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Full Length Research Paper

Chloride transport in undisturbed soil columns of the loess Plateau

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In soils containing preferential flow paths, both water and solute can move preferentially, bypassing much of the soil matrix. The object of this study was to examine the effect of preferential solute transport in Changwu (loamy soil) soil and Ansai soil (sandy soil) containing macroporosity. Miscible displacement experiments were conducted with 5 undisturbed soil columns (19.45 cm diameter, 43.5 cm long). Breakthrough curves (BTC's) of Chloride were measured under water-saturated steady flow conditions. The data were simulated using three conceptual models. The results show that two-flow region model described the preferential solute transport much better than the two-region model and the convection dispersion equation (CDE), especially there were humps in the tailing side. Moreover, distinct double peaks were apparent with the increase of pore water velocity in a loamy soil column. In addition, high pore water velocity and small mass transfer coefficient between the two-flow regions enhanced the development of double BTC peaks.

Key words: Breakthrough curves, convection dispersion equation, two-region model, two-flow region model.

INTRODUCTION

Investigation of water and solute movement on the Loess Plateau have indicated that in soil containing macropores, both water and solutes can move preferentially through macropores, bypassing much of the soil matrix. Flow through macrospores can allow surface-applied agricultural chemicals to move rapidly through the root zone, posing problems of economic loss and deteriorating the wicked environment on the Loess Plateau. An understanding of the preferential solute transport is needed to predict the movement of chemicals and to improve techniques for managing the applications of such chemicals on the Loess Plateau.

Detection of preferential flow in soils is commonly achieved by applying a chemical tracer to the soil surface and measuring its concentration over time at a certain depth to produce a breakthrough curve (BTC) (Haws et al., 2004). Asymmetry and tailing of BTC's under water saturated conditions have been reported by Biggar and Nielsen (1962), DeSmedt (1979), Rao et al. (1980a) and Nkedi-Kizza et al. (1982). Furthermore, because of the heterogeneity and complexity of soil-solute interactions, mathematical models describing solute transport in soils are becoming conceptually intricate. In the earliest model, all the water was assumed to be mobile with physical equilibrium existing in the system. Later, the model was developed based on chemical and physical nonequilibrium into the two-region model (Skopp and Warrick, 1974) and two-site model (Selim et al., 1976). During the last 2 decades, emphasis has been focused on the two-region or mobile-immobile concept. In the model, soil water was partitioned into mobile and immobile regions; convective diffusive solute transport was limited to the mobile water region. Transfer between the two water regions was assumed to occur at a rate proportional to the difference in chloride concentration

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between the two-regions. Skopp (1981) developed a two-flow domain model based on the mobile-immobile concept, in which soil water was divided into two regions, but neither of the water regions have a zero flow rate, limited interactions between the two domains were assumed. The two-flow domain model, which may be referred to as a dual-porosity concept, has been used in solute transport modeling to account for contribution of large pores on preferential flow (Ma and Selim, 1995).

Although Skopp et al. (1981) proposed a two-flow region model to describe solute transport in soils, to our knowledge, only Ma (1995) fitted their experimental data with this method in three soil types. As a result, the objectives of this study were: (1) to study the BTC's in large undisturbed loamy and sandy soil columns and to (2) comparing the fitted BTC's with CDE, two-region model and two-flow region model and the parameters.

THEORY

Classical convection dispersion equation

The most commonly used model in solute transport is the classical convection dispersion equation (CDE), which is characterized by a convection term with an average pore water velocity v (cm/h) and a hydrodynamic dispersion coefficient D (cm^2/h):

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

Where C is solute transport concentration (μg/ml), t is time (h), x is the spatial coordinate (cm), and R is a retardation factor. Two-region model:

$$\theta_m \frac{\partial C_m}{\partial t} + \theta_{im}\frac{\partial C_{im}}{\partial t} = \theta_m D \frac{\partial^2 C_m}{\partial x^2} - v_m \theta_m \frac{\partial C_m}{\partial x} \quad (2)$$

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

$$\theta = \theta_m + \theta_{im} \quad (4)$$

where θ_m is the volumetric water content in the mobile region, (L^3 L^{-3}), θ_{im} is the volumetric water content in the immobile region, (L^3 L^{-3}); C_m is the concentration in the mobile region, (M L^{-3}); C_{im} is the concentration in the immobile region, (M L^{-3}); *D* is the dispersion coefficient, (L^2 T^{-1}); v_m is the pore-water velocity in the mobile region, (L T^{-1}); *t* is the flow time, (T); x is the flow distance, (L); and α is the dimensionless mass transfer coefficient, (T^{-1}).

Interpreting effluent concentrations to be flux-averaged concentrations, the assumed initial and boundary conditions for this study were:

$$C = 0 \text{ or } C_m = 0;\ 0 < x < l\ ,\ t = 0 \quad (5)$$

$$VC_0 = K \text{ or } V_m C_0 = -D_m \frac{\partial C_m}{\partial x} + V_m C_m;\ x = 0,\ t > 0 \quad (6)$$

$$\frac{\partial C}{\partial x} = 0 \text{ or } \frac{\partial C_m}{\partial x} = 0;\ x = l,\ t > 0 \quad (7)$$

Where C is the relative concentration in the soil column, C_m is the relative concentration in the soil column's mobile region, D and D_m are the dispersion coefficients of the soil column and of the mobile region, V and V_m (C, C_m) are the soil water velocities (solute concentrations) in the soil column and in the mobile region of the soil column, l is the length of the soil column (cm), t is the time. Using the derived values of the dispersion coefficient (*D* or D_m) from each of the fitted solute transport models, we calculated the dispersivity, λ, (L) using the following equation:

$$\lambda = D/v \quad (8)$$

Under saturated conditions, and for non-interacting solutes and relatively high rates of water flow, the dispersion coefficient, *D*, and the flow rate exhibit a linear relationship (DeSmet and Wierenga, 1984). At lower flow rates, *D* decreases with the flow rate in a non-linear fashion (Bear, 1972). This relationship is defined in a one-dimensional system in the form:

$$D = D_0\tau + \lambda v \quad (9)$$

Where, D_0 is the molecular diffusion coefficient, τ is the tortuousity, λ is dispersivity, and v is the pore-water velocity. Values of *D* were estimated by fitting Equation 2 to the solute breakthrough curve for each batch of leachate samples; λ was calculated from Equation 8; and if the term '$D_0\tau$' in Equation 9 was small enough with respect to D_0 it could be ignored (Schulin et al., 1987). To calculate the fraction of mobile soil solution, we used:

$$\phi = \theta_m / \theta = \beta R - f(R-1) \quad (10)$$

Table 1. Selected properties of the soil used in this study.

Soil	pH	Organic matter (g/kg)	Clay (%)	Silt (%)	Sand (%)
Changwu soil	8.2	11.2	32.5	21.2	46.3
Ansai soil	7.6	0.31	13.9	26.1	60.0

in which the fraction of adsorption sites, f, in the mobile region needs to be known; θ_m is the volumetric water content of the mobile region; θ is the total volumetric soil water content; β is the mobile-immobile partition coefficient; and R is the retardation factor. Using the first approximation of Nkedi-Kizza et al. (1983), we assumed that $\phi = f$ so that, from Equation 10, one obtains: $\phi = \beta$. To calculate the absolute mass transfer coefficient, α, we used:

$$\omega = \alpha L / q \tag{11}$$

Where, ω is the mass transfer coefficient; q is the Darcy flux; and L is the length of the soil column.

Two-flow region model

The two-flow region model divides soil water into two regions based on their flow velocity. Both water regions have a non-zero flow rate. Without loss of generality, we denoted the fast flow region as A and the slow region as B. The soil system was characterized by velocity (V_A,V_B),water content (θ_A, θ_B), solute concentration (C_A,C_B), and dispersion coefficient (D_A,D_B). The two domains are related by an interaction term Γsuch that:

$$\Gamma = \alpha(C_A - C_B) \tag{12}$$

The convection-dispersion equation in the two domains can be written as Skopp et al. (1981):

$$\frac{\partial C_A}{\partial t} = D_B \frac{\partial^2 C_A}{\partial x^2} - V_A \frac{\partial C_A}{\partial x} - \frac{\alpha}{\theta}(C_A - C_B) \tag{13}$$

$$\frac{\partial C_B}{\partial t} = D_A \frac{\partial^2 C_B}{\partial x^2} - V_B \frac{\partial C_B}{\partial x} - \frac{\alpha}{\theta}(C_B - C_A) \tag{14}$$

Where a is the first-order transfer coefficient between the two water flow domains (h^{-1}). The initial and boundary conditions for the fast (i=A) and slow (i=B) flow domains are:

$$C_i = 0 \quad t = 0,\ 0<x<L \tag{15}$$

$$V_i C_0 = -D_i(\partial C_i / \partial x) + V_i C_i \quad x = 0,\ 0<t<t_p \tag{16}$$

$$0 = -D_i(\partial C_i / \partial x) + V_i C_i \quad x = 0,\ t>t_p \tag{17}$$

$$\partial C_i / \partial x = 0 \quad x = L,\ t>0 \tag{18}$$

By introducing the following ratios:

$$f = \frac{\theta_A}{\theta} \text{ and } \gamma = \frac{V_A}{V_B} \tag{19}$$

We have:

$$V = fV_A + (1-f)V_B \text{ and}$$
$$C = \frac{C_A f\gamma + C_B(1-f)}{f\gamma + (1-f)} \tag{20}$$

Where, θ is the total volumetric water content , V is average pore water velocity (cm/h), C is the average solute transport concentration ($\mu g/ml$). Moreover, D_A and D_B was assumed to be a linear function of V_A and V_B, respectively (Skopp et al., 1981):

$$D_A = D_0 + \lambda V_A \text{ and } D_B = D_0 + \lambda V_B \tag{21}$$

Where λ is the dispersivity (cm), D_0 is molecular diffusion in water (cm^2/h).

MATERIALS AND METHODS

Two kinds of undisturbed soil columns were excavated from A and B horizon (10 to 60 cm depth) in Changwu field station and Ansai field station of Shaanxi Province, on the Loess Plateau of China, respectively. Their characteristics and soil classification are given in Table 1.

The particle size distribution was determined by sieving in combination with the pipette method; pH was measured with a pH meter in the extract of saturated paste; organic matter was determined by potassium dichromate titration.

In the laboratory, undisturbed soil columns were trimmed to the desired dimensions and the side walls were coated with the layer of paraffin wax. The experimental conditions for each soil column are listed in Table 2. Transport experiments of chloride in undisturbed

Table 2. Dimensions and physical properties of the three soil columns used.

Soil	K_{sat} (cm/h)	Diameter (cm)	Length (cm)	Bulk (g/cm^3)	Total porosity*	θ_s (cm^3/cm^3)
Changwu soil	0.514±0.514	19.45	43.50	1.270±0.0006	0.529±0.0002	0.424±0.0128
Ansai soil	8.340±1.451	19.45	43.50	1.430±0.045	0.460±0.0012	0.387±0.0521

* ± Refer to mean standard error.

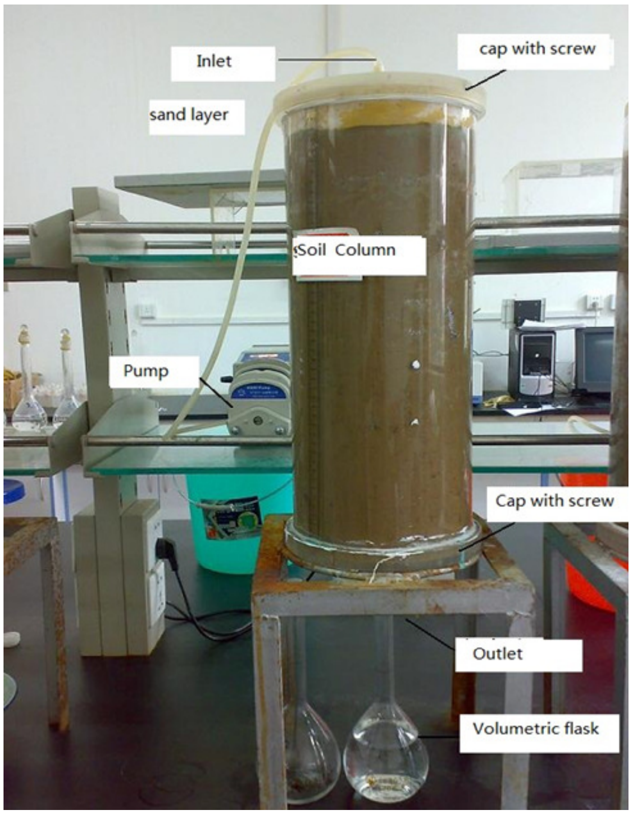

Figure 1. Experiment setup.

soil columns were carried out using miscible displacement method (Selim et al., 1987). Before conducting the solute transport experiment, each soil column was put into the distilled water for saturation. After that, both ends of the soil column were capped with filter discs from quick setting porous cement, and each disc has an outlet (Figure 1). The inlet at the top of the column was connected to a peristaltic pump which will control the desired input water velocity and solution pulse time while the effluent at the bottom was connected to the volumetric flask to collect the outflow. The effluent samples were then titrated with silver nitrate solution to determine the chloride ion (Cl^-) concentrations.

Two types of breakthrough curves (BTCs) were generated. The first type referred to as small pulse BTC (or impulse) was generated by introducing a small chloride pulse into each soil column (Cl 0.1 pore volume). This small chloride pulse was recovered by about four pore volumes of distilled water after pulse application. The second type of chloride BTC's, denoted as large pulse BTC, was generated in the same way as the first type except that the duration of the input chloride pulse was longer (about 0.5 pore volume). The small pulse was used to predicted the model validation and the large number BTC's were used to derived model parameters for further study on the preferential solute transport.

RESULTS AND DISCUSSION

Chloride transport in a small pulse experiment

Figure 2 presents the effect of the pore water velocity on the small pulse BTCs in Changwu soil columns (A and B) with two different pore water velocities of 3.34 and 1.67 cm/h and Ansai soil columns (C, D and E) with threee different pore water velocity of 3.34, 6.69 and 8.35 cm/h and the comparisons of the experimental data with the fitted curves determined by CDE, the two-region model and two-flow region model. In Figure 2, all the small pulse BTCs showed tailing of the desorption side, respectively. A hump had also been observed on the tailing side of the Changwu soil columns, which became pronounced as the velocity decreased, and it is mainly resulted of a nonuniform flow domain in the soil columns. In Changwu soil columns, they may consist of two overlapping pore domains, with solute transport relative fast in one domain and slow in the other domain. The solution transferred in the first domain will come quickly to the bottom of the soil column and the reach to the first pump value. Later, the solute transferred in the slow region finally reached to the bottom of the soil column and another pump value was generated. Moreover, the bimodal peaks did not appear in Ansai soil column of which the soil belongs to sandy soil. Therefore, it is apparent that the shape of BTCs is dependent on soil types as well as pore water velocity. Similar results were obtained by Ma and Selim (1995) who studied on the preferential solute transport in disturbed aggregate soil columns.

From Figure 2, we also could find that the breakthrough curves changed greatly with the increase of pore water velocities. In all five soil columns, with the increase of pore water velocities, the peak value appeared much earlier but the peak values changed differently in different soil types. In column A and B, the peak values decreased with the increase of pore water velocities, while the opposite is true in columns C, D and E.

In order to study further on the effect of preferential solute transport on the breakthrough curves, we fitted small pulse experimental data with CDE (sign multiplication), two-region model (triangle) and two-flow region model (circle) (Figure 1). As shown in Figure 1, the

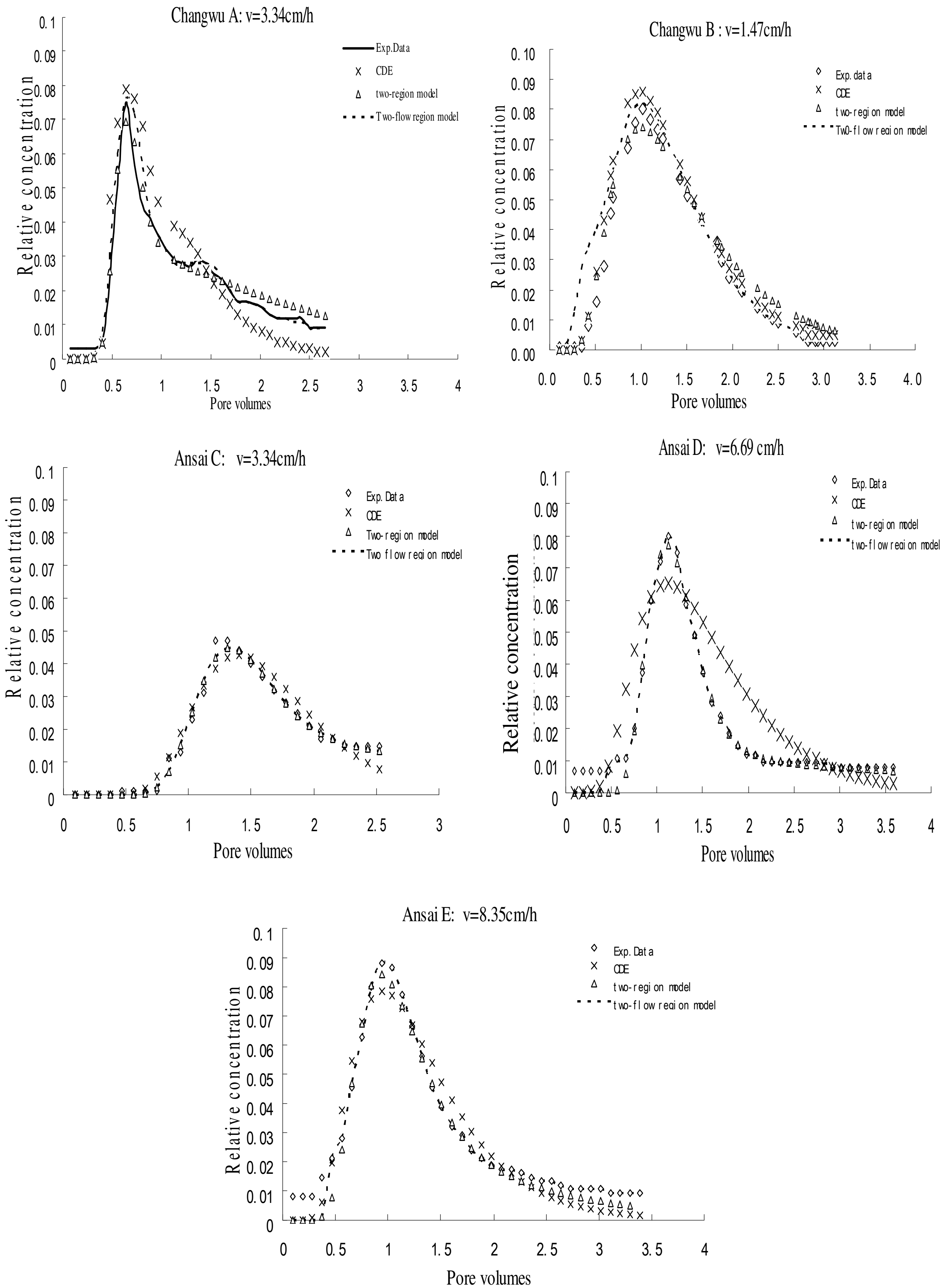

Figure 2. Small pulse BTCs from Changwu soil (A, B), Ansai soil (C, D, E).

two-flow region model improved the overall descriptions of BTCs compared with the two region model and CDE, especially the hump and bimodal peaks in soil column A with a pore water of 3.34 cm/h. When there was no bimodal in the BTCs, CDE and two-region model could describe the experimental data as well as the two-flow model.

Chloride transport in a large pulse experiment

Figure 3 shows the large pulse chloride BTCs for Changwu soil columns with pore water velocity of 3.34 and 1.47 cm/h, and Ansai soil columns with pore water velocity of 3.34, 6.69 and 8.35 cm/h, respectively. All three BTCs showed an obvious early breakthrough and tailing phenomena but the second hump was only observed on the tailing side in Changwu soil columns (A and B), which was much more pronounced with the increase of pore water velocity. All the first pumps of soil column appeared when the pore volume was more than 1.0. From Figure 3, we could also found that the shapes of the breakthrough curves of Ansai soil columns varied with the increase of pore water velocities. With the increase of pore water velocity, the peak value decreased and the hump appeared much earlier, form 2.0 pore volume to1.5 pore volume.

From Figure 3, we also can found the hump on the tailing side became much more significant in column A and B which showed the obvious effect of solute in fast region and slow region. The distance between first peak and second peak did not changed a lot with the changing of pore water velocity which is opposite in the small pulse experiment.

The large pulse BTCs were also fitted with two-flow region model, two-region model and CDE shown in Figure 2. As shown in Figure 2, the two-flow region model improved the overall descriptions of BTCs compared with the two-region model and CDE especially the double peaks and the extensive tailing. In contrast, CDE is capable of predicted only umimodal BTCs. Moreover, although all the models successfully predicted the position of the first peak, the two-region model and CDE predicted the peak height a little lower.

Parameters derived by CDE, two-region model and two-flow region model

To further examine the effect of preferential flow on solute transport, the large pulse BTCs were fitted by CDE, two-region model and two-flow region model. Fitted parameters and their coefficient of determination are listed in Table 3.

The values for dispersivity, λ (cm), are an indication of the distance traveled by the solute. To further compare all the models, the values of dispersivity λ were calculated from fitted D values. From Table 3, we can see that the dispersivity of Changwu soil columns calculated by two-flow region model, two-region model and CDE are all larger than those of Ansai soil columns. This also indicated that the solute transport paths in Changwu soil columns are much more torturous. From Table 3, it also could be found that the calculated λ values with CDE were obvious larger than those derived by two-flow region model and two-region model, especially in Changwu soil columns. This is expected to because in the CDE, the process of solute exchange between mobile and immobile regions is combined into an equivalent dispersivity incorporating the two processes of hydrodynamic dispersion and diffusional exchange, which are treated separately by the two-region model (Wang and Shao, 2007). Thus, the dispersivity values in the two region model and two-flow region model account for only part of the dispersion phenomenon (the other part is defined by the absolute mass transfer coefficient, α) described by the CDE and are thus always smaller than the dispersivity values obtained with the CDE.

From Table 3, it also could be found that the dispersivity coefficient increased with the increase of pore water velocity which may support the use of Equation 9. Estimated λ's also indicated that regardless of the model used, Changwu soil columns have larger λ than those of Ansai soil columns.

In the two-region model, Cl^- is transported by a convective–dispersive process in the mobile domain and enters and leaves the immobile domain by a first-order process. Conceptually, the assumption of first-order exchange kinetics is only valid for dead-end pores with a neck of negligible volume and with large-capacity dead-end volumes of negligible extension (Coats and Smith, 1964). However, in the two-flow region model, Cl^- is also transported in both the fast and slow region under different pore water velocities; furthermore first-order process also occurred between fast region and slow region. Both the two-region model and two-flow region model gave the fitted data of mass transfer coefficient (α) and mobile fraction (FM) (Table 1). According to van Genuchten and Dalton (1986) and Schulin et al. (1987), for any other pore geometry, the differences in the mass transfer coefficient, a compound parameter, depends not only on the pore space geometry, the solute diffusivity, and the relative magnitude of the mobile region, but also on the pore water velocity. In this experiment, the values of mass transfer coefficient decrease with the increase of pore water velocities which will further enhance the development of double BTC peaks (Table 3). Ma and Selim (1995) studies on a disturbed aggregate soil column and pointed that with the increase of pore water velocity, a small solute exchange between the two-domain will be obtained. Griffioen et al. (1998) discussed many examinations of published data and found that linear variation of the transfer rate with mobile fluid

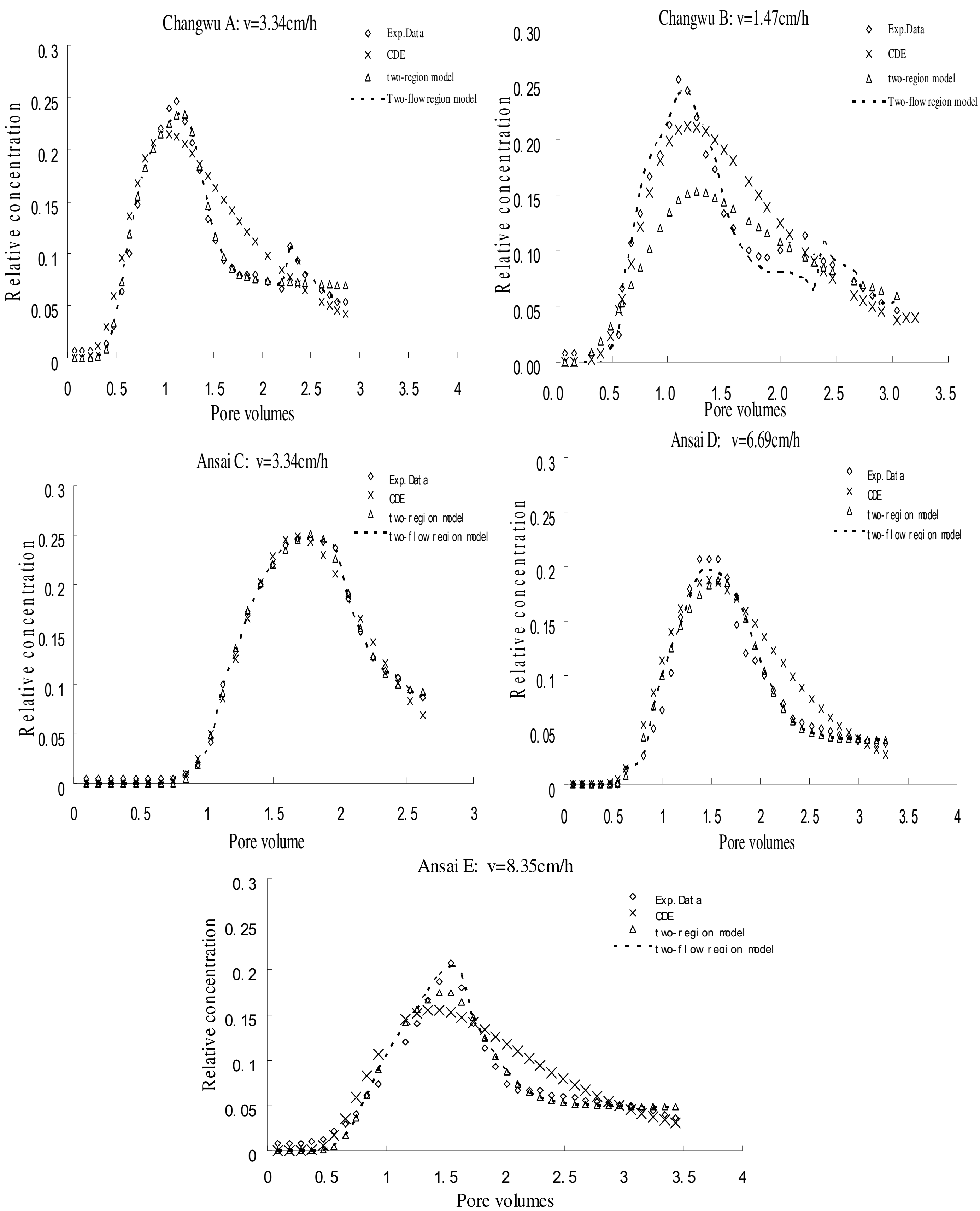

Figure 3. Large pulse BTCs from Changwu soil (A, B), Ansai soil (C, D, E).

Table 3. Fitted parameters from the Large pulse BTCs with CDE, two-region model and two-flow region model.

	Two-flow region model					
	SSQ	**Λ (cm)**	**D (cm^2/h)**	**α**	**γ**	**β**
Changwu A	4.23E-02	0.432	1.442±1.005	0.0023±0	2.123±0.021	0.533±0.030
Changwu B	1.18E-02	0.621	1.035±2.340	0.0021±0.0001	2.780±0.320	0.499±0.036
Ansai C	2.24E-02	0.330	2.200±1.045	0.0012±0	1.037±0.151	0.628±0.026
Ansai D	2.32 E-02	0.153	2.247±0,967	0.0005±0.0001	1.320±0.0074	0.762±0.024
Ansai E	6.60E-02	0.148	2.482±1.456	0.0002±0.0001	1.799±0.0156	0.855±0.041
	Two-region model					
	SSQ	**λ**	**D**	**β**	**α**	
Changwu A	3.96E-03	1.020	3.401±1.346	0.88±0.089	0.135±0.125	
Changwu B	1.57E-02	1.112	1.854±1.023	0.77±0.078	0.203±0.142	
Ansai C	7.03E-04	0.369	2.458±1.072	0.80±0.047	0.218±0.085	
Ansai D	7.51E-03	0.326	4.350±0.570	0.91±0.038	0.574±0.156	
Ansai E	4.38E-03	0.341	6.080±1.356	0.90±0.069	0.729±0.131	
	CDE					
	SSQ	**λ**	**D**			
Changwu A	6.23E-02	1.773	5.912±0.789			
Changwu B	4.15E-02	3.097	5.162±0.932			
Ansai C	2.57E-03	0.455	3.035±0.125			
Ansai D	1.87E-02	0.339	4.523v±1.721			
Ansai E	3.95E-02	0.368	6.086±1.899			

*Mean particle density was determined to be 2.65 g/cm^3, *γ: mobile fraction, α: mass transfer coefficient, * ± refer to mean standard error.

velocity was the dominate trend. Comparisons of the mass transfer coefficient fitted by two-flow region model and two-region model show that the mass transfer coefficient values calculated by two-flow region model are quite smaller than those fitted by later. It mainly because that the mass transfer coefficient in two-region model contained both the mass. This is to be expected because, in the two-region model, the process of solute exchange between fast and slow regions and the solute transfer in the slow region are combined into an equivalent mass transfer coefficient incorporating the two processes of hydrodynamic dispersion and diffusional exchange, which are treated separately by the two flow region model.

The parameter β of the two-region model represents the fraction of solute present in the mobile region under equilibrium conditions. From Table 3, it could be found that the values of β in Ansai soil column are much larger than those in Changwu soil columns, especially in Ansai Ш soil column; β is nearly equal to 1.0. As β approaches to 1, the contribution of the fast flow region becomes negligible and the resulting BTC approaches a symmetrically unimodal distribution (Ma, 1995).

The relative magnitude of flow velocity in the two-flow region model (γ) is also critical for the shapes of dual peaks. As shown in Figure 2 and Table 3, when γ =1, or nearly equal to 1, the two-flow region could be reduced to one-region model and no doubt peaks were expected. In Changwu soil columns, values of γ were lager than 1 and double peaks BTCs were obtained. Furthermore, with the increase of pore water velocity, the values of γ decreased from 2.780 to 2.123. However, with further increase of γ, the two peaks will disappear and changed into the bimodal. It was illustrated for a γ of 10 in laboratory experiment where the hump disappeared.

Conclusions

The authors examined the preferential solute transport in undisturbed soil columns with a miscible experiment. We found that distinct two peaks were obtained in BTCs of Changwu soil columns (loamy soil). With the increase of pore water velocity, the pump on the tailing side became much more obviously and the first peak appeared at a much smaller pore volume. The two-flow region model was superior to the CDE and two-region model in chloride BTCs description, especially on the tailing side. Both of the dispersivity and dispersivity coefficient of Changwu soil columns calculated by all the three models are all larger than those of Ansai soil columns. Furthermore, the λ values derived by the CDE and two-region model and mass transfer coefficient fitted by the two-region model were obvious larger than those obtained by two-flow region model.

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