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Study of penetration behavior of PCB-DNAPL in a sand layer by a column experiment

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HIGHLIGHTS

- Infiltration performance of PCB-DNAPL in a sand layer was investigated.
- Residual PCB-DNAPL under saturated and unsaturated conditions was measured.
- Residual PCB-DNAPL in the PCB–air phase was more abundant than in the PCB–water phase.
- PCB-DNAPL was not directly transported but dissolved in an aqueous phase.
- PCB infiltration rate is governed by the degree of PCB saturation in a sand layer.

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ABSTRACT

To better understand the infiltration performances of high concentration PCB oils (KC-300 and KC-1000 polychlorinated biphenyl (PCB) mixtures), representative dense non-aqueous phase liquid (DNAPL), under both saturated and unsaturated conditions, we conducted experiments on a sand column filled with Toyoura Standard Sand. When PCB oil with the volume comparable to the total porosity in the column was supplied, the residual PCB concentrations under PCB–water conditions were 4.9×10^4 mg kg^{-1} in KC-300 and 3.9×10^4 mg kg^{-1} in KC-1000. Under PCB–air conditions, residual PCB concentrations were 6.0×10^4 mg kg^{-1} and 2.4×10^5 mg kg^{-1} in the upper and lower parts for KC-300 and 3.6×10^4 mg kg^{-1} and 1.5×10^5 mg kg^{-1} in those for KC-1000, respectively, while the rest of the PCBs were infiltrated. On the other hand, when a small amount of PCB oil with the volume far smaller than the total porosity in the column was supplied, the original PCBs were not transported via water permeation. However, lower-chlorinated PCB congeners—e.g., di- or tri-chlorinated biphenyls—preferentially dissolved and were infiltrated from the bottom of the column. These propensities on PCB oil infiltration can be explained in conjunction with the degree of PCB saturation in the sand column.

1. Introduction

Polychlorinated biphenyls (PCBs), representative dense non-aqueous phase liquid (DNAPL), are toxic persistent organic pollutants (POPs), and regarded as compounds that must be completely decomposed (Van den Berg et al., 2006). PCBs have been detected in sediments in Tokyo Bay (Shimizu et al., 2002), in the vacant lots of chemical plants that utilize PCBs (Donato et al., 2006), and at landfill sites where inappropriate PCB treatments have been conducted (Weber et al., 2011). Forty years since the use of PCB was banned, discoveries of cases where PCB-related equipment has been overlooked are increasing, especially in Japan. This has led to an awareness of increased potential risk levels of PCB-contaminated soils and sediments (Hosomi, 1993).

The transport of PCB wastes and storage of high concentration PCB oil, e.g. capacitors and transformers, represent the greatest risk for further contamination of soil and groundwater. The spill of a maximum of 21 m^3 of PCB and transformer oil containing trichlorobenzene (TCB) at the Regina Canada chemical plant diffused horizontally over a wide surface area and vertically to a soil depth of 9 m. These spilled PCBs were reported to be in an oil–liquid phase (Roberts et al., 1982). Another study investigated the adsorption of dissolved PCBs and chlorinated benzene onto a layer of silty

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clay and the associated transport delays (Anderson and Pankow, 1986). The authors concluded that the transport rates of PCBs in the silty layer are lowered in the presence of chlorinated benzenes, which prevents the PCBs from diffusing into the lower layer unless there is a crack in that layer. Given these findings, it is important to better understand the behavior of PCB oil in the ground in order to evaluate soils contaminated with PCB oil.

Numerous studies have investigated the physicochemical properties of PCBs (Mackkay et al., 1992), soils and water contaminated with low concentrations of PCBs (Girvin et al., 1997; Manz et al., 2001), behaviors of atmospheric PCBs in the environment (Mills et al., 2004), and the potential for predicting the dynamics of PCBs in the environment (Nakanishi and Ogura, 2008). Batch experiments on adsorption onto the soil layer of PCBs and chlorobenzenes at the ppb level have revealed that the amount of organic carbon in soil is clearly correlated with the distribution constant, and that compounds with a higher number of chlorine molecules in their biphenyl structures have a higher affinity for soil surface adsorption (Paya-Perez et al., 1991). A continuous column experiment, where soil was filled with low concentrations of PCBs, indicated that rainfall might accelerate the degree of contamination due to diffusion of PCBs into the lower soil layer and thus change the PCB congeners (Higashino et al., 2008). However, few studies have addressed the penetration behavior of high concentration PCB oil (capacitors and transformers as representatives, containing high percentages of PCB as compositions), which assumes that spills due to cracks or breakages of an electrical capacitors or transformers accidently occur. These equipments have been kept for more than forty years since the use of PCBs was banned, implying that a chance of such spills unfortunately gets increasing. Moreover, there are no reports on the parameterization of high concentration PCB oil by soil column experiments. Given this context, data on the PCB infiltration propensities in the ground layer are highly required, under an assumption where PCB spills from either a capacitor or transformer. Hence, we undertook this study of two PCB oils (KC-300 and KC-1000) to investigate the basic characteristics of PCBs and to better understand the infiltration properties of high concentration PCB oil in a sand column filled with Toyoura Standard Sand, based on several scenarios where large and small amounts of high concentration PCB oils were spilled under saturated or dry conditions.

2. Materials and methods

2.1. Polychlorinated biphenyl

Two commonly used PCBs in Japan, KC-300 and KC-1000, were used in this study. The concentrations of PCBs, TCBs and dioxins, and viscosities of PCBs, measured by a viscosity meter (Visco Elite-L, Fungilab, Barcelona, Spain), are shown in Table 1. The viscosities at 20 °C, at which all the column experiments were performed, are 0.043 and 0.020 Pa s for KC-300 and KC-1000, respectively. The PCBs were stained with Sudan IV (Tokyo Chemical Industry Co., Ltd., Tokyo, Japan) to allow visible observation of PCB infiltration.

2.2. Sand

Toyoura Standard Sand, used in this experiment, has a consistent particle diameter ranging from 0.1 to 0.2 mm, contains lower amounts of organic carbon, and has less capacity for adsorption.

2.3. Overview of the column experiment

A vertical one-dimensional column experiment was performed to confirm the transport behaviors of PCB oil in soil. Six acrylic columns, length of 70 cm and inner diameter of 5 cm, were constructed to ensure that our study tested two different combinations of phases for the PCB oil–water condition (saturated test: Run 1 (KC-300), Run 2 (KC-1000), Run 5 (KC-300), and Run 6 (KC-1000)) and PCB oil–air (unsaturated test: Run 3 (KC-300) and Run 4 (KC-1000)). Schematic diagrams of the column and the operational conditions are shown in Fig. 1 and Table 2, respectively. Toyoura Standard Sand, with an 80% relative density and bulk density of 1574 kg m^{-3}, filled each column up to 50 cm. Runs 1, 2, 5, and 6 each consisted of a water supply tank with a constant water level and a soil layer column connected with a resin tube and a connector to maintain a constant water pressure and ensure the saturation of the soil layer in the column. A set amount of PCBs was supplied from the top of the column and the permeate at the bottom of the column was collected on a recovery tray. The weight and volume of the recovered material were measured by an electric balance (GF-3000, AND Inc., Tokyo, Japan) and a cylinder. The tray and the balance with the column were placed into an acrylic closed box to avoid any PCB emission to the outside. All the samples—i.e., the vessel on the balance, the PCBs, and the water—were measured every minute. Because Runs 1 and 2 considered the weights of the PCBs and the water, the weight of the PCBs and the height of the PCB layer were recorded every 5 min to enable the calculation of the PCBs' from the cross-sectional area (144 cm^2), the density, and the PCB layer height. The weight of the water was measured by subtracting the obtained PCB weight from the sum of the PCBs and the water.

2.4. Column experiment

To investigate the large amount of PCB oil in the vertical direction, PCB infiltration experiments were conducted in Runs 1–4, as shown in Table 3. In Runs 1 and 2, water was filled in the column up to 5 cm above the top of the sand layer. Then, 400 g of high concentration PCB oil (KC-300 or KC-1000), comparable with volume of the pore space in the sand layer, was supplied by a tube pump with the valve below the sand column completely closed (Fig. 1). During this period, PCB infiltration into the sand layer was not observed. Water was supplied from the water tank to fill the headspace of the column and a tube was connected with the water tank

Table 1
Composition and characteristics of PCBs, TCBs, and dioxins for KC-300 and KC-1000.

Property		PCB (KC-300)	PCB (KC-1000)	Toyoura Standard Sand
Composition	PCB content	97%[a] (95%)[b]	65%[a] (63%)[b]	230 pg g^{-1}[a]
	TCB content	ND [-]	31%[a]	<100 pg g^{-1}[a]
	Dioxin content	2.0 μg-TEQ g^{-1}[a]	12 μg-TEQ g^{-1}[a]	0.001 pg-TEQ g^{-1}[a]
Characteristics	Density	1320 kg m^{-3}	1460 kg m^{-3}	2651 kg m^{-3}
	Viscosity (20 °C)	0.043 Pa s	0.020 Pa s	–

[a] Numbers represent percentages by HRGC-HRMS.
[b] Numbers in parentheses represent percentages by GC-ECD; ND represents no data.