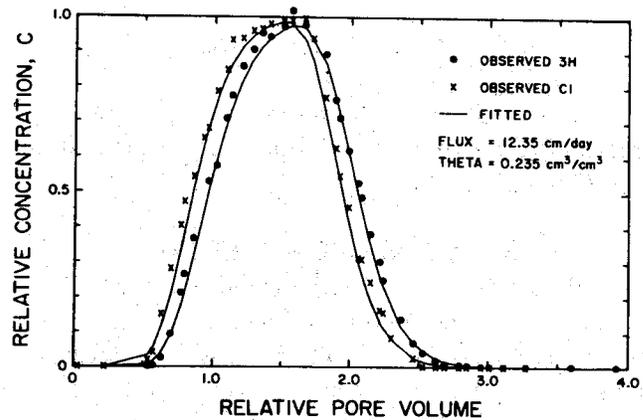


Fig. 2. Observed and calculated chloride and tritium distributions from leaching at a flux of 12.35 cm/day. The solid lines were computed with the one-region model [equation (1)].

Observed and calculated effluent curves for experiment #4 are shown in Figure 2. Data for the other column studies were very similar. In each case the symmetrically shaped experimental curves agreed closely with curves calculated with equation (1).

Table 1 shows that while the dispersion coefficient is an increasing function of the pore-water velocity, the dispersivity is not much affected by the velocity. A plot of D versus v yields an approximately straight line with a slope equal to the mean dispersivity. An average value of 0.84 was obtained for the four displacement experiments involving two tracers each.

The retardation factor, R , of chloride was less than that of tritium (Figure 2, Table 1). This difference in retardation, which averaged 0.16 pore volumes for the four experiments, is most likely the result of anion exclusion. The clay and organic matter contents of the soil material used in this study were determined by pipette analysis to be only 4.7 and <1%, respectively. Thus, even though clay content and organic matter content,

the main contributors to anion exclusion (Bolt, 1979), are relatively small, there is significant anion exclusion indicating that chloride (and possibly also bromide) is not an ideal tracer for this unsaturated sandy soil.

Note that the retardation factor for tritium is generally less than one (Table 1). Values for the retardation factor were obtained by fitting a solution of equation (1) to the column effluent data. This procedure implies that equation (1) is valid, and that all water in the columns participates equally in the transport process. This is not necessarily the case (van Genuchten and Wierenga, 1976), resulting in values of R different than one.

The laboratory column effluent data were also analyzed with the two-region (mobile-immobile) transport model given by equations (3a, b). Agreement between observed and calculated curves of experiments #1 and #2 was only slightly better than that obtained with equation (1). A better match is expected because two additional parameters (ϕ and α) were included in the optimization process. A nearly constant immobile water content of about 4% by volume was obtained. The estimated mass transfer coefficients were small (generally less than 0.1 l/day), indicating relatively little diffusion into and out of immobile liquid regions during the residence time in the column. Hence, there were only minor improvements in the calculated curves based on equations (3a, b), as compared to those based on equation (1).

Large Column Experiment

The results of the large column experiment are presented in Figures 3 through 6. Figure 3 shows the chloride concentration distributions for the 0.82, 1.25, and 2.2-m depths (upper half) and for the 3.1, 4.0, and 5.0-m depths (lower half). Note that the data are well-behaved; peak concentrations decrease, and breakthrough curves shift to the right with depth. One exception is the chloride peak concentration at 5.0-m depth, which shows a higher peak concentration than at 4.0 m or at 3.1 m. Such behavior is not expected. It could result from uneven lateral distribution of the solute during transport through the column or from differences in soil physical properties associated with uneven packing at this depth.

The solid lines in Figure 3 were calculated with equation (1) using parameters that were determined for each depth separately. The calculated chloride curves fit the observed data well, except for the tailing ends of the curves at the

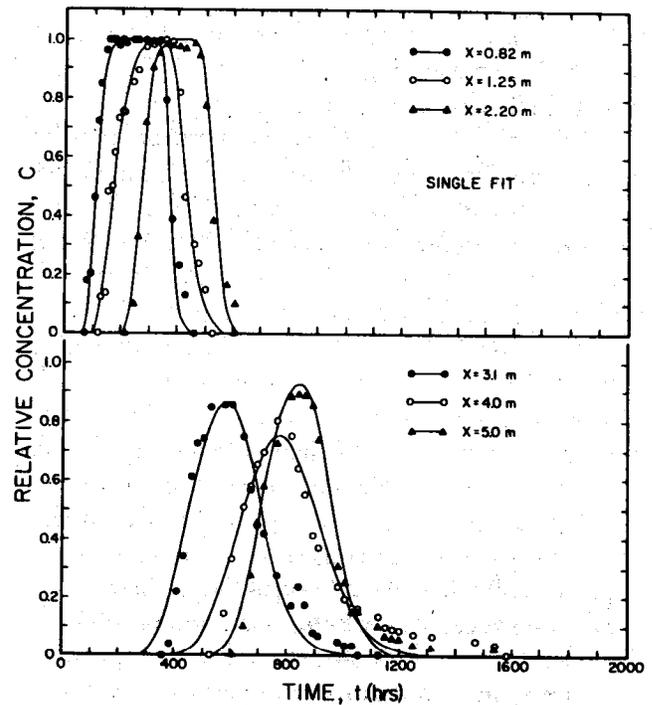


Fig. 3. Observed and fitted chloride concentration distributions. The solid lines were individually fit with the one-region model.

deeper depths. Table 2 lists the optimized parameter values for the dispersion coefficients and retardation factors for each of the six curves in Figure 3. The dispersion coefficient shows a tendency to increase with depth to 400 cm, whereas the retardation factor varies much less with depth. The average of the six fitted dispersion coefficients was 49 cm²/day, which is lower than the global value for D of 65 cm²/day obtained by fitting all six depths simultaneously. The average retardation factor of 0.77 was nearly identical to the global value of 0.78. Figure 4 presents the observed and fitted chloride distributions for all six depths using the global values for D and R . The fit between observed and predicted data is not as

Table 2. Values of the Dispersion Coefficient (D) and Retardation Factor (R) Obtained by Fitting Equation (1) to the Observed Chloride Concentration Distributions for Each Depth and For All Six Depths Simultaneously

x , cm	D , cm ² /day	R	r^2
82	25	0.74	0.98
125	53	0.76	0.97
220	21	0.66	0.98
320	79	0.80	0.95
400	82	0.88	0.95
500	34	0.76	0.98
82-500*	65	0.78	0.88

* Simultaneous fit at six depths.

good as for the individual fits (Figure 3), but is reasonable considering the size of the column and the difficulty of packing it uniformly.

The observed chloride concentration distributions were also analyzed with the two-region model given by equations (3a, b). Use of this model resulted in a significantly better description of the data, notably of the considerable tailing at the deeper depths. This is evidenced in part by the higher r^2 -values (Table 3) obtained by fitting equations (3a, b), as opposed to those obtained by fitting equation (1). While the individual (local) fits were much better, use of the two-region model did not materially improve the global fit using data from all six depths simultaneously. For example, the r^2 for the simultaneous fit only increased from 0.88 to 0.89 when the one-region equilibrium model was replaced with the two-region mobile-immobile model. A likely reason for this lack of improvement is the presence of a spatially variable velocity field in the column. Flow variability was probably caused by uneven packing, leading to spatial variations in local bulk densities, water contents, and hence, flow velocities. Flow heterogeneity was evidenced by significant differences in first arrival times of the chloride tracer at the three suction cups or candles located near the bottom of

Table 3. Values of the Parameters D , R , ϕ and α Obtained by Fitting Equations (3a, b) to the Observed Chloride Concentration Distributions for Each Depth, for Six Depths Simultaneously, and for the Lower Three Depths Simultaneously

x , cm	D , cm^2/day	R	ϕ	α , $10^{-6}/\text{day}$	r^2
82	16.0	0.73	0.95	11	0.98
125	1.0	0.80	0.79	26	0.99
220	10.0	0.68	0.94	3	0.99
310	11.0	0.84	0.85	8	0.98
400	11.0	0.95	0.84	7	0.99
500	11.0	0.79	0.93	3	0.99
82-500*	11.0	0.81	0.84	13	0.89
310-500**	17.0	0.85	0.88	6	0.88

* Simultaneous fit at six depths.

** Simultaneous fit at three depths.

the lysimeter (Figure 5). This variability may also have caused the shift to the right of the breakthrough curve at the 4-m depth, as compared to the calculated curve which typifies a homogeneous flow field. This shift is a first-order effect that masks most of the second-order tailing effects of mobile-immobile water in a least-squares parameter optimization analysis of the data.

In Figure 6, measured concentration distributions are compared with calculated curves using two-region model parameters obtained by simultaneously fitting the concentration distributions at only the three lower depths. The general shapes of the curves at the lower three depths are described better with the two-region than with the one-region model. However, the observed and calculated curves are displaced with respect to each other, because of presumed areal variations in flow velocity across the column.

The α -values obtained by fitting equations (3a, b) to the large column data are extremely

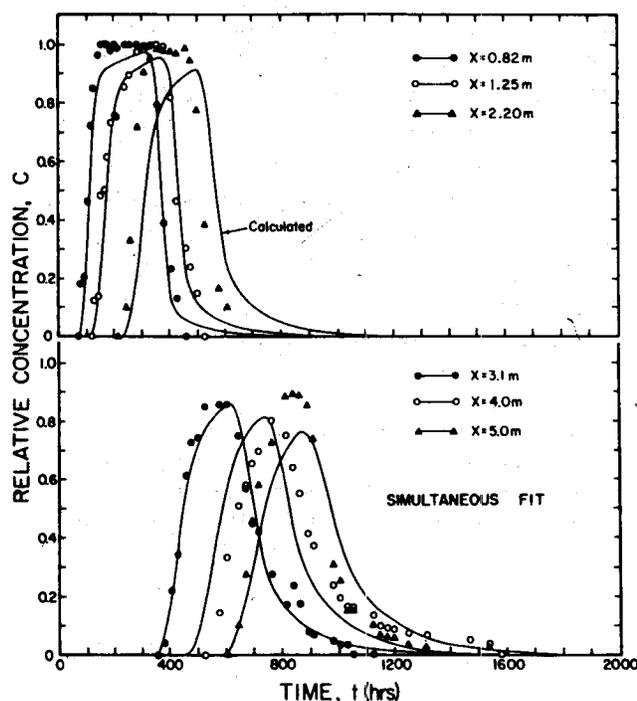


Fig. 4. Observed and computed chloride concentration distributions for the 6-m deep lysimeter. The solid lines were calculated with D and R values obtained by fitting all six measured chloride concentration distributions simultaneously.

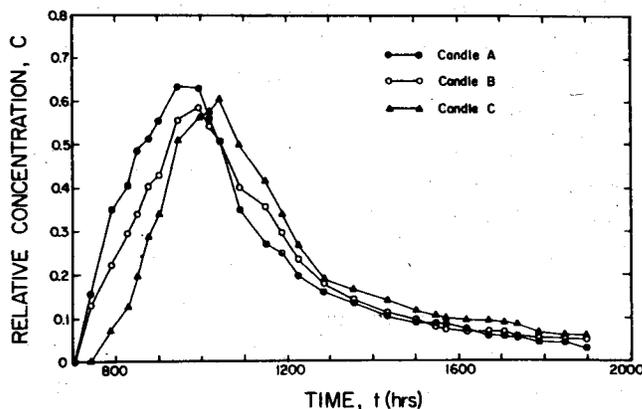


Fig. 5. Observed chloride breakthrough curves for the effluent from the 6-m lysimeter.

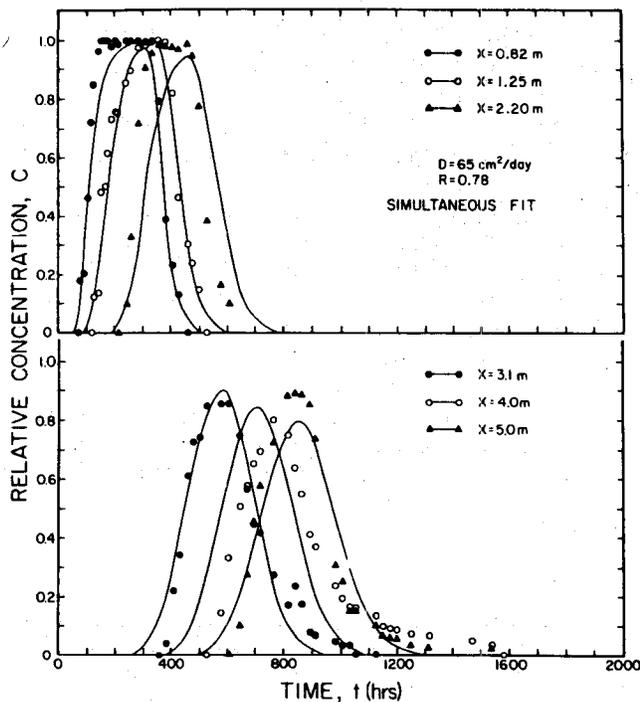


Fig. 6. Observed and fitted chloride concentration distributions. The solid lines were predicted with the two-region model using parameters derived from the simultaneous fit at 3.1, 4.0, and 5.0 m.

small (Table 3), suggesting slow exchange between mobile and immobile liquid regions. These small α -values are consistent with the small α -values (not shown) estimated from the symmetrical curves.

The dispersion coefficients computed with the two-region model (Table 3) are relatively independent of depth and much smaller than calculated with the equilibrium model. Whereas the average D value computed with the equilibrium model was $65 \text{ cm}^2/\text{day}$ (simultaneous fit at the six depths), the two-region model value was only $11 \text{ cm}^2/\text{day}$. Based on a water content of $0.161 \text{ cm}^3/\text{cm}^3$, a flux of $1.9 \text{ cm}/\text{day}$ and a pore-water velocity of $11.8 \text{ cm}/\text{day}$, the dispersivity ($\epsilon = D/v$) is 5.5 cm for the equilibrium model and 0.93 cm for the two-region model. Thus, the two-region model requires dispersivity values that are typical for small columns. In contrast, retardation factors calculated with the two-region model (0.81) are very similar to those estimated with the equilibrium model (0.78) using the experimental data from all six depths.

CONCLUSIONS

Comparing small column effluent concentration distributions with data from a large column showed greater D values for the large column. Dispersivity was about 5 cm in the large column but

only about 1 cm in the small column. There was no clear evidence of an increase in the dispersion coefficient with depth in the large column. While the dispersivity in the large column was about five times larger than in the small columns, values for the retardation factor were essentially the same. Anion exclusion resulted in retardation factors of around 0.8 for the small columns as well as for the large column. The data showed that anions such as chloride or bromide can move considerably faster than noninteracting tracers such as tritium. There was evidence of areal variability in the flow regime for the large column, probably a result of non-uniform packing and associated variability in the water content.

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