DISTRIBUTION OF CHLORINATED ORGANIC COMPOUNDS IN THE ATMOSPHERE

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ABSTRACT

Polychlorinated dibenzo-p-dioxins(PCDDs), polychlorinated dibenzofurans(PCDFs), polychlorinated biphenyls(PCBs), chlordanes and DDE were measured in both the vapor and particle-bound phases. The daily variation of these compounds in urban air was discussed with meteorological factors in summer and winter. The vapor-to-particle(V/P) ratios for these compounds were depended on the vapor pressure of each compounds and the meteorological condition. The behavior and transport of these compounds in the atmosphere were closely related to wet and dry deposition.

INTRODUCTION

PCDDs, PCDFs, PCBs and chlordanes have been observed in various environmental samples. Few data are available on the levels of these compounds in urban air [1]. Although there has been many interesting information in recent studies [2-5], none of these studies reported the daily variability. As to the number of sampling for these compounds, few times a month is not enough to grasp the variability with various conditions. We felt that it was important to study the daily variability in concentrations of each congeners in both the vapor and particle-bound phases.

The preliminary study [6] showed that the atmospheric levels of these compounds have a tendency to increase on stagnant and cloudy days. In order to eliminate the effect of regional and seasonal variability, we investigated the levels of these compounds at a single location in the same season [1].

Daily variability in atmospheric levels of these compounds from June 1988 to July 1988 (summer) and from December 1988 to January 1989 (winter) were discussed with meteorological factors: temperature, cloud amount, wind speed, rainfall, visibility, humidity and total suspended particulate.

Table 1. Meteorological factors

	Jun.-Jul. 1988 Range	(Mean)	Dec.-Jan. 1988.89 Range	(Mean)
Temperature (°C)	20.2 - 27.8	(24.2)	3.6 - 12.7	(7.8)
Humidity (%)	62 - 93	(76.6)	54 - 89	(65.3)
Cloud (Max=10)	2.5 - 10	(8.8)	0.3 - 10	(6.8)
Wind speed (m/s)	1.6 - 9.3	(3.3)	1.8 - 8.6	(3.5)
Rain (mm)	0 - 54.5	(5.2)	0 - 7.5	(0.95)
Visibility (km)	8.0 - 27.5	(15.5)	13.5 - 42.5	(22.9)
TSP (ug/m^3)	25.7 - 204	(75.3)	22.4 - 169	(78.5)

TSP:Total suspended particulate

Table 2. Atmospheric levels (pg/m^3)

	Jun.-Jul. 1988 Range	(Mean)	Dec.-Jan. 1988.89 Range	(Mean)
PCDFs	< 0.1 - 22.4	(3.3)	< 0.1 - 10.6	(1.0)
PCDDs	< 0.1 - 28.2	(5.4)	< 0.1 - 6.8	(1.0)
PCBs	169 - 1479	(446)	2.2 - 2310	(536)
t-CHL	207 - 2406	(687)	7.0 - 538	(157)
c-CHL	176 - 1934	(581)	8.3 - 452	(134)

Table 3. Correlation matrix (Dec.1988 - Jan.1989)

R.T	575 t-CHL	593 c-CHL	606 nona	501 52	559 91	585 101	618 DDE	654 149	857 BA	1090 BF	1199 HepCDF	1216 HepCDD
t-CHL	1	0.994	0.995	0.619	0.737	0.753	0.650	0.625	0.453	0.451	0.249	0.128
c-CHL		1	0.987	0.643	0.769	0.796	0.697	0.669	0.419	0.411	0.269	0.143
nona			1	0.616	0.713	0.730	0.623	0.600	0.468	0.455	0.276	0.150
52				1	0.907	0.875	0.620	0.882	0.108	0.128	0.159	0.083
91					1	0.963	0.781	0.966	0.081	0.096	0.165	0.100
101						1	0.835	0.962	0.000	0.007	0.153	0.104
DDE							1	0.761	0.029	0.026	0.124	0.113
149								1	0.049	0.020	0.159	0.137
BA									1	0.949	0.302	0.290
BF										1	0.247	0.247
HepCDF											1	0.669
HepCDD												1

t-CHL: trans-chlordane; c-CHL: cis-chlordane; nona: trans-nonachlor; 52: 2,5,2',5'-tetra chlorobiphenyl(CB); 91: 2,3,6,2',4'-penta CB; 101:2,4,5,2',5'-penta CB; DDE: p,p'-DDE; 149: 2,3,6,2',4',5'-hexa CB; BA: benz(a)antrathene; BF:benzo(b)fluoranthene+B(j)F+B(k)F; HepCDF: 1,2,3,4,6,7,8-hepta chloro dibenzofuran; HepCDD: 1,2,3,4,6,7,9-hepta chloro dibenzo-p-dioxin.

EXPERIMENTAL

Air samples were collected every 24hrs using high volume air sampler located on the roof of our laboratory. Quartz microfibre filter(QMF) QM-A for high volume air sampling were obtained from Whatman and cleaned before use by ashing for 24 hrs. The polyurethane foam plugs(PUF) washed with hot water followed by acetone with Soxhlet extractor for 12 hrs. Internal standard were full labeled PCDDs and half labeled PCDFs with ^{13}C obtained from Cambridge Isotope Laboratories

The JEOL DX-303 mass spectrometer equipped with post acceleration type high sensitive ion detector and a Hewlet-Packard 5890A GC were used. Data acquisition and analysis carry out by the DA-5100 computer system. Two or three ions from each congener groups were monitored. The GC/MS condition are same as previous study[1]

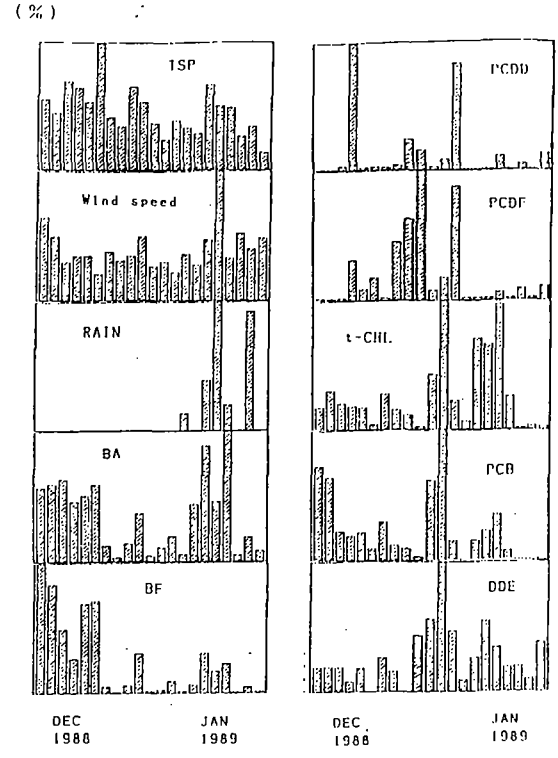

Fig.1 Daily variation (Dec.1988 ~ Jan.1989)
Abbreviations are defined in the Table 3

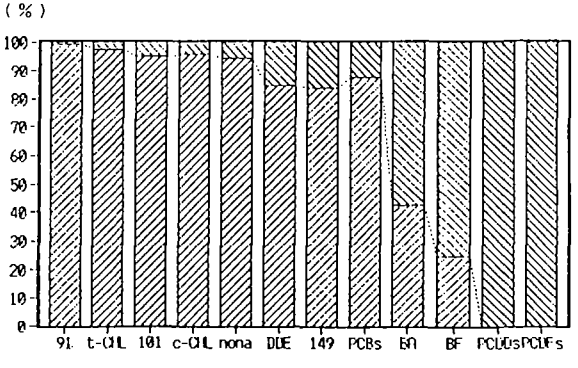

Fig.2 Vapor-to-particle ratios (Dec.1988 ~ Jan.1989)
Abbreviations are defined in the Table 3

RESULTS AND DISCUSSION

The seasonal difference of meteorological conditions are shown in Table 1. The comparison of atmospheric levels of these compounds are shown in Table 2. We reported that the correlation coefficient between PCDD and PCDF is higher than the correlation coefficients between PCDD and the other compounds. These compounds were divided into two groups according to the behavior of the chemicals in environment. One was PCDD/PCDF group, and another was the chlordanes/PCB/DDE group. Temporal variations in atmospheric concentrations of these compounds in the urban air showed that the meteorological factors such as rain have played important roles on the behavior and transport of these compounds.

As a result of continuous measurement in winter, the similar tendency was observed as for the correlation coefficient and meteorological effect. Correlation matrix of concentrations of these compounds in winter are shown in Table 3. The correlation coefficients between the congener/isomer having similar vapor pressure is relatively higher than those having different value.

Fig. 1 shows the daily variability in atmospheric levels of these compounds with meteorological condition. The chlordane/PCB/DDE which exist primarily in the vapor phase resembles each other in the behavior.

Bidleman pointed out that the V/P ratio was controlled by semivolatile organic compound vapor pressure and the total suspended particle concentration [7]. Despite artifact problems in high-volume sampling (overestimation or underestimation of V/P ratio), it is true that we can get important information from the apparent V/P ratio. The extent of migration from QMF to PUF was negligible for our study.

Fig. 2 shows the vapor-to-particle ratios in winter. Atmospheric temperatures ranged 20.2 - 27.8°C (av. 24.2°C) in summer, 3.6 - 12.7°C (av. 7.8°C) in winter. V/P ratio in winter is lower than in summer. These ratios in summer and winter are as follows: PCB :24, 7.2; PCDD :0.33, <0.001 ; PCDF :0.18, <0.001; t-CHL : 49, 19 : DDE : 19, 2.5 respectively.

V/P ratio depended on temperature of sampling date and vapor pressure of these compounds. V/P ratio decrease as temperature and vapor pressure decrease.

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