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Wet Deposition of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofuran in a Rural Area of Taiwan

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ABSTRACT

The annual variations of wet deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in atmosphere were measured at two sites (A and B) near two municipal solid waste incinerators (MSWIs) in southern Taiwan. Results showed that particle scavenging dominates in the wet deposition processes for the removal of PCDD/Fs from the atmosphere, the highest value was observed at the highest chlorinated congener. The ambient temperature and the amount of precipitation played an important role in the variation of PCDD/F deposition fluxes. It was found that temperature was inversely associated with the existence of particulate PCDD/Fs, indicating PCDD/Fs are scavenged most efficiently in cold weather. PCDD/F wet deposition fluxes in rainy seasons (from June to August) were significantly higher than those in dry seasons (from December to February), revealing a positive relationship between wet deposition flux and monthly rainfall. Additionally, the annual total (dry + wet) deposition fluxes of PCDD/Fs were 149 ng/m^2-year (5.02 ng I-TEQ/m^2-year) and 177 ng/m^2-year (5.11 ng I-TEQ/m^2-year) for sites A and B, respectively, revealing that dry deposition was more dominant than the wet deposition for the atmospheric deposition of PCDD/Fs. Since atmosphere deposition is believed to be the main transfer pathway of PCDD/Fs into food chains, its impact on human exposure to PCDD/Fs is of great importance.

Keywords: Polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs); Gas-Particle partitioning; Wet deposition; Scavenging ratio.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are two groups of persistent, semi-volatile, and toxicologically significant contaminants. They are highly resistant to biodegradation in the environment, become concentrated in the food chain, and accumulate in fatty tissues of animals and humans (Smith and Gangolli, 2002). Industrial or combustion processes are usually considered as main sources of PCDD/Fs as well as polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and polycyclic aromatic hydrocarbons (PAHs) (Lai *et al.*, 2007; Chomanee *et al.*, 2009; Shi *et al.*, 2009, Wu *et al.*, 2009; Chung *et al.*, 2010; Artha *et al.*, 2011). In Taiwan, several combustion processes have been identified as significant sources of atmospheric PCDD/F, including waste incinerators (Wang *et al.*, 2008; Wang *et al.*, 2009; Huang *et al.*, 2011), secondary aluminium smelters, sinter plants (Kuo *et al.*, 2011), electric arc furnaces (Lee *et al.*, 2004; Lee *et al.*, 2005; Chiu *et al.*, 2011), power plants (Lin *et al.*, 2010b; Wu *et al.*, 2010), crematories (Chiu *et al.*, 2011), joss paper incinerators (Hu *et al.*, 2009), and woodchip-fuelled boilers (Chen *et al.*, 2011). Once released into the atmosphere, PCDD/Fs can be transported over long distances from the combustion sources (Eitzert and Hites, 1989) and enter terrestrial and aquatic environments through deposition (Jurado *et al.*, 2004).

Deposition of PCDD/Fs in air can be divided into dry deposition (gaseous, particulate) and wet deposition, both processes contribute significantly to the removal of atmospheric PCDD/Fs (Koester and Hites, 1992). Dry deposition, including both gaseous adsorption at the air-surface interface and airborne particles, comes into contact with a surface (Lohmann and Jones, 1998). Wet deposition

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is the removal of atmospheric particles by precipitation scavenging of rain and cloud droplets(Eitzert and Hites, 1989). Precipitation scavenging of particles accounts for the vast majority of the wet deposition for the removal of semi-volatile organic compounds from the atmosphere, which strongly influences their long-range transport potential and overall persistence (Scheringer, 1997).

Recently, several studies describing the atmospheric PCDD/Fs deposition from relevant sources in Taiwan were reported. Atmospheric dry deposition fluxes of total PCDD/Fs were found to range from 5.07 to 56.8 pg I-TEQ/m^2-day in the ambient air in the vicinity of municipal solid waste incinerators (Wu *et al.*, 2009). Lin *et al.* (2010a) investigated dry and wet deposition of PCDD/Fs on a drinking water treatment plant. The total deposition flux (dry + wet) of PCDD/Fs entering the drinking water treatment plant was 1439 ng/m^2-year, and dry deposition contributed approximately 7.3 times higher than wet deposition. In the study of Wang *et al.* (2010), dry and wet depositions of PCDD/Fs were sampled seasonally in the ambient air among different kinds of areas (a commercial suburban area, an industrial area, a coastal rural area, and am agricultural rural area). The annual total deposition ranged from 115 to 310 ng/m^2-year, and the highest was found in the industrial area. The dry deposition flux observed was significantly higher than the wet deposition flux, indicating that dry deposition is the major PCDD/F removal mechanism in the air. In the author's previous study, the atmospheric dry deposition fluxes of PCDD/Fs in the vicinity of two municipal solid waste incinerators (MSWIs) were investigated (Huang *et al.*, 2011). Calculated dry deposition fluxes of total PCDD/Fs ranged from 0.0274–0.718 ng I-TEQ/m^2-month and were found to decrease as temperature increased. The dry deposition velocities of atmospheric particles (0.48–0.91 cm/s) were similar to that in the vicinity of MSWI in southern Taiwan (0.44–0.68 cm/s) (Wu *et al.*, 2009), but slightly higher than those in urban areas of Korea (0.49 cm/s) (Moon *et al.*, 2005). However, the significance of the wet deposition for PCDD/Fs has seldom been addressed.

In this study, the gas-particle partitioning of PCDD/Fs as well as the atmospheric wet deposition fluxes of PCDD/Fs in the vicinity of two MSWIs situated in southern Taiwan were investigated seasonally. Scavenging ratios of PCDD/Fs in air were determined by model calculations. Annual atmospheric wet deposition fluxes of PCDD/Fs during July 2009 to June 2010 were compared with those of dry deposition.

MATERIALS AND METHODS

PCDD/F Sampling

Sites A and B with maximum ground concentration of PCDD/F from the emissions of two MSWIs, respectively, were found by the Industrial Source Complex Short Term Model (ISCST). Total of eight ambient samples in each area were collected simultaneously during July 2009 and January 2010. All meteorological information for sampling sites during the periods from July 2009 to June 2010 was obtained from the Meteorological Bureau in Kaohsiung City. The maximum and minimum temperatures at sampling areas were 29.4°C (in September) and 19.9°C (in January), with an average of 25.4°C. The annual precipitation in this area ranged from 0.5 mm (in December) to 934.5 mm (in August) and the mean wind speed ranged from 1.84 to 3.35 m/s. The total TSP concentrations were found to vary in the range of 43 to 166 μg/m^3 during the sampling periods in this area and their corresponding PM_{10} concentrations were calculated according to a factor TSP: PM_{10} = 1.24:1 (Sheu *et al.*, 1996). The basic information for these two MSWIs and meteorological information for the sampling areas were listed in our previous work (Huang *et al.*, 2011).

Ambient air samples were collected using a PS-1 sampler (Graseby Anderson, GA, USA), following the revised U.S. EPA Method TO9A. Each sample was collected continuously on three consecutive days, yielding a sampling volume of about 972 m^3. The PS-1 sampler was equipped with a quartz fiber filter for sampling particle-phase compounds, and a glass cartridge that contained PUF for sampling gas-phase ones. A known amount of surrogate standard was spiked to check the collection efficiency of the sampling train before the sampling was conducted. To ensure that the collected samples were free of contamination, one field blank was completed. The recoveries of the PCDD/Fs surrogate standards were 90–122%, falling within the required 70–130%.

Analyses of PCDD/Fs

Analyses of PCDD/F samples were performed in the Super Micro Mass Research and Technology Center in Cheng Shiu University, certified by the Taiwan EPA for analyzing PCDD/Fs. Each sample was spiked with a known standard and extracted for 24 h. Then, the extract was concentrated and treated with sulfuric acid, followed by a series of cleanup and fraction procedures (Wang *et al.*, 2003). The standard solution was added to the sample before PCDD/F analysis to ensure recovery during analysis. A high resolution gas chromatography with a mass spectrometer (HRGC/MS) was used to determine the concentrations of seventeen individual PCDD/Fs. The HRGC (Hewlett Packard 6970 Series gas, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, and film thickness = 0.25 μm) and splitless injection (J&W Scientific, CA, USA). The oven temperature was programmed as follows: initial temperature at 150°C (held for 1 min), increasing to 220°C at 30˚C/min (held for 12 min), then to 240°C at 1.5 °C/min (held for 5 min), and finally to 310°C at 1.5 °C/min (held for 20 min). Helium was used as the carrier gas. The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode was set to ion monitoring with resolving power at 10,000. The electron energy and the source temperature were set at 35 eV and 250°C, respectively. The recoveries for the seven individual PCDD/Fs compounds ranged from 75 to 118%, and the method detection limits ranged from 0.0001 to 0.0035 ng/Nm^3 (Wang *et al.*, 2010).

Atmospheric Wet Deposition of PCDD/Fs

The wet deposition flux of PCDD/Fs is associated with both vapor dissolution into rain and the removal of suspended particulates by precipitation. Scavenging ratio is defined as the PCDD/F concentrations of the dissolved phase in the raindrop divided by those of the gas phase in the air during the precipitation event. The gas scavenging ratio of PCDD/Fs, S_g, can be estimated as follows (Ligocki *et al.*, 1985a):

$$S_g = \frac{RT}{H} \tag{1}$$

$$S_g = \frac{C_{rain,\,dis.}}{C_g} \tag{2}$$

where S_g is the gas scavenging ratio of PCDD/Fs (dimensionless), R is the universal gas constant (82.06 × 10^{-6} m^3 atm/mol-K), T is ambient temperature (K), and H is the Henry constant (m^3 atm/mol). $C_{rain,dis}$ is the dissolved-phase concentration of PCDD/Fs in the raindrop, and C_g is the concentration of PCDD/Fs in the gas phase.

The particle scavenging ratio of PCDD/Fs, S_p, can be calculated by

$$S_p = \frac{C_{rain,\,particle}}{C_p} \tag{3}$$

where S_p is the particle scavenging ratio of PCDD/Fs (dimensionless), $C_{rain,particle}$ is the particle-phase concentration of PCDD/Fs in the raindrop, and C_p is the concentration of PCDD/Fs in the particle phase.

Total scavenging of precipitation (S_{tot}) is defined as the sum of gas and particle scavenging, which can be calculated as follows (Bidleman, 1988; Ligocki *et al.*, 1985b):

$$S_{tot} = S_g\,(1-\Phi) + S_p \times \Phi \tag{4}$$

where S_{tot} is the total scavenging ratio of PCDD/Fs (dimensionless) and Φ is the fraction of the total air concentration bound to particles.

Due to the lack of measured data for the particle scavenging ratio of PCDD/Fs, average Bloomington air and rain concentrations measured by Eitzer and Hites (1989) were adopted in several studies (Wu *et al.*, 2009; Lin *et al.*, 2010a; Wang *et al.*, 2010). Suppose the seventeen 2,3,7,8-substituted PCDD/Fs were distributed among different particle sizes and particles in the atmosphere were washed out uniformly, the same value of S_p should be obtained (Lin *et al.*, 2010a). However, Eitzer and Hites (1989) demonstrate the wide range of S_p values. The possible reason is the congener that has dissolved in a raindrop to be readsorbed by the particle scavenged, and vice versa (Wu *et al.*, 2009). The above description suggests that the S_p values of seventeen congeners which have lowest solubility for PCDD and PCDF should be more accurate. Accordingly, the average S_p of OCDD and OCDF (i.e. 42,000) measured by Eitzer and Hites (1989) was also used here for calculation of total precipitation scavenging. Based on gas and particle scavenging ratios, the dissolved and particle phase concentrations of raindrops, $C_{rain,dis.}$ and $C_{rain,particle}$, can be calculated by Eqs. (2) and (3), respectively. Additionally, the total wet deposition flux (F_W) contributed from both gas and particle phases were calculated by precipitation and days of precipitation during July 2009 to June 2010 (Table 1).

RESULTS AND DISCUSSION

Gas-particle Partitioning of PCDD/Fs

The PCDD/F concentrations in the gas and particle phases (C_g and C_p), as shown in Tables 2a–2b and 3a–3b, were determined based on the gas-particle partitioning and the total PCDD/F concentration in ambient air presented in the author's previous study (Huang *et al.*, 2011). The fraction of PCDD/Fs bound to particles (Φ) of TCDD/F, PCDD/F, HCDD/F, and OCDD/F ranged from 0.0100–0.1350, 0.0392–0.4690, 0.1760–0.9670, and 0.8890–0.9940, respectively, for the ambient air of site A (Tables 2a–2b), while those of site B ranged from 0.0147–0.1860, 0.0564–0.5630, 0.2370–0.9770, and 0.9190–0.9960, respectively (Tables 3a–3b). It was found that Φ increased as the number of chlorinated substitutes increased; the higher chlorinated congeners (and particularly HCDD/F and OCDD/F) were predominant in the particle phase. Additionally, the total PCDD was found predominantly associated with particles at both sites (Φ of PCDD ranged from 0.778–0.968 at the two sites), probably due to lower vapor pressures for the PCDDs (Rordorf, 1989).

The gas-particle partitioning of PCDD/Fs in air has been shown to correlate highly with meteorological factors; it could be affected by domestic heating and temperature inversion in winter or photodegradation and OH radical reaction in summer (Lohmann *et al.*, 1999; Ogura *et al.*, 2001). The total PCDD/Fs bound to particles increased with decreasing temperature, the highest level of Φ of total PCDD/Fs was observed in winter, while the lowest one in summer for both sites. The seasonal variation of Φ that

Table 1. Precipitation and days of precipitation during July, 2009 to June, 2010.

Month	Precipitation (mm)	Days of precipitation (d)
July. 2009	200.5	19
Aug. 2009	934.5	14
Sep. 2009	134.5	13
Oct. 2009	48.5	3
Nov. 2009	9.0	2
Dec. 2009	0.5	2
Jan. 2010	1.5	3
Feb. 2010	10.5	4
Mar. 2010	1.0	2
Apr. 2010	48.0	9
May. 2010	188.0	7
June. 2010	257.0	13
Total	1833.5	91

Table 2a. Estimated monthly fluctuations of gas-particle concentrations of PCDD/Fs in the ambient air of sampling site A.

PCDD/Fs	Jul. 2009			Aug. 2009			Sep. 2009			Oct. 2009			Nov. 2009			Dec. 2009		
	C_p[a]	C_g[b]	Φ[c]	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ
2,3,7,8-TeCDD	0.0001	0.0039	0.0195	0.0001	0.0047	0.0173	0.0001	0.0060	0.0218	0.0003	0.0044	0.0547	0.0003	0.0037	0.0714	0.0006	0.0039	0.1330
1,2,3,7,8-PeCDD	0.0007	0.0068	0.0952	0.0008	0.0083	0.0839	0.0012	0.0104	0.1040	0.0032	0.0106	0.2350	0.0035	0.0084	0.2950	0.0063	0.0073	0.4630
1,2,3,4,7,8-HxCDD	0.0017	0.0032	0.3450	0.0019	0.0041	0.3100	0.0028	0.0049	0.3620	0.0071	0.0046	0.6060	0.0069	0.0032	0.6830	0.0094	0.0021	0.8210
1,2,3,6,7,8-HxCDD	0.0031	0.0055	0.3580	0.0033	0.0070	0.3230	0.0050	0.0083	0.3750	0.0142	0.0087	0.6200	0.0137	0.0060	0.6960	0.0186	0.0038	0.8290
1,2,3,7,8,9-HxCDD	0.0022	0.0036	0.3850	0.0024	0.0046	0.3480	0.0036	0.0054	0.4020	0.0118	0.0064	0.6470	0.0113	0.0044	0.7200	0.0152	0.0028	0.8460
1,2,3,4,6,7,8-HpCDD	0.0301	0.0098	0.7550	0.0348	0.0135	0.7210	0.0472	0.0145	0.7640	0.1250	0.0138	0.9000	0.1110	0.0086	0.9290	0.1320	0.0046	0.9660
OCDD	0.1360	0.0083	0.9430	0.1630	0.0120	0.9310	0.2110	0.0124	0.9440	0.3280	0.0068	0.9800	0.2850	0.0040	0.9860	0.3270	0.0020	0.9940
2,3,7,8-TeCDF	0.0004	0.0282	0.0131	0.0004	0.0342	0.0117	0.0007	0.0436	0.0148	0.0020	0.0525	0.0374	0.0023	0.0447	0.0488	0.0049	0.0486	0.0919
1,2,3,7,8-PeCDF	0.0016	0.0299	0.0514	0.0017	0.0365	0.0454	0.0028	0.0461	0.0566	0.0082	0.0515	0.1370	0.0091	0.0425	0.1760	0.0177	0.0409	0.3020
2,3,4,7,8-PeCDF	0.0027	0.0360	0.0691	0.0029	0.0440	0.0610	0.0045	0.0554	0.0757	0.0154	0.0712	0.1780	0.0170	0.0577	0.2270	0.0319	0.0531	0.3750
1,2,3,4,7,8-HxCDF	0.0091	0.0316	0.2230	0.0098	0.0394	0.1980	0.0149	0.0480	0.2380	0.0396	0.0471	0.4570	0.0403	0.0346	0.5380	0.0605	0.0248	0.7090
1,2,3,6,7,8-HxCDF	0.0071	0.0236	0.2320	0.0077	0.0295	0.2060	0.0117	0.0358	0.2470	0.0392	0.0444	0.4690	0.0397	0.0325	0.5500	0.0592	0.0230	0.7200
1,2,3,7,8,9-HxCDF	0.0009	0.0019	0.3280	0.0010	0.0024	0.2950	0.0015	0.0029	0.3450	0.0083	0.0058	0.5880	0.0081	0.0041	0.6660	0.0112	0.0027	0.8090
2,3,4,6,7,8-HxCDF	0.0078	0.0194	0.2860	0.0084	0.0245	0.2560	0.0127	0.0294	0.3020	0.0477	0.0407	0.5390	0.0473	0.0290	0.6200	0.0673	0.0195	0.7750
1,2,3,4,6,7,8-HpCDF	0.0444	0.0320	0.5820	0.0499	0.0425	0.5400	0.0705	0.0477	0.5960	0.2000	0.0490	0.8030	0.1830	0.0316	0.8530	0.2260	0.0181	0.9260
1,2,3,4,7,8,9-HpCDF	0.0075	0.0026	0.7430	0.0086	0.0036	0.7080	0.0117	0.0038	0.7520	0.0428	0.0051	0.8940	0.0382	0.0031	0.9240	0.0454	0.0017	0.9640
OCDF	0.0442	0.0040	0.9170	0.0525	0.0058	0.9010	0.0686	0.0060	0.9190	0.1530	0.0047	0.9700	0.1330	0.0028	0.9790	0.1540	0.0014	0.9910
PCDDs	0.1740	0.0410	0.8100	0.2060	0.0542	0.7920	0.2710	0.0619	0.8140	0.4890	0.0552	0.8990	0.4320	0.0382	0.9190	0.5080	0.0265	0.9510
PCDFs	0.1260	0.2090	0.3750	0.1430	0.2620	0.3530	0.2000	0.3190	0.3850	0.5560	0.3720	0.5990	0.5180	0.2830	0.6470	0.6780	0.2340	0.7440
PCDDs/PCDFs ratio	1.3900	0.1960	0.8760	1.4400	0.2070	0.8750	1.3600	0.1940	0.8750	0.8800	0.1480	0.8560	0.8330	0.1350	0.8600	0.7500	0.1130	0.8690
Total PCDD/Fs	**0.3000**	**0.2500**	**0.5450**	**0.3490**	**0.3160**	**0.5240**	**0.4710**	**0.3810**	**0.5530**	**1.0500**	**0.4270**	**0.7100**	**0.9500**	**0.3210**	**0.7470**	**1.1900**	**0.2600**	**0.8200**
PCDDs (pg I-TEQ/Nm^3)	0.0016	0.0086	0.1540	0.0017	0.0106	0.1410	0.0026	0.0132	0.1620	0.0068	0.0118	0.3650	0.0066	0.0093	0.4150	0.0097	0.0085	0.5330
PCDFs (pg I-TEQ/Nm^3)	0.0045	0.0303	0.1290	0.0049	0.0373	0.1160	0.0075	0.0465	0.1380	0.0244	0.0578	0.2970	0.0251	0.0458	0.3530	0.0400	0.0407	0.4960
PCDDs/PCDFs ratio	0.3480	0.2840	0.5510	0.3560	0.2840	0.5570	0.3420	0.2840	0.5460	0.2770	0.2030	0.5760	0.2650	0.2040	0.5650	0.2420	0.2090	0.5370
Total PCDD/Fs TEQ (pg I-TEQ/Nm^3)	**0.0061**	**0.0389**	**0.1350**	**0.0066**	**0.0479**	**0.1210**	**0.0100**	**0.0597**	**0.1440**	**0.0311**	**0.0695**	**0.3090**	**0.0317**	**0.0552**	**0.3650**	**0.0497**	**0.0492**	**0.5030**

[a] C_p: the concentration of PCDD/Fs in the particle phase; [b] C_g: the concentration of PCDD/Fs in the gas phase; [c] Φ: the fraction of PCDD/Fs bound to particles.

Table 2b. Estimated monthly fluctuations of gas-particle concentrations of PCDD/Fs in the ambient air of sampling site A.

PCDD/Fs	Jan. 2010			Feb. 2010			Mar. 2010			Apr. 2010			May. 2010			Jun. 2010		
	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ
2,3,7,8-TeCDD	0.0006	0.0038	0.1350	0.0002	0.0026	0.0718	0.0004	0.0043	0.0868	0.0004	0.0072	0.0514	0.0001	0.0052	0.0236	0.0001	0.0036	0.0148
1,2,3,7,8-PeCDD	0.0061	0.0069	0.4690	0.0025	0.0059	0.3010	0.0049	0.0093	0.3420	0.0032	0.0111	0.2260	0.0011	0.0089	0.1130	0.0005	0.0065	0.0731
1,2,3,4,7,8-HxCDD	0.0090	0.0019	0.8250	0.0049	0.0022	0.6940	0.0087	0.0033	0.7280	0.0056	0.0038	0.5990	0.0026	0.0040	0.3870	0.0013	0.0033	0.2810
1,2,3,6,7,8-HxCDD	0.0180	0.0036	0.8330	0.0099	0.0041	0.7060	0.0174	0.0061	0.7390	0.0100	0.0063	0.6130	0.0046	0.0069	0.4010	0.0023	0.0056	0.2930
1,2,3,7,8,9-HxCDD	0.0146	0.0026	0.8490	0.0081	0.0030	0.7300	0.0143	0.0045	0.7610	0.0071	0.0040	0.6400	0.0033	0.0044	0.4290	0.0017	0.0037	0.3170
1,2,3,4,6,7,8-HpCDD	0.1260	0.0043	0.9670	0.0791	0.0057	0.9330	0.1340	0.0083	0.9420	0.0684	0.0077	0.8990	0.0420	0.0114	0.7860	0.0256	0.0113	0.6930
OCDD	0.3130	0.0019	0.9940	0.2020	0.0026	0.9870	0.3400	0.0038	0.9890	0.2700	0.0056	0.9800	0.1840	0.0094	0.9510	0.1240	0.0103	0.9230
2,3,7,8-TeCDF	0.0048	0.0466	0.0935	0.0016	0.0317	0.0488	0.0033	0.0527	0.0596	0.0019	0.0526	0.0349	0.0006	0.0377	0.0160	0.0003	0.0262	0.0100
1,2,3,7,8-PeCDF	0.0173	0.0390	0.3070	0.0065	0.0300	0.1790	0.0128	0.0486	0.2090	0.0078	0.0524	0.1300	0.0026	0.0397	0.0616	0.0012	0.0281	0.0392
2,3,4,7,8-PeCDF	0.0311	0.0506	0.3810	0.0123	0.0407	0.2320	0.0238	0.0653	0.2670	0.0126	0.0612	0.1710	0.0043	0.0475	0.0825	0.0019	0.0340	0.0529
1,2,3,4,7,8-HxCDF	0.0584	0.0233	0.7150	0.0291	0.0239	0.5480	0.0527	0.0365	0.5910	0.0347	0.0429	0.4470	0.0140	0.0405	0.2570	0.0066	0.0310	0.1760
1,2,3,6,7,8-HxCDF	0.0572	0.0217	0.7250	0.0287	0.0224	0.5610	0.0519	0.0342	0.6030	0.0269	0.0317	0.4590	0.0110	0.0302	0.2660	0.0052	0.0232	0.1840
1,2,3,7,8,9-HxCDF	0.0108	0.0025	0.8130	0.0059	0.0028	0.6770	0.0104	0.0042	0.7120	0.0031	0.0023	0.5800	0.0014	0.0024	0.3700	0.0007	0.0019	0.2660
2,3,4,6,7,8-HxCDF	0.0650	0.0184	0.7800	0.0341	0.0200	0.6310	0.0609	0.0301	0.6690	0.0276	0.0244	0.5310	0.0118	0.0246	0.3250	0.0058	0.0194	0.2290
1,2,3,4,6,7,8-HpCDF	0.2170	0.0169	0.9280	0.1310	0.0212	0.8600	0.2240	0.0312	0.8780	0.1160	0.0292	0.7990	0.0639	0.0384	0.6240	0.0358	0.0350	0.5060
1,2,3,4,7,8,9-HpCDF	0.0436	0.0016	0.9650	0.0272	0.0021	0.9290	0.0462	0.0031	0.9380	0.0171	0.0021	0.8930	0.0104	0.0030	0.7750	0.0063	0.0030	0.6790
OCDF	0.1470	0.0013	0.9910	0.0946	0.0018	0.9810	0.1600	0.0027	0.9830	0.0892	0.0028	0.9700	0.0600	0.0046	0.9290	0.0397	0.0050	0.8890
PCDDs	0.4880	0.0249	0.9510	0.3070	0.0260	0.9220	0.5200	0.0397	0.9290	0.3650	0.0455	0.8890	0.2380	0.0503	0.8260	0.1550	0.0443	0.7780
PCDFs	0.6530	0.2220	0.7460	0.3710	0.1970	0.6530	0.6460	0.3090	0.6770	0.3370	0.3010	0.5280	0.1800	0.2690	0.4010	0.1040	0.2070	0.3330
PCDDs/PCDFs ratio	0.7480	0.1120	0.8690	0.8280	0.1320	0.8620	0.8050	0.1290	0.8620	1.0800	0.1510	0.8770	1.3200	0.1870	0.8760	1.5000	0.2140	0.8750
Total PCDD/Fs	**1.1400**	**0.2470**	**0.8220**	**0.6770**	**0.2230**	**0.7530**	**1.1700**	**0.3480**	**0.7700**	**0.7020**	**0.3470**	**0.6690**	**0.4180**	**0.3190**	**0.5670**	**0.2590**	**0.2510**	**0.5070**
PCDDs (pg I-TEQ/Nm^3)	0.0094	0.0081	0.5370	0.0048	0.0066	0.4210	0.0086	0.0105	0.4500	0.0052	0.0142	0.2690	0.0023	0.0113	0.1720	0.0012	0.0082	0.1290
PCDFs (pg I-TEQ/Nm^3)	0.0388	0.0387	0.5010	0.0181	0.0321	0.3600	0.0333	0.0512	0.3940	0.0175	0.0489	0.2640	0.0070	0.0397	0.1490	0.0033	0.0290	0.1030
PCDDs/PCDFs ratio	0.2420	0.2090	0.5370	0.2640	0.2040	0.5640	0.2570	0.2050	0.5570	0.2980	0.2900	0.5070	0.3370	0.2840	0.5420	0.3660	0.2840	0.5630
Total PCDD/Fs TEQ (pg I-TEQ/Nm^3)	**0.0482**	**0.0467**	**0.5080**	**0.0228**	**0.0387**	**0.3710**	**0.0419**	**0.0617**	**0.4040**	**0.0228**	**0.0631**	**0.2650**	**0.0093**	**0.0510**	**0.1540**	**0.0046**	**0.0372**	**0.1090**

Table 3a. Estimated monthly fluctuations of gas-particle concentrations of PCDD/Fs in the ambient air of sampling site B.

PCDD/Fs	Jul. 2009			Aug. 2009			Sep. 2009			Oct. 2009			Nov. 2009			Dec. 2009		
	C_p[a]	C_g[b]	Φ[c]	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ
2,3,7,8-TeCDD	0.0001	0.0021	0.0228	0.0001	0.0027	0.0217	0.0001	0.0036	0.0284	0.0004	0.0044	0.0775	0.0005	0.0039	0.1050	0.0009	0.0039	0.1860
1,2,3,7,8-PeCDD	0.0005	0.0039	0.1100	0.0006	0.0051	0.1030	0.0010	0.0066	0.1320	0.0039	0.0086	0.3090	0.0044	0.0069	0.3900	0.0071	0.0055	0.5630
1,2,3,4,7,8-HxCDD	0.0012	0.0019	0.3830	0.0014	0.0025	0.3610	0.0023	0.0030	0.4270	0.0075	0.0034	0.6910	0.0076	0.0023	0.7670	0.0096	0.0014	0.8730
1,2,3,6,7,8-HxCDD	0.0023	0.0034	0.3960	0.0027	0.0046	0.3750	0.0043	0.0055	0.4410	0.0152	0.0064	0.7030	0.0152	0.0044	0.7770	0.0192	0.0026	0.8790
1,2,3,7,8,9-HxCDD	0.0017	0.0023	0.4250	0.0021	0.0031	0.4020	0.0033	0.0037	0.4690	0.0133	0.0050	0.7270	0.0132	0.0034	0.7970	0.0165	0.0020	0.8910
1,2,3,4,6,7,8-HpCDD	0.0317	0.0087	0.7840	0.0397	0.0122	0.7650	0.0562	0.0132	0.8100	0.1550	0.0118	0.9290	0.1440	0.0072	0.9520	0.1650	0.0039	0.9770
OCDD	0.2110	0.0109	0.9510	0.2690	0.0158	0.9450	0.3640	0.0163	0.9570	0.4180	0.0059	0.9860	0.3810	0.0035	0.9910	0.4270	0.0018	0.9960
2,3,7,8-TeCDF	0.0003	0.0213	0.0154	0.0004	0.0273	0.0147	0.0007	0.0363	0.0194	0.0029	0.0511	0.0534	0.0036	0.0453	0.0726	0.0072	0.0474	0.1310
1,2,3,7,8-PeCDF	0.0013	0.0208	0.0599	0.0016	0.0268	0.0564	0.0028	0.0352	0.0731	0.0107	0.0468	0.1870	0.0128	0.0393	0.2460	0.0229	0.0353	0.3930
2,3,4,7,8-PeCDF	0.0022	0.0250	0.0804	0.0026	0.0323	0.0755	0.0045	0.0421	0.0972	0.0196	0.0624	0.2390	0.0230	0.0513	0.3100	0.0392	0.0437	0.4730
1,2,3,4,7,8-HxCDF	0.0061	0.0180	0.2530	0.0074	0.0236	0.2370	0.0120	0.0293	0.2910	0.0483	0.0396	0.5500	0.0510	0.0287	0.6400	0.0698	0.0191	0.7850
1,2,3,6,7,8-HxCDF	0.0059	0.0165	0.2620	0.0071	0.0217	0.2460	0.0116	0.0269	0.3010	0.0461	0.0360	0.5620	0.0484	0.0259	0.6520	0.0658	0.0171	0.7940
1,2,3,7,8,9-HxCDF	0.0005	0.0009	0.3650	0.0006	0.0012	0.3450	0.0010	0.0014	0.4090	0.0125	0.0060	0.6750	0.0127	0.0042	0.7530	0.0162	0.0026	0.8640
2,3,4,6,7,8-HxCDF	0.0073	0.0155	0.3200	0.0088	0.0204	0.3020	0.0141	0.0249	0.3620	0.0596	0.0350	0.6300	0.0612	0.0245	0.7140	0.0801	0.0155	0.8370
1,2,3,4,6,7,8-HpCDF	0.0359	0.0220	0.6210	0.0444	0.0300	0.5960	0.0656	0.0337	0.6600	0.2550	0.0432	0.8550	0.2430	0.0275	0.8980	0.2870	0.0153	0.9490
1,2,3,4,7,8,9-HpCDF	0.0054	0.0016	0.7730	0.0068	0.0022	0.7530	0.0096	0.0024	0.8000	0.0494	0.0040	0.9250	0.0459	0.0025	0.9490	0.0527	0.0013	0.9760
OCDF	0.0345	0.0027	0.9280	0.0439	0.0039	0.9190	0.0597	0.0040	0.9370	0.2220	0.0047	0.9790	0.2030	0.0028	0.9860	0.2280	0.0014	0.9940
PCDDs	0.2480	0.0333	0.8820	0.3160	0.0461	0.8730	0.4310	0.0518	0.8930	0.6130	0.0455	0.9310	0.5650	0.0315	0.9470	0.6450	0.0211	0.9680
PCDFs	0.0995	0.1440	0.4080	0.1240	0.1900	0.3950	0.1820	0.2360	0.4350	0.7270	0.3290	0.6880	0.7040	0.2520	0.7360	0.8680	0.1990	0.8140
PCDDs/PCDFs ratio	2.5000	0.2310	0.9150	2.5500	0.2430	0.9130	2.3700	0.2190	0.9150	0.8440	0.1380	0.8590	0.8030	0.1250	0.8650	0.7430	0.1060	0.8750
Total PCDD/Fs	**0.3480**	**0.1780**	**0.6620**	**0.4390**	**0.2360**	**0.6510**	**0.6130**	**0.2880**	**0.6800**	**1.3400**	**0.3740**	**0.7820**	**1.2700**	**0.2830**	**0.8170**	**1.5100**	**0.2200**	**0.8730**
PCDDs (pg I-TEQ/Nm^3)	0.0013	0.0049	0.2130	0.0017	0.0064	0.2040	0.0025	0.0083	0.2330	0.0079	0.0103	0.4330	0.0081	0.0084	0.4910	0.0111	0.0073	0.6010
PCDFs (pg I-TEQ/Nm^3)	0.0036	0.0210	0.1470	0.0044	0.0273	0.1390	0.0072	0.0351	0.1700	0.0306	0.0508	0.3760	0.0329	0.0408	0.4470	0.0483	0.0339	0.5870
PCDDs/PCDFs ratio	0.3690	0.2350	0.6110	0.3760	0.2360	0.6150	0.3510	0.2350	0.5990	0.2570	0.2030	0.5590	0.2450	0.2060	0.5440	0.2290	0.2160	0.5150
Total PCDD/Fs TEQ (pg I-TEQ/Nm^3)	**0.0050**	**0.0260**	**0.1600**	**0.0060**	**0.0337**	**0.1520**	**0.0097**	**0.0433**	**0.1830**	**0.0384**	**0.0611**	**0.3860**	**0.0410**	**0.0492**	**0.4550**	**0.0593**	**0.0413**	**0.5900**

[a] C_p: the concentration of PCDD/Fs in the particle phase; [b] C_g: the concentration of PCDD/Fs in the gas phase; [c] Φ: the fraction of PCDD/Fs bound to particles.

Table 3b. Estimated monthly fluctuations of gas-particle concentrations of PCDD/Fs in the ambient air of sampling site B.

PCDD/Fs	Jan. 2010			Feb. 2010			Mar. 2010			Apr. 2010			May. 2010			Jun. 2010		
	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ	C_p	C_g	Φ
2,3,7,8-TeCDD	0.0008	0.0035	0.1760	0.0003	0.0026	0.1000	0.0005	0.0040	0.1120	0.0004	0.0047	0.0728	0.0001	0.0036	0.0357	0.0001	0.0029	0.0267
1,2,3,7,8-PeCDD	0.0061	0.0050	0.5480	0.0029	0.0047	0.3830	0.0048	0.0070	0.4080	0.0030	0.0072	0.2980	0.0012	0.0063	0.1630	0.0008	0.0053	0.1260
1,2,3,4,7,8-HxCDD	0.0084	0.0013	0.8660	0.0050	0.0015	0.7660	0.0080	0.0023	0.7800	0.0049	0.0023	0.6840	0.0026	0.0027	0.4920	0.0018	0.0025	0.4160
1,2,3,6,7,8-HxCDD	0.0168	0.0025	0.8730	0.0102	0.0029	0.7760	0.0161	0.0043	0.7900	0.0092	0.0040	0.6960	0.0050	0.0048	0.5070	0.0034	0.0045	0.4300
1,2,3,7,8,9-HxCDD	0.0144	0.0019	0.8850	0.0088	0.0023	0.7960	0.0140	0.0033	0.8090	0.0068	0.0026	0.7210	0.0037	0.0032	0.5350	0.0026	0.0031	0.4580
1,2,3,4,6,7,8-HpCDD	0.1450	0.0036	0.9760	0.0961	0.0048	0.9530	0.1500	0.0070	0.9550	0.0871	0.0068	0.9280	0.0589	0.0104	0.8490	0.0451	0.0109	0.8050
OCDD	0.3760	0.0016	0.9960	0.2540	0.0022	0.9910	0.3970	0.0034	0.9920	0.5080	0.0072	0.9860	0.3680	0.0123	0.9680	0.2940	0.0135	0.9560
2,3,7,8-TeCDF	0.0060	0.0421	0.1240	0.0023	0.0304	0.0689	0.0040	0.0470	0.0776	0.0025	0.0476	0.0498	0.0009	0.0361	0.0243	0.0005	0.0294	0.0181
1,2,3,7,8-PeCDF	0.0194	0.0319	0.3780	0.0083	0.0265	0.2390	0.0141	0.0402	0.2600	0.0092	0.0422	0.1780	0.0035	0.0345	0.0914	0.0021	0.0286	0.0692
2,3,4,7,8-PeCDF	0.0335	0.0397	0.4570	0.0150	0.0346	0.3030	0.0252	0.0522	0.3260	0.0145	0.0487	0.2300	0.0057	0.0410	0.1210	0.0035	0.0342	0.0923
1,2,3,4,7,8-HxCDF	0.0607	0.0177	0.7750	0.0338	0.0194	0.6360	0.0545	0.0284	0.6570	0.0302	0.0258	0.5390	0.0143	0.0270	0.3460	0.0094	0.0240	0.2800
1,2,3,6,7,8-HxCDF	0.0573	0.0158	0.7840	0.0322	0.0175	0.6480	0.0518	0.0257	0.6680	0.0287	0.0233	0.5520	0.0138	0.0247	0.3580	0.0090	0.0221	0.2910
1,2,3,7,8,9-HxCDF	0.0142	0.0024	0.8560	0.0085	0.0028	0.7510	0.0134	0.0041	0.7670	0.0022	0.0011	0.6670	0.0012	0.0013	0.4740	0.0008	0.0012	0.3980
2,3,4,6,7,8-HxCDF	0.0699	0.0144	0.8290	0.0407	0.0165	0.7110	0.0651	0.0242	0.7290	0.0328	0.0200	0.6210	0.0166	0.0224	0.4240	0.0111	0.0204	0.3520
1,2,3,4,6,7,8-HpCDF	0.2520	0.0143	0.9460	0.1620	0.0183	0.8990	0.2550	0.0267	0.9050	0.1150	0.0199	0.8520	0.0713	0.0280	0.7180	0.0523	0.0280	0.6510
1,2,3,4,7,8,9-HpCDF	0.0464	0.0012	0.9740	0.0307	0.0016	0.9500	0.0480	0.0024	0.9530	0.0150	0.0013	0.9240	0.0101	0.0019	0.8410	0.0077	0.0020	0.7940
OCDF	0.2010	0.0013	0.9930	0.1350	0.0018	0.9870	0.2110	0.0027	0.9870	0.0844	0.0018	0.9790	0.0606	0.0030	0.9520	0.0482	0.0033	0.9360
PCDDs	0.5680	0.0194	0.9670	0.3780	0.0210	0.9470	0.5900	0.0311	0.9500	0.6190	0.0347	0.9470	0.4390	0.0434	0.9100	0.3470	0.0427	0.8900
PCDFs	0.7600	0.1810	0.8080	0.4690	0.1690	0.7350	0.7420	0.2540	0.7450	0.3340	0.2320	0.5910	0.1980	0.2200	0.4740	0.1450	0.1930	0.4280
PCDDs/PCDFs ratio	0.7470	0.1070	0.8750	0.8050	0.1240	0.8670	0.7950	0.1230	0.8660	1.8500	0.1500	0.9250	2.2200	0.1970	0.9180	2.4000	0.2210	0.9160
Total PCDD/Fs	**1.3300**	**0.2000**	**0.8690**	**0.8470**	**0.1900**	**0.8160**	**1.3300**	**0.2850**	**0.8240**	**0.9530**	**0.2660**	**0.7820**	**0.6370**	**0.2630**	**0.7080**	**0.4920**	**0.2360**	**0.6760**
PCDDs (pg I-TEQ/Nm^3)	0.0096	0.0066	0.5920	0.0054	0.0057	0.4870	0.0086	0.0085	0.5020	0.0054	0.0092	0.3680	0.0028	0.0079	0.2630	0.0020	0.0067	0.2280
PCDFs (pg I-TEQ/Nm^3)	0.0417	0.0308	0.5750	0.0217	0.0275	0.4420	0.0354	0.0413	0.4620	0.0187	0.0384	0.3280	0.0085	0.0337	0.2020	0.0056	0.0286	0.1630
PCDDs/PCDFs ratio	0.2300	0.2150	0.5170	0.2460	0.2050	0.5450	0.2430	0.2070	0.5410	0.2860	0.2400	0.5440	0.3320	0.2350	0.5850	0.3560	0.2350	0.6020
Total PCDD/Fs TEQ (pg I-TEQ/Nm^3)	**0.0513**	**0.0375**	**0.5780**	**0.0271**	**0.0331**	**0.4500**	**0.0441**	**0.0499**	**0.4690**	**0.0241**	**0.0477**	**0.3360**	**0.0114**	**0.0416**	**0.2150**	**0.0076**	**0.0353**	**0.1770**

was shown herein was caused mainly because of the variation of ambient temperatures among the different seasons (average of 20.6°C in winter and 28.9°C in summer). Accordingly, the relatively higher PCDD/Fs in the particle phase during winter was observed, which is similar to that obtained in many studies (Oh *et al*., 2001; Xu *et al*., 2009; Wang *et al*., 2010).

Scavenging Ratio

According to the method described in a previous section (Atmospheric wet deposition of PCDD/Fs), the calculated scavenging ratios of PCDD/Fs in the ambient air of sites A and B were listed in Tables 4 and 5, respectively. Results indicated that S_g values ranged from 5.46×10^2 (HxCDD) to 2.48×10^4 (OCDF) in ambient air for both site, but for which there is no consistent association between S_g values and level of chlorination. The total scavenging ratios for TCDD/F, PCDD/F, HCDD/F, and OCDD/F ranged from 1.35–5.42 × 10^3, 6.35–24.7 × 10^3, 9.69–40.7 × 10^3, and 39.0–41.8 × 10^3, respectively, for the ambient air of site A, and those of site B ranged from 1.64–8.42 × 10^3, 6.99–27.7 × 10^3, 11.3–41.1 × 10^3, and 39.9–41.9 × 10^3, respectively. It is also noted that the highest scavenging ratio of OCDD/F is similar to that measured at two different sites (Clinton Drive and Lang Road) in Houston (3.15×10^4) (Correa *et al*., 2006). The total scavenging ratio of PCDD/Fs (S_{tot}) in this study was close to the typical ratio of semivolatile organic compounds between 10^4 and 10^5 as shown in previous studies (Eitzer and Hites, 1989; Koester and Hites, 1992; Lohmann and Jones, 1998; McLachlan and Sellström, 2009). Additionally, the S_{tot} values increased with increasing number of chlorinated substitutes, which is consistent with the observations of gas-particle partitioning of PCDD/Fs in air.

Relative contributions of particle scavenging were calculated by dividing $S_p \times \Phi$ by S_{tot} as summarized in Tables 4 and 5. Accordingly, particle scavenging dominates in the wet deposition processes for the removal of PCDD/Fs from the atmosphere. The highest value of particle scavenging was observed at the highest chlorinated congener, approximately 98.6% for OCDD/F. The mean particle scavenging of PCDD/Fs were 87.3%, 78.0%, 86.6%, and 93.5% observed in spring, summer, fall, and winter, respectively, indicating a larger fraction of PCDD/Fs is associated with particles at lower temperatures. Therefore, many semivolatile organic compounds in the atmosphere are expected be scavenged most efficiently in cold weather (Koester and Hites, 1992).

Wet Deposition Flux of PCDD/Fs

The mean wet deposition fluxes of total PCDD/F in particle phase at site A were 1.5900 ng/m^2-month (0.0404 ng I-TEQ/m^2-month), 6.3400 ng/m^2-month (0.1200 ng I-TEQ/m^2-month), 1.7200 ng/m^2-month (0.0440 ng I-TEQ/m^2-month), and 0.1320 ng/m^2-month (0.0047 ng I-TEQ /m^2-month), respectively, with a total wet deposition flux of 1.6700 ng/m^2-month (0.0566 ng I-TEQ/m^2-month), 6.7600 ng/m^2-month (0.0922 ng I-TEQ/m^2-month), 1.7900 ng/m^2-month (0.0594 ng I-TEQ/m^2-month), and 0.1350 ng/m^2-month (0.0054 ng I-TEQ/m^2-month) in spring, summer, fall, and winter (Tables 6a and 6b). Similarly, the one at site B was 2.3400 ng/m^2-month (0.0468 ng I-TEQ/m^2-month), 8.4800 ng/m^2-month (0.1200 ng I-TEQ/m^2-month), 2.2200 ng/m^2-month (0.0495 ng I-TEQ/m^2-month), and 0.1630 ng/m^2-month (0.0055 ng I-TEQ/m^2-month), respectively, with a total wet deposition flux of 2.400 ng/m^2-month (0.0599 ng I-TEQ/m^2-month), 8.8100 ng/m^2-month (0.1780 ng I-TEQ/m^2-month), 2.2900 ng/m^2-month (0.0614 ng I-TEQ/m^2-month), and 0.1660 ng/m^2-month (0.0060 ng I-TEQ/m^2-month) in spring, summer, fall, and winter (Tables 7a and 7b). Results reveal that the particle bond deposition contributed 90.3%–98.2% of total wet deposition flux at both sites, which is similar to the observations of dry deposition flux of PCDD/Fs in air reported in the author's previous work (Huang *et al*., 2011).

With regard to the relative contribution of individual PCDD/Fs, OCDD was predominant among seventeen 2,3,7,8-substituted PCDD/Fs, accounting for 27.1%–59.2% of the total wet flux, followed by 1,2,3,4,6,7,8-HpCDF and OCDF. This is probably because the higher chlorinated PCDD/Fs are more closely associated with the fine particles and thus were effectively scavenged by wet deposition (Kaupp and McLachlan, 1999; Moon *et al*., 2005). Moreover, it has been shown that wet deposition is the major pathway responsible for the deposition of the higher chlorinated PCDD/Fs to a bare soil in rural Germany (Schröder *et al*., 1997). The PCDD/F profiles characterized in this study were similar to those reported previously (Wu *et al*., 2009; Lin *et al*., 2010a; Chang *et al*., 2011), but different from Ren's study (Ren *et al*., 2007). The discrepancy in congener profiles can be attributed to the different sources of PCDD/Fs among sampling sites.

The total wet PCDD/F deposition fluxes at all sampling locations were higher in summer than in winter (Figs. 1 and 2). Because of the great variations in precipitation, wet depositions differ greatly from season to season. In rainy seasons (from June to August), the average precipitation was 464 mm much higher than the dry seasons (from December to February) at 4.2 mm. Since precipitation is more effective in scavenging particle bond PCDD/Fs, the fluxes of total PCDD/Fs in rainy seasons were 51 and 53 times as high as those in dry seasons at sites A and B, respectively. A positive relationship between wet deposition flux and monthly rainfall was observed, revealing that wet deposition fluxes of total PCDD/F are affected by meteorological conditions. Consequently, precipitation is a significant mechanism for the removal of PCDD/Fs from atmosphere (Kaupp and McLachlan, 1999; Correa *et al*., 2006).

Annual Total Deposition Flux of PCDD/Fs

The annual total deposition flux of PCDD/Fs was calculated as the sum of annual dry and wet deposition fluxes, where the results of the dry deposition were reported in our earlier work (Huang *et al*., 2011) and those of wet deposition were obtained in the previous section. Within July 2009 to June 2010, the annual total (dry + wet) deposition fluxes of PCDD/Fs were 149 ng/m^2-year and

Table 4. Scavenging ratios of PCDD/Fs in the ambient air of sampling site A.

PCDD/Fs	S_g [a]	S_p [b]	Jul. 2009		Aug. 2009		Sep. 2009		Oct. 2009		Nov. 2009		Dec. 2009		Jan. 2010		Feb. 2010		Mar. 2010		Apr. 2010		May. 2010		Jun. 2010	
			S_{tot} [c]	% P [d]	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P
2,3,7,8-TeCDD	741	42000	1544	52.9	1455	50.0	1639	55.8	2998	76.6	3686	81.3	6216	89.7	6310	89.8	3705	81.4	4322	84.3	2861	75.4	1714	57.8	1353	46.0
1,2,3,7,8-PeCDD	9346	42000	12458	32.1	12091	29.2	12732	34.2	17027	58.0	18993	65.3	24469	79.5	24658	79.9	19190	66.0	20509	70.0	16743	56.8	13034	36.4	11738	26.2
1,2,3,4,7,8-HxCDD	546	42000	14843	97.6	13412	97.2	15539	97.8	25686	99.2	28859	99.4	34566	99.7	34729	99.7	29303	99.4	30716	99.5	25362	99.1	16600	98.0	12188	96.8
1,2,3,6,7,8-HxCDD	546	42000	15392	97.7	13927	97.3	16092	97.9	26258	99.2	29384	99.4	34927	99.7	35084	99.7	29824	99.5	31195	99.5	25944	99.2	17173	98.1	12677	97.0
1,2,3,7,8,9-HxCDD	546	42000	16517	98.0	14988	97.6	17222	98.1	27377	99.3	30396	99.5	35606	99.8	35752	99.8	30827	99.5	32113	99.6	27083	99.3	18337	98.3	13690	97.3
1,2,3,4,6,7,8-HpCDD	1946	42000	32195	98.5	30823	98.2	32568	98.6	38013	99.5	39140	99.6	40647	99.8	40687	99.8	39328	99.7	39667	99.7	37960	99.5	33434	98.8	29716	98.0
OCDD	3623	42000	39786	99.5	39337	99.4	39847	99.5	41217	99.8	41466	99.9	41763	100.0	41771	100.0	41514	99.9	41570	99.9	41219	99.8	40112	99.6	39008	99.4
2,3,7,8-TeCDF	1650	42000	2180	25.3	2124	23.2	2247	27.7	3158	49.7	3618	56.6	5360	72.0	5423	72.4	3621	56.7	4054	61.7	3060	48.0	2295	29.2	2055	20.5
1,2,3,7,8-PeCDF	4902	42000	6806	31.7	6584	28.9	7000	34.0	9966	57.5	11425	64.7	16116	78.8	16288	79.1	11533	65.1	12652	69.4	9726	56.2	7186	36.0	6354	25.9
2,3,4,7,8-PeCDF	4902	42000	7464	38.9	7162	35.8	7709	41.2	11512	65.0	13331	71.6	18825	83.7	19022	84.0	13489	72.1	14804	75.7	11230	63.8	7959	43.5	6861	32.4
1,2,3,4,7,8-HxCDF	1704	42000	10692	87.6	9689	85.9	11272	88.5	20100	95.4	23374	96.6	30293	98.4	30504	98.4	23794	96.8	25505	97.3	19712	95.2	12053	89.5	8802	84.1
1,2,3,6,7,8-HxCDF	1704	42000	11045	88.2	10009	86.5	11635	89.0	20600	95.6	23880	96.8	30717	98.5	30924	98.5	24304	96.9	25996	97.4	20215	95.5	12439	90.0	9096	84.7
1,2,3,7,8,9-HxCDF	1704	42000	14927	92.3	13580	91.2	15596	92.9	25416	97.2	28553	98.0	34303	99.1	34468	99.1	28988	98.1	30413	98.4	25088	97.2	16602	93.5	12424	90.0
2,3,4,6,7,8-HxCDF	1704	42000	13221	90.8	11999	89.5	13864	91.4	23441	96.7	26682	97.6	32935	98.8	33118	98.9	27119	97.7	28670	98.0	23085	96.5	14792	92.2	10940	88.0
1,2,3,4,6,7,8-HpCDF	1736	42000	25157	97.1	23491	96.6	25753	97.3	34066	99.0	36065	99.3	39023	99.7	39102	99.7	36375	99.3	37081	99.4	33919	99.0	26878	97.6	22113	96.1
1,2,3,4,7,8,9-HpCDF	1736	42000	31641	98.6	30225	98.3	32035	98.7	37746	99.5	38939	99.7	40546	99.8	40589	99.8	39137	99.7	39501	99.7	37687	99.5	32933	98.8	29079	98.1
OCDF	24752	42000	40565	94.9	40291	93.9	40610	95.1	41483	98.2	41644	98.8	41840	99.4	41845	99.5	41674	98.9	41713	99.0	41482	98.2	40773	95.7	40084	93.1

[a] Gas scavenging ratio, dimensionless; [b] Particle scavenging ratio, dimensionless; [c] The sum of gas and particle scavenging, dimensionless, $S_{tot} = S_g (1 - \Phi) + S_p \times \Phi$; [d] The percentage of particle scavenging, calculated by dividing $S_p \times \Phi$ by S_{tot}.

Table 5. Scavenging ratios of PCDD/Fs in the ambient air of sampling site B.

PCDD/Fs	S_g [a]	S_p [b]	Jul. 2009		Aug. 2009		Sep. 2009		Oct. 2009		Nov. 2009		Dec. 2009		Jan. 2010		Feb. 2010		Mar. 2010		Apr. 2010		May. 2010		Jun. 2010	
			S_{tot} [c]	% P [d]	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P	S_{tot}	% P
2,3,7,8-TeCDD	741	42000	1683	57.0	1635	55.7	1914	62.4	3939	82.6	5077	86.9	8423	92.8	8015	92.4	4882	86.3	5366	87.7	3744	81.6	2214	67.7	1842	60.9
1,2,3,7,8-PeCDD	9346	42000	12948	35.7	12724	34.1	13656	40.6	19427	66.7	22096	74.2	27740	85.3	27232	84.5	21868	73.6	22677	75.6	19074	65.6	14677	46.7	13454	39.2
1,2,3,4,7,8-HxCDD	546	42000	16409	97.9	15530	97.8	18242	98.3	29197	99.4	32341	99.6	36715	99.8	36435	99.8	32280	99.6	32892	99.6	28888	99.4	20945	98.7	17781	98.2
1,2,3,6,7,8-HxCDD	546	42000	16981	98.1	16084	97.9	18826	98.4	29705	99.5	32772	99.6	36987	99.8	36719	99.8	32718	99.6	33305	99.7	29409	99.4	21545	98.7	18364	98.3
1,2,3,7,8,9-HxCDD	546	42000	18147	98.3	17215	98.1	20005	98.6	30687	99.5	33591	99.7	37494	99.8	37251	99.8	33551	99.7	34090	99.7	30414	99.5	22742	98.9	19545	98.5
1,2,3,4,6,7,8-HpCDD	1946	42000	33353	98.7	32575	98.6	34396	98.9	39167	99.6	40080	99.8	41085	99.9	41034	99.9	40108	99.8	40218	99.8	39123	99.6	35963	99.2	34176	98.9
OCDD	3623	42000	40103	99.6	39853	99.5	40340	99.7	41457	99.9	41649	99.9	41841	100	41833	100	41661	99.9	41675	99.9	41457	99.9	40747	99.7	40299	99.6
2,3,7,8-TeCDF	1650	42000	2273	28.5	2244	27.6	2431	33.4	3804	58.9	4581	66.6	6956	79.4	6652	78.3	4430	65.3	4782	68.2	3661	57.2	2630	38.8	2381	32.0
1,2,3,7,8-PeCDF	4902	42000	7124	35.3	6993	33.9	7612	40.3	11828	66.3	14021	73.6	19487	84.7	18921	83.9	13761	72.9	14536	75.0	11508	65.0	8292	46.3	7468	38.9
2,3,4,7,8-PeCDF	4902	42000	7882	42.8	7701	41.2	8506	48.0	13785	73.0	16401	79.4	22459	88.5	21870	87.8	16133	78.8	16995	80.6	13421	71.9	9392	54.1	8325	46.6
1,2,3,4,7,8-HxCDF	1704	42000	11884	89.3	11261	88.5	13408	91.0	23847	96.8	27492	97.8	33337	98.9	32917	98.8	27341	97.7	28183	97.9	23439	96.7	15659	92.9	13004	90.6
1,2,3,6,7,8-HxCDF	1704	42000	12268	89.8	11624	89.0	13820	91.4	24343	96.9	27959	97.9	33683	99.0	33276	98.9	27816	97.8	28640	98.0	23942	96.8	16115	93.2	13411	91.0
1,2,3,7,8,9-HxCDF	1704	42000	16416	93.4	15588	92.9	18180	94.5	28901	98.1	32052	98.7	36505	99.4	36216	99.3	31982	98.7	32609	98.8	28585	98.0	20783	95.7	17733	94.2
2,3,4,6,7,8-HxCDF	1704	42000	14610	92.1	13854	91.4	16304	93.4	27080	97.7	30458	98.4	35450	99.2	35114	99.2	30360	98.4	31071	98.5	26726	97.6	18807	94.8	15867	93.1
1,2,3,4,6,7,8-HpCDF	1736	42000	26733	97.5	25755	97.3	28320	97.9	36179	99.3	37906	99.5	39959	99.8	39845	99.8	37923	99.5	38181	99.6	36054	99.3	30654	98.4	27956	97.8
1,2,3,4,7,8,9-HpCDF	1736	42000	32847	98.8	32042	98.7	33942	99.0	38971	99.7	39942	99.8	41016	99.9	40961	99.9	39970	99.8	40089	99.8	38922	99.7	35583	99.2	33709	98.9
OCDF	24752	42000	40765	95.6	40613	95.1	40922	96.2	41640	98.8	41765	99.2	41893	99.6	41887	99.6	41772	99.2	41783	99.3	41639	98.8	41179	97.1	40893	96.1

Table 6a. Estimated monthly fluctuations of wet deposition flux of PCDD/Fs in the ambient air of sampling site A (ng/m^2-month).

PCDD/Fs	Jul. 2009			Aug. 2009			Sep. 2009			Oct. 2009			Nov. 2009			Dec. 2009		
	dissolved[a]	particle[b]	F_w[c]	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w
2,3,7,8-TeCDD	0.0006	0.0006	0.0012	0.0033	0.0033	0.0065	0.0006	0.0008	0.0014	0.0002	0.0005	0.0007	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
1,2,3,7,8-PeCDD	0.0127	0.0060	0.0188	0.0727	0.0299	0.1030	0.0131	0.0068	0.0199	0.0048	0.0066	0.0114	0.0007	0.0013	0.0020	0.0001	0.0001	0.0002
1,2,3,4,7,8-HxCDD	0.0004	0.0143	0.0147	0.0021	0.0726	0.0747	0.0004	0.0156	0.0159	0.0001	0.0144	0.0145	0.0001	0.0026	0.0026	0.0001	0.0002	0.0002
1,2,3,6,7,8-HxCDD	0.0006	0.0258	0.0264	0.0036	0.1310	0.1350	0.0006	0.0280	0.0286	0.0002	0.0289	0.0292	0.0001	0.0052	0.0052	0.0001	0.0004	0.0004
1,2,3,7,8,9-HxCDD	0.0004	0.0188	0.0191	0.0023	0.0956	0.0980	0.0004	0.0203	0.0207	0.0002	0.0240	0.0242	0.0001	0.0043	0.0043	0.0001	0.0003	0.0003
1,2,3,4,6,7,8-HpCDD	0.0038	0.2540	0.2570	0.0245	1.3700	1.3900	0.0038	0.2670	0.2700	0.0013	0.2540	0.2560	0.0002	0.0420	0.0422	0.0001	0.0028	0.0028
OCDD	0.0060	1.1500	1.1500	0.0407	6.3900	6.4300	0.0061	1.1900	1.2000	0.0012	0.6680	0.6690	0.0001	0.1080	0.1080	0.0001	0.0069	0.0069
2,3,7,8-TeCDF	0.0093	0.0032	0.0125	0.0527	0.0159	0.0686	0.0097	0.0037	0.0134	0.0042	0.0042	0.0084	0.0007	0.0009	0.0015	0.0001	0.0001	0.0001
1,2,3,7,8-PeCDF	0.0294	0.0137	0.0431	0.1670	0.0680	0.2350	0.0304	0.0156	0.0460	0.0123	0.0166	0.0289	0.0019	0.0034	0.0053	0.0001	0.0004	0.0005
2,3,4,7,8-PeCDF	0.0354	0.0225	0.0579	0.2010	0.1120	0.3130	0.0365	0.0256	0.0621	0.0169	0.0314	0.0484	0.0026	0.0064	0.0090	0.0001	0.0007	0.0008
1,2,3,4,7,8-HxCDF	0.0108	0.0764	0.0872	0.0628	0.3830	0.4450	0.0110	0.0844	0.0954	0.0039	0.0807	0.0846	0.0005	0.0152	0.0157	0.0001	0.0013	0.0013
1,2,3,6,7,8-HxCDF	0.0081	0.0600	0.0680	0.0470	0.3010	0.3480	0.0082	0.0662	0.0744	0.0037	0.0799	0.0836	0.0005	0.0150	0.0155	0.0001	0.0012	0.0013
1,2,3,7,8,9-HxCDF	0.0007	0.0078	0.0085	0.0038	0.0396	0.0435	0.0007	0.0085	0.0092	0.0005	0.0170	0.0174	0.0001	0.0031	0.0031	0.0001	0.0002	0.0002
2,3,4,6,7,8-HxCDF	0.0066	0.0655	0.0722	0.0390	0.3300	0.3690	0.0067	0.0719	0.0786	0.0034	0.0972	0.1010	0.0004	0.0179	0.0183	0.0001	0.0014	0.0014
1,2,3,4,6,7,8-HpCDF	0.0111	0.3740	0.3850	0.0689	1.9600	2.0300	0.0111	0.3980	0.4100	0.0041	0.4070	0.4110	0.0005	0.0691	0.0696	0.0001	0.0048	0.0048
1,2,3,4,7,8,9-HpCDF	0.0009	0.0627	0.0636	0.0058	0.3370	0.3430	0.0009	0.0660	0.0669	0.0004	0.0873	0.0877	0.0001	0.0144	0.0145	0.0001	0.0010	0.0010
OCDF	0.0200	0.3720	0.3920	0.1340	2.0600	2.2000	0.0201	0.3880	0.4080	0.0057	0.3120	0.3170	0.0006	0.0504	0.0510	0.0001	0.0032	0.0032
PCDDs	0.0245	1.4700	1.4900	0.1490	8.0900	8.2400	0.0249	1.5300	1.5600	0.0080	0.9970	1.0000	0.0011	0.1630	0.1640	0.0001	0.0107	0.0107
PCDFs	0.1320	1.0600	1.1900	0.7820	5.6100	6.3900	0.1350	1.1300	1.2600	0.0550	1.1300	1.1900	0.0078	0.1960	0.2040	0.0004	0.0142	0.0146
PCDDs/PCDFs ratio	0.1850	1.3900	1.2500	0.1910	1.4400	1.2900	0.1840	1.3600	1.2300	0.1450	0.8800	0.8460	0.1380	0.8330	0.8060	0.1260	0.7500	0.7340
Total PCDD/Fs	**0.1570**	**2.5300**	**2.6800**	**0.9320**	**13.7000**	**14.6000**	**0.1600**	**2.6600**	**2.8200**	**0.0630**	**2.1300**	**2.1900**	**0.0089**	**0.3590**	**0.3680**	**0.0004**	**0.0249**	**0.0253**
PCDDs TEQ	0.0071	0.0132	0.0203	0.0407	0.0682	0.1090	0.0073	0.0144	0.0217	0.0026	0.0138	0.0164	0.0004	0.0025	0.0029	0.0001	0.0002	0.0002
PCDFs TEQ	0.0229	0.0380	0.0608	0.1300	0.1910	0.3220	0.0235	0.0421	0.0656	0.0107	0.0497	0.0604	0.0016	0.0095	0.0111	0.0001	0.0008	0.0009
PCDDs/PCDFs ratio	0.3120	0.3480	0.3350	0.3120	0.3560	0.3380	0.3110	0.3420	0.3310	0.2450	0.2770	0.2710	0.2420	0.2650	0.2610	0.2330	0.2420	0.2420
Total PCDD/Fs TEQ (ng I-TEQ/m^2-month)	**0.0300**	**0.0512**	**0.0812**	**0.1710**	**0.2600**	**0.4310**	**0.0309**	**0.0565**	**0.0874**	**0.0133**	**0.0635**	**0.0768**	**0.0020**	**0.0120**	**0.0140**	**0.0001**	**0.0010**	**0.0011**

[a] dessolved: the dissolved phase wet deposition flux of PCDD/Fs in the raindrop; [b] particle: the particle phase wet deposition flux of PCDD/Fs in the raindrop; [c] F_w: total wet deposition flux.

Table 6b. Estimated monthly fluctuations of wet deposition flux of PCDD/Fs in the ambient air of sampling site A (ng/m^2-month).

PCDD/Fs	Jan. 2010			Feb. 2010			Mar. 2010			Apr. 2010			May. 2010			Jun. 2010		
	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w
2,3,7,8-TeCDD	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0003	0.0008	0.0010	0.0007	0.0010	0.0017	0.0007	0.0006	0.0013
1,2,3,7,8-PeCDD	0.0001	0.0004	0.0005	0.0006	0.0011	0.0017	0.0001	0.0002	0.0003	0.0050	0.0065	0.0115	0.0157	0.0090	0.0246	0.0155	0.0055	0.0210
1,2,3,4,7,8-HxCDD	0.0001	0.0006	0.0006	0.0001	0.0022	0.0022	0.0001	0.0004	0.0004	0.0001	0.0113	0.0114	0.0004	0.0202	0.0206	0.0005	0.0138	0.0143
1,2,3,6,7,8-HxCDD	0.0001	0.0011	0.0011	0.0001	0.0044	0.0044	0.0001	0.0007	0.0007	0.0002	0.0201	0.0203	0.0007	0.0363	0.0370	0.0008	0.0250	0.0258
1,2,3,7,8,9-HxCDD	0.0001	0.0009	0.0009	0.0001	0.0036	0.0036	0.0001	0.0006	0.0006	0.0001	0.0142	0.0143	0.0005	0.0262	0.0267	0.0005	0.0183	0.0189
1,2,3,4,6,7,8-HpCDD	0.0001	0.0080	0.0080	0.0001	0.0349	0.0350	0.0001	0.0057	0.0057	0.0007	0.1380	0.1390	0.0042	0.3320	0.3360	0.0057	0.2770	0.2820
OCDD	0.0001	0.0197	0.0198	0.0001	0.0890	0.0891	0.0001	0.0143	0.0143	0.0010	0.5450	0.5460	0.0064	1.4500	1.4600	0.0096	1.3300	1.3400
2,3,7,8-TeCDF	0.0001	0.0003	0.0004	0.0005	0.0007	0.0013	0.0001	0.0001	0.0002	0.0042	0.0038	0.0080	0.0117	0.0048	0.0165	0.0111	0.0029	0.0140
1,2,3,7,8-PeCDF	0.0003	0.0011	0.0014	0.0015	0.0029	0.0044	0.0002	0.0005	0.0008	0.0123	0.0158	0.0281	0.0366	0.0206	0.0571	0.0354	0.0124	0.0478
2,3,4,7,8-PeCDF	0.0004	0.0020	0.0023	0.0021	0.0054	0.0075	0.0003	0.0010	0.0013	0.0144	0.0254	0.0398	0.0438	0.0337	0.0776	0.0428	0.0205	0.0633
1,2,3,4,7,8-HxCDF	0.0001	0.0037	0.0037	0.0004	0.0128	0.0132	0.0001	0.0022	0.0023	0.0035	0.0699	0.0734	0.0130	0.1100	0.1230	0.0136	0.0717	0.0853
1,2,3,6,7,8-HxCDF	0.0001	0.0036	0.0037	0.0004	0.0126	0.0130	0.0001	0.0022	0.0022	0.0026	0.0543	0.0568	0.0097	0.0865	0.0962	0.0102	0.0564	0.0666
1,2,3,7,8,9-HxCDF	0.0001	0.0007	0.0007	0.0001	0.0026	0.0026	0.0001	0.0004	0.0004	0.0002	0.0063	0.0065	0.0008	0.0111	0.0118	0.0008	0.0075	0.0084
2,3,4,6,7,8-HxCDF	0.0001	0.0041	0.0041	0.0004	0.0150	0.0154	0.0001	0.0026	0.0026	0.0020	0.0556	0.0576	0.0079	0.0935	0.1010	0.0085	0.0625	0.0710
1,2,3,4,6,7,8-HpCDF	0.0001	0.0137	0.0137	0.0004	0.0576	0.0580	0.0001	0.0094	0.0095	0.0024	0.2350	0.2370	0.0125	0.5040	0.5170	0.0156	0.3870	0.4020
1,2,3,4,7,8,9-HpCDF	0.0001	0.0027	0.0028	0.0001	0.0120	0.0120	0.0001	0.0019	0.0020	0.0002	0.0344	0.0346	0.0010	0.0822	0.0832	0.0013	0.0681	0.0695
OCDF	0.0001	0.0093	0.0093	0.0005	0.0417	0.0422	0.0001	0.0067	0.0068	0.0033	0.1800	0.1830	0.0214	0.4740	0.4950	0.0317	0.4290	0.4600
PCDDs	0.0001	0.0308	0.0309	0.0009	0.1350	0.1360	0.0001	0.0219	0.0220	0.0073	0.7360	0.7430	0.0286	1.8800	1.9100	0.0333	1.6700	1.7100
PCDFs	0.0010	0.0411	0.0422	0.0063	0.1630	0.1700	0.0010	0.0271	0.0281	0.0451	0.6800	0.7250	0.1580	1.4200	1.5800	0.1710	1.1200	1.2900
PCDDs/PCDFs ratio	0.1250	0.7480	0.7320	0.1370	0.8280	0.8020	0.1350	0.8050	0.7830	0.1610	1.0800	1.0200	0.1810	1.3200	1.2100	0.1940	1.5000	1.3300
Total PCDD/Fs	**0.0012**	**0.0719**	**0.0731**	**0.0072**	**0.2990**	**0.3060**	**0.0011**	**0.0490**	**0.0501**	**0.0524**	**1.4200**	**1.4700**	**0.1870**	**3.3000**	**3.4900**	**0.2040**	**2.7900**	**3.0000**
PCDDs TEQ	0.0001	0.0006	0.0006	0.0003	0.0021	0.0024	0.0000	0.0004	0.0004	0.0028	0.0105	0.0133	0.0088	0.0185	0.0273	0.0087	0.0132	0.0218
PCDFs TEQ	0.0002	0.0024	0.0027	0.0013	0.0080	0.0093	0.0002	0.0014	0.0016	0.0091	0.0353	0.0444	0.0282	0.0549	0.0831	0.0278	0.0359	0.0637
PCDDs/PCDFs ratio	0.2330	0.2420	0.2410	0.2420	0.2640	0.2610	0.2400	0.2570	0.2550	0.3060	0.2980	0.3000	0.3110	0.3370	0.3280	0.3120	0.3660	0.3430
Total PCDD/Fs TEQ (ng I-TEQ/m^2-month)	**0.0003**	**0.0030**	**0.0033**	**0.0016**	**0.0101**	**0.0117**	**0.0002**	**0.0018**	**0.0020**	**0.0119**	**0.0459**	**0.0578**	**0.0370**	**0.0734**	**0.1100**	**0.0365**	**0.0491**	**0.0855**

Table 7a. Estimated monthly fluctuations of wet deposition flux of PCDD/Fs in the ambient air of sampling site B (ng/m^2-month).

PCDD/Fs	Jul. 2009			Aug. 2009			Sep. 2009			Oct. 2009			Nov. 2009			Dec. 2009		
	dissolved[a]	particle[b]	F_w [c]	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w
2,3,7,8-TeCDD	0.0003	0.0004	0.0007	0.0019	0.0024	0.0043	0.0004	0.0006	0.0010	0.0002	0.0008	0.0009	0.0000	0.0002	0.0002	0.0000	0.0000	0.0000
1,2,3,7,8-PeCDD	0.0073	0.0041	0.0114	0.0443	0.0229	0.0672	0.0082	0.0056	0.0138	0.0039	0.0079	0.0118	0.0006	0.0017	0.0023	0.0000	0.0001	0.0002
1,2,3,4,7,8-HxCDD	0.0002	0.0100	0.0102	0.0013	0.0564	0.0577	0.0002	0.0128	0.0130	0.0001	0.0153	0.0154	0.0000	0.0029	0.0029	0.0000	0.0002	0.0002
1,2,3,6,7,8-HxCDD	0.0004	0.0190	0.0194	0.0023	0.1080	0.1100	0.0004	0.0243	0.0247	0.0002	0.0310	0.0312	0.0000	0.0058	0.0058	0.0000	0.0004	0.0004
1,2,3,7,8,9-HxCDD	0.0003	0.0146	0.0148	0.0016	0.0826	0.0842	0.0003	0.0185	0.0188	0.0001	0.0271	0.0272	0.0000	0.0050	0.0050	0.0000	0.0003	0.0003
1,2,3,4,6,7,8-HpCDD	0.0034	0.2670	0.2700	0.0222	1.5600	1.5800	0.0035	0.3170	0.3210	0.0011	0.3150	0.3170	0.0001	0.0543	0.0544	0.0000	0.0035	0.0035
OCDD	0.0079	1.7800	1.7800	0.0535	10.6000	10.6000	0.0079	2.0600	2.0600	0.0010	0.8520	0.8530	0.0001	0.1440	0.1440	0.0000	0.0090	0.0090
2,3,7,8-TeCDF	0.0070	0.0028	0.0098	0.0422	0.0160	0.0582	0.0081	0.0041	0.0121	0.0041	0.0059	0.0100	0.0007	0.0013	0.0020	0.0000	0.0002	0.0002
1,2,3,7,8-PeCDF	0.0204	0.0112	0.0316	0.1230	0.0630	0.1860	0.0232	0.0157	0.0389	0.0111	0.0219	0.0330	0.0017	0.0048	0.0066	0.0001	0.0005	0.0006
2,3,4,7,8-PeCDF	0.0246	0.0184	0.0430	0.1480	0.1040	0.2520	0.0278	0.0256	0.0534	0.0148	0.0400	0.0549	0.0023	0.0087	0.0110	0.0001	0.0008	0.0009
1,2,3,4,7,8-HxCDF	0.0062	0.0513	0.0575	0.0376	0.2890	0.3260	0.0067	0.0679	0.0746	0.0033	0.0984	0.1020	0.0004	0.0193	0.0197	0.0000	0.0015	0.0015
1,2,3,6,7,8-HxCDF	0.0057	0.0495	0.0552	0.0346	0.2790	0.3130	0.0062	0.0654	0.0715	0.0030	0.0939	0.0969	0.0004	0.0183	0.0187	0.0000	0.0014	0.0014
1,2,3,7,8,9-HxCDF	0.0003	0.0044	0.0047	0.0019	0.0247	0.0266	0.0003	0.0056	0.0060	0.0005	0.0256	0.0261	0.0001	0.0048	0.0049	0.0000	0.0003	0.0003
2,3,4,6,7,8-HxCDF	0.0053	0.0614	0.0666	0.0325	0.3460	0.3790	0.0057	0.0799	0.0856	0.0029	0.1210	0.1240	0.0004	0.0231	0.0235	0.0000	0.0017	0.0017
1,2,3,4,6,7,8-HpCDF	0.0076	0.3030	0.3100	0.0487	1.7400	1.7900	0.0079	0.3700	0.3780	0.0036	0.5200	0.5240	0.0004	0.0919	0.0923	0.0000	0.0060	0.0060
1,2,3,4,7,8,9-HpCDF	0.0006	0.0456	0.0462	0.0036	0.2660	0.2700	0.0006	0.0544	0.0549	0.0003	0.1010	0.1010	0.0000	0.0174	0.0174	0.0000	0.0011	0.0011
OCDF	0.0132	0.2900	0.3040	0.0890	1.7200	1.8100	0.0133	0.3370	0.3510	0.0057	0.4520	0.4580	0.0006	0.0765	0.0772	0.0000	0.0048	0.0048
PCDDs	0.0198	2.0900	2.1100	0.1270	12.4000	12.5000	0.0209	2.4300	2.4600	0.0066	1.2500	1.2600	0.0009	0.2140	0.2150	0.0000	0.0135	0.0136
PCDFs	0.0909	0.8380	0.9290	0.5610	4.8500	5.4100	0.0997	1.0300	1.1300	0.0494	1.4800	1.5300	0.0070	0.2660	0.2730	0.0003	0.0182	0.0185
PCDDs/PCDFs ratio	0.2180	2.5000	2.2700	0.2260	2.5500	2.3100	0.2090	2.3700	2.1800	0.1340	0.8440	0.8210	0.1270	0.8030	0.7850	0.1150	0.7430	0.7320
Total PCDD/Fs	**0.1110**	**2.9300**	**3.0400**	**0.6880**	**17.2000**	**17.9000**	**0.1210**	**3.4600**	**3.5800**	**0.0560**	**2.7300**	**2.7900**	**0.0079**	**0.4800**	**0.4880**	**0.0003**	**0.0318**	**0.0321**
PCDDs TEQ	0.0041	0.0113	0.0154	0.0248	0.0647	0.0895	0.0046	0.0142	0.0188	0.0022	0.0160	0.0182	0.0003	0.0031	0.0034	0.0000	0.0002	0.0002
PCDFs TEQ	0.0159	0.0305	0.0463	0.0957	0.1720	0.2680	0.0178	0.0405	0.0583	0.0094	0.0623	0.0717	0.0014	0.0124	0.0139	0.0001	0.0010	0.0011
PCDDs/PCDFs ratio	0.2590	0.3690	0.3320	0.2590	0.3760	0.3340	0.2580	0.3510	0.3230	0.2310	0.2570	0.2540	0.2270	0.2450	0.2430	0.2180	0.2290	0.2280
Total PCDD/Fs TEQ (ng I-TEQ/m^2-month)	**0.0200**	**0.0417**	**0.0617**	**0.1210**	**0.2370**	**0.3570**	**0.0225**	**0.0547**	**0.0771**	**0.0116**	**0.0783**	**0.0899**	**0.0017**	**0.0155**	**0.0172**	**0.0001**	**0.0013**	**0.0013**

Table 7b. Estimated monthly fluctuations of wet deposition flux of PCDD/Fs in the ambient air of sampling site B (ng/m^2-month).

PCDD/Fs	Jan. 2010			Feb. 2010			Mar. 2010			Apr. 2010			May. 2010			Jun. 2010		
	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w	dissolved	particle	F_w
2,3,7,8-TeCDD	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0002	0.0007	0.0009	0.0005	0.0011	0.0016	0.0006	0.0009	0.0014
1,2,3,7,8-PeCDD	0.0001	0.0004	0.0005	0.0005	0.0013	0.0017	0.0001	0.0002	0.0003	0.0032	0.0061	0.0094	0.0111	0.0097	0.0208	0.0128	0.0083	0.0211
1,2,3,4,7,8-HxCDD	0.0001	0.0005	0.0005	0.0001	0.0022	0.0022	0.0001	0.0003	0.0003	0.0001	0.0099	0.0100	0.0003	0.0206	0.0209	0.0004	0.0193	0.0196
1,2,3,6,7,8-HxCDD	0.0001	0.0011	0.0011	0.0001	0.0045	0.0045	0.0001	0.0007	0.0007	0.0001	0.0186	0.0187	0.0005	0.0391	0.0395	0.0006	0.0366	0.0372
1,2,3,7,8,9-HxCDD	0.0001	0.0009	0.0009	0.0001	0.0039	0.0039	0.0001	0.0006	0.0006	0.0001	0.0137	0.0138	0.0003	0.0295	0.0299	0.0004	0.0279	0.0283
1,2,3,4,6,7,8-HpCDD	0.0001	0.0091	0.0092	0.0001	0.0424	0.0425	0.0001	0.0063	0.0063	0.0006	0.1760	0.1760	0.0038	0.4650	0.4690	0.0055	0.4870	0.4920
OCDD	0.0001	0.0237	0.0237	0.0001	0.1120	0.1120	0.0001	0.0167	0.0167	0.0013	1.0200	1.0200	0.0084	2.9000	2.9100	0.0126	3.1700	3.1800
2,3,7,8-TeCDF	0.0001	0.0004	0.0005	0.0005	0.0010	0.0015	0.0001	0.0002	0.0002	0.0038	0.0050	0.0088	0.0112	0.0071	0.0183	0.0125	0.0059	0.0183
1,2,3,7,8-PeCDF	0.0002	0.0012	0.0015	0.0014	0.0037	0.0050	0.0002	0.0006	0.0008	0.0099	0.0185	0.0284	0.0318	0.0274	0.0592	0.0360	0.0229	0.0589
2,3,4,7,8-PeCDF	0.0003	0.0021	0.0024	0.0018	0.0066	0.0084	0.0003	0.0011	0.0013	0.0115	0.0293	0.0407	0.0378	0.0446	0.0824	0.0431	0.0376	0.0807
1,2,3,4,7,8-HxCDF	0.0001	0.0038	0.0039	0.0003	0.0149	0.0153	0.0001	0.0023	0.0023	0.0021	0.0609	0.0630	0.0087	0.1130	0.1220	0.0105	0.1010	0.1120
1,2,3,6,7,8-HxCDF	0.0001	0.0036	0.0037	0.0003	0.0142	0.0145	0.0001	0.0022	0.0022	0.0019	0.0580	0.0599	0.0079	0.1090	0.1170	0.0097	0.0975	0.1070
1,2,3,7,8,9-HxCDF	0.0001	0.0009	0.0009	0.0001	0.0037	0.0038	0.0001	0.0006	0.0006	0.0001	0.0044	0.0045	0.0004	0.0091	0.0095	0.0005	0.0085	0.0090
2,3,4,6,7,8-HxCDF	0.0001	0.0044	0.0044	0.0003	0.0180	0.0182	0.0001	0.0027	0.0028	0.0016	0.0661	0.0678	0.0072	0.1310	0.1380	0.0090	0.1200	0.1290
1,2,3,4,6,7,8-HpCDF	0.0001	0.0159	0.0159	0.0003	0.0716	0.0720	0.0001	0.0107	0.0108	0.0017	0.2310	0.2330	0.0091	0.5630	0.5720	0.0125	0.5640	0.5770
1,2,3,4,7,8,9-HpCDF	0.0001	0.0029	0.0029	0.0001	0.0135	0.0136	0.0001	0.0020	0.0020	0.0001	0.0303	0.0304	0.0006	0.0798	0.0805	0.0009	0.0833	0.0842
OCDF	0.0001	0.0126	0.0127	0.0005	0.0597	0.0601	0.0001	0.0089	0.0089	0.0022	0.1700	0.1720	0.0141	0.4790	0.4930	0.0211	0.5200	0.5410
PCDDs	0.0001	0.0358	0.0359	0.0007	0.1670	0.1670	0.0001	0.0248	0.0249	0.0055	1.2500	1.2500	0.0249	3.4700	3.4900	0.0328	3.7500	3.7800
PCDFs	0.0008	0.0479	0.0487	0.0055	0.2070	0.2120	0.0008	0.0312	0.0320	0.0348	0.6740	0.7090	0.1290	1.5600	1.6900	0.1560	1.5600	1.7200
PCDDs/PCDFs ratio	0.1160	0.7470	0.7360	0.1270	0.8050	0.7870	0.1260	0.7950	0.7790	0.1580	1.8500	1.7700	0.1930	2.2200	2.0700	0.2110	2.4000	2.2000
Total PCDD/Fs	**0.0009**	**0.0837**	**0.0846**	**0.0062**	**0.3730**	**0.3800**	**0.0009**	**0.0560**	**0.0569**	**0.0403**	**1.9200**	**1.9600**	**0.1540**	**5.0300**	**5.1800**	**0.1880**	**5.3100**	**5.5000**
PCDDs TEQ	0.0000	0.0006	0.0006	0.0003	0.0024	0.0026	0.0001	0.0004	0.0004	0.0018	0.0108	0.0126	0.0062	0.0224	0.0286	0.0072	0.0214	0.0286
PCDFs TEQ	0.0002	0.0026	0.0028	0.0011	0.0096	0.0107	0.0002	0.0015	0.0017	0.0072	0.0378	0.0450	0.0241	0.0674	0.0916	0.0277	0.0602	0.0879
PCDDs/PCDFs ratio	0.2190	0.2300	0.2290	0.2270	0.2460	0.2440	0.2260	0.2430	0.2420	0.2510	0.2860	0.2800	0.2570	0.3320	0.3120	0.2580	0.3560	0.3250
Total PCDD/Fs TEQ (ng I-TEQ/m^2-month)	**0.0002**	**0.0032**	**0.0035**	**0.0014**	**0.0119**	**0.0133**	**0.0002**	**0.0019**	**0.0021**	**0.0090**	**0.0486**	**0.0576**	**0.0303**	**0.0898**	**0.1200**	**0.0349**	**0.0816**	**0.1160**

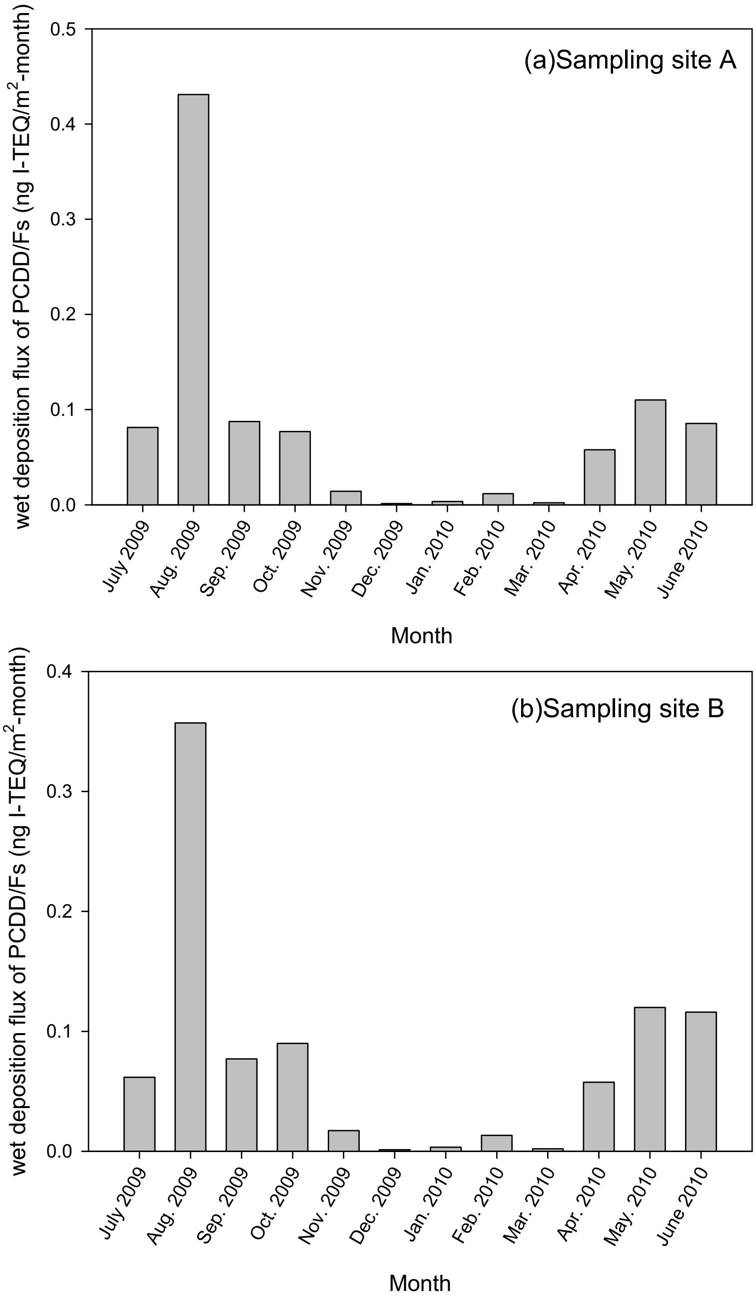

Fig. 1. Estimated monthly fluctuations of wet deposition flux of PCDD/Fs in the ambient air of sampling areas A and B (ng I-TEQ/m^2-month).

177 ng/m^2-year for sites A and B, respectively, and wet deposition contributed 20.8% and 23.2% of those. With regard to TEQ, the total deposition fluxes of PCDD/Fs were 5.02 ng I-TEQ/m^2-year and 5.11 ng I-TEQ/m^2-year for sites A and B, respectively, and wet deposition contributed 19.2% and 21.8% of those. The above results reveal that dry deposition was more dominant than the wet deposition for the atmospheric deposition of PCDD/Fs. Results also showed that the total deposition flux was highest in winter, which is consistent with the observations reported in other studies (Ogura *et al.*, 2001; Moon *et al.*, 2005). The increase of total deposition flux in winter may be due to the inversion layers that reduced atmospheric dilution (Oka *et al.*, 2006). Additionally, the annual total deposition fluxes were comparable to those measured in the urban area (68–228 ng/m^2-year) and suburban area (38–252 ng/m^2-year) in Korea (Moon *et al.*, 2005) and several locations (1.0–14.9 ng I-TEQ/m^2-year) in Germany (Wallenhorst *et al.*, 1997). However, the annual PCDD/F deposition fluxes from Tokyo, Yokohama, Tsukuba, and Tanzawa in Japan (160–3500 ng/m^2-year) were about 1 to 24 times higher than those in this study (Ogura *et al.*, 2001). Atmospheric deposition is the key process governing the transfer of PCDD/Fs into food chains (Welsch-Pausch and McLachlan, 1998), and therefore, its impact on human exposure to PCDD/Fs is of great importance.

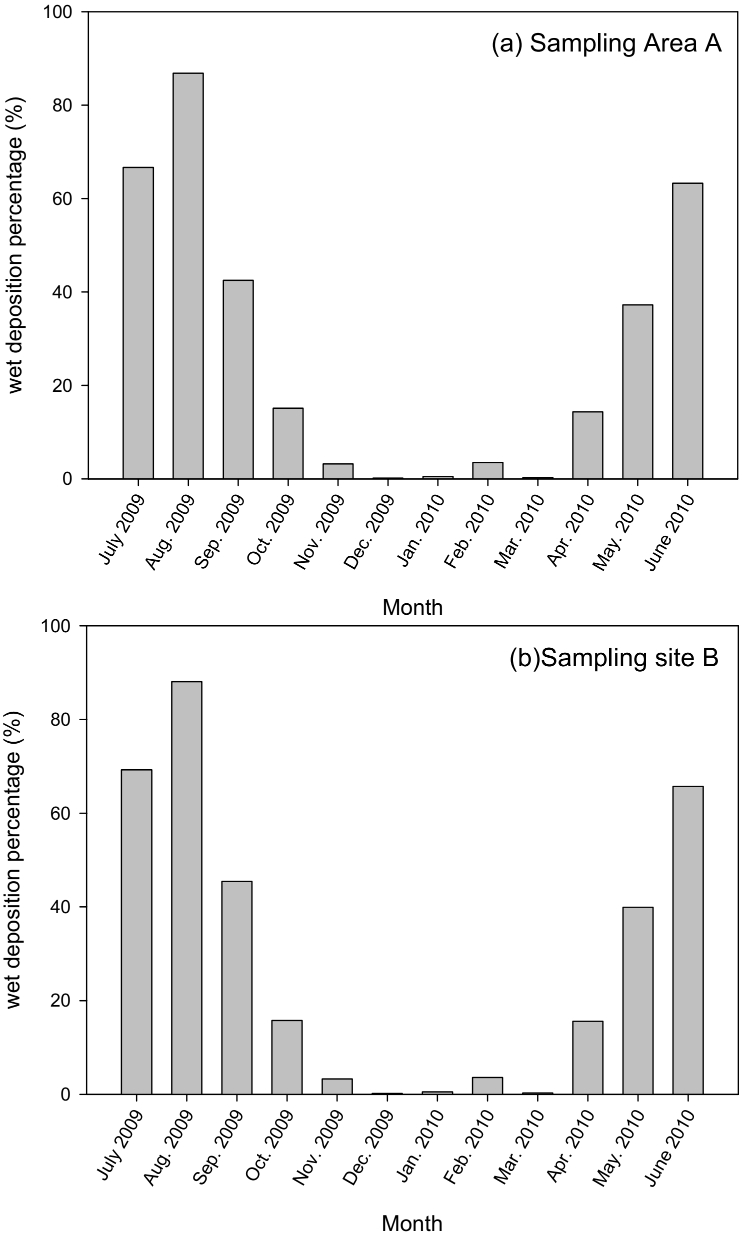

Fig. 2. Estimated monthly fluctuations of wet deposition flux percentage of total PCDD/Fs deposition in the ambient air of sampling sites A and B (%).

CONCLUSIONS

This study investigated the annual variation of wet deposition of PCDD/F in the atmosphere near the two MSWIs in southern Taiwan. The gas-particle partitioning of PCDD/Fs in air has been shown to correlate highly with meteorological factors; temperature was inversely associated with the existence of particulate PCDD/Fs. Seasonal variation of particulate PCDD/Fs observed herein was caused mainly because of the variation of ambient temperatures among the different seasons (average of 20.6°C in winter and 28.9°C in summer). It was found that a larger fraction of PCDD/Fs is associated with particles at lower temperature, indicating semivolatile organic compounds in the atmosphere are expected be scavenged most efficiently in cold weather. Similar to the observations of dry deposition flux of PCDD/Fs, particle bond deposition contributed 90.3%–98.2% of total wet deposition flux. Precipitation also played an important role in the seasonal variation of PCDD/F deposition fluxes; fluxes in rainy seasons (from June to August) were 51–53 times as high as those in dry seasons (from December to February). Moreover, the annual total (dry + wet) deposition fluxes of PCDD/Fs were 149 ng/m2-year (5.02 ng I-TEQ/m^2-year) and 177 ng/m^2-year (5.11 ng I-TEQ/m^2-year) for sites A and B, respectively, revealing that dry deposition was more dominant than the wet deposition for the atmospheric deposition of PCDD/Fs. Since atmosphere deposition is believed to be the main transfer pathway of PCDD/ Fs into food chains, its impact on human exposure to PCDD/Fs is of great importance.

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