

The Interference of Organic Matter in the Characterization of Aquifers Contaminated with LNAPLs by Partitioning Tracer Method

LNAPLs 오염 지반에 분배성 추적자 시험법 적용 시 유기물질의 영향에 관한 연구

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요 지

분배성 추적자 시험법은 LNAPLs(light nonaqueous phase liquids)로 오염된 지반을 조사하는데 아주 유용한 방법이다. 하지만 토양 내 유기물질로 흡착되는 분배성 추적자는 잠재적으로 분배성 추적자 시험법의 정확성에 영향을 끼칠 수 있다. 연구 결과, 추적자의 액상-LNAPL 간 분배 계수는 선형 관계를 보였다. 토양의 흡착능력을 평가하기 위해 흡착 등은 실험을 수행한 결과, Freundlich 흡착 등은 양상과 거의 일치하였고, 추적자의 흡착 정도는 토양 내 유기물질 함량이 증가함에 따라 증가하였다. 또한, 토양 유기물의 흡착능에 따른 잠재적 영향을 판단하고, 추적자 시험법에 의한 LNAPLs 예측의 오차를 수정하기 위해 서로 다른 유기물 함량을 가진 4개의 컬럼 실험을 수행하였다. 컬럼 실험 결과, 오염물질이 없더라도 주문진 표준사와 유기물질이 섞인 컬럼에서는 추적자의 분리 현상이 발생하였다. 오염물질로 케로진을 주입한 이후에 다시 추적자 시험법을 수행하여 파괴곡선을 구한 결과, 토양 유기물질에 대한 추적자의 흡착으로 인해 추적자의 지연계수(R)가 커졌고 LNAPLs의 오염도가 과대평가되었다. 또한 컬럼 실험 결과를 바탕으로 유기물 함량과 LNAPLs의 예측도 사이의 관계식을 제안하였다.

Abstract

Partitioning tracer method is a useful tool to characterize large domains of the aquifers contaminated with light nonaqueous phase liquids (LNAPLs). Sorption of the partitioning tracers to the organic matter content of soil can potentially influence the efficacy of partitioning tracer method. LNAPL-water partitioning coefficients of tracers (K_{nw}), measured by static method, showed linear relationship. Sorption isotherm tests were conducted to evaluate the sorption capacity of the soils packed in the columns and the results were appropriately represented by Freundlich sorption isotherm. The sorption of tracers proportionally increased with the increase of the organic matter content of the soil. Laboratory experiments were conducted in four columns each packed with soils of different organic matter contents to determine the potential interference effects of sorption to soil organic matter content and correction factors for the errors in estimation of LNAPLs by partitioning tracer method. Though there were no contaminants added, breakthrough curves from columns packed with mixture of Jumunjin standard sand and organic matter showed separation of tracers. Columns were then contaminated to residual saturation with kerosene and breakthrough curves were obtained. The results show that sorption of tracers to soil organic matter leads to an increase in the retardation factor (R) and hence, to an overestimation of the saturation of LNAPLs. A relation between the percentage of organic matter content and the corresponding percentage error in the estimation of NAPLs has been developed.

Keywords : Column Test, LNAPLs Monitoring, Partitioning Tracer Test, Soil Organic Matter, Sorption Test, Tracer Test

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1. Introduction

Sub-Surface contamination by light nonaqueous phase liquids (LNAPLs) has proven to be a formidable challenge for environmental engineers and its presence is often the single most important factor limiting remediation of sites contaminated by organic compounds (National Research Council, 1994). Potential for further contamination is also a concern when considering the presence of LNAPLs sources in the subsurface (e.g. underground storage tanks and oil/gas pipelines). Proper characterization, which involves the location, composition and quantification of LNAPLs, is required for accurate risk assessments and effective remediation (Jacksons and Pickens, 1994). As many LNAPLs are both sparingly soluble and highly mobile, assessing their time-varying concentrations and sub-surface distribution can be extremely difficult, particularly in complex, near-surface industrial environment. Furthermore, considering the various constraints to mass transfer and the generally low maximum contaminant levels, LNAPLs are now widely accepted to be a long-term source of both vapor phase and ground water contamination (National Research Council, 1994).

Light NAPLs remain floating on the top of the capillary fringe but the fluctuation of the water table creates a smear zone of LNAPLs at the residual saturation within the upper portion of the saturated subsurface. It is due to these unusual behaviors that make the detection and quantification of LNAPLs extremely difficult in the subsurface with the point sampling techniques of site characterization like core sampling, cone penetrometer, and geophysical logging (Cain et al., 2000). The sample of these methods has relatively small volume of the subsurface and thus, accurate characterization of the given domain is very difficult without a cost prohibitive amount of sampling. Thus, partitioning tracer method has been proposed as a means to characterize the sites contaminated with LNAPLs. This method, with a particular advantage over the point sampling techniques covers large volume of the subsurface, produces more reliable estimates of the quantity and distribution of LNAPLs (Jin et al., 1995). Partitioning tracer method is based on performing a tracer

test in the subsurface of the site, potentially contaminated with LNAPLs. Chemical tracers with known NAPL-water partition coefficients are injected into the subsurface to detect the presence of LNAPLs and to estimate LNAPLs' saturation within the zone swept by the tracers. The LNAPLs reversibly retain the partitioning tracers with respect to nonpartitioning tracers causing the former to lag behind the later. The extent of separation depends on the residence time of tracers, which are a function of its partition coefficient and the saturation of LNAPLs. Thus, the magnitude of measured separation of the partitioning tracers can be translated into quantifying NAPLs present within the zone swept through by the tracers (Jin et al., 1995).

The partition tracer method can be used as an innovative and effective technique for the detection and quantification of LNAPLs' contamination in the subsurface as well as to evaluate the remediation performance. However, the results can be affected by many factors such as rate limited transfer, subsurface heterogeneities, multiphase retention, biochemical degradation, and sorption on to the soil organic matter of chemical tracers, which leads to inappropriate characterization of the site under consideration (Brusseau et al., 2003). Hence, partition tracer method needs to be evaluated to determine the effect of influencing factors. The purpose of this paper is to present the effect of sorption of the chemical tracers to the soil organic matter on partition tracer method from column scale experiments in laboratory. A comparison of the saturation of LNAPLs determined from partition tracer test conducted in a column packed with Jumunjin standard sand and columns packed with different weight ratios of Jumunjin standard sand and organic matter provided a measure of the effect of sorption.

2. Theory and Analysis Techniques

2.1 Method of Moments

It has been shown that the method of moment's theory can be used to determine LNAPLs' saturations in a subsurface, given the difference in mean residence times between two different tracers by using partitioning tracer method (Jin et al., 1995). Partitioning tracer method is

based on the chromatographic separation of two or more selected tracers as they flow with ground water through the LNAPLs' contaminated aquifer. The partition coefficient (K_{nw} , unitless) of a partitioning tracer quantifies the fraction of tracer in LNAPLs' phases and water phase at equilibrium (Jin et al., 1995). It is defined as the ratio of the concentration of tracer in the LNAPLs phase (C_n , unit: mg/L) to the concentration of tracer in the water phase (C_w , unit: mg/L), or

$$K_{nw} = C_n / C_w \quad (1)$$

Nonpartitioning tracers have a partition coefficient of zero with respect to LNAPLs, whereas the partitioning tracers have partition coefficients with non zero positive value. A set of partitioning and nonpartitioning tracers is selected to get the greatest degree of separation between tracer pairs in a reasonably short period of time. The magnitude of retardation is a function of LNAPLs saturation and partition coefficient (Brusseau et al., 2003). The R value (unitless) determined from the tracer test is equated to the mass-balance definition of R , given for aqueous-phase transport as:

$$\frac{t_p}{t_n} = R = 1 + \left(\frac{\rho_b}{\theta_w} \right) K_d + \left[\frac{S_n}{(1 - S_n)} \right] K_{nw} \quad (2)$$

Where, t_p is the travel time of the partitioning tracer, t_n is the travel time for the nonpartitioning tracer, ρ_b is bulk density of porous media (unit: g/cm³), θ_w is the water-filled porosity (unitless), K_d is the water-aquifer solids partition coefficient (unit: cm³/g), and S_n is the effective LNAPLs' saturation (unitless) degree.

The previous researchers have neglected the second term on the right side of the equation (2) assuming that liquid-liquid partitioning is much greater than the liquid-solid partitioning. It was further supported that the large volume of the LNAPLs in the subsurface would cause the liquid - liquid partitioning to dominate tracers' retardation (Cain et al., 2000). No previous research has been carried out to support this assumption. We have considered the second term i.e. sorption, in our calculations to evaluate the authenticity of this assumption. The sorption can occur with organic matter or clay material.

But, this study focuses on the effect of organic matter. The detailed procedure for the quantification of LNAPLs using partitioning tracer theory can be found in Jin et al. (1994, 1995).

The total volume of LNAPLs in subsurface may often be underestimated using partitioning tracer method because of factors such as rate-limited transfer, bypass flow and mass loss, but the contrary can be true in case of sorption of the tracers to soil organic matter (Hatfield and Stauffer, 1993). We have demonstrated how the saturation of LNAPLs is an overestimate of the true value because of sorption to the soil organic matter using columns packed with selected soils of known sorption capacities.

3. Material and Methods

3.1 Composition of Soil

Jumunjin standard sand was packed in column 1 and mixture of Jumunjin standard sand and organic fertilizer in columns 2-4 in ratios 19:1, 9:1, and 4:1 respectively. Organic fertilizer, passing No. 10 sieve (0.200 mm opening size) and retained by No. 40 sieve (0.420 mm opening size), was used to represent the soil organic matter. The organic matter content in the organic fertilizer and Jumunjin standard sand was determined by the loss-on-ignition method (Veres, 2002) and was found to be 5.35% and 0.50% respectively. X-Ray Fluorescence (XRF) analysis and X-ray Diffraction (XRD) analysis of Jumunjin standard sand and organic fertilizers were conducted and are given in Table 1. Jumunjin standard sand and its mixture with organic fertilizer in different ratios were used to ascertain the effect of concentration of the organic matter content on tracers' breakthrough curves. Jumunjin standard sand was used in column marked as number "1" and the columns marked as "2, 3 and 4 were packed with the mixture of organic fertilizer and Jumunjin standard sand with organic matter content of 2.64, 5.29 and 10.58, respectively.

3.2 Tracers

Methanol and chloride ions were used as non-

Table 1. Composition of Jumunjin standard sand and organic fertilizer from XRF and XRD analysis

Jumunjin standard sand				Organic fertilizer			
XRF analysis		XRD analysis		XRF analysis		XRD analysis	
Weight (%)		Weight (%)		Weight (%)		Weight (%)	
SiO ₂	88.50	Quartz	74.70	SiO ₂	60.04	Quartz	43.80
Al ₂ O ₃	6.59	Plagioclase	7.20	Al ₂ O ₃	11.85	Plagioclase	7.70
TiO ₂	0.08	K-feldspar	18.10	TiO ₂	0.57	K-feldspar	14.10
Fe ₂ O ₃	0.04	Muscovite	0.00	Fe ₂ O ₃	6.55	Muscovite	16.40
MgO	0.02	Calcite	0.00	MgO	1.98	Calcite	2.70
CaO	0.21	Goethite	0.00	CaO	7.34	Goethite	15.10
Na ₂ O	0.12			Na ₂ O	0.54		
K ₂ O	3.81			K ₂ O	3.78		
MnO	0.03			MnO	0.37		
P ₂ O ₅	0.02			P ₂ O ₅	1.33		
Loss on ignition	0.50			Loss on ignition	5.35		

partitioning tracers while 2-ethyl-1-butanol, 4-methyl-2-pentanol, 1-pentanol, 1-hexanol, and 2,3-dimethyle-2-pentanol were used as partitioning tracers in the column experiments and were chosen to yield breakthrough curves in a reasonably short time, and yet they ensured good separation of the partitioning and nonpartitioning tracers (Varandarajan and Garry Pope, 1998). Kerosene dyed with Sudan IV was used as a representative LNAPLs in columns packed with selected soils.

3.3 Batch-Partitioning Experiments

Batch-partitioning experiments were conducted to determine kerosene-water partition coefficients (K_{nw}) for the group of selected tracers. Batch partitioning tests, in 20 mL septa-capped vials with equal volumes of kerosene and water (10 mL each), were conducted with methanol, 2-ethyl-1-butanol, 4-methyl-2-pentanol, 1-pentanol, 1-hexanol, and 2,3-dimethyle-2-pentanol ranging in concentration from 50 to 800 mg/L. Vigorous mixing on an orbital mixer for thirty one hours equilibrated the vials. Following equilibration, a 2 mL aqueous sample was collected via syringe after centrifuging the sample in the centrifuge for 30 minutes at 3500 rpm, and placed into a 2 mL septa vial for alcohol analysis with a Hewlett-Packard (HP) 6890 gas chromatograph (GC). The GC was equipped with a 30.0 m long by 0.25 mm PAG capillary column (Supelco 2-4223) and a flame ionization detector (FID). The FID signal was acquired and integrated with personal

computer (PC) using HP Chemstation software.

3.4 Sorption Isotherm Experiments

Sorption isotherm experiments were conducted to determine soil-water partition coefficients of tracers and the sorption capacity of the selected soils. The tests were conducted in 20 mL, septa-capped vials with 4 grams of the selected soil and measured amount of the tracer solution to make the head space almost zero. Vigorous mixing on an orbital mixer for 48 hours equilibrated the vials. Following equilibration, a 2 mL aqueous sample was collected via syringe after centrifuging the sample in the centrifuge for 40 minutes at 3800 rpm, and placed into a 2 mL septa vial for alcohol analysis with GC.

3.5 Column-Scale Experiments

Figure 1 shows the schematic diagram of the equipment setup for column experiment. Jumunjin standard sand and its mixture with organic fertilizer were packed under dynamic compaction in the glass columns, each 40 cm long and with an inner diameter of 3.5 cm.

Packed columns were saturated with DI (deionized) water at a constant flow rate of 0.1 ml/min after purging by CO₂ gas to remove the air bubbles and get full saturation of the packed soils. The flow rate was slow enough to give sufficient time for reversible reaction to occur between tracers and the other media. Sodium azide

solution was injected to kill the bacteria in the packed columns as they may biodegrade the chemical tracers. Tracer test in the columns was conducted before and after contamination with kerosene. A known volume of dyed kerosene was injected as a representative of the LNAPLs contamination. Residual contamination was insured by continuously injecting DI water until there was no movement of the dyed kerosene. The method of volume measurement was adopted to determine the volume of residual kerosene saturation in the columns which involves the measurement of the volume of the injected kerosene and the volume produced during the DI water flood (Jin et al., 1995). The tracers' pulse was 0.15 pore volume for all the columns. Effluents were collected every hour and were analyzed with GC.

4. Results and Discussions

4.1 Batch Tests and Tracer Screening

The results of batch-partitioning experiments conducted

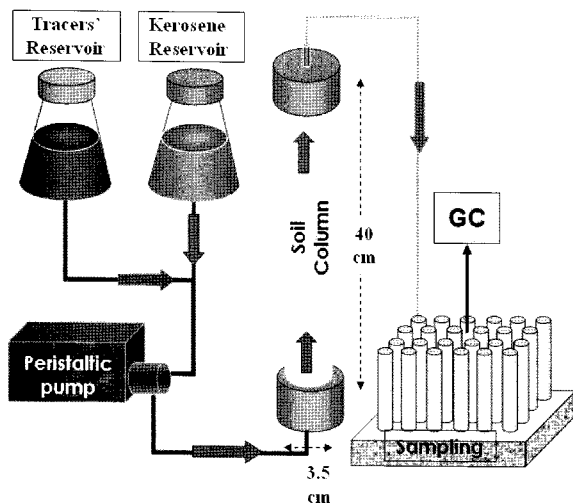


Fig. 1. Schematic diagram of column setup and sampling

to determine kerosene-water partition coefficients for the selected group suite of alcohol tracers are shown in Table 2. The Results in Figure 2 indicate that kerosene-water partitioning is linear with respect to alcohol tracers' concentrations employed in this study. Measured partition coefficients, reflected as the slope of the linear trend, are constant with increasing aqueous tracer concentration. The retention times from our GC analysis for methanol, 2-ethyl-1-butanol, 4-methyl-2-pentanol, 1-pentanol, 1-hexanol, and 2,3-dimethyl-2-pentanol were 1.64, 7.13, 4.76, 4.78, 7.14, and 8.27 respectively. The results from batch tests and a column test were screened to determine the most suitable group of tracers for producing breakthrough responses in a reasonably short time and yet ensuring good separation of tracers. 1-Pentanol and 2,3-dimethyl-2-pentanol were discarded because of its similar retention time with 4-methyl-2-pentanol and 2-ethyl-1-butanol respectively and hence, their peaks cannot be differentiated in the GC. Another reason for discarding 2,3-dimethyl-2-pentanol was that it was restrained for unreasonably long

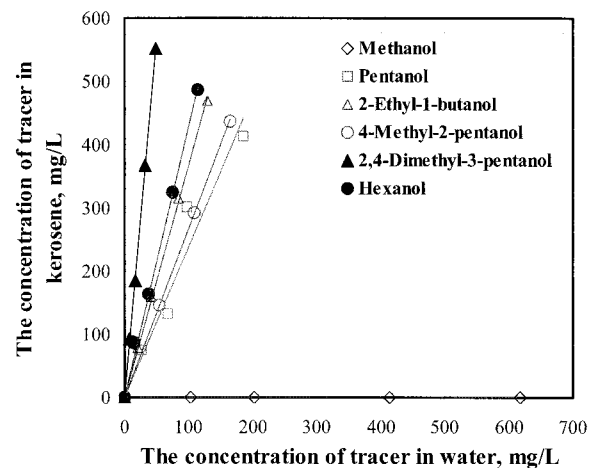


Fig. 2. Partitioning of tracers between kerosene and water

Table 2. Kerosene-water partitioning coefficients of tracers

Tracer	Formula	Molar mass (g/mol)	K_{nw}	R^2
Methanol	CH_3OH	32.04	0.003	0.9204
1-Pentanol	$\text{CH}_3(\text{CH}_2)_4\text{OH}$	88.15	2.276	0.9233
1-Hexanol	$\text{CH}_3(\text{CH}_2)_5\text{OH}$	102.17	4.293	0.9958
2-Ethyl-1-butanol	$(\text{C}_2\text{H}_5)_2\text{CHCH}_2\text{OH}$	102.18	3.656	0.9987
4-Methyl-2-pentanol	$(\text{CH}_3)_2\text{CHCH}_2\text{CH}(\text{OH})\text{CH}_3$	102.18	2.677	0.9713
2,4-Dimethyl-3-pentanol	$(\text{CH}_3)_2\text{CHCH}(\text{OH})\text{CH}(\text{CH}_3)_2$	116.20	11.257	0.9996

time in the columns packed with mixture of selected soils. Measured partition coefficients were used in conjunction with measured partitioning tracer retardations to predict kerosene volume within zone swept by the tracers.

4.2 Sorption Isotherm Experiments

The results of sorption isotherm experiments conducted to determine soil-water partition coefficients for the selected group of alcohols are shown in Table 3. Freundlich sorption isotherm is used which is mathematically expressed as below:

$$S=KC^N \quad (3)$$

Where, S is the mass of the tracers sorbed per unit dry mass of solid in mg/kg, C is the concentration of the tracers in solution at equilibrium in mg/L, and K is the distribution coefficient in L/kg.

The equation (3) is linearized by plotting log of C verses log S. The slope of the straight line is N and the intercept is equal to log K. It is the most commonly used isotherm in contaminant migration analysis and is generally valid at low contaminant concentration ranges. Results indicate the solute concentration sorbed onto the soil and the concentration of the tracer in solution phase in equilibrium. The capacity of the soil to remove the traces i.e. solutes is a function of the concentration of the solute within the same test soil. The sorptive process

is rapid initially but an equilibrium condition of solute is reached with the amount sorbed onto the soil within certain duration. The results clearly demonstrate that the sorption capacity of Jumunjin standard sand is negligible and that its mixture with organic fertilizer shows considerable sorption capacity. The sorption of tracers increased proportionally to the percentage of soil organic matter content.

4.3 Column Experiments

The breakthrough curves obtained from column 1 (packed with Jumunjin standard sand) show no separation of tracers as given in Figure 3, which indicate that there is no partitioning of tracers to the media swept through

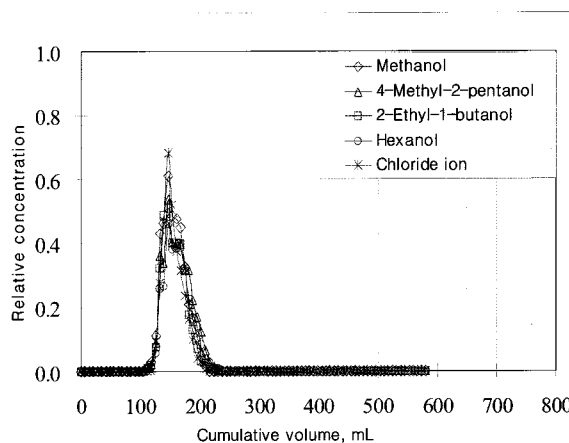


Fig. 3. Breakthrough curves from pre-contaminated column 1 which has 0% organic matter content

Table 3. Results from sorption isotherm experiments using Freundlich isotherm

Column number	Organic matter content (%)	Tracers	K_f	N	R^2
1	0.00	4-Methyl-2-pentanol	1.7814	0.3011	0.7646
		Hexanol	2.3799	0.1809	0.9765
		2-Ethyl-1-butanol	-0.9913	1.3293	0.9990
2	2.64	4-Methyl-2-pentanol	1.7814	0.3011	0.9773
		Hexanol	2.3799	0.1809	0.6671
		2-Ethyl-1-butanol	-0.9913	1.3293	0.9187
3	5.29	4-Methyl-2-pentanol	1.7814	0.3011	0.9514
		Hexanol	2.3799	0.1809	0.9351
		2-Ethyl-1-butanol	-0.9913	1.3293	0.9772
4	7.93	4-Methyl-2-pentanol	1.7814	0.3011	0.8518
		Hexanol	2.3799	0.1809	0.9907
		2-Ethyl-1-butanol	-0.9913	1.3293	0.9845

by the tracers and hence no contamination was detected. The breakthrough curves from the pre-contaminated columns 3~5 which contain known percentage of organic matter content, show significant separation of tracers and a marked increase in the separation of tracers was observed with the increase in the percentage of organic matter content as shown in the Figures 4~6. It is evident from this phenomenon that partitioning of tracers occurred only between water and organic matter content of soil as the columns were precontaminated and that Jumunjin standard sand caused no separation of tracers. A situation like this can be quite misleading as it can be taken for contamination in the subsurface or, at least, exaggerate the quantity of the contaminants.

The breakthrough curves from the post-contaminated columns are given in Figures 7~10 and measured versus

predicted volumes of kerosene are shown for homogeneously distributed residual saturation of kerosene in different columns in Table 4. The retardation factor is

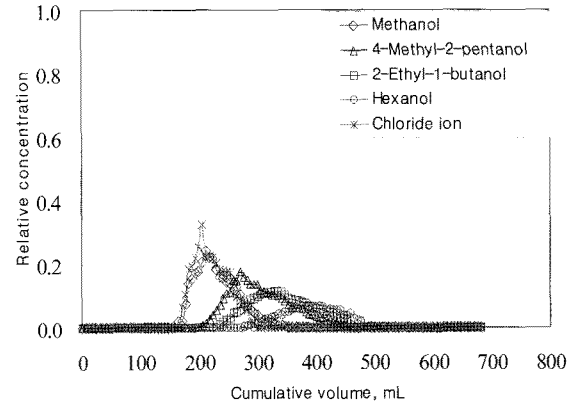


Fig. 6. Breakthrough curves from pre-contaminated column 4 which has 10.58% organic matter content

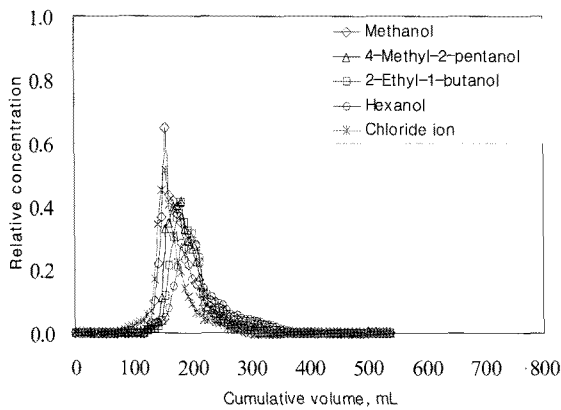


Fig. 4. Breakthrough curves from pre-contaminated column 2 which has 2.64% organic matter content

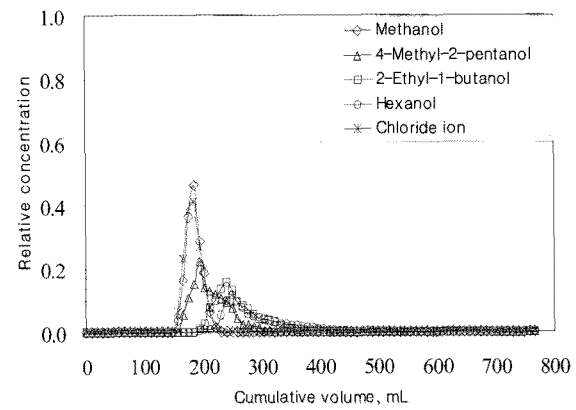


Fig. 7. Breakthrough curves from post-contaminated column 1 which has 0% organic matter content

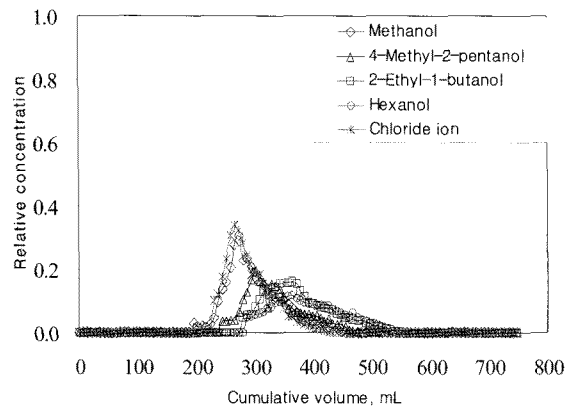


Fig. 5. Breakthrough curves from pre-contaminated column 3 which has 5.29% organic matter content

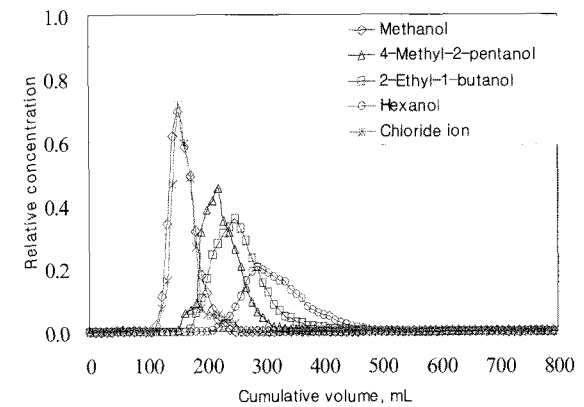


Fig. 8. Breakthrough curves from post-contaminated column 2 which has 2.64% organic matter content

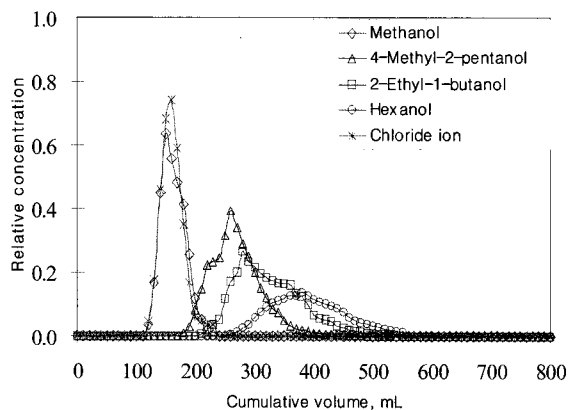


Fig. 9. Breakthrough curves from post-contaminated column 3 which has 5.29% organic matter content

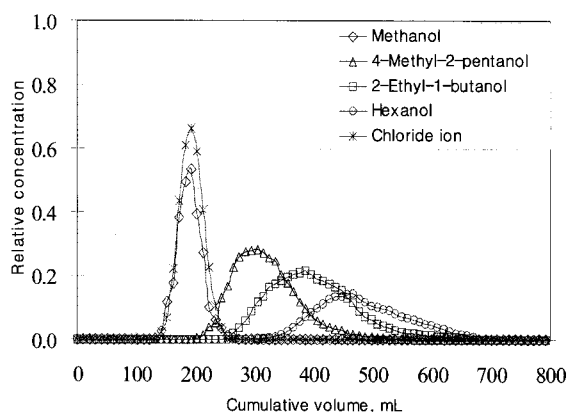


Fig. 10. Breakthrough curves from post-contaminated column 4 which has 10.58% organic matter content

between 1.2 and 4 for all the partitioning tracers which are desirable for partitioning tracer method to have appropriate estimation of kerosene in the subsurface. The magnitude of retardation is a function of the kerosene saturation and the partition coefficients for column 1 as the partitioning of tracer to the Jumunjin standard stand is negligible but in the cases of columns 2~4, sorption of the tracers to the soil organic contents also contributes to the magnitude of retardation. To get appropriate

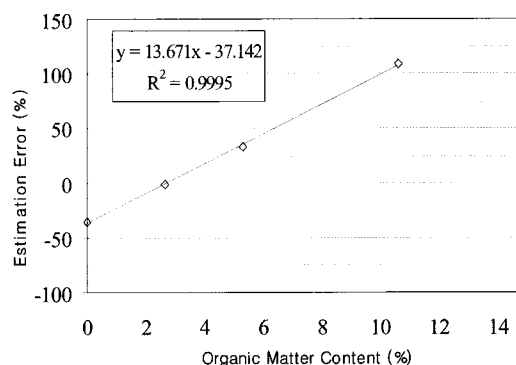


Fig. 11. The error in estimation of kerosene by partition tracer method caused by the organic matter content in the soil

estimation of the kerosene saturation, it is imperative to determine the appropriate correction factor. The hexanol breakthrough curve was retarded more with 4-methyl-2-pentanol and 2-ethyl-1-butanol. The methanol and chloride ions were used as nonpartitioning tracers and were not retarded at all. The predicted values by partitioning tracer method vary linearly from under estimation to overestimation with the increase in organic matter contents of the soil as shown in Figure 10. This significant difference is due to the sorption of tracers to the organic matter in the columns. Based on these results, the error in the estimated values of kerosene can be corrected for the known percentage of the organic matter content. From this data we have been able to express the error estimation as a function of organic content of the soil.

$$\text{Estimation Error (\%)} = 13.7 \times [\text{Organic Matter Contents}] - 37.5 \quad (4)$$

Thus, knowing only the organic content in the soil will enable us to determine the error estimation from now on.

Table 4. The percentage error in predicted values of kerosene by partitioning tracer method

Column Number	Organic matter content (%)	Measured contamination (ml)	Average estimated contamination (ml)	Percentage error
1	0.00	28.01	17.97	-35.83
2	2.64	29.35	28.87	-1.65
3	5.29	34.72	46.33	33.44
4	10.58	27.48	57.30	108.51

5. Conclusions

Most of the current subsurface characterization methods provide measurements for very small spatial domains, even for point values. While such methods can provide accurate data for small scales, their use for characterizing larger domains is generally constrained by sample-size limitations. Thus, partition tracer method that provides measurements at larger scales has been developed to complement the point-sampling methods. The experimental and theoretical basis for these tracer techniques is well established. However, there is a major concern about its application in the subsurface organic soil to provide reliable in-situ measures of the relative quantities of LNAPLs due the sorption of tracers to the organic matter. In our study, we only test with the mixture of organic fertilizer and Jumunjin standard soil to evaluate the interference of organic soil in partitioning tracer method. The tracers tests conducted in the pre-contaminated columns, with known amount of organic matter, demonstrate that tracers can be retarded with marked separation. Thus, the accurate quantity estimation of LNAPL using partitioning tracer test should be considered by conducting partitioning tracer test without soils containing organic matter. The presence of organic matter caused a linear increase in the overestimation of the predicted values and thus can be corrected for the known percentage of the organic matter in the subsurface soil under given conditions.

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