DISTRIBUTION OF PCDD/Fs WITH ATMOSPHERIC PARTICLE SIZE

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Introduction

Particle size is a major determining factor in the atmospheric behavior of aerosol particles and controls the residence time and removal mechanisms of aerosol-bound contaminants. Particle size also plays a critical role in human health effects. Study of the size distribution pattern of airborne constituents can give helpful information about their origin and atmospheric environment. Studies of PCDD/Fs have been hindered by sampling artifacts and the trace levels of PCDD/Fs in the environment, hence, only three studies have been made of the distribution of PCDD/Fs with particle size. These studies reported the largest amounts of particle-bound PCDD/Fs on fine particles (Kaupp et al, 1994; Kurogawa et al,1998; Kaupp et al 2000). However, the result for the distribution of PCDD/Fs homologue with particle size was different. It is therefore difficult to define the general distribution of PCDD/Fs with particle size.

The present work was undertaken to investigate the dependence of the concentration of PCDD/Fs on particle size in urban and suburban areas. The concentration and particle mass of PCDD/Fs in size-segregated airborne particulate matter were measured using a six-stage high volume cascade impactor during summer and fall conditions. We discuss the size distributions of PCDD/Fs and compare our results with those from previous studies.

Methods and Materials

Particle samples were taken from the ambient air at two sites during summer and fall 2000. Sampling was performed on the roof of a building over 10 m high at both sites. The locations of the sites are as follows: (a) Site A; an incineration area with a municipal solid waste incinerator (MSWI) and industrial waste incinerator.. (b) Site B; a suburban area within 10 km of steel industry. There was no known point source within 10 km of this site. Three particle size samples were taken at Site A during summer and fall 2000, and one particle size sample was taken at Site B during fall 2000. Airborne particles were collected using a six-stage high volume cascade impactor at a flow rate of 560 l/min (cascade impactor; SA236, Graseby Andersen, high volume air sampler; DHA-1000S, SIBATA). The particles were separated into the following size ranges: less than 0.41, 0.41-0.73, 0.73-1.4, 1.4-2.1, 2.1-4.2, 4.2-10.2, and greater than 10.2 μm in aerodynamic diameter (D_{ae}).

Sample preparation was done according to the US EPA method 1613. PCDD/Fs were analyzed by high-resolution gas chromatography / high-resolution mass spectrometry (Hewlett-Packard Model 6890 series Ⅱ/ JMS 700T) with a DB-5MS column (60m, 0.25 mm i.d. 0.25 um film thickness).

Results and Discussion

Particle size distribution

Table 1 shows the particle concentration with particle size in each sample. The total particle concentration ranged from 62.68 to 160.71 μg/m^3 at Site A and was 39.75 μg/m^3 at Site B. The total particle concentration in A1 was about two or three times lower than that of other samples collected at Site A because of rain before sampling. The total particle concentration at Site B was much lower than at Site A because Site B was a cleaner area than Site A, as described above.

The simple distribution of particle size is presented in Figure 1. The fractions were obtained by dividing the concentration of each impactor stage by the total concentration. As shown in Figure 1, the particle size distribution was very similar for both sites, although the total particle concentrations were different. The largest mass of particles was collected in the back filter (D_{ae} < 0.41μm), and more than 50% of particles were in the < 1.4 μm size class.

Both of the sampling sites used in this study show this characteristic distribution pattern for urban particles, regardless of the total particle concentration.

Table 1. Particle concentrations with respect to particle size

(unit ; ug/m3)

stage	range (um)	A1	A2	A3	B1
1	10.2 ~	7.66	16.73	19.31	1.94
2	4.2 ~ 10.2	9.89	7.92	38.10	6.80
3	2.1 ~ 4.2	5.43	27.44	19.18	4.29
4	1.4 ~ 2.1	6.83	14.84	19.05	4.12
5	0.73 ~ 1.4	7.79	16.73	17.53	3.88
6	0.41 ~ 0.73	5.87	15.38	9.43	3.71
BF	~ 0.41	19.21	57.57	38.10	15.01
total	-	62.68	156.61	160.71	39.75

PCDD/F distribution with particle size

The concentration of PCDD/Fs with particle size are ranging from 0.169 pg I-TEQ/m^3 to 0.664 pg I-TEQ/ m^3 (8.15 pg/ m^3 ~ 32. 56 pg/ m^3) for Site A and with a value of 0.124 pg I-TEQ/ m^3 (5.78 pg/ m^3) for Site B. The PCDD/F concentration in the A1 sample was lower than that of the other samples collected from Site A. The lowest level of PCDD/Fs was observed at Site B, which was due to the same reason cited above for the case of particle concentration. The distribution of PCDD/F concentration with particle size is presented in Figure 1. In the present study, more than 60% of the PCDD/Fs were associated with particles of diameters D_{ae} < 0.41 μm, and 90% of the PCDD/Fs were on particles with diameters D_{ae} < 2.1 μm, regardless of sampling site. At both sites the fraction of PCDD/F concentration was higher than the fraction of particle concentration for D_{ae} < 1.4 μm, whereas the reverse was observed for D_{ae} > 1.4 μm (Figure 1).

From these results, most of the PCDD/Fs are associated with fine particles, which is in accordance with previous studies.

PCDD/Fs Homologue distribution with particle size

We presented simple homologue distribution of PCDD/Fs with particle size in Site A (Figure 4). The fraction of homologues was calculated by dividing the concentration of each impactor stage by the total concentration, and this concentration was calculated as pg/g on each stage. The fraction of less chlorinated dioxin and furan (ex;TCDD/Fs, PeCDD/Fs) increased with increasing particle size, but the opposite was true for high chlorinated dioxin and furans (ex;HxCDD/Fs, HpCDD/Fs, OCDD/Fs). The less chlorinated dioxin/furans (tetra, penta CDD/Fs) are more prevalent on coarse particles (> 1 ~ 2 μm) and the high chlorinated dioxin/furans (hexa, hepta, octa CDD/Fs) are more prevalent on fine particles.

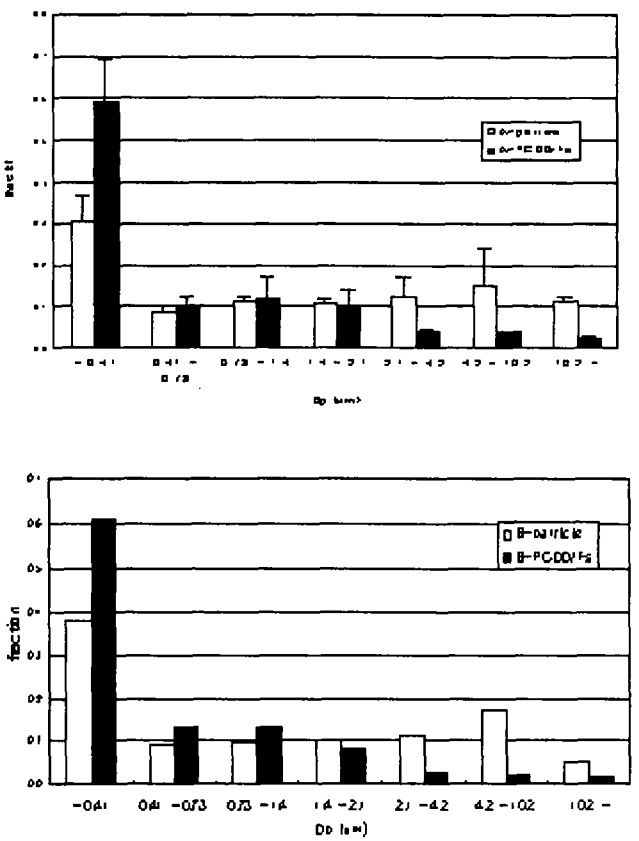

Figure 1. Distribution of particles and PCDD/Fs with respect to particle size

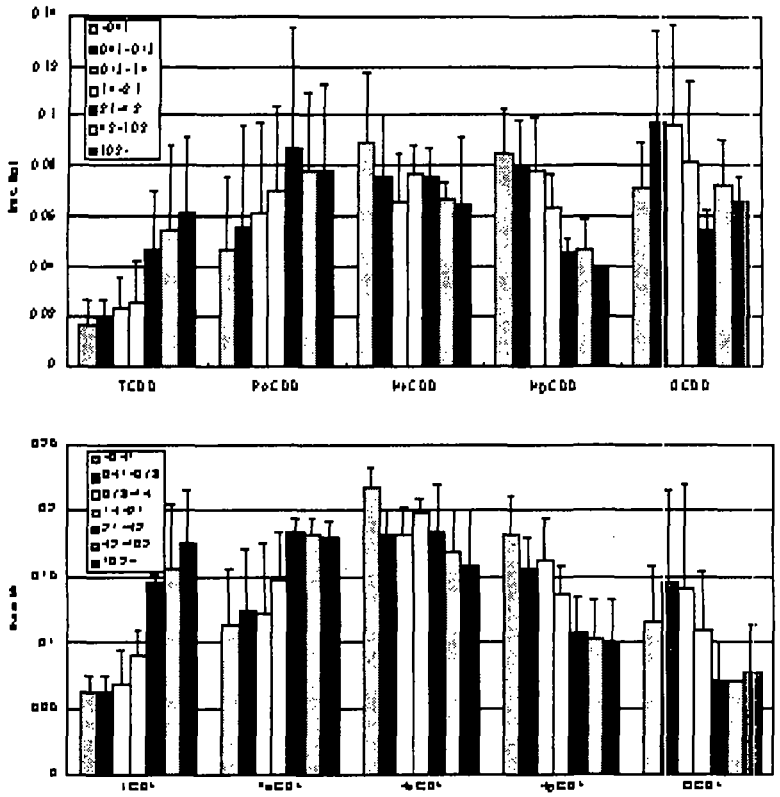

Figure 2. PCDD/Fs homologue patterns PCDD/Fs with respect to particle size

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