Korean J. Soil Sci. & Fert.(2000)
Vol. 33(2) : 71~78

Preferential Flow as Tested by Breakthrough Curves of Cl^- and Cu^{2+} from Saturated Undisturbed Soil Core Samples under Steady Flow Conditions

Sun-Ho Yoo*, Kyung-Hwa Han*, Hee-Myong Ro** and Gwang-Hyun Han*

ABSTRACT

Preferential flow has recently been the subject of increasing interest because these phenomena contribute to solute transport in soils. Commonly, preferential flow paths are associated with macropores or highly structured soils. We presented an analysis of the measured breakthrough curves (BTCs) of Cl^- and Cu^{2+} ions to test the occurrence of preferential flow in soils using miscible displacement technique under steady flow conditions. We also analyzed soil water retention curves and from this curves induced cumulative pore size distribution of undisturbed soils, which sampled from Apl, B1, and C horizons of Songjeong series soils (the fine loamy, mesic family of Typic Hapludults). In this study, miscible displacement experiment on C horizon was excluded, because it is structureless sandy loam with saturated hydraulic conductivity of 5.2 cm hr^{-1}. The saturated hydraulic conductivity of Apl horizon was 2.0 cm hr^{-1}, which was about 7 times higher than that of B1 horizon (0.27 cm hr^{-1}). Cumulative pore size distribution predicted that Apl horizon had more macropores (pore diameter larger than 49㎛, equivalent to -6 kPa of soil matric potential) than B1 horizon. The hydrodynamic dispersion coefficient from chloride BTCs was estimated as 1.3 cm2 hr^{-1} for B1 and 34 cm^2 hr^{-1} for Apl horizon. However the retardation factors of B1 and Apl horizon were significantly different, i.e. 1 and 0.6, respectively, which means that there was distinct partition between mobile water and immobile phase in Apl horizon. The copper retardation effect of Apl horizon was less than that of B1 horizon, even though cation exchange capacity of Apl horizon was higher than that of B1 horizon. Thus, breakthrough curves of Cl^- and Cu^{2+} obviously showed the probability that preferential flow would occur in Apl horizon.

Key words : Preferential flow, Miscible displacement, Breakthrough curves, Macropore, Pore size distribution.

Introduction

Water and solute transport through macroporous soils are very important subject in the fields of environment as well as agriculture because the flow of chemical-containing water through preferential pathway can threaten groundwater quality. Macropores have the capacity to carry enormous amounts of water at velocities that greatly exceed those in the surrounding matrix (Beven and Germann, 1982). In macroporous soils, therefore, the transport of water and solute can be described as two flow region, of which one is macropore flow and the other is matrix flow

---

* College of Agriculture and Life Sciences, Seoul National University, Suwon, 441-744, Korea
** Dept. of Horticultural Env. National Horticultural Research Institute, Suwon, Korea

※ This research was funded by ARPC (Agricultural Research and Development Promotion Center).

(Beven and Germann, 1982; White, 1985; van Genuchten and Wierenga, 1976; Bouma, 1991).

In particular, water flow through continuous macropores is called preferential flow. The preferential flow has been studied in various soils and conditions. The rapid leaching of water and chemicals (Phillips et al., 1995) and the channeling effects on their transport (Tyler and Thomas, 1981) are of major concerns in most no-till macroporous soils, while the film-flow down the channel walls is important in cracked clay soils (Bouma et al., 1981). Li and Ghodrati (1994) observed the significant preferential movement of NO_3^- through the decayed root channels.

The extent of solute leaching by preferential flow in undisturbed soils depends on the location of solute, the ratio of macropore to matrix flow, the saturated hydraulic conductivity of the matrix, the antecedent water content of soil, and the rate of solute diffusion between the mobile and immobile water volumes (Bouma, 1991; Shipitalo et al., 1990; White, 1985). As such, preferential flow through soils has been studied using non-reactive tracer, which is not common in the adsorbed phase of soil using miscible displacement technique (Bouma, 1991; Shipitalo et al., 1993; White, 1985). In general, Br^- and Cl^- anions have been used as tracers to study water flow in soil cores, lysimeters, and the field soils, since the mobility of cation species is much lower than that of anion species due to adsorption to the exchange site of soil particles (White, 1985). Moreover, the breakthrough curves (BTCs) of cations applied to the surface of soil surface are also influenced by preferential flow due to presence of macropores in soils (Shipitalo et al., 1993).

Since the analysis of BTCs can provide informations on the characteristics of water and solute transport through soil (Bouma, 1991), we presented an analysis of the measured breakthrough curves of Cl^- and Cu^{2+} ions to test the occurrence of preferential flow in soils using miscible displacement technique under steady flow conditions. In this analysis, we measured soil water retention curves and estimated cumulative pore size distributions of undisturbed soil samples collected from Ap1, B1, and C horizons of Songjeong series (the fine loamy, mesic family of Typic Hapludults).

Materials and Methods

1. Soil profile characteristics

Undisturbed soil core samples were collected from Ap1, B1, and C horizons of Songjeong series (the fine loamy, mesic family of Typic Hapludults) located in apple orchard, College of Agriculture and Life science, Seoul National University, in Suwon.

Ap1 (0 - 17 cm): brown to dark brown (7.5YR 4/4) fine gravelly sandy clay loam; moderate fine granular structure; friable, slightly sticky and slightly plastic; common fine quartz; many fine grass and tree roots; abrupt smooth boundary.

B1 (17 - 55 cm): yellowish red (7.5YR 5/6) fine gravelly sandy clay loam; weak fine to medium subangular blocky structure; thin patch clay cutan; friable, slightly sticky and slightly plastic; common fine mica and common fine quartz; many fine grass roots; clear smooth boundary.

B2 (55 - 79 cm): reddish yellow to yellowish red (7.5YR 5.5/6) fine gravelly sandy loam; weak fine to medium subangular blocky structure; friable, slightly sticky and slightly plastic; common fine mica and quartz grits; many fine to coarse roots; gradual wavy boundary (excluded in this study).

C horizon (79 - 120 cm): reddish yellow (7.5YR 7/6) sandy loam; granite saprolite; firm, nonsticky and nonplastic; many fine quartz grits; few coarse roots.

Even though this study site can be classified as Sonjeong series because of clay content higher than 0.18 kg kg^{-1}, its physical properties are rather similar to those of Yesan series (the coarse loamy, mesic family of Typic Dystrochrepts). The B2 horizon was regarded as a mixed horizon of B1 and C horizons.

2. Measured water-retention and saturated hydraulic conductivity

To measure hydraulic properties of the soil, five undisturbed cores (5.4 cm in diameter by 3 cm long) were taken from Ap1, B1, and C horizons of Songjeong series soil. Prior to measurements, the cores were saturated with 0.005 M $CaCl_2$ solution from the bottom and then allowed to be equilibrated for more than 24 h. Five cores from each horizon were used to determine saturated hydraulic conductivity (K_s) in the laboratory by the constant-head method. The K_s value for each horizon was the arithmetic mean of five cores from the same horizon.

The same cores used to determine K_s were further used for water-retention curves in the laboratory by the multi-step outflow method. Water-retention was determined successively at -2, -4, -6, -10 (or -15), -33, -50, and -100 kPa of matric potential using Tempe pressure cell. At each measurement span, the changes in volume of outflow was measured with time. At the end of entire span for each sample core, the bulk density and water content of the soil was determined. Saturated water content (θs) and water content at -2, -4, -6, -10 (or -15), -33, -50, and -100 kPa matric potential were calculated using total volume of outflow, final water content, and bulk density.

3. Estimation of unsaturated hydraulic conductivity function

The measured water-retention data were fitted to the equation of Campbell (1974) to obtain an empirically derived parameter describing the slope of the relationship between soil water tension and relative saturation (θ/θ_s):

$$h(\theta) = h_a\left(\frac{\theta}{\theta_s}\right)^{-b} \qquad (1)$$

where h is the soil water tension, ha is the air entry pressure, and θ is volumetric water content at a given soil water tension.Unsaturated hydraulic conductivity function was calculated from water-retention and saturated hydraulic conductivity using the equation (Campbell, 1974):

$$K(\theta) = K_a\left(\frac{\theta}{\theta_s}\right)^{2b+3} \qquad (2)$$

where K is hydraulic conductivity at a given soil water content θ.

4. Estimation of particle-size distribution

Mean pore diameter (㎛) at a given soil water tension was estimated from water-retention using the following equation (Danielson and Sutherland, 1986):

$$d_p = 4\sigma \times 10^5/\rho_w gh) \qquad (3)$$

where σ is surface tension of water (0.0724 J m^{-2} at 25℃), ρ_w is density of water (1 Mg m^{-3}), g is gravitational acceleration (9.8 N kg^{-1}), and h is the soil water tension expressed in cm of water.

5. Miscible displacement experiment

Miscible displacement experiments were performed using undisturbed cores (7.4 cm in diameter by 7.4 cm long) sampled from Ap1 and B1. Each soil core was sealed with fritted glass plates at both ends and connected to a Mariotte bottle at the upper end of the column and open to the atmosphere at the column terminus. Prior to experiments, soil cores were slowly saturated with 0.005 M CaSO4 from the bottom. After a steady-state flux of $CaSO_4$ solution was established, a 0.005 M $CuCl_2$ solution was introduced into the column. Effluent samples were collected using fraction collector (SMI. Model no. 12063) and analyzed for Ca^{2+} using AAS (Atomic Absorption Spectrophotometer) and for Cl^- using ion chromatography.

6. Breakthrough curve analysis

The measured breakthrough curves were analyzed using the following equation (van Genuchten and Wierenga, 1986) under constant concentration boundary at surface:

$$C/C_0 = \frac{1}{2}\, erfc[\frac{R-p}{(\frac{4DRp}{vL})^{1/2}}] + \frac{1}{2}\exp(\frac{vL}{D})\, erfc[\frac{R+p}{(\frac{4DRp}{vL})^{1/2}}] \quad (4)$$

where C is the solution concentration (ML^{-3}) at a given pore volume, C_0 is the concentration of incoming solution, t is time (T), D is the hydrodynamic dispersion coefficient (L^2T^{-1}), R is the retardation factor (dimensionless), ν is the pore water velocity (LT^{-1}) calculated from q/θ (q: water flux, θ: volumetric water content), p is the number of pore volume, and L is the length of the column.

The hydrodynamic dispersion coefficient and retardation factor of solutes during transport were estimated by fitting Eq. (4) to the measured chloride BTCs, and the goodness of the fit was obtained by minimizing sum of error square (Singh et al., 1991).

Results and Discussions

1. Soil physical properties and pore size distribution

The bulk density of B1 horizon was higher than that of Ap1 and B1 horizon. And clay content and sand content of B1 horizon was higher than those of Ap1 horizon. This could be due to thin patch clay cutan of B1 horizon. Thin patch clay cutan was reported as an indication that water has flowed with sufficient force to redistribute fine particles of sediment (Beven and Germann, 1982). The distribution of cutans in the profile may be interpreted in terms of connectivity and ability of the macropores to conduct water (Beven and Germann, 1982). In this soil profile, the clay cutan was widely distributed in B1 and B2 horizon. The organic content was highest in Ap1 horizon and very low in B1 and C horizon. C horizon had low clay content.

The saturated hydraulic conductivity was lower in B1 than Ap1 horizon, and was highest in C horizon. According to high saturated hydraulic conductivity of C horizon, it can be considered that the limiting horizons of water and solute transport is Ap1 and B1 horizon.

The parameter b of Campbell model is related with slope of semi-log water retention curves. As b is bigger, decreased water content relative to unit change of soil water potential was smaller. So large b value may represent large water retention capacity. Because the b value of B1 horizon was highest and that of C horizon lowest, it could be considered

Table 1. The physical properties of Ap1, B1 and C horizon

Horizon	Bulk density (g cm^{-3})	* Organic matter (g kg^{-1})	Gravel (%)	** Particle size distribution (%)			Textural class	Saturated hydraulic conductivity (cm hr^{-1})
				Sand	Silt	Clay		
Ap1	1.35	15.6	10.87	54.40	22.51	23.09	SCL	2.00
B1	1.54	1.3	9.76	58.58	15.21	26.21	SCL	0.27
C	1.32	0.7	14.57	73.26	19.37	7.37	SL	5.24

* Organic matter : Walkley-Black method

** Particle size distribution : Pipet method

Table 2. The chemical properties of Ap1, B1 and C horizon

Horizon	Cation exchage capacity (cmol$^+$ kg^{-1})	** Exchageable cation (cmol$^+$ kg^{-1})					Base saturation (%)
		Ca	Mg	K	Na	Cu	
Ap1	15.15	2.18	1.50	1.12	0.04	*ND	31.90
B1	12.60	062	0.51	0.34	0.05	*ND	12.09
C	8.84	0.27	0.29	0.13	0.04	*ND	8.24

* ND : Not detect

** Exchangeable cations : 1N ammonium acetate extraction method that the water retention capacity was highest in B1 horizon and lowest in C horizon. The maximum pore size in each horizon can be calculated from air entry pressure (h_a). The pore diameter equivalent to h_a of Ap1, B1 and C horizon was 466.4 ㎛, 348.81 ㎛ and 230.95 ㎛, respectively, by using eq. (3). This suggests that Ap1 horizon can have more macropores than other horizons. In cumulative pore distribution (Fig. 1), pores larger than 148 ㎛ in Ap1 horizon occupied about 13% of total porosity, which is about two times of that of B1 horizon. Macropore and macroporosity were defined as the pore size and porosity more than equivalent pore size of - 6 kPa soil water potential (McDonald, 1967). Therefore in measured data, the - 6 kPa could be set as the boundary of macropore and micropore. By this definition, macroprosity of Ap1 and B1 horizon were 24% and 14% of total porosity, respectively. It can be considered that the granular structure of Ap1 horizon and the subangular structure of B1 horizon contributed to macroporosity.

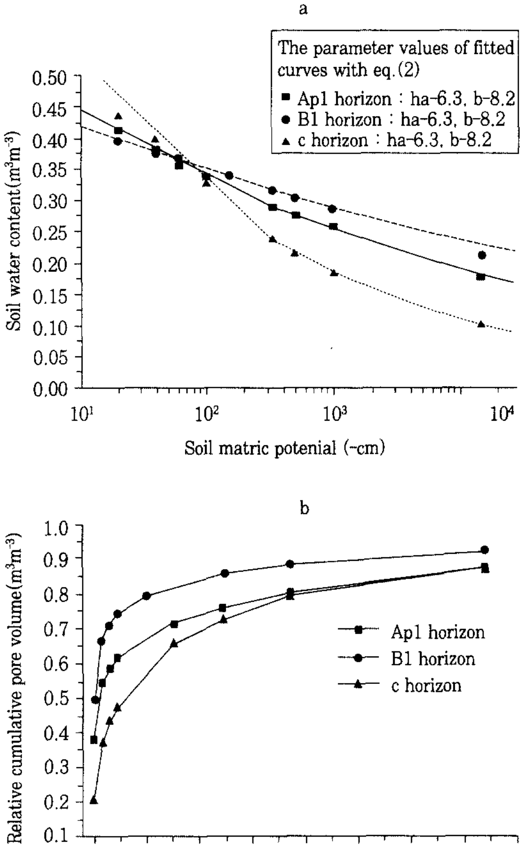

Fig. 1. The water retention curves(a) and cumulative pore size distribution(b) of Apl, B1, C horizon.

2. Copper and chloride BTCs of saturated undisturbed cores

Miscible displacement was carried out by using undisturbed cores of Apl and B1 horizon except C horizon which has little limitation of transport processes. Water flux of Ap1 and B1 cores was controlled less than saturated hydraulic conductivity because ponding water would hardly occur due to good drainage of soil profile.

Chloride BTCs of Ap1 and B1 core is shown in figure 2. At one pore volume, the relative concentration of BTC in B1 core was about 0.5 whereas in Apl core was 0.85. The hydrodynamic dispersion coefficient (D) of Ap1 core was about forty times larger than that of B1 core (Table 3.). The retardation factor (R) of B1 core was 1, where suggest that immobile fraction of pore water did not exist in B1 core. But in Ap1 core having 0.60 of the retardation factor, immobile fraction of pore water was 40% of total porosity. Initial breakthrough was also about eight times faster in Ap1 than

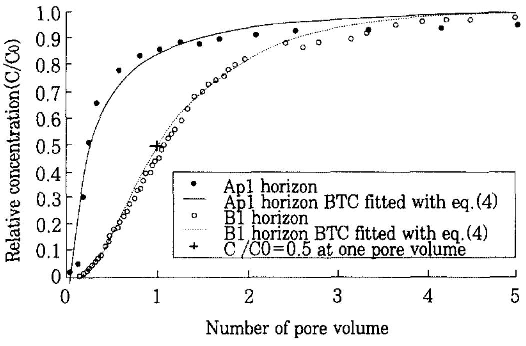

Fig. 2. The chloride breakthrough curves of undisturbed cores from Apl and B1 horizon.

Table 3. The parameters of chloride breakthrough curves(BTCs)

Horizon	Ap1	B1
Water flux (cm hr^{-1})	1.43	0.25
Porosity (m^3 m^{-3})	0.49	0.42
Average pore water velocity, v (cm hr^{-1})	3.10	0.59
Hydrodynamic dispersion coefficient, D(cm^2 hr^{-1})	33.95	1.29
Retardation factor, R	0.60	1.00
Immobile pore water fraction, (1-R)	0.40	0.00

B1 core. In the type of breakthrough curves, the chloride BTC of Ap1 core had large non-symmetry and slope of initial chloride concentration. Bouma (1991) reported that the type such as chloride BTC of Ap1 corresponded with occurrence of macropores and the type such as chloride BTC of B1 core with a heterogeneous pore system.

Tailing of BTC at more than three pore volume was more significant in Ap than in B1 core. This phenomenon indicates that the increase of relative concentration can be limited by an effect of diffusion-limited exchange between the immobile intrasolution and the rapidly percolating interaggregate solutions (Sollins et al., 1988).

The mobility of copper was less than chloride because cation adsorbs soil particle surface having negative charge. So the relative concentration of copper BTCs was much less than that of chloride BTCs at same pore volume. Figure 3 shows that initial breakthrough of copper was faster in Ap1 core than in B1 core and increase rate of relative concentration was also larger in Ap1 core than in B1 core. This suggests that the retardation effect of copper was greater in Ap1 core than in B1 core even though the cation exchange capacity of Ap1 horizon was higher than that of B1 horizon.

Thus, BTCs of Cl^- and Cu^{2+} obviously showed the probability that preferential flow would occur in Ap1 horizon. But in B1 horizon of clay accumulated layer preferential flow did not occur. So preferential flow through whole soil profile could rarely occur because it could be limited by B1 horizon even though it occur in Ap1 horizon.

Preferential water flow caused by aggregation and macropore development may serve to prevent nutrient leaching in many ecosystems, for example, forest soils (Solins et al., 1988). But in cultivated macroporous soil applied agrochemicals such as fertilizers and pesticides, when heavy rainfall or excessive irrigation happens, these dissolved agrochemicals can leach rapidly as preferential flow (Edward et al., 1992). Especially, the chemical-containing water leaching by preferential flow would be significant in macroporous soils which do not have a limited layer, for example, Bt horizon.

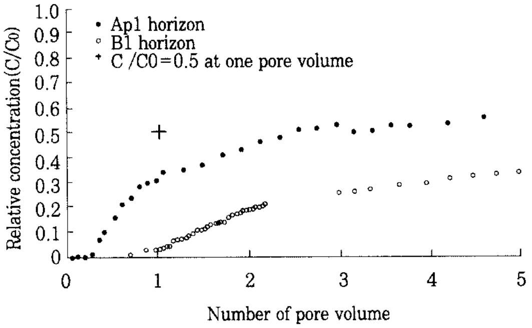

Fig. 3. The copper breakthrough curves of undisturbed cores from Ap1 and B1 horizon.

References

Beven, K. and P. Germann. 1982. Macropores and water flow in soils. Water Resource Res. 18:1311-1325.

Booltink, H. W. G. and J. Bouma. 1991. Physical and morphological characterization of bypass flow in a well-structured clay soil. Soil Sci. Soc. Am. J. 55:1249-1254.

Bouma, J. 1991. Influence of soil macroporosity on environmental quality. Advances-in-Agronomy(USA). 46:1-37.

Bouma, J., L.W. Dekker and C.J. Muilwijk. 1981. A field method for measuring short-circuiting in clay soils. J. Hydrol. 52:347-354.

Campbell, G. S. 1974. A simple method for determining unsaturated conductivity from moisture retention data. Soil Sci. 117:311-314.

Chen, C. and R. J. Wagenet. 1992. Simulation of water and chemicals in macropore soils Part 1. Representation of the equivalent macropore influence and its effect on soil water flow. J. Hydrol. 130:105-126.

Danielson, R. E. and P. L. Sutherland. 1986. Porosity In methods of soil analysis, part 1. A. Klute(ed.). Monograph No. 9, ASA, Madison, WI.

Edwards, W. M., W. A. Shipitalo, M. J. Dick and L. B. Owens. 1992. Rainfall intensity affects transport of water and chemicals through macropores in no-till soil. Soil Sci. Soc. Am. J. 56:52-58.

Jardine, P. M., G. K. Jacobs. and G. V. Wilson. 1993. Unsaturated Transport Processes in undisturbed heterogeneous porous media : 1. Inorganic contaminants. Soil Sci. Soc. Am. J. 50:945-943.

Kirkham, D. and W. L. Powers. 1972. Chapter 8. Miscible displacement. Advanced soil physics. p379-427. Wiley-Interscience.

Li, Y. and M. Ghodrati. 1994. Preferential transport of nitrate through soil columns containing root channels. Soil Sci. Soc. Am. J. 58: 653-659.

Marion, J. M., D. Or, D. E. Rolston, M. L. Kavvas and J. W. Biggar. 1994. Evaluation of methods for determining soil water retentivity and unsaturated hydraulic conductivity. Soil Sci. 158:1-13.

Mcdonald, P. M. 1967. Disposition of soil moisture held in temporary storage in large pores. Soil Sci. 103(2):139-143.

Phillips, R. E., V. L. Quisenberry and J. M. Zeleznik. 1995. Water and solute movement in an undisturbed, macroporous column : Extraction pressure effects. Soil Sci. Soc. Am. J. 59: 707-712.

Shipitalo, M. J. and W. M. Edwards. 1993. Seasonal patterns of water and chemical movement in tilled and no-tilled column lysimeters. Soil Sci. Soc. Am. J. 57:218-223.

Shipitalo, M. J., W. M. Edwards, W. A. Dick and L. B. Owens. 1990. Initial storm effects on macropore transport of surface-applied chemicals in no-till soil. Soil Sci. Soc. Am. J. 54:1530-1536.

Singh, P. and R. S. Kanwar. 1991. Preferential solute transport through macropores in large undisturbed saturated soil columns. J. Environ. Qual. 20:295-300.

Sollins, P. and R. Radulovich. 1988. Effects of soil physical structure on solute transport in a weathered tropical soil. Soil Sci. Soc. Am. J. 52:1168-1173.

Tyler, D. D. and G. W. Thomas. 1981. Chloride movement in undisturbed soil columns. Soil Sci. Soc. Am. J. 45: 459-461.

van Genuchten, M. Th. and P. J. Wierenga. 1976. Mass transfer studies in sorbing porous media Ⅰ. Analytical solutions. Soil Sci. Soc. Am. J. 40:473-480.

van Genuchten, M. Th. and P. J. Wierenga. 1986. Solute dispersion coefficients and retardation factors In methods of soil analysis, part1. A. Klute(ed.). Monograph No. 9, ASA, Madison, WI.

White, R. E. 1985. The influence of macropores on the transport of dissolved and suspended matter through soil. Advanced Soil Science 3:95-120.

韓土肥誌(2000)
Vol. 33(2) : 71~78

Preferential Flow as Tested by Breakthrough Curves of Cl^- and Cu^{2+} from Saturated Undisturbed Soil Core Samples under Steady Flow Conditions

Sun-Ho Yoo*, Kyung-Hwa Han*, Hee-Myong Ro** and Gwang-Hyun Han*

포화 불교란 토양시료의 Cl^- 및 Cu^{2+} 출현곡선에 의한 preferential flow의 검증

류순호* · 한경화* · 노희명** · 한광현*

사과 과수원 토양(송정통, the fine loamy, mesic family of Typic Hapludults)을 층위별로 채취하여 토양 수리적 특성과 누적공극분포를 조사하고 표토인 Ap1층과 점토집적층인 B1층에서 각각 토양코아시료(지름 7.4 cm, 높이 7.4 cm)를 채취하여 포화조건에서 Cl^- 와 Cu^{2+}로 혼성치환실험을 수행하고 그 출현곡선으로부터 選擇流를 검증하고자 하였다. Ap1층과 B1층은 사질 식양토로 같은 토성이나 Ap1층의 포화수리전도도는 2.0 cm hr^{-1}로 B1층(0.27 cm hr^{-1})의 약 7배에 달하였다. C층은 포화수리전도도 5.2 cm hr^{-1} 인 구조가 없는 사양토로 물과 용질의 이동을 제한하는 역할이 가장 낮을 것으로 판단되었다. 누적 공극분포에서 Ap1층과 B1층의 대공극량(토양수분포텐셜 - 6 kPa에 해당하는 지름 49㎛ 이상의 공극)은 각각 총 공극량의 24%, B1층은 14%이었다. 염소출현곡선에서 Ap1층 코아와 B1층 코아의 수력학적 분산계수는 각각 34 cm^2 hr^{-1}, 1.3 cm^2 hr^{-1}이었고 이때 B1층 코아의 지연계수는 1인 반면 Ap1층 코아는 0.6으로 부동수분과 유동수분의 분배가 일어났다. 양이온 치환용량이 Ap1층이 B1층보다 높음에도 불구하고 Ap1층 코아의 구리 지연효과가 B1층보다 더 작게 나타났다. 이 결과는 포화수리전도도와 대공극 함량이 큰 Ap1층에서 選擇流가 일어날 수 가능성을 보여 주었다. 그러나 투수성이 좋은 모재층을 가진 이 토양단면에서 Ap1층에서 選擇流가 일어난다 하더라도 B1층에 의해 제한될 것으로 사료된다.

Key words : Preferential flow, Miscible displacement, Breakthrough curves, Macropore, Pore size distribution.

---

* 서울대학교 농업생명과학대학 (College of Agriculture and Life Sciences, Seoul National University, Suwon, 441-744, Korea)
** 농촌진흥청 원예연구소 원예환경과 (Dept. of Horticultural Env. National Horticultural Research Institute)

※ 본 연구는 농림기술관리센터에서 주관하는 농림기술개발연구과제 중 첨단농업기술개발사업의 일환으로 수행되었으므로 연구비 지원에 감사드립니다.