

LEACHING BEHAVIOR OF POLYCHLORINATED BIPHENYL CONGENERS IN SOIL

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Abstract

To investigate the behavior of polychlorinated biphenyls (PCBs) in soil due to rain and ground water, Kanechlor-400 as PCBs was added to soil, and the mobility of PCBs affected by percolating water was examined for four months. As a result, the eluted amount of PCBs was only 0.047% of the loaded PCBs and it was determined that the lower PCBs leached dependently on the number of substituted chlorine. However, although PCB congeners in a homologue have few differences in their chemical and physical properties, the behavior of each congener in the effluent water was found to be dissimilar. Therefore, the congener distribution may be also influenced by the chlorine substitution position and/or degradation. These results suggest that a congener profile of PCBs in a contaminated site could possibly change that of PCBs in the spread site.

Introduction

Polychlorinated biphenyls (PCBs) have been implicated in the global environmental problem. Their major sources have been determined to be technical PCBs and combustion. Because PCBs are long-lasting and semi-volatile, they result in widespread pollution. Especially, soil is considered to be a primary reserve media of PCBs due to leaks, spills, and illegal dumping of PCB waste. The current environmental levels of PCBs in various environmental media in Japan seem to be declining. However, sites of heavily contaminated soil are occasionally discovered and the cause of contamination in each case needs to be clarified. Therefore, the requirement of each specific profile of PCBs emission sources has been raised. Namely, in order to estimate the identification and contribution ratio of emission sources, statistical analysis has been frequently attempted using the profiles of individual emission sources^{1,2}. However, although PCBs are extremely slow to degrade and highly adsorbent in soil, the soil-water partition coefficient values of PCBs are different due to substituted chlorine numbers that have been reported^{3,4}. Furthermore, there have been few reports indicating that each homologue of PCBs reveals a difference in mobility by percolated water in soil⁵. Moreover, the behavior of all congeners of PCBs has not been sufficiently defined. The purpose of this study is to investigate the behavior of leaching PCB congeners in soil due to rain or ground water, and to estimate the profile change of PCBs in the environment.

Materials and Methods

Figure 1 shows an illustration of the experimental soil column. The sample soil (ca. <2mm) was analyzed for PCB concentration, ignition loss and particle-size distribution. The top layer of the non-loaded soil, the middle layer of soil loaded with 200mg of PCBs (Kanechlor (KC)- 400), and the bottom layer of non-loaded soil were packed in the glass column. Purified water was poured into the soil column at 14 - 70ml/day for four months. PCBs in effluent water were trapped by polyurethane foam (PUF) spiked with labeled PCBs. The PUF was changed once a month and soxhlet extraction was conducted. After cleaning up the extract, congener-specific analyses of PCBs in KC-400, non-loaded soil and effluent water samples of each month were carried out using

HRGC-HRMS (HP6890-JEOL JMS-700) fitted with HT8-PCB (60m x 0.25mm i.d., SGE).

Results and Discussion

The characteristics of the soil were 7.3% of ignition loss and the particle size distribution was measured to be $75\mu\text{m} >$: 58.5%, $75\mu\text{m} - 2\text{mm}$: 41.4%, $2\text{mm} <$: 0.16%. For four months, 4,289ml of water was passed through the soil column, and the monthly water volume from one month to four months was 1,025ml, 2,088ml, 442ml, and 734ml. The total volume of water poured into the soil column was equivalent to 1.7 years of the average annual precipitation in Tokyo.

The total and the homologue amounts of loaded KC-400 as PCBs are shown in Table 1, along with the PCB content in the soil and the effluent water. During the experimental period, the eluted PCB amount was only 0.047% of loaded PCBs.

The monthly PCB amounts in the effluent

water seemed to be independent on the variation of the flow rate of percolating water and tended to increase with time. Regarding the homologue distribution of KC-400, $T_4\text{CB}$ accounted for 57.6%, and $P_5\text{CB} - D_{10}\text{CB}$ accounted for 28.6 % of the total PCBs. On the other hand, the ratios of $T_4\text{CB}$ and $P_5\text{CB} - D_{10}\text{CB}$ of the total

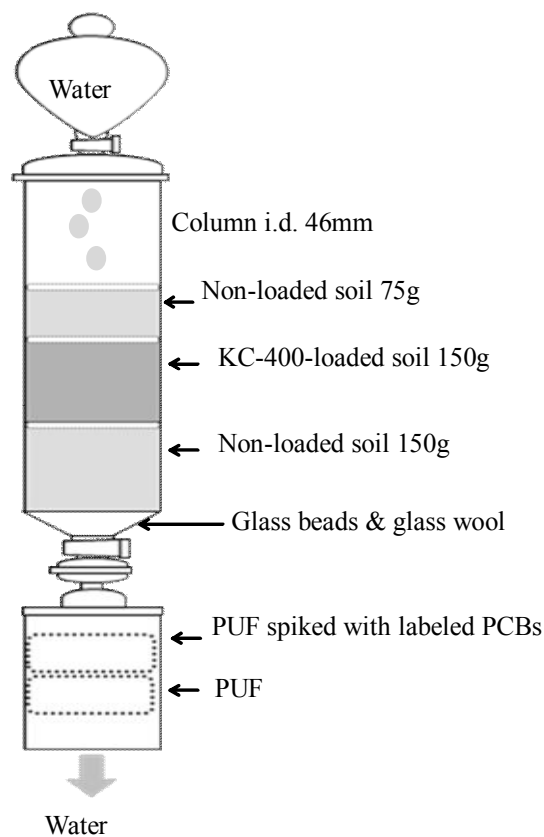


Figure 1 Schematic of soil column

Table 1 The total and the homologue amounts of loaded PCBs and the PCB content in the soil and the effluent water

		(ng)									
	total PCB	$M_1\text{CB}$	$D_2\text{CB}$	$T_3\text{CB}$	$T_4\text{CB}$	$P_5\text{CB}$	$H_6\text{CB}$	$H_7\text{CB}$	$O_8\text{CB}$	$N_9\text{CB}$	$D_{10}\text{CB}$
soil*	1660	Tr.**	13	28	102	350	655	240	78	160	14
KC-400	200000	Tr.**	1770	25800	115000	53800	3400	Tr.**	N.D.	N.D.	N.D.
total effluent	94.7	6.3	27.4	23.4	26.0	10.7	0.92	0.06	Tr.**	N.D.	N.D.
1month	5.8	0.08	1.4	1.1	2.1	1.0	0.10	Tr.**	Tr.**	N.D.	N.D.
2month	19.4	0.39	4.6	6.0	6.2	1.9	0.18	Tr.**	Tr.**	N.D.	N.D.
3month	34.9	1.7	12.8	9.4	8.0	2.7	0.25	Tr.**	Tr.**	N.D.	N.D.
4month	34.6	4.1	8.5	6.8	9.8	5.0	0.39	0.04	N.D.	N.D.	N.D.

* The total & the homologue amounts of PCBs in non-loaded soil (375g) , **trace level of PCBs

content in the non-loaded soil were 6.8% and 90.7%, respectively. However, the PCB content in the non-loaded soil was quite low in comparison to the loaded amount of KC-400. Therefore, the homologue distribution of PCBs in the effluent water was hardly influenced by indigenous PCBs in the soil. In the effluent PCBs, large ratios of lower chlorinated PCBs were obtained in the experiment, as follows: M₁CB -T₃CB accounted for 60.5%, and T₄CB accounted for 27.6%. Thus, the mobility of each PCB homologue in the effluent water was confirmed to relate with the soil - water partition coefficient values.

The ratios of the concentrations of 209 congeners against total PCBs are shown in Figure 2. As an overall trend,

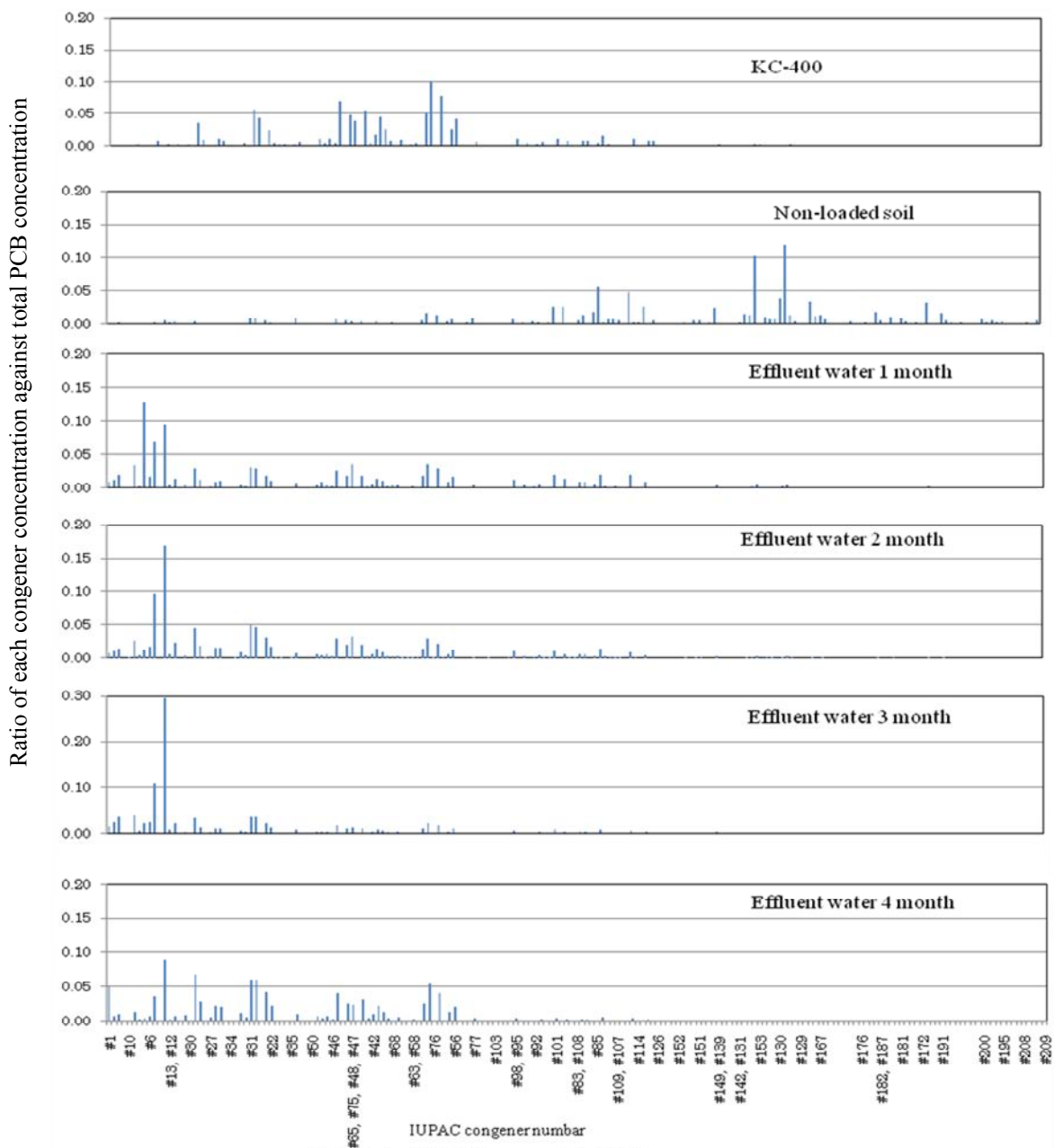


Figure 2 The profiles of PCB congeners in loaded KC-400, non-loaded soil and the effluent waters

the mobility of PCB congeners in the soil was generally observed to have a tendency to absorb more of the highly chlorinated PCBs. However, individual congeners, which have the same number of chlorine molecules, did not always behave in the same manner. The monthly variation of the eluted amount of predominant PCBs is shown in Table 2. Though congeners in M₁CB in KC-400 were under the quantitative limit, these congeners were observed to increase. For example, the breakthrough amount of congener #1 increased 8-fold from 3 to 4 months. On the other hand, the amount of congener #3 has increased from 1 to 3 months and slightly decreased on 4th month. The increase and decrease of eluted amount of congener #11 (D₂CB) was also obtained similar behavior during the experiment period. Since PCB congeners in a homologue are known to have similar chemical/physical properties⁴, the soil-water partition coefficient of PCB congeners may be affected by the chlorine substitution position as well as the number of chlorination of the biphenyl. In addition, Tucker et al. reported that lower PCBs were more biodegradable. Therefore, the congener distribution of PCBs in the effluent water is speculated to be due to the differences of adsorption of each congener in soil and the degradation, such as anaerobic dechlorination, which may promote a change in the congener profile. Consequently, almost all PCBs emitted in soil adsorb on soil strongly. However, when percolating water, such as rainfall and ground water, leaches PCBs from a contaminated site, the profile of PCBs in the site where it is spread appears to be different.

Table 2 Monthly amount of predominant PCBs in the effluent water

congener			(pg)			
			1month	2month	3month	4month
2	#1	(M ₁ CB)	Tr*.	92	370	3060
4	#3	(M ₁ CB)	78	170	830	620
2,4	#7	(D ₂ CB)	540	160	520	240
3,3'	#11	(D ₂ CB)	400	2250	7370	4740
2,2',5	#18	(T ₃ CB)	200	1030	1560	1280
2,4',5	#31	(T ₃ CB)	220	1120	1700	1140
2,3',4',5	#70	(T ₄ CB)	290	720	1140	1470
3,3',4,4'	#77	(T ₄ CB)	Tr.*	34	70	86
2,3,3',4,4'	#105	(H ₅ CB)	66	100	170	380
2,3',4,4',5	#118	(H ₅ CB)	140	198	320	570

* trace level of PCBs

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