Characterization of Atmospheric Dry Deposition of Polychlorinated Dibenzo-p-dioxins/Dibenzofuran in a Rural Area of Taiwan

Chun-Jen Huang[1], Kang-Shin Chen[1*], Yi-Chieh Lai[2*], Lin-Chi Wang[3], Guo-Ping Chang-Chien[3]

[1] *Institute of Environmental Engineering, National Sun Yat-Sen University, 70 Lienhai Road, Kaohsiung 80424, Taiwan*
[2] *Department of Bioenvironmental Engineering, Chung Yuan Christian University, 200 Chung-Pei Road, Chung-Li 32023, Taiwan*
[3] *Department of Chemical and Materials Engineering, Cheng Shiu University, 840 Chengcing Road, Niaosong Township, Kaohsiung County 833, Taiwan*

ABSTRACT

The characteristics of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and the variation of the gas-particle partitioning of PCDD/Fs near two municipal solid waste incinerators (MSWIs) located in southern Taiwan were investigated. In order to better understand the mechanism of dry deposition, the atmospheric dry deposition flux and velocity of PCDD/Fs were calculated. It was found that the mean atmospheric PCDD/F concentrations (0.0386–0.106 pg I-TEQ/Nm^3) were comparable to those detected in the vicinity of MSWIs in Taiwan, but significantly lower than those in a highly industrialized urban area (0.15 pg I-TEQ/Nm^3) located in southern Taiwan. The relatively higher atmospheric PCDD/F concentrations was found in winter than in summer, probably because of several loss process including photolysis, chemical reactivity, wet and dry deposition, and scavenging by vegetation. The calculated total dry deposition flux of PCDD/Fs ranged from 0.0274–0.718 ng I-TEQ/m^2-month, and the atmospheric deposition flux in winter tended to be higher than those in summer. The results also indicated that dry deposition velocities of atmospheric particles for each month ranged from 0.52–0.91 cm/s (mean = 0.63 cm/s) and 0.48–0.73 cm/s (mean = 0.55 cm/s) in sites A and B, respectively, which were similar to that for the ambient air near two MSWIs in Taiwan, but slightly higher than those in urban area of Korea. In addition, the dry deposition of PCDD/Fs was mainly contributed by particle-phase at both sampling areas during the estimated period. The above results demonstrated that the dominant mechanism of dry deposition was particle phase deposition.

Keywords: Polychlorinated dibenzo-p-dioxins/Dibenzofurans (PCDD/Fs); Gas-Particle partitioning; Dry deposition; Velocity.

INTRODUCTION

Polychlorinated dibenzo-p-dioxins and dibenzofurans (also known as PCDD/Fs or dioxin) have received much public concern over the last decade due to their potential adverse health effects, such as reproductive difficulties and increased risk of cancer. Since PCDD/Fs are persistent, lipophilic, and bioaccumulative, they can be slowly reduced by photodegradation (Eitzert *et al.*, 1989). As a result, they can remain in the environment for long periods and tend to accumulate in food chains. PCDD/Fs are semi-volatile organic compounds, which are similar to PAHs, PCBs and

PBDD/Fs and exited in both gas and particle phases in the ambient air and in the stack flue gases (Lee *et al.*, 1996; Lai *et al.*, 2007; Chen *et al.*, 2011). In general, PCDD/Fs released to the atmosphere are mainly from anthropogenic activities, particularly from various forms combustions or other thermal processes involving organic matters and chlorine (Rappe, 1993; Wang *et al.*, 2003). The important sources of PCDD/Fs have been reported as the waste incinerators (Wang *et al.*, 2008; Wu *et al.*, 2009), power plants (Lin *et al.*, 2007; Lin *et al.*, 2010b; Wu *et al.*, 2010), electric arc furnaces (EAFs) (Lee *et al.*, 2004; Lee *et al.*, 2005; Chiu *et al.*, 2011), secondary aluminum smelters (ALS), crematories (Chiu *et al.*, 2011), vehicles (Chuang *et al.*, 2010), and woodchip-fuelled boilers (Chen *et al.*, 2011).

Atmosphere is a major medium for the transport of pollutants from combustion sources to terrestrial and aquatic environments (Jurado *et al.*, 2004; Fang *et al.*, 2011; Oh *et al.*, 2011; Yeh *et al.*, 2011). Lohmann *et al.* (2000) reported that PCDD/Fs in air are primarily subjected to dispersion and deposition. Dry deposition depends significantly on the gas-particle distribution of PCDD/Fs (Oh *et al.*, 2001).

---

* Corresponding author. Tel.: +886-7-5254406;
  Fax: +886-7-5254406
  E-mail address: shin@mail.nsysu.edu.tw (K.S. Chen)
* Corresponding author. Tel.: +886-3-2654902;
  Fax: +886-3-2654949
  E-mail address: yclai@cycu.edu.tw (Y.C. Lai)

Gas-phase (vapor phase) PCDD/Fs were observed to be depleted from the atmosphere due to photochemical degradation reactions (Tysklind *et al.* 1993), while particle-bound PCDD/Fs deposition accounted for most of the atmospheric flux to the ecosystem (Lohmann and Jones, 1998). Furthermore, deposition of PCDD/Fs in air can be divided into dry deposition (gaseous, particulate) and wet deposition, both deposition processes contribute significantly to the removal of atmospheric PCDD/Fs (Koester and Hites, 1992).

Several studies have been carried out in Taiwan to investigate atmospheric PCDD/F deposition from relevant sources. Shih *et al.* (2006) reported the atmospheric dry deposition flux of total PCDD/Fs in rural area averaged 150 pg/m^2/day, and the calculated dry deposition velocity of total PCDD/Fs was 0.42 cm/s. The mean dry deposition fluxes of total PCDD/Fs ranged from 9.81–29.1 pg I-TEQ/m^2-day for the four investigated sites (a commercial suburban area, an industrial area, a coastal area, and an agricultural rural area), while those of wet deposition ranged from 19.2–47.8 pg I-TEQ/L (Wang *et al.*, 2010). Results indicated that the dry deposition of PCDD/Fs is more important than the wet deposition of PCDD/Fs to the total PCDD/F distribution in the environment. In addition, significant influence of atmospheric deposition of PCDD/Fs on tap drinking water has been addressed (Lin *et al.*, 2010a). Therefore, knowledge of the characteristics of PCDD/Fs deposition is essential for observing subsequent fate of PCDD/Fs in the environment.

In order to clarify the important features, this study investigates the atmospheric PCDD/F concentration and the variation of the gas-particle partitioning of PCDD/Fs in the vicinity of two municipal solid waste incinerators (MSWIs) located in a rural area of Taiwan. The monthly dry deposition fluxes were determined by model calculations. Estimated monthly dry deposition velocities of PCDD/Fs were then calculated by using the PCDD/F concentrations and dry deposition fluxes data.

MATERIALS AND METHODS

PCDD/F Sampling

Two municipal solid waste incinerators (MSWIs) situated in a rural area of southern Taiwan were taken for the treatment of municipal solid wastes generated from the whole city with a population of 1.24 millions. The basic information for these two MSWIs was shown in Wu *et al.* (2009). 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). As a result, a total of 16 ambient air samples were collected at two sites (Sites A and B) twice separately, during July 2009 and January 2010. Mean temperature and total suspended particulate (TSP) concentration during July 2009 and January 2010 are listed in Table 1. Meteorological information and TSP concentrations during the periods from July 2009 to June 2010 are given in Table 2. All meteorological information for sampling sites was obtained from the Meteorological Bureau in Kaohsiung City.

PCDD/F concentrations in ambient air samples were collected simultaneously by using a PS-1 sampler (Graseby

Table 1. Mean temperature and total suspended particulate (TSP) concentration during the sampling periods.

Sampling site	Sampling Period		Mean Temp. (°C)	TSP (µg/m^3)
A	July	20–23, 2009	31.0	43
	January	18–20, 2010	22.3	142
B	July	27–30, 2009	31.3	34
	January	19–21, 2010	25.0	166

Table 2. Meteorological information (temperature, wind speed and sunny days) and total suspended particulate (TSP) concentrations (µg/m^3) during the periods from July 2009 to June 2010.

Sampling Period	Mean Temp. (°C)	TSP (µg/m^3) Sampling site		Mean Wind Speed (m/s) Sampling site		Sunny Days (day)
		A	B	A	B	
July, 2009	29.2	45	43	2.68	2.31	12
August, 2009	29.2	56	56	3.35	2.85	17
September, 2009	29.4	72	77	2.11	1.84	18
October, 2009	26.8	128	150	2.08	2.06	28
November, 2009	23.9	110	135	2.07	2.18	29
December, 2009	20.3	126	152	2.23	2.36	29
January, 2010	19.9	120	133	2.38	2.31	28
February, 2010	21.5	77	89	2.56	2.28	27
March, 2010	23.7	132	141	2.62	2.19	29
April, 2010	24.9	90	105	2.55	2.30	22
May, 2010	27.8	62	77	2.70	2.20	24
une, 2010	28.3	42	61	2.52	1.95	18

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 m³. 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. Prior to sampling, a known amount of surrogate standard (SS) was spiked to check the collection efficiency of the sampling train. 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. 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 method detection limits of the seventeen individual PCDD/Fs for ambient air samples ranged from 0.0001 to 0.0035 ng/Nm³. The recoveries for the seven individual PCDD/Fs compounds were 75–118% (Wang *et al.*, 2010).

Gas-particle Partitioning

Particle and gas concentrations were calculated by gas-particle partitioning multiplying total PCDD/Fs concentrations. The Eqs. (1)–(3), based research by Pankow (1991), Pankow (1994), and Pankow and Bidleman, (1992) have been used to calculate the gas-particle partitioning of semivolatile organic compounds (especially for PAHs). Since PCDD/Fs and PAHs are all semivolatile organic compounds, equations which have been successfully used to describe gas-particle partitioning of PCDD/Fs by several researchers (Lohmann and Jones, 1998; Wu *et al.*, 2009; Xu *et al.*, 2009; Lin *et al.*, 2010a; Wang *et al.*, 2010).

$$K_p = \frac{F/\mathrm{TSP}}{A} \qquad (1)$$

where K_p (m³/μg) is a temperature-dependent partitioning constant, TSP (μg/m³) is the total suspended particle concentration, and F (pg/m³) and A (pg/m³) are the associated particulate and gaseous concentrations of PCDD/Fs, respectively.

When the log K_p is regressed against the logarithm of the subcooled liquid vapor pressure log P_L^o, the partitioning constant can be calculated as follows (Yamassaki *et al.*, 1982):

$$\log K_p = m_r \log P_L^o + b_r \qquad (2)$$

where m_r is the slope and b_r is the y-intercept of the trend line.

P_L^o was then calculated as follows (Hung *et al.*, 2002):

$$\log P_L^o = \frac{-1.34\,(RI)}{T} + 1.67 \times 10^{-3}\,(RI) - \frac{1320}{T} + 8.087 \qquad (3)$$

where RI is the gas chromatographic retention indexes derived by Donelly *et al.* (1987) and Hale *et al.* (1985), and T is ambient temperature (K).

Complete datasets on the gas-particle partitioning of PCDD/Fs in Taiwan has been reported by Chao *et al.* (2004), giving values for $m_r = -1.29$ and $b_r = -7.2$ with $R^2 = 0.94$.

Atmospheric Dry Deposition of PCDD/Fs

The atmospheric dry deposition flux of PCDD/Fs is a combination of both gas- and particle- phase fluxes, which is given by

$$F_T = F_g + F_p \qquad (4)$$

$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \qquad (5)$$

where F_T is the total PCDD/F deposition flux contributed from both gas and particle phases, F_g and F_p are the PCDD/F deposition flux contributed by the gas phase and particle phase, respectively, C_T is the measured concentration of total PCDD/F in air, $V_{d,T}$ is the dry deposition velocity of total PCDD/Fs, C_g and C_p are the calculated PCDD/F concentrations in the gas phase and particle phase, respectively, and $V_{d,g}$ and $V_{d,p}$ are the dry deposition velocities of PCDD/Fs in the gas phase and particle phase, respectively.

Dry deposition velocities of total PCDD/Fs have been presented by Shih *et al.* (2006), which were 0.45, 0.52, 0.32, and 0.39 cm/s in spring, summer, autumn, and winter, respectively. Values were also adopted in this study for the calculation of the dry deposition flux of total PCDD/Fs. Dry deposition of gaseous PCDD/Fs is mainly by diffusion. Because of the lack of measured data for PCDD/Fs, a selected value (0.010 cm/s) for gaseous polycyclic aromatic hydrocarbon (PAH) dry deposition velocity, reported by Sheu *et al.* (1996) and used by Lee *et al.* (1996), is also used here for the calculation of PCDD/F dry deposition flux contributed by its gas phase.

RESULTS AND DISCUSSION

Concentrations of PCDD/Fs in the Ambient Air

Mean PCDD/F concentrations in the ambient air of two sampling sites are shown in Table 3. The mean total PCDD/F TEQ concentrations in the ambient air of site A were 0.0348 pg I-TEQ/Nm^3 (RSD = 49.3%) and 0.106 pg I-TEQ/Nm^3 (RSD = 17.3%) in July 2009 and January 2010, respectively. For site B, those in July 2009 and January 2010 were 0.0386 I-TEQ/Nm^3 (RSD = 17.1%) and 0.0791 pg I-TEQ/Nm^3 (RSD = 8.22%), respectively. Both values were much lower than the Japanese ambient air quality standard (JAQS) of 0.6 pg I-TEQ/Nm^3 for PCDD/Fs (JAQS, 1999). The above results revealed that the atmospheric concentrations were similar to the results of Shih *et al.* (2006), which indicated that total atmospheric concentrations collected from rural area in southern Taiwan were 0.027, 0.016, 0.024, 0.063 pg I-TEQ/Nm^3 in spring, summer, fall, and winter, respectively. The PCDD/F values are comparable to those detected in the vicinity of two MSWIs in southern Taiwan (Hsieh *et al.*, 2009; Wu *et al.*, 2009), but lower than those in the highly industrialized urban area also located in southern Taiwan (0.15 pg I-TEQ/Nm^3) as reported by Lee *et al.* (2004). When compared with worldwide levels, PCDD/F I-TEQ concentrations found in this study are in the low range of those analyzed in the vicinity of MSWI in Porto, Lisbon, and Madeira, Portugal (0.130 pg I-TEQ/Nm^3) (Oh *et al.*, 2006) and in Bucheon, Korea (0.22–1.16 pg I-TEQ/Nm^3) (Coutinho *et al.*, 2007). As can be seen from the atmospheric concentrations between the two seasons, the total I-TEQ concentration in winter (January 2010) was 2–3 times higher than in summer (July 2009), respectively, which has been found in previous studies (Shih *et al.*, 2006; Lee *et al.*, 2009; Lin *et al.*, 2010b). As shown in Table 1, the significantly higher TSP concentration was measured at both sites during January 2010. The PCDD/Fs bound to suspended particles would thus increase and then led to the relatively higher PCDD/F concentrations in the ambient air, particularly during winter. Additionally, it has been reported that the atmospheric PCDD/F concentrations varied with the seasons because of several loss process including photolysis, chemical reactivity, wet and dry deposition, and scavenging by vegetation (Duarte-Davidson *et al.*, 1997).

The congener profiles of the seventeen 2,3,7,8 chlorinated substituted PCDD/Fs (mean ± SD) detected in air of two sampling sites are shown in Fig. 1. The profiles were calculated according to the fraction (%) of each congener to total PCDD/F mass concentration. Similar PCDD/F congener profiles were observed in 16 ambient air samples (n = 16). OCDD was the dominant congener, followed by

Table 3. Mean PCDD/F concentrations in the ambient air of sampling sites A and B, respectively.

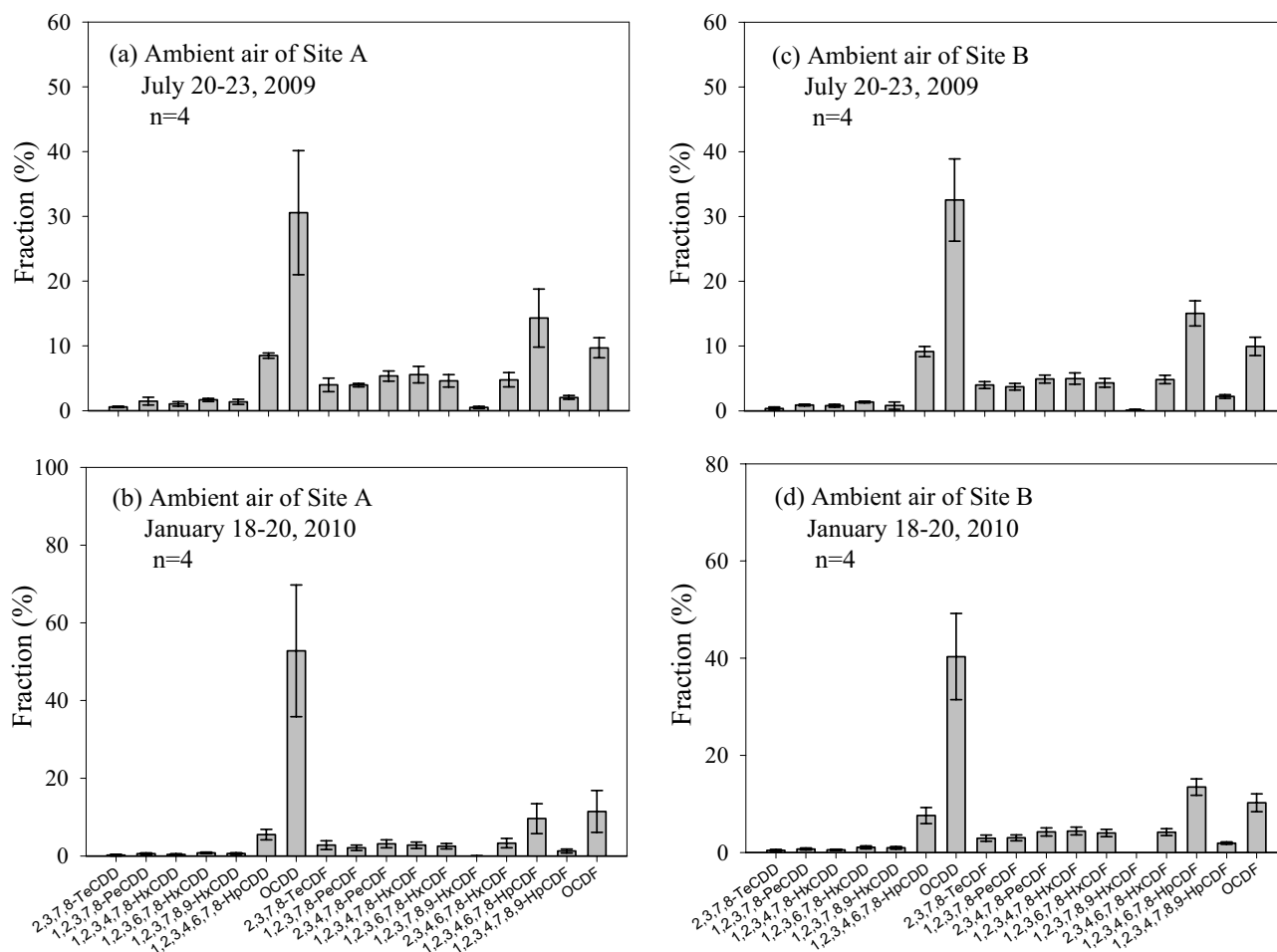| PCDD/Fs	A				B			
	July 20–23, 2009		January 18–20, 2010		July 27–30, 2009		January 19–21, 2010	
	Mean (pg/Nm^3) n = 4	RSD (%)	Mean (pg/Nm^3) n = 4	RSD (%)	Mean (pg/Nm^3) n = 4	RSD (%)	Mean (pg/Nm^3) n = 4	RSD (%)
2,3,7,8-TeCDD	0.00286	31.5	0.00478	10.4	0.00274	10.0	0.00380	10.0
1,2,3,7,8-PeCDD	0.00583	44.4	0.0147	29.3	0.00546	19.5	0.00995	3.28
1,2,3,4,7,8-HxCDD	0.00378	41.0	0.0122	20.0	0.00385	25.1	0.00866	2.52
1,2,3,6,7,8-HxCDD	0.00657	41.4	0.0241	19.7	0.00710	26.2	0.0173	3.92
1,2,3,7,8,9-HxCDD	0.00441	37.3	0.0192	19.6	0.00507	23.1	0.0146	3.14
1,2,3,4,6,7,8-HpCDD	0.0302	35.2	0.146	22.0	0.0505	19.4	0.134	19.9
OCDD	0.116	48.0	0.350	15.7	0.307	64.5	0.341	24.2
2,3,7,8-TeCDF	0.0220	49.2	0.0567	10.6	0.0271	8.34	0.0429	9.48
1,2,3,7,8-PeCDF	0.0240	38.1	0.0622	11.4	0.0275	15.8	0.0457	9.70
2,3,4,7,8-PeCDF	0.0306	60.7	0.0908	17.8	0.0339	18.2	0.0651	13.8
1,2,3,4,7,8-HxCDF	0.0305	37.5	0.0909	16.4	0.0300	19.4	0.0699	8.69
1,2,3,6,7,8-HxCDF	0.0235	45.5	0.0874	14.2	0.0278	21.4	0.0652	8.74
1,2,3,7,8,9-HxCDF	0.00218	54.0	0.0149	35.2	0.00183	18.6	0.0148	5.63
2,3,4,6,7,8-HxCDF	0.0214	60.4	0.0929	19.5	0.0283	29.4	0.0751	13.2
1,2,3,4,6,7,8-HpCDF	0.0596	50.6	0.262	23.2	0.0725	27.8	0.238	7.22
1,2,3,4,7,8,9-HpCDF	0.00758	34.7	0.0500	14.1	0.00878	20.1	0.0426	13.5
OCDF	0.0393	76.5	0.166	18.8	0.0475	18.4	0.181	9.08
PCDDs	0.169	43.9	0.571	17.9	0.382	49.0	0.528	20.0
PCDFs	0.261	44.3	0.974	17.7	0.305	18.0	0.840	6.07
PCDDs/PCDFs ratio	0.649	18.4	0.587	2.59	1.35	67.0	0.635	24.9
Total PCDD/Fs	0.430	43.1	1.55	17.7	0.687	21.6	1.37	5.09
PCDDs (pg I-TEQ/Nm^3)	0.00766	37.7	0.0195	20.0	0.00788	12.7	0.0145	4.81
PCDFs (pg I-TEQ/Nm^3)	0.0272	52.9	0.0861	16.9	0.0307	18.1	0.0646	10.9
PCDDs/PCDFs ratio	0.297	15.4	0.225	4.68	0.259	7.22	0.227	15.7
Total PCDD/Fs TEQ (pg I-TEQ/Nm^3)	0.0348	49.3	0.106	17.3	0.0386	17.1	0.0791	8.22

Fig. 1. Congener Profiles of PCDD/Fs in ambient air of two sampling sites.

1,2,3,4,6,7,8-HpCDF, OCDF, and 1,2,3,4,6,7,8-HpCDD, which are consistent with those found in other studies (Shih *et al.*, 2006; Wang *et al.*, 2008; Hsieh *et al.*, 2009).

Gas-particle Partitioning of PCDD/Fs

The total TSP concentrations were found to vary in the range of 43 to 166 µg/m³ during the sampling periods at sites A and B (Table 1) and their corresponding PM$_{10}$ concentrations were calculated according to a factor TSP: PM$_{10}$ = 1.24:1 (Sheu *et al.*, 1996). The relationship between PCDD/F concentration and PM$_{10}$ value during the sampling periods were estimated in the regression analysis as presented in Fig. 3. It was demonstrated that the PCDD/F concentrations were strongly related to PM$_{10}$ values, and the correlation coefficient was as high as 0.9438. Based on the regression model and environmental conditions stated above (Table 2), the subcooled liquid vapour pressure (P_L^o) and gas-particle partitioning constant (K_p) for individual PCDD/F congeners in the ambient air can be calculated and then gas-particle partitioning can be determined. Tables 4 and 5 list the monthly fluctuations of gas-particle partitioning of total PCDD/Fs in the ambient air of sampling sites A and B, respectively. The mean particulate fractions of TCDD/F, PCDD/F, HCDD/F, and OCDD/F in the ambient air of site A ranged from 1.2–13.5%, 3.9–46.9%, 17.6–96.5%, and

88.9–99.4%, respectively. Those of site B ranged from 1.5–18.6%, 5.6–56.3%, 23.7–97.4%, and 91.9–99.6%, respectively. Results from both sampling sites show that atmospheric PCDD/Fs tended to be distributed between the gaseous and particulate phases based on molecular weight; the higher chlorinated congener occupied a higher particulate fraction. Moreover, PCDD homologues tended to be more associated with particles than the equivalent PCDFs, probably due to the slightly lower vapour pressures of PCDDs (Rordorf, 1989). The above findings are comparable with those reported earlier (Lohmann *et al.*, 1998; Chao *et al.*, 2004; Wu *et al.*, 2009; Xu *et al.*, 2009; Lin *et al.*, 2010a).

Vapor pressure, a property strongly related to temperature, has been reported as the main factor influencing partition of semivolatile organic compounds (e.g. PAHs) (Pankow, 1987). Due to the variation of ambient temperatures during July 2009 and January 2010, from 19.9°C–21.5°C in winter and 29.2°C–29.4°C in summer, the PCDD/Fs bound to particles was found to increase with decreasing temperature. As a result, the relatively higher PCDD/Fs in the particle phase during winter were observed. Additionally, results show that the total PCDD/Fs is dominated by the particle-phase, while total I-TEQ is dominated by its gas phase.

Table 4. Estimated monthly fluctuations of gas-particle partitioning of total PCDD/Fs in the ambient air of sampling site A.

PCDD/Fs	Jul. 2009		Aug. 2009		Sep. 2009		Oct. 2009		Nov. 2009		Dec. 2009		Jan. 2010		Feb. 2010		Mar. 2010		Apr. 2010		May. 2010		Jun. 2010	
	P[a] (%)	G[b] (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)
2,3,7,8-TeCDD	1.95	98.05	1.73	98.27	2.18	97.82	5.47	94.53	7.14	92.86	13.27	86.73	13.5	86.5	7.18	92.82	8.68	91.32	5.14	94.86	2.36	97.64	1.48	98.52
1,2,3,7,8-PeCDD	9.52	90.48	8.39	91.61	10.36	89.64	23.51	76.49	29.54	70.46	46.31	53.69	46.89	53.11	30.14	69.86	34.18	65.82	22.64	77.36	11.28	88.72	7.31	92.69
1,2,3,4,7,8-HxCDD	34.49	65.51	31.04	68.96	36.17	63.83	60.65	39.35	68.3	31.7	82.07	17.93	82.46	17.54	69.37	30.63	72.78	27.22	59.86	40.14	38.73	61.27	28.08	71.92
1,2,3,6,7,8-HxCDD	35.81	64.19	32.28	67.72	37.5	62.5	62.03	37.97	69.57	30.43	82.94	17.06	83.32	16.68	70.63	29.37	73.94	26.06	61.27	38.73	40.11	59.89	29.26	70.74
1,2,3,7,8,9-HxCDD	38.53	61.47	34.84	65.16	40.23	59.77	64.72	35.28	72.01	27.99	84.57	15.43	84.93	15.07	73.05	26.95	76.15	23.85	64.02	35.98	42.92	57.08	31.71	68.29
1,2,3,4,6,7,8-HpCDD	75.52	24.48	72.09	27.91	76.45	23.55	90.05	9.95	92.86	7.14	96.62	3.38	96.72	3.28	93.33	6.67	94.17	5.83	89.91	10.09	78.61	21.39	69.33	30.67
OCDD	94.28	5.72	93.13	6.87	94.44	5.56	97.98	2.02	98.62	1.38	99.39	0.61	99.41	0.59	98.74	1.26	98.89	1.11	97.98	2.02	95.13	4.87	92.28	7.72
2,3,7,8-TeCDF	1.31	98.69	1.17	98.83	1.48	98.52	3.74	96.26	4.88	95.12	9.19	90.81	9.35	90.65	4.88	95.12	5.96	94.04	3.49	96.51	1.6	98.4	1	99
1,2,3,7,8-PeCDF	5.14	94.86	4.54	95.46	5.66	94.34	13.65	86.35	17.59	82.41	30.23	69.77	30.69	69.31	17.88	82.12	20.9	79.1	13.01	86.99	6.16	93.84	3.92	96.08
2,3,4,7,8-PeCDF	6.91	93.09	6.1	93.9	7.57	92.43	17.82	82.18	22.72	77.28	37.53	62.47	38.06	61.94	23.15	76.85	26.69	73.31	17.06	82.94	8.25	91.75	5.29	94.71
1,2,3,4,7,8-HxCDF	22.31	77.69	19.82	80.18	23.75	76.25	45.66	54.34	53.78	46.22	70.95	29.05	71.47	28.53	54.82	45.18	59.07	40.93	44.7	55.3	25.69	74.31	17.62	82.38
1,2,3,6,7,8-HxCDF	23.19	76.81	20.62	79.38	24.65	75.35	46.9	53.1	55.04	44.96	72	28	72.52	27.48	56.09	43.91	60.29	39.71	45.94	54.06	26.65	73.35	18.35	81.65
1,2,3,7,8,9-HxCDF	32.82	67.18	29.48	70.52	34.48	65.52	58.85	41.15	66.63	33.37	80.9	19.1	81.31	18.69	67.71	32.29	71.25	28.75	58.04	41.96	36.98	63.02	26.61	73.39
2,3,4,6,7,8-HxCDF	28.59	71.41	25.56	74.44	30.18	69.82	53.95	46.05	61.99	38.01	77.51	22.49	77.96	22.04	63.08	36.92	66.92	33.08	53.07	46.93	32.49	67.51	22.93	77.07
1,2,3,4,6,7,8-HpCDF	58.17	41.83	54.03	45.97	59.65	40.35	80.29	19.71	85.26	14.74	92.6	7.4	92.8	7.2	86.03	13.97	87.78	12.22	79.93	20.07	62.44	37.56	50.6	49.4
1,2,3,4,7,8,9-HpCDF	74.27	25.73	70.75	29.25	75.25	24.75	89.43	10.57	92.4	7.6	96.39	3.61	96.5	3.5	92.89	7.11	93.79	6.21	89.29	10.71	77.48	22.52	67.91	32.09
OCDF	91.65	8.35	90.06	9.94	91.92	8.08	96.99	3.01	97.93	2.07	99.07	0.93	99.1	0.9	98.1	1.9	98.33	1.67	96.99	3.01	92.87	7.13	88.86	11.14

[a] P: Particle phase. [b] G: Gas phase.

Table 5. Estimated monthly fluctuations of gas-particle partitioning of total PCDD/Fs in the ambient air of sampling site B.

PCDD/Fs	Jul. 2009		Aug. 2009		Sep. 2009		Oct. 2009		Nov. 2009		Dec. 2009		Jan. 2010		Feb. 2010		Mar. 2010		Apr. 2010		May. 2010		Jun. 2010	
	P[a] (%)	G[b] (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)	P (%)	G (%)
2,3,7,8-TeCDD	2.28	97.72	2.17	97.83	2.84	97.16	7.75	92.25	10.51	89.49	18.62	81.38	17.63	82.37	10.04	89.96	11.21	88.79	7.28	92.72	3.57	96.43	2.67	97.33
1,2,3,7,8-PeCDD	11.02	88.98	10.34	89.66	13.19	86.81	30.86	69.14	39.04	60.96	56.32	43.68	54.77	45.23	38.34	61.66	40.82	59.18	29.78	70.22	16.31	83.69	12.57	87.43
1,2,3,4,7,8-HxCDD	38.27	61.73	36.15	63.85	42.69	57.31	69.11	30.89	76.7	23.3	87.25	12.75	86.58	13.42	76.55	23.45	78.03	21.97	68.37	31.63	49.21	50.79	41.58	58.42
1,2,3,6,7,8-HxCDD	39.65	60.35	37.48	62.52	44.1	55.9	70.34	29.66	77.74	22.26	87.91	12.09	87.26	12.74	77.61	22.39	79.03	20.97	69.63	30.37	50.66	49.34	42.98	57.02
1,2,3,7,8,9-HxCDD	42.46	57.54	40.21	59.79	46.94	53.06	72.71	27.29	79.72	20.28	89.13	10.87	88.54	11.46	79.62	20.38	80.92	19.08	72.05	27.95	53.54	46.46	45.83	54.17
1,2,3,4,6,7,8-HpCDD	78.41	21.59	76.47	23.53	81.01	18.99	92.93	7.07	95.21	4.79	97.72	2.28	97.59	2.41	95.27	4.73	95.55	4.45	92.82	7.18	84.93	15.07	80.46	19.54
OCDD	95.1	4.9	94.46	5.54	95.72	4.28	98.6	1.4	99.09	0.91	99.59	0.41	99.57	0.43	99.13	0.87	99.16	0.84	98.6	1.4	96.77	3.23	95.61	4.39
2,3,7,8-TeCDF	1.54	98.46	1.47	98.53	1.94	98.06	5.34	94.66	7.26	92.74	13.15	86.85	12.4	87.6	6.89	93.11	7.76	92.24	4.98	95.02	2.43	97.57	1.81	98.19
1,2,3,7,8-PeCDF	5.99	94.01	5.64	94.36	7.31	92.69	18.67	81.33	24.59	75.41	39.32	60.68	37.79	62.21	23.89	76.11	25.97	74.03	17.81	82.19	9.14	90.86	6.92	93.08
2,3,4,7,8-PeCDF	8.04	91.96	7.55	92.45	9.72	90.28	23.95	76.05	31	69	47.33	52.67	45.74	54.26	30.28	69.72	32.6	67.4	22.97	77.03	12.11	87.89	9.23	90.77
1,2,3,4,7,8-HxCDF	25.27	74.73	23.72	76.28	29.05	70.95	54.96	45.04	64	36	78.5	21.5	77.46	22.54	63.63	36.37	65.72	34.28	53.94	46.06	34.64	65.36	28.05	71.95
1,2,3,6,7,8-HxCDF	26.22	73.78	24.63	75.37	30.08	69.92	56.19	43.81	65.16	34.84	79.36	20.64	78.35	21.65	64.8	35.2	66.85	33.15	55.19	44.81	35.77	64.23	29.06	70.94
1,2,3,7,8,9-HxCDF	36.52	63.48	34.46	65.54	40.89	59.11	67.5	32.5	75.31	24.69	86.36	13.64	85.65	14.35	75.14	24.86	76.7	23.3	66.71	33.29	47.35	52.65	39.79	60.21
2,3,4,6,7,8-HxCDF	32.03	67.97	30.16	69.84	36.24	63.76	62.98	37.02	71.36	28.64	83.75	16.25	82.91	17.09	71.12	28.88	72.88	27.12	62.1	37.9	42.45	57.55	35.15	64.85
1,2,3,4,6,7,8-HpCDF	62.08	37.92	59.65	40.35	66.02	33.98	85.54	14.46	89.83	10.17	94.93	5.07	94.65	5.35	89.87	10.13	90.51	9.49	85.23	14.77	71.82	28.18	65.12	34.88
1,2,3,4,7,8,9-HpCDF	77.26	22.74	75.27	24.73	79.99	20.01	92.48	7.52	94.89	5.11	97.56	2.44	97.42	2.58	94.96	5.04	95.25	4.75	92.35	7.65	84.06	15.94	79.41	20.59
OCDF	92.82	7.18	91.94	8.06	93.73	6.27	97.91	2.09	98.63	1.37	99.38	0.62	99.34	0.66	98.68	1.32	98.74	1.26	97.9	2.1	95.23	4.77	93.56	6.44

[a] P: Particle phase; [b] G: Gas phase.

Dry Deposition Flux of PCDD/Fs

Based on the Eqs. (4) and (5), the dry deposition flux of PCDD/Fs was calculated. In this study, the term C_T was calculated according to the regression model shown in Fig. 2; C_g and C_p were determined based on the gas-particle partitioning shown in Tables 4 and 5; $V_{d,T}$ and $V_{d,g}$ were assumed; and then the unknown $V_{d,p}$ can be determined. The estimated monthly fluctuations of dry deposition fluxes of PCDD/Fs in the ambient air of sampling sites A and B were listed in Tables 6 and 7, respectively. Atmospheric dry deposition fluxes of total PCDD/Fs in site A ranged from 0.186–0.718, 0.0406–0.0653, 0.118–0.431, and 0.323–0.698 ng I-TEQ/m^2-month, with an average of 0.416, 0.0518, 0.325, and 0.558 ng I-TEQ/m^2-month in spring, summer, fall, and winter, respectively. Those in site B ranged ranged from 0.181–0.708, 0.0274–0.0607, 0.0925–0.506, and 0.355–0.769 ng I-TEQ/m^2-month, with an average of 0.400, 0.0455, 0.360, and 0.593 ng I-TEQ/m^2-month in spring, summer, fall, and winter, respectively. The above findings are similar to Wu's research, which reported the mean dry deposition fluxes of total PCDD/Fs were 18.0 and 23.5 pg I-TEQ/m^2-day in the ambient air near MSWI-GS and MSWI-RW located in southern Taiwan (Wu *et al.*, 2009).

Results revealed that the total dry deposition flux for both sampling sites reached the highest level in winter and the lowest level in summer, the total dry deposition flux was found to decrease as the temperature increased (Fig. 3). The observed findings are similar to those reported by Shih *et al.* (2006). Authors indicated that temperature influences the amount of PCDD/Fs that are bound to particles and subsequently dry deposit. Results also shown that approximately 90% of dry deposition fluxes PCDD/Fs were contributed by particle-phase deposition in both sampling sites. It was demonstrated that the dry deposition of PCDD/Fs was primarily contributed by the particle phase. This is probably because of the significantly higher deposition velocity of particle-phase PCDD/Fs (0.32–0.52 cm/s) than that of gas-phase velocity (0.010 cm/s).

Additionally, higher chlorinated congeners were dominant in the deposition flux for all seasons. The dry deposition flux was most dominated by OCDD, followed by OCDF, 1,2,3,4,6,7,8-HpCDF, and 1,2,3,4,6,7,8-HpCDD. This pattern is very similar to congener profiles of ambient concentrations in this study and same trends have been mentioned in previous studies (Shih *et al.*, 2006; Wu *et al.*, 2009).

Dry Deposition Velocity of PCDD/Fs

Deposition velocity is a function of various parameters related to gas-particle partitioning of PCDD/Fs in ambient air, particle size distribution, atmospheric conditions, surface roughness, and may vary seasonally (Chi *et al.*, 2009). In order to better understand the dry deposition process, the dry deposition velocities of individual PCDD/Fs were calculated and the monthly fluctuations of dry deposition velocities of total PCDD/Fs in the ambient air of sites A and B were hown in Table 8. Dry deposition velocities of total PCDD/Fs which were assumed to be about 0.45, 0.52, 0.32, and 0.39 cm/s in spring, summer, autumn, and winter, respectively (Shih *et al.*, 2006). In this study, the estimated deposition velocities of atmospheric particles ($V_{d,p}$) for each month ranged from 0.52–0.91 cm/s (mean = 0.63 cm/s) and 0.48–0.73 cm/s (mean = 0.55 cm/s) in sites A and B, respectively. The highest and lowest values of atmospheric particles were observed in May 2010 and November 2009, respectively. The deposition velocities in particle phase were similar to that for the ambient air near two MSWIs (0.44–0.68 cm/s) (Wu *et al.*, 2009), but slightly higher than those in urban site of Korea (0.49 cm/s) as reported by Moon *et al.* (2005). The differences among these deposition velocities can be attributed to the discrepancy in sampling site and particle size distribution.

CONCLUSIONS

The mean atmospheric PCDD/F concentrations in this investigation were comparable to those detected in the
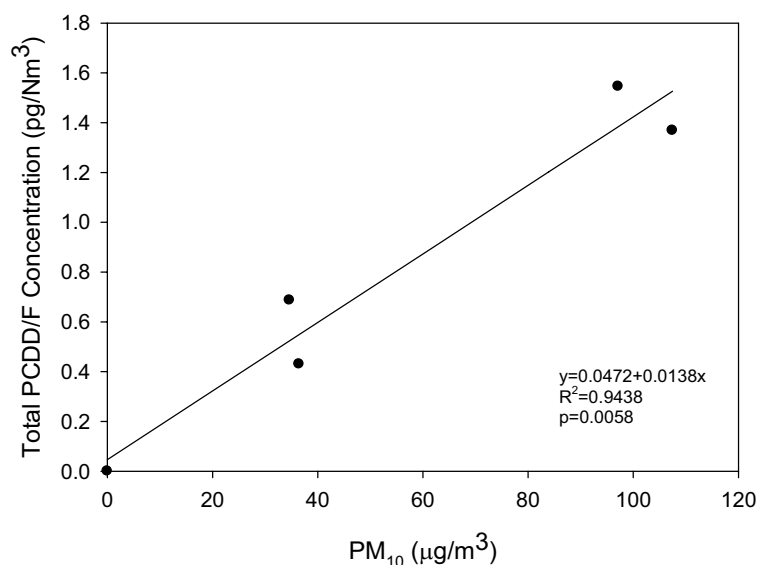

y=0.0472+0.0138x
R^2=0.9438
p=0.0058

Fig. 2. Regression of PM$_{10}$ and total PCDD/F concentration during the sampling periods.

Table 6. Estimated monthly fluctuations of dry deposition fluxes of PCDD/Fs in the ambient air of sampling site A (ng/m²-month).

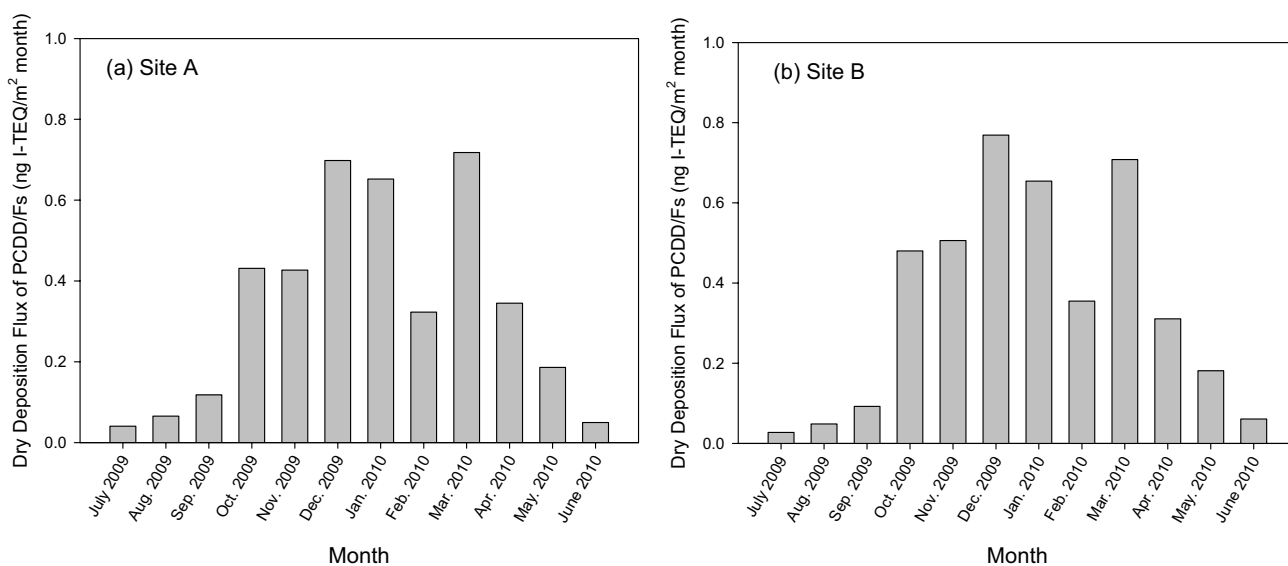| PCDD/Fs	Jul. 2009		Aug. 2009		Sep. 2009		Oct. 2009		Nov. 2009		Dec. 2009		Jan. 2010		Feb. 2010		Mar. 2010		Apr. 2010		May. 2010		Jun. 2010	
	$F_{d,T}$[a]	P %[b]	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %	$F_{d,T}$	P %
2,3,7,8-TeCDD	0.000865	53.53	0.00142	51.41	0.00238	60.873	0.00441	76.018	0.00463	79.965	0.00928	89.38	0.00872	89.564	0.00820	82.035	0.00800	86.392	0.00704	80.682	0.00343	68.805	0.00109	48.30
1,2,3,7,8-PeCDD	0.00500	85.93	0.0079	84.63	0.0147	88.99	0.0457	94.43	0.0479	95.62	0.0883	97.94	0.0828	97.98	0.0363	96.23	0.0838	97.21	0.0496	95.75	0.0233	92.04	0.00591	83.08
1,2,3,4,7,8-HxCDD	0.0106	96.82	0.0169	96.43	0.0308	97.54	0.0949	98.83	0.0900	99.12	0.130	99.61	0.121	99.61	0.0683	99.26	0.147	99.44	0.0831	99.14	0.0491	98.29	0.0129	96.04
1,2,3,6,7,8-HxCDD	0.0190	97.00	0.0308	96.62	0.0553	97.67	0.191	98.90	0.181	99.16	0.258	99.63	0.240	99.64	0.137	99.30	0.294	99.48	0.147	99.18	0.0881	98.39	0.0232	96.25
1,2,3,7,8,9-HxCDD	0.0138	97.33	0.0221	96.98	0.0400	97.92	0.159	99.02	0.149	99.26	0.209	99.67	0.195	99.68	0.113	99.38	0.241	99.54	0.104	99.27	0.0636	98.56	0.0170	96.65
1,2,3,4,6,7,8-HpCDD	0.182	99.45	0.309	99.36	0.516	99.56	1.67	99.80	1.45	99.85	1.82	99.94	1.68	99.94	1.09	99.88	2.26	99.91	1.00	99.85	0.795	99.70	0.249	99.29
OCDD	0.821	99.90	1.44	99.88	2.30	99.92	4.36	99.96	3.71	99.97	4.50	99.99	4.17	99.99	2.78	99.98	5.71	99.98	3.96	99.97	3.48	99.94	1.19	99.86
2,3,7,8-TeCDF	0.00518	43.55	0.00860	41.63	0.0139	51.26	0.0398	68.09	0.0411	72.75	0.0800	84.75	0.0752	84.97	0.0298	75.19	0.0693	80.95	0.0379	73.61	0.0194	59.55	0.00664	38.55
1,2,3,7,8-PeCDF	0.0129	75.88	0.0206	74.09	0.0373	80.76	0.121	89.63	0.129	91.76	0.255	95.95	0.239	96.06	0.0968	92.78	0.228	94.63	0.125	92.03	0.0574	85.64	0.0154	71.75
2,3,4,7,8-PeCDF	0.0198	81.19	0.0316	79.53	0.0580	85.16	0.223	92.26	0.236	93.84	0.453	97.07	0.426	97.13	0.178	94.68	0.416	96.05	0.196	94.07	0.0905	89.10	0.0236	77.61
1,2,3,4,7,8-HxCDF	0.0578	94.34	0.0917	93.69	0.170	95.62	0.538	97.88	0.533	98.38	0.839	99.26	0.783	99.28	0.406	98.62	0.894	98.98	0.515	98.42	0.272	96.92	0.0689	92.98
1,2,3,6,7,8-HxCDF	0.0453	94.59	0.0718	93.97	0.133	95.83	0.533	97.99	0.526	98.45	0.821	99.30	0.766	99.32	0.400	98.69	0.879	99.03	0.400	98.50	0.213	97.06	0.0540	93.32
1,2,3,7,8,9-HxCDF	0.00578	96.60	0.00925	96.16	0.0169	97.35	0.112	98.75	0.107	99.05	0.156	99.57	0.145	99.58	0.0813	99.20	0.175	99.40	0.0463	99.07	0.0269	98.17	0.00703	95.75
2,3,4,6,7,8-HxCDF	0.0488	95.86	0.0778	95.37	0.143	96.79	0.645	98.47	0.624	98.83	0.933	99.47	0.869	99.49	0.474	99.02	1.03	99.27	0.408	98.86	0.229	97.77	0.0588	94.87
1,2,3,4,6,7,8-HpCDF	0.270	98.78	0.446	98.60	0.775	99.04	2.67	99.55	2.39	99.67	3.12	99.86	2.90	99.86	1.80	99.73	3.78	99.79	1.71	99.67	1.21	99.35	0.351	98.45
1,2,3,4,7,8,9-HpCDF	0.0451	99.41	0.0762	99.32	0.128	99.53	0.571	99.79	0.498	99.84	0.625	99.93	0.580	99.93	0.375	99.87	0.777	99.90	0.250	99.84	0.197	99.68	0.0613	99.24
OCDF	0.266	99.84	0.464	99.82	0.748	99.87	2.04	99.94	1.74	99.96	2.12	99.98	1.96	99.98	1.30	99.97	2.68	99.97	1.31	99.96	1.13	99.92	0.384	99.80
PCDDs	1.05	99.60	1.82	99.56	2.96	99.67	6.52	99.79	5.63	99.83	7.01	99.91	6.50	99.91	4.23	99.86	8.75	99.89	5.35	99.84	4.50	99.77	1.50	99.54
PCDFs	0.777	97.21	1.30	97.04	2.22	97.77	7.49	98.80	6.82	98.96	9.40	99.38	8.74	99.39	5.15	99.11	10.9	99.29	5.00	98.85	3.45	98.39	1.03	96.87
PCDDs/PCDFs ratio	1.35	87.64	1.41	87.43	1.33	87.52	0.871	85.60	0.826	86.05	0.746	86.91	0.744	86.98	0.822	86.25	0.801	86.19	1.07	87.73	1.30	87.59	1.46	87.51
Total PCDD/Fs	*1.83*	*98.58*	*3.12*	*98.51*	*5.19*	*98.86*	*14.0*	*99.26*	*12.5*	*99.36*	*16.4*	*99.60*	*15.2*	*99.61*	*9.37*	*99.45*	*19.7*	*99.56*	*10.3*	*99.36*	*7.95*	*99.17*	*2.53*	*98.45*
PCDDs (I-TEQ)	0.0103	91.37	0.0169	90.80	0.0298	93.13	0.0927	96.94	0.0887	97.36	0.136	98.44	0.127	98.46	0.0670	97.72	0.146	98.21	0.0793	96.60	0.0466	94.97	0.0130	90.21
PCDFs (I-TEQ)	0.0303	89.62	0.0484	88.71	0.0883	91.81	0.339	95.87	0.338	96.60	0.562	98.19	0.525	98.22	0.256	97.08	0.572	97.76	0.266	96.51	0.139	94.09	0.0366	87.70
PCDDs/PCDFs I-TEQ ratio	0.342	55.06	0.348	55.63	0.338	54.63	0.274	57.71	0.263	56.50	0.242	53.66	0.241	53.66	0.262	56.41	0.256	55.63	0.298	50.68	0.334	54.27	0.356	56.31
Total PCDD/Fs (I-TEQ)	*0.0406*	*90.06*	*0.0653*	*89.24*	*0.118*	*92.15*	*0.431*	*96.10*	*0.427*	*96.77*	*0.698*	*98.24*	*0.652*	*98.27*	*0.323*	*97.20*	*0.718*	*97.84*	*0.345*	*96.52*	*0.186*	*94.29*	*0.0496*	*88.34*

[a] $F_{d,T}$: Total deposition flux of PCDD/Fs; [b] P %: The ratio of dry deposition flux contributed by the particle-phase of PCDD/Fs.

Table 7. Estimated monthly fluctuations of dry deposition fluxes of PCDD/Fs in the ambient air of sampling site B (ng/m²-month).

PCDD/Fs	Jul. 2009 $F_{d,T}$[a]	P %[b]	Aug. 2009 $F_{d,T}$	P %	Sep. 2009 $F_{d,T}$	P %	Oct. 2009 $F_{d,T}$	P %	Nov. 2009 $F_{d,T}$	P %	Dec. 2009 $F_{d,T}$	P %	Jan. 2010 $F_{d,T}$	P %	Feb. 2010 $F_{d,T}$	P %	Mar. 2010 $F_{d,T}$	P %	Apr. 2010 $F_{d,T}$	P %	May. 2010 $F_{d,T}$	P %	Jun. 2010 $F_{d,T}$	P %
2,3,7,8-TeCDD	0.000466	52.85	0.000835	52.06	0.00150	62.51	0.00554	80.77	0.00643	84.93	0.0125	92.11	0.0103	91.76	0.00432	85.99	0.00896	88.83	0.00549	83.82	0.00275	72.99	0.00104	56.31
1,2,3,7,8-PeCDD	0.00282	85.60	0.00495	84.96	0.00983	89.65	0.0487	95.71	0.0548	96.85	0.0922	98.50	0.0779	98.44	0.0382	97.16	0.0776	97.75	0.0395	96.55	0.0199	93.43	0.00643	87.11
1,2,3,4,7,8-HxCDD	0.00609	96.75	0.0107	96.52	0.0206	97.70	0.0918	99.11	0.0915	99.37	0.123	99.71	0.106	99.70	0.0650	99.45	0.127	99.56	0.0621	99.30	0.0401	98.61	0.0134	97.10
1,2,3,6,7,8-HxCDD	0.0116	96.93	0.0204	96.71	0.0390	97.82	0.186	99.16	0.184	99.41	0.246	99.73	0.212	99.72	0.131	99.48	0.256	99.58	0.116	99.34	0.0759	98.68	0.0255	97.26
1,2,3,7,8,9-HxCDD	0.00885	97.25	0.0156	97.05	0.0296	98.06	0.162	99.25	0.160	99.47	0.211	99.76	0.182	99.75	0.114	99.54	0.221	99.63	0.0860	99.42	0.0573	98.83	0.0194	97.55
1,2,3,4,6,7,8-HpCDD	0.159	99.43	0.288	99.38	0.500	99.59	1.88	99.85	1.73	99.90	2.10	99.95	1.82	99.95	1.23	99.91	2.37	99.93	1.09	99.88	0.893	99.76	0.331	99.49
OCDD	1.05	99.89	1.94	99.88	3.23	99.92	5.06	99.97	4.58	99.98	5.46	99.99	4.74	99.99	3.26	99.98	6.26	99.99	6.37	99.98	5.57	99.95	2.15	99.90
2,3,7,8-TeCDF	0.00386	42.94	0.00696	42.27	0.0120	52.95	0.0472	73.82	0.0540	78.99	0.104	88.53	0.0852	88.04	0.0360	80.28	0.0741	84.13	0.0404	77.59	0.0211	64.51	0.00853	46.46
1,2,3,7,8-PeCDF	0.00876	75.37	0.0155	74.55	0.0301	81.80	0.141	91.99	0.164	93.99	0.301	97.06	0.252	96.93	0.113	94.52	0.233	95.67	0.123	93.47	0.0597	88.02	0.0200	77.75
2,3,4,7,8-PeCDF	0.0135	80.75	0.0238	80.01	0.0468	85.99	0.253	94.03	0.290	95.57	0.512	97.86	0.431	97.77	0.201	95.98	0.411	96.82	0.191	95.16	0.0940	90.96	0.0308	82.70
1,2,3,4,7,8-HxCDF	0.0322	94.20	0.0564	93.84	0.111	95.89	0.594	98.39	0.620	98.84	0.896	99.47	0.768	99.44	0.439	98.97	0.868	99.18	0.384	98.72	0.222	97.48	0.0722	94.82
1,2,3,6,7,8-HxCDF	0.0310	94.46	0.0543	94.12	0.107	96.08	0.566	98.46	0.589	98.90	0.846	99.49	0.725	99.47	0.417	99.02	0.823	99.22	0.365	98.78	0.213	97.60	0.0695	95.06
1,2,3,7,8,9-HxCDF	0.00268	96.50	0.00471	96.26	0.00906	97.53	0.153	99.05	0.154	99.32	0.208	99.69	0.179	99.68	0.109	99.40	0.213	99.52	0.0278	99.25	0.0177	98.50	0.00591	96.88
2,3,4,6,7,8-HxCDF	0.0379	95.77	0.0665	95.49	0.129	97.01	0.729	98.84	0.742	99.17	1.03	99.62	0.883	99.61	0.526	99.27	1.03	99.41	0.415	99.08	0.255	98.18	0.0842	96.22
1,2,3,4,6,7,8-HpCDF	0.181	98.74	0.324	98.64	0.587	99.11	3.10	99.66	2.93	99.76	3.67	99.90	3.17	99.89	2.09	99.80	4.03	99.83	1.44	99.74	1.09	99.47	0.386	98.87
1,2,3,4,7,8,9-HpCDF	0.0271	99.39	0.0492	99.33	0.0857	99.56	0.599	99.84	0.553	99.89	0.674	99.95	0.584	99.95	0.394	99.90	0.759	99.92	0.189	99.87	0.153	99.74	0.0567	99.45
OCDF	0.172	99.84	0.316	99.82	0.530	99.88	2.69	99.96	2.44	99.97	2.91	99.99	2.53	99.99	1.74	99.98	3.33	99.98	1.06	99.97	0.919	99.93	0.353	99.85
PCDDs	1.24	99.72	2.28	99.70	3.83	99.79	7.43	99.85	6.81	99.88	8.24	99.94	7.15	99.93	4.85	99.90	9.33	99.92	7.77	99.92	6.66	99.86	2.55	99.74
PCDFs	0.510	97.07	0.918	96.96	1.65	97.77	8.87	99.10	8.53	99.26	11.1	99.55	9.61	99.54	6.06	99.35	11.8	99.46	4.24	98.96	3.04	98.50	1.09	97.24
PCDDs/PCDFs ratio	2.43	102.73	2.48	102.82	2.32	102.07	0.838	100.76	0.798	100.63	0.740	100.38	0.744	100.39	0.800	100.55	0.792	100.46	1.83	100.96	2.19	101.39	2.34	102.57
Total PCDD/Fs	*1.75*	*98.95*	*3.20*	*98.92*	*5.48*	*99.18*	*16.3*	*99.44*	*15.3*	*99.54*	*19.4*	*99.72*	*16.8*	*99.71*	*10.9*	*99.59*	*21.1*	*99.66*	*12.0*	*99.58*	*9.70*	*99.44*	*3.63*	*98.99*
PCDDs (I-TEQ)	0.00716	92.85	0.0128	92.63	0.0236	94.55	0.0976	97.44	0.0993	97.88	0.143	98.72	0.122	98.69	0.0700	98.12	0.138	98.45	0.0690	97.46	0.0445	96.31	0.0155	93.28
PCDFs (I-TEQ)	0.0202	89.21	0.0356	88.74	0.0689	92.09	0.382	96.78	0.406	97.49	0.626	98.64	0.532	98.60	0.285	97.75	0.570	98.18	0.242	96.99	0.136	94.87	0.0452	90.18
PCDDs/PCDFs I-TEQ ratio	0.355	104.08	0.360	104.38	0.342	102.68	0.256	100.68	0.244	100.41	0.229	100.08	0.230	100.09	0.245	100.37	0.242	100.27	0.285	100.49	0.327	101.51	0.344	103.44
Total PCDD/Fs (I-TEQ)	*0.0274*	*90.16*	*0.0484*	*89.77*	*0.0925*	*92.72*	*0.480*	*96.92*	*0.506*	*97.56*	*0.769*	*98.66*	*0.654*	*98.61*	*0.355*	*97.82*	*0.708*	*98.24*	*0.311*	*97.09*	*0.181*	*95.23*	*0.0607*	*90.97*

[a] $F_{d,T}$: Total deposition flux of PCDD/Fs; [b] P %: The ratio of dry deposition flux contributed by the particle-phase of PCDD/Fs.

Fig. 3. Estimated monthly fluctuations of dry deposition fluxes of total PCDD/Fs (ng I-TEQ/m²- month) in ambient air of two sampling sites.

Table 8. Monthly fluctuation of dry deposition velocity.

Month			Jul. 2009	Aug. 2009	Sep. 2009	Oct. 2009	Nov. 2009	Dec. 2009	Jan. 2010	Feb. 2010	Mar. 2010	Apr. 2010	May. 2010	Jun. 2010
$V_{d,g}$ (cm/s)			0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
$V_{d,T}$ (cm/s)			0.32	0.32	0.39	0.39	0.39	0.45	0.45	0.45	0.52	0.52	0.52	0.32
$V_{d,p}$[a]	Sampling	A	0.58	0.60	0.70	0.55	0.52	0.55	0.55	0.59	0.67	0.77	0.91	0.62
(cm/s)	Site	B	0.48	0.49	0.57	0.50	0.48	0.51	0.52	0.55	0.63	0.66	0.73	0.47

[a] $V_{d,p} = (C_T \times V_{d,T} - C_g \times V_{d,g})/C_p$, calculated by total concentration of 17 congeners.

vicinity of MSWIs in Taiwan, but significantly lower than those in the highly industrialized urban area located in southern Taiwan. The relatively higher atmospheric PCDD/F concentrations was found in winter than in summer, probably because of several loss process including photolysis, chemical reactivity, wet and dry deposition, and scavenging by vegetation. The observed total dry deposition flux for both sampling sites was found to decrease as the temperature increased. This was attributed to the fact that temperature influences the amount of PCDD/Fs that are bound to particles and subsequently dry deposit. Calculated dry deposition velocities of atmospheric particles (0.48–0.91 cm/s) were similar to that for the ambient air near two MSWIs in southern Taiwan (0.44–0.68 cm/s), but slightly higher than those in urban area of Korea (0.49 cm/s). The differences among these deposition velocities can be attributed to the discrepancy in sampling site and particle size distribution. In addition, results shown that approximately 90% of dry deposition fluxes PCDD/Fs were contributed by particle-phase deposition, therefore the dominant mechanism of dry deposition was particle phase deposition.

REFERENCES

Chao, M.R., Hu, C.W., Chen, Y.L., Chang-Chien, G.P., Lee, W.J., Chang, L.W., Lee, W.S. and Wu, K.Y. (2004). Approaching Gas-particle Partitioning Equilibrium of Atmospheric PCDD/Fs with Increasing Distance from an Incinerator: Measurements and Observations on Modeling. *Atmos. Environ.* 38: 1501–1510.

Chen, W.S., Shen, Y.H., Hsieh, T.Y., Lin, C.W., Wang. L.C. and Chang-Chien, G.P. (2011). Fate and Distribution of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in a Woodchip-fuelled Boiler. *Aerosol Air Qual. Res.* 11: 282–289.

Chi, K.H., Liu, K.T., Chang, S.H. and Chang, M.B. (2009). Atmospheric Deposition of PCDD/Fs Measured via Automated and Traditional Samplers in Northern Taiwan. *Chemosphere* 77: 1184–1190.

Chiu, J.C., Shen, Y.H., Li, H.W., Lin, L.F., Wang, L.C. and Chang-Chien, G.P. (2011). Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from an Electric Arc Furnace, Secondary Aluminum Smelter, Crematory and Joss Paper Incinerators. *Aerosol Air Qual. Res.* 11: 13–20.

Chuang, S.C., Chen, S.J., Huang, K.L., Wu, E.M.Y., Chang-Chien, G.P. and Wang, L.C. (2010). Gas/Particle Partitioning of Dioxins in Exhaust Gases from Automobiles. *Aerosol Air Qual. Res.* 10: 489–496.

Coutinho, M., Pereira, M. and Borrego, C. (2007). Monitoring of Ambient Air PCDD/F Levels in Portugal. *Chemosphere* 67: 1715–1721.

Donnelly, J.R., Munslow, W.D., Mitchum, R.K. and Sovocool, G.W. (1987) Correlation of Structure with Retention Index for Chlorinated Dibenzo-*p*-dioxins. *J. Chromatogr.* 392: 51–63.

Duarte-Davidson, R., Sewart, A., Alcock, R.E., Cousins, I.T. and Jones, K.C. (1997). Exploring the Balance between Sources, Deposition, and the Environmental Burden of PCDD/Fs in the U.K. Terrestrial Environment: An Aid to Identifying Uncertainties and Research Needs. *Environ. Sci. Technol.* 31: 1–11.

Eitzert, B.D. and Hites, R.A. (1989). Atmospheric Transport and Deposition of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans. *Environ. Sci. Technol.* 23: 1396–1401.

Fang, G.C., Lin, C.C., Huang, J.H. and Huang, Y.L. (2011). Measurement of Ambient Air Arsenic (As) Pollutant Concentration and Dry Deposition Fluxes in Central Taiwan. *Aerosol Air Qual. Res.* 11: 218–229.

Hale, M.D., Hileman, F.D., Mazer, T., Shell, T.L., Noble, R.W. and Brooks, J.J. (1985). Mathematical Modeling of Temperature Programmed Capillary Gas Chromatographic Retention Indexes for Polychlorinated Dibenzofurans. *Anal. Chem.* 57: 640–648.

Hsieh, L.T., Wang, Y.F., Kuo, G.H., Wang, L.C. and Chang-Chien, G.P. (2009). Cluster Analysis for Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans Concentrations in Southern Taiwan. *J. Air Waste Manage. Assoc.* 59: 1474–1480.

Hung, H., Blanchard, P., Poole, G., Thibert, B. and Chiu, C.H. (2002). Measurement of Particle-Bound Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in Arctic Air at Alert, Nunavut, Canada. *Atmos. Environ.* 36: 1041–1050.

Jurado, E., Jaward, F.M., Lohmann, R., Jones, K.C., Simo, R. and Dachs, J. (2004). Atmospheric Dry Deposition of Persistent Organic Pollutants to the Atlantic and Inferences for the Global Oceans. *Environ. Sci. Technol.* 38: 5505–5513.

Koester, C. and Hites, R.A. (1992). Wet and Dry Deposition of Chlorinated Dioxins and Furans. *Environ. Sci. Technol.* 26: 1375–1382.

Lai, Y.C., Lee, W.J., Li, H.W., Wang, L.C. and Chang-Chien, G.P. (2007). Inhibition of Polybrominated Dibenzo-*p*-dioxin and Dibenzofuran Formation from the Pyrolysis of Printed Circuit Boards. *Environ. Sci. Technol.* 41: 957–962.

Lee, W.J., Lin Lewis, S.J., Chen, Y.Y., Wang, Y.F., Sheu, H.L., Su, C.C. and Fan, Y.C. (1996). Polychlorinated Biphenyls in the Ambient Air of Petroleum Refinery, Urban and Rural Areas. *Atmos. Environ.* 30: 2371–2378.

Lee, W.J., Shih, S.I., Li, H.W., Lin, L.F., Yu, K.M., Lu, K., Wang, L.C., Chang-Chien, G.P., Fang, K. and Lin, M. (2009). Assessment of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans Contribution from Different Media to Surrounding Duck Farms. *J. Hazard. Mater.* 163: 1185–1193.

Lee, W.S., Chang-Chien G.P., Wang, L.C., Lee, W.J., Tsai, P.J., Wu, K.Y. and Lin, C. (2004). Source Identification of PCDD/Fs for Various Atmospheric Environments in a Highly Industrialized City. *Environ. Sci. Technol.* 38: 4937–4944.

Lee, W.S., Chang-Chien, G.P., Wang, L.C., Lee, W.J., Wu, K.Y. and Tsai, P.J. (2005). Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Stack Gases of Electric Arc Furnaces and Secondary Aluminum Smelters. *J. Air Waste Manage. Assoc.* 55: 219–26.

Lin, L.F., Lee, W.J., Li, H.W., Wang, M.S. and Chang-Chien, G.P. (2007). Characterization and Inventory of PCDD/F Emissions from Coal-fired Power Plants and other Sources in Taiwan. *Chemosphere* 68: 1642–1649.

Lin, L.F., Shih, S.I., Su, J.W., Shih, M., Lin, K.C., Wang, L.C. and Chang-Chien, G.P. (2010a). Dry and Wet Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans on the Drinking Water Treatment Plant. *Aerosol Air Qual. Res.* 10: 378–390.

Lin, W.Y., Wu, Y.L., Tu, L.K., Wang, L.C. and Lu, X. (2010b). The Emission and Distribution of PCDD/Fs in Municipal Solid Waste Incinerators and Coal-fired Power Plant. *Aerosol Air Qual. Res.* 10: 519–532.

Lohmann, R. and Jones, K.C. (1998). Dioxins and Furans in Air and Deposition: A Review of Levels, Behavior and Processes. *Sci. Total Environ.* 219: 53–81.

Lohmann, R., Lee, R.G.M., Green, N.J.L. and Jones, K.C. (2000). Gas-particle Partitioning of PCDD/Fs in Daily Air Samples. *Atmos. Environ.* 34: 2529–2537.

Ministry of the Environment Japan. (1999). Environmental Quality Standards in Japan-Air Quality. http://www.env.go.jp/en/air/aq/aq.html

Moon, H.B., Lee, S.J., Choi, H.G. and Ok, G. (2005). Atmospheric Deposition of Polychlorinated Dibenzo-*p*-dioxins (PCDDs) and Dibenzofurans (PCDFs) in Urban and Suburban Areas of Korea. *Chemosphere* 58: 1524–1534.

Oh, J.E., Choi, J.S. and Chang, Y.S. (2001). Gas/Particle Partitioning of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Atmosphere; Evaluation of Predicting Models. *Atmos. Environ.* 35: 4125–4134.

Oh, J.E., Choi, S.D., Lee, S.J. and Chang, Y.S. (2006). Influence of a Municipal Solid Waste Incinerator on Ambient Air and Soil PCDD/Fs Levels. *Chemosphere* 64: 579–587.

Oh, M.S., Lee, T.J. and Kim, D.S. (2011). Quantitative Source Apportionment of Size-segregated Particulate Matter at Urbanized Local Site in Korea. *Aerosol Air Qual. Res.* 11: 247–264.

Pankow, J.F. (1987). Review and Comparative Analysis of the Theories on Partitioning between the Gas and Aerosol Particulate Phases in the Atmosphere. *Atmos. Environ.* 21: 2275–2283.

Pankow, J.F. (1991). Commom y-intercept and Single Compound Regressions of Gas-particle Partitioning Data vs. 1/T. *Atmos. Environ.* 25: 2229–2239.

Pankow, J.F. (1994). An Adsorption Model of Gas/Particle Partitioning of Organic Compounds in the Atmosphere.

Atmos. Environ. 28: 185–188.

Pankow, J.F. and Bidleman, T.F. (1992). Interdependence of the Slopes and Intercepts from Log-log Correlations of Measured Gas-particle Partitioning and Vapor Pressure-I Theory and Analysis of Available Data. *Atmos. Environ.* 26: 1071–1080.

Rappe, C. (1993). Sources of Exposure, Environmental Concentrations and Exposure Assessment of PCDDs and PCDFs. *Chemosphere* 27: 211–225.

Rordorf, B.F. (1989). Prediction of Vapor Pressures, Boiling Points and Enthalpies of Fusion for Twenty-Nine Halogenated Dibenzo-*p*-Dioxins and Fifty-Five Dibenzofurans by a Vapor Pressure Correlation Method. *Chemosphere* 18: 783–788.

Sheu, H.L. and Lee, W.J. (1996). Dry Deposition of Polycyclic Aromatic Hydrocarbons in Ambient Air. *J. Environ. Eng.* 122: 1101–1112.

Shih, M., Lee, W.S., Chang-Chien, G.P., Wang, L.C., Hung, C.Y. and Lin, K.C. (2006). Dry Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in Ambient Air. *Chemosphere* 62: 411–416.

Tysklind, M., Fängmark, I., Marklund, S., Lindskog, A., Thaning, L. and Rapple, C. (1993). Atmospheic Transport and Transformation of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans. *Environ. Sci. Technol.* 27: 2190–2197.

Wang, J.B., Wang, M.S., Wu, E.M.Y., Chang-Chien, G.P. and Lai, Y.C. (2008). Approaches Adopted to Assess Environmental Impacts of PCDD/F Emissions from a Municipal Solid Waste Incinerator. *J. Hazard. Mater.* 152: 968–975.

Wang, L.C., Lee, W.J., Lee, W.S., Chang-Chien, G.P. and Tsai, P.J. (2003). Effect of Chlorine Content in Feeding Wastes of Incineration on the Emission of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofurans. *Sci. Total Environ.* 302: 185–198.

Wang, Y.F., Hou, H.C., Li, H.W., Lin, L.F., Wang, L.C., Chang-Chien, G.P. and You, Y.S. (2010). Dry and Wet Depositions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in the Atmosphere in Taiwan. *Aerosol Air Qual. Res.* 10: 378–390.

Wu, Y.L., Li, H.W., Chien, C.H., Lai, Y.C. and Wang, L.C. (2010). Monitoring and Identification of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in the Ambient Central Taiwan. *Aerosol Air Qual. Res.* 10: 463–471.

Wu, Y.L., Lin, L.F., Hsieh, L.T., Wang, L.C. and Chang-Chien, G.P. (2009). Atmospheric Dry Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in the Vicinity of Municipal Solid Waste Incinerators. *J. Hazard. Mater.* 162: 521–529.

Xu, M.X., Yan, J.H., Lu, S.Y., Li, X.D., Chen, T., Ni, M.J., Dai, H.F., Wang, F. and Cen, K.F. (2009). Gas/Particle Partitioning of Atmospheric PCDD/Fs in a Satellite Town in Eastern China. *Chemosphere* 76: 1540–1549.

Yamassaki, H., Kuwata, K. and Miyamoto, H. (1982). Effects of Ambient Temperature on Aspects of Airborne PolycyclicAromatic Hydrocarbons. *Environ. Sci. Technol.* 16: 189–194.

Yeh, S.H., Lai, C.H., Lin, C.H., Chen, M.J., Hsu, H.T., Lin, G.X., Lin, T.T. and Huang, Y.W. (2011). Estimating Cancer Risk Increment from Air Pollutant Exposure for Sewer Workers Working in an Industrial City. *Aerosol Air Qual. Res.* 11: 120–127.