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Effects of macropore continuity on water movement and solute transport in a loessial soil

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Abstract

Flow through macropores, created by soil pedogenesis and biological activity, play important roles in soil water and chemical transport on the Loess Plateau. Numerous studies have examined individual macropores and the effects of their size on solute transport, but few have assessed the effects of macropore continuity and of neighboring macropores. This paper describes a laboratory investigation of the effects of macropores with varying degrees and types of continuity on the transport and distribution of solutes in loessial soil columns. Soil columns (2-D, 60 cm high) containing standardized artificial macropores were used to obtain breakthrough curves (BTCs) for input solutions containing 1190 mg/L KBr, and 100 mg/L FD&C Blue #1 (a food dye) under a constant hydraulic head of 8 cm. The types of macropore were: open at both the surface and bottom of the soil column (O-O); open at the surface-closed at the bottom (O-C); and closed at the surface-open at the bottom (C-O). Columns with no macropores served as a control. As expected, in the O-O column the solution reached the bottom rapidly, bypassing most of the soil matrix. The presence of an O-C macropore resulted in weak retardation and much deeper penetration of the bromide and FD&C Blue #1 than in the control columns, but the C-O macropore had little effect on either BTCs or solute distributions. In further tests where neighboring macropores were present, an inclined macropore strongly affected solute concentrations in the profile. Therefore, the type of macropore and the presence of neighboring macropores, all affect soil water flows and solute infiltration parameters.

Keywords: macropore continuity; breakthrough curve; CDE; solute distribution.

Abbreviations: BTC : breakthrough curve ; CDE: convection dispersion equation; FD&C Blue #1: Food, Drugs, & Cosmetic Dye Blue number one; PV: pore volume; KBr: Potassium bromide.

Introduction

Water flow and solute transport mechanisms have received increasing attention in recent years due to environmental regarding potential soil and groundwater concerns contamination caused by human activities and rapid economic development occurring on the Loess Plateau (Jia. et al., 1993; Zhou and Shao, 2010). Preferential flow through macropores that were formed by biological activity and structural cracking might increase contamination and the movement of solutes in general (Beven and Germann, 1982; Starr et al, 1978, 1986; Richard and Steenhuis, 1988; Hallberg, 1989; Rice et al., 1991; Ghodrati and Jury, 1992). Macropore flow has been investigated in the field using hydrometric methods, but interpretation of results acquired under field conditions has been complicated by the complexity of natural soil systems and difficulties associated with assessing hydraulic properties in the field (Buttle and Leigh, 1997). Intact soil, in which macropores rarely occur, has also been investigated in some studies (Elrick and French, 1966; Gish and Shiirmohammad, 1991; Edwards et al., 1993; Pearson et al., 1996; Ashraf et al., 1997). In addition, macropores have been morphologically characterized using computed tomography scans and paint injection techniques, but substantial uncertainties regarding effects of their characteristics remain, and it may not be possible to simulate unsaturated flow conditions adequately using undisturbed soil cores. Therefore, the effects of macropores have been investigated in laboratory experiments that have mostly used repacked soil columns with artificial macropores of known dimensions, open at the soil surface and oriented parallel to the direction of water and solute flow (Gish and Shiromhammad, 1991; Steenhuis et al., 1994; Jury et al., 1997; Jarvis, 2007). Munyankusi et al. (1994) concluded that knowing the number and size of visible surface macropores was not sufficient to model water and solute transport through macroporous soils, but claimed that information about the length and the continuity of macropores was also needed. Furthermore, little is known about the effects of other types of macropores on water flow and solute transport, or the interactive effects of multiple macropores. In most studies on the impact of macropores on preferential flow, a conservative tracer has been used for breakthrough characterization (Gish and Shirmohammad, 1991; Jose et al., 2004). However, important characteristics of contaminants include their adsorption-desorption parameters, and the effects of interactions between these parameters and macropore continuity or tortuosity on solute transport are poorly understood. Breakthrough curves (BTCs) have been were widely used to characterize the presence or absence of macropores, since this is the most practical and straight forward method (Pearson et al., 1996; Ashraf et al., 1997). Simple parameters such as the dispersion coefficient have been used to describe the macropore population and the solute transport paths. The objective of the study presented here was to characterize the effects of macropores having different degrees and types of continuity within 2-D columns on infiltration, BTCs, transport parameters and distributions of solutes in the soil profile, as well as considering the influence of neighboring macropores.

Results and discussion

Effects of macropore continuity types on the infiltration of bromide and FD&C Blue #1

Figure 4 presents the observed infiltration rate of bromide and the depth of the wetting front of FD&C Blue #1 as a function of time for columns with two types of macropore (O-C, C-O) and the control. Since the solutes passed directly through the O-O macropore to the outflow of the soil column without entering the soil matrix, the effect of this type of macropore on the infiltration process was not examined. The data presented in the figure show that smooth infiltration curves and wetting fronts were obtained for both the control and the columns of soil containing macropores. However, the infiltration was notably more rapid for the O-C macropore case than for either the C-O macropore or no macropore case. Furthermore, the effects of macropore type on the bromide infiltration rate and the wetting front of FD&C Blue #1 became relatively more pronounced with time. These findings indicate that macropores close to the soil surface had relatively little effect on infiltration rates when compared to the control. Additionally, when macropore flow occurred, the relative fractions of soil and macropores involved governed the infiltration parameters. We also compared the effects of four macropore types on the wetting fronts of bromide and FD&C Blue #1. As shown in Fig 4, the wetting front of the bromide conservative tracer moved more rapidly to the bottom of the columns than FD&C Blue #1 due to the weaker adsorption of bromide. Furthermore, the wetting fronts of both bromide and FD&C Blue #1 advanced more rapidly in the presence of O-C macropores than in the presence of C-O or when there were no macropores; there were no significant differences between the latter two cases. This was consistent with expectations as the macropores in the C-O columns remained empty during the infiltration process, while the bromide and FD&C Blue #1 both migrated rapidly through the O-O macropores, with little retardation by absorption by, or dispersion into, the soil matrix through which they passed. Water flow in the columns with the O-C type macropore used in this study could be conceptually described by flows through two regions. The first region was the macropore domain that appeared to conduct water when it was saturated and that included the macropore wall surface area, which influenced flow and transport kinetics via the exchange of water between the macropore and the soil matrix. The conductivity in this domain ranged from the matrix-saturated hydraulic conductivity, when the macropore was empty, to a maximum value when the macropore was filled with water. The second region was the soil matrix, in which flow and solute transport parameters were assumed to be identical to those of the homogenous soil (Chen and Wagenet, 1992). Allaire-Leung et al. (2000) also studied the effect of macropore continuity on soil water movement and photographed the infiltration process. These photographs showed that the water moved rapidly through the macropores and bypassed most of the soil matrix, with very little absorption of water occurring beyond the immediate vicinity of the macropore walls until the macropore was filled with water. Thus, in this present study, the C-O type macropore had little effect on the infiltration rate and the wetting front of both tracers when the macropore was empty, due to the small soil matric and gravitational potentials. All

these observations proved that the assumption often used in modeling, that water almost instantaneously reaches the lower end of surface open macropores under ponding conditions, was reasonable (Allaire-Leung et al., 2000).

Effect of macropore continuity types on solute transport

As shown in Fig 5, the bromide and the FD&C Blue #1 tracers both passed directly through the artificial macropores in soil columns with O-O and O-C type macropores (directly to the bottom of the column in the former case). This flow was more rapid than through any other columns. Consequently, the BTCs for the O-O column were linear. Due to the displacement between the macropore and the soil matrix in the O-C column, the bromide concentration started to decrease slightly more slowly, after about 1 PV of effluent, than in the C-O and control columns where the bromide concentration started to decrease at about 1.5 PV. This indicated that bromide moved slowly through the soil matrix to the bottom. Figure 5 also indicates that the relative bromide concentration was the same in the first effluent samples collected from the columns with all the tested types of macropore, including the control, as in the input solution. This was consistent with expectations, since there was no solute to displace at the wetting front because the soil was dry. Thus, the bromide concentration remained the same as it moved down the column. In further accordance with expectations, the bromide and FD&C Blue #1 concentrations rose to input levels and subsequently declined sooner in the outflows from the O-C column than in the outflows from the C-O and the control columns. However, the O-C treatment BTCs took longer to attain zero relative concentration than the other treatments. This may have been due to two processes occurring in the O-C columns. Firstly, dispersion into the matrix below the artificial macropore (from 0.45 to 0.6 m depth) and adjacent to it may have led to higher bromide and FD&C Blue #1 concentrations in these parts of the matrix. Secondly, displacement of the first solution that contained bromide by the second bromide-free solution may have resulted in more diffusion and mixing. There were no significant differences between the BTCs obtained for the C-O and control columns, for either bromide or FD&C Blue #1. This suggested that solute movement through the soil matrix was more important than through the macropores in C-O columns. To explore the effect of macropore continuity types on solute transport further, we used the convection-dispersion equation under unsaturated conditions to fit our experimental data. The parameters obtained are presented in Table 1, and as shown in Fig 5 the equation well-described the bromide and FD&C Blue #1 transport in the soil columns containing macropores, further indicating that solute transport through the soil matrix was much more important than transport through the macropores of all tested types. Table 1 lists the dispersion coefficients and retardation factors of the two tracers fitted by the CDE, and also the sum of the squares of residuals (SSQ) values, which indicate that the CDE well-predicted the values for the relative concentrations of bromide. Dispersion coefficients provide indications of the distance traveled by a given solute in a given time. Thus, the values for the two tracers would be smaller when the path taken by a solute in a column was more tortuous (Table 1). The dispersion coefficients of the tracers were substantially larger for the O-C column than for the C-O and control columns, presumably because the O-C macropore provided a vertical channel from the top of the column that made the flow path less tortuous (Elrick and French, 1966). Furthermore, as a result of the strong chemical sorptivity of FD&C Blue #1, the dispersion coefficients were smaller than

Table 1. Dispersion	coefficients (D)	and retardation	factors (R)	obtained by	the convection	n-dispersion	equation ((CDE) fc	or columns
with each tested type	of macropore.								

		Macropore types	SSQ	D	R
		C-0	0.0015	0.00107	0.528
	FD&C Blue#1	O-C	0.00172	0.00287	0.279
CDE		control	0.126	0.0019	0.656
		C-O	0.00119	0.0130	0.201
	Bromide	O-C	0.00373	21.8	0.00104
		control	0.00137	0.00479	0.219

SSQ: sum of the squares of the residuals; C-O and O-C: macropores closed (C) or open (O) at the top or base, respectively; control, without macropores

Table 2. Dispersion coefficients and retardation factors obtained by the convection-dispersion equation for columns with or without (control) macropores.

		Soil column types	SSQ	D	R
		control	0.00176	1.00E-03	3.84E-01
Br		macropores	0.00581	7.17E-02	3.04E-01
	FD&C	control	0.120	1.00E-03	7.58E-01
]	Blue #1	macropores	0.145	1.80E-02	5.26E-01
SSQ: sum of the squares of the residuals					

those obtained for the bromide BTCs. In contrast to the dispersion coefficients, the retardation factors were smaller for the O-C columns than for the C-O and control columns, presumably due to the higher water velocity in the O-C columns. Nkedi-Kizza et al. (1983) also obtained lower retardation factors when pore water velocities were higher. Thus, at higher pore water velocities, solute mixing appears to be less complete and solutes move more rapidly through the porous medium or macropores. Presumably, at higher velocities, the larger pores were filled more quickly, and water and solutes move more readily through them, bypassing parts of the soil matrix (Tyler and Thomas, 1981; Bouma and Anderson, 1977). Table 1 also shows that there were no marked differences in the dispersion coefficients and retardation factors obtained for the C-O and control columns, further suggesting that macropores that are not open at the surface have little effect on solute transport parameters.

Effects of neighboring macropores on solute transport and distributions

Effects of neighboring macropores on solute transport

The preceding section discussed observations from the experiment with columns containing different types of single macropores as well as macropore-free controls. However, macropores rarely occur in isolation in nature. Typically, multiple macropores are usually present and they may interact with each other. Therefore, in Experiment 2 we examined the effects of neighboring macropores on bromide and FD&C Blue #1 transport through, and their distributions within, the soil profile. As shown in Fig 6, the neighboring macropores of both types (vertical and inclined) strongly affected the shape of the bromide BTCs and advanced the initial breakthrough time of both solutes. We also fitted our experimental results using the CDE, with similar results to those described in the previous section (Table 2); higher dispersion coefficients and lower retardation factors were obtained for the column containing the macropores than for the control.

Effects of neighboring macropores on solute distribution in the soil profile

The BTC results, as described above, do not allow definitive conclusions to be drawn regarding the influence of neighboring

macropores. Therefore, we also examined the effects of the neighboring vertical and inclined macropores on bromide and FD&C Blue #1 distributions in the soil columns by measuring their concentrations along four vertical profiles (A, B, C and D in Fig 3). As shown in Fig 7, the inclined macropore had marked effects on the bromide distribution. The concentration of bromide rapidly declined with depth in the column containing macropores along the A, C and D profiles. However, along the B profile it first increased with soil depth, peaked at a depth (~12.5 cm) corresponding to a depth just below that of the end of the inclined macropore (~10.6 cm), and then decreased to negligible levels at about 20 cm depth. In contrast, there was little difference in the concentrations for profile C, reflecting the influence of a vertical macropore on solute distribution, and those of the control profiles (left and right, Fig 3). Figure 7 also shows that the FD&C Blue #1 concentration slowly increased with increases in depth, corroborating its slow transport rates, and that it was lower in profile B than in the other profiles. The inclined macropore directed the water flow path and, thus, the movement of bromide towards the neighboring macropore. The presence of this type of neighboring macropore in the soil profile could thereby lead to interactions between macropores that could in turn enhance the importance of macropores for solute transport. Allaire-Leung et al. (2000) studied the effect of inclined C-C macropores and found that even this type of inclined macropore in the profile may be an important factor affecting the solute flow path associated with neighboring macropores with consequences for the solution flow path at deeper depths. The concentrations of FD&C Blue #1 were lower near the upper ends of the macropores than at the lower ends, which indicated that the inclined macropores and the soil beside them inclined macropore were saturated with the bromide and water that diluted the FD&C Blue #1 concentration when it was input.

Materials and methods

Soil

The soil used in this study was collected from the B horizon of a loamy soil (sand, silt and clay proportions were 46.3%, 21.2% and 32.5%, respectively; pH, 8.2; organic matter content, 11.2 g/kg) in a field that had been used to cultivate wheat at the Changwu field station, Shaanxi, on the Loess Plateau. The



Fig 1. Positions of macropores of varying continuity in the soil columns used in Experiment 1: O-O, C-O and O-C are macropores closed (C) or open (O) at the top or base, respectively.



Fig 2. Positions of the macropores in the soil column used in Experiment 2.



Fig 3. Soil sampling positions down four profiles (A, B, C, D) where macropores were present, and two profiles in the control in Experiment 2.

saturated and residual water contents were 0.424 kg/kg and 0.048 kg/kg, respectively. The cation exchange capacity is 14.0 cmol/kg and the exchangeable Na percentage is 0.27%. The soil was air-dried and passed through a 2-mm sieve.

Experiment setup

The effects of various types and configurations of macropores, as well as of neighboring macropores, on water and solute movement were studied in two experiments. In Experiment 1, soil columns (60 cm long) were created in 2-D rectangular plexiglas chambers (internal dimensions: width 40 cm, height 70 cm, mean thickness 1 cm) with, or without, an artificial macropore varying in extent and position in the column. Three types of macropore were created: (i) open at both the soil column surface and the lower boundary (O-O); (ii) open at the surface and closed at the lower boundary (O-C); and (iii) closed at the surface and open at the lower boundary (C-O). Columns with no macropores served as controls. The macropores were constructed from a stainless steel mesh (with 40 holes cm⁻²), and were straight with a square cross-section $(1.0 \times 1.0 \text{ cm}^2)$, and were all 45 cm long, except those used in the O-O columns, which were 60 cm long (Fig 1). Hence, in this experiment 'continuity' was controlled by having the macropores open or closed at the upper and lower boundaries of the soil column. The setup of Experiment 2 was similar, except that the column chamber was 100 cm wide and 30 cm high, and contained three artificial O-C macropores (one inclined at 45° and two vertical, all 15 cm long) to study the effects of neighboring macropores on solute distributions. Columns with no macropores served as controls. All three macropores had perforated bases covered by coarse filter paper. The soil was uniformly packed in the columns to obtain a bulk density of 1.27 g/cm³. The initial soil moisture content was 0.0048 kg/kg. The surfaces of the soil columns were covered with filter paper to reduce disturbance by inflowing solutions. To obtain BTCs, a miscible displacement procedure was applied to simulate water and solute transport into an initially air-dry soil under ponded conditions while maintaining a constant hydraulic head of 0.08 m at the upper boundary with free drainage at the lower boundary. The input solution contained 1190 mg/L KBr (potassium bromide) and 100 mg/L FD&C Blue #1 (Food Drugs & Cosmetic Dyes Blue number 1). After adding 1.5 pore volumes (PV) of this solution, the input solution was changed to a solution of 100 mg/L FD&C Blue #1 alone. In both experiments, all the effluent was collected in 50 ml volumetric flasks at the lower boundary of each soil column until a total of 6 PV of the applied solutions had passed through it. After collecting all the effluent, the soil columns were quickly dismantled and sampled to evaluate the concentrations of KBr and FD&C Blue #1 along vertical profiles. In Experiment 1, 40 soil samples (2.5 g) were taken at equi-depth intervals adjacent to each side of the macropore and a further 20 soil samples were taken at positions 10 cm away from the macropore; for control columns, which had no macropores, soil samples were taken from the corresponding positions (Fig 2-a). In Experiment 2, 50 soil samples (2.5 g) were taken along four vertical profiles (designated A-D, one beside a vertical macropore (B), two passing through the mid-points between the tops of the two adjacent macropores (A and C), and one



Fig 4 Effects of macropores with varying continuity (C-O and O-C are macropores closed (C) or open (O) at the top or base, respectively; control is without macropores) on bromide and FD&C Blue #1 infiltration in Experiment 1.



+ O-O experimental data; \triangle O-C experimental data \diamond C-O experimental data \times Control experimental data ---data fitted by CDE

Fig 5. Effect of macropores of varying continuity (O-O, C-O and O-C are macropores closed (C) or open (O) at the top or base, respectively; control is without macropores) on bromide and FD&C Blue #1 breakthrough curves.



△ Soil columns containing macropores □ Soil columns without macropores





Fig 7. Effects of neighboring macropores on bromide and FD&C Blue #1 distributions in soil profiles (A, B, C, D, and control left and right).

passing through the mid-point between another vertical macropore and the side-wall of the chamber (D), as illustrated in Fig 3); in the control columns, soil samples were taken from positions corresponding to one of the vertical macropores and the inclined macropore (Fig 2-b). Each experiment was conducted in two replications. To extract the tracers, each soil sample was shaken with 25 ml of deionized water on a reciprocating shaker for 24 hours and centrifuged at 6000 rpm for 10 min, after which the supernatant solution was transferred to a clean test tube. The concentration of bromide in the supernatent solution was measured using a bromide-specific electrode. The FD&C Blue #1 concentration in the supernatant solution was determined from the absorbance at 630 nm using a UV/Visible spectrophotometer.

Conclusions

We examined the effects of macropore continuity and neighboring macropores on infiltration, solute transport and solute distributions in soil columns. We found that the macropore types that were open at the soil surface accelerated infiltration and solute transport. Smooth BTCs, which were well-fitted by the CDE under unsaturated conditions, were obtained for all soil columns containing either one macropore (with varying continuity) or three macropores with different orientations. We also found that both macropore continuity type and the presence of neighboring inclined macropores strongly affected BTCs and solute concentrations in the soil profile. The presence of neighboring inclined macropores could generate interactions between macropores and enhance their importance to solute transport.

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