Dry and Wet Depositions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in the Atmosphere in Taiwan

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ABSTRACT

The study has investigated the annual variation of the dry and wet depositions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) among different kinds of areas. A commercial suburban area, an industrial urban area, a coastal rural area and an agricultural rural area were chosen to measure the PCDD/F depositions in each season. The mean TEQ concentration, dry deposition flux and wet deposition in the industrial urban area all had the highest values, at 0.0958 pg I-TEQ/Nm^3, 29.1 pg I-TEQ/m^2-d and 47.8 pg/L, respectively. Additionally, the annual total deposition flux of PCDD/Fs in the commercial, industrial, coastal and agricultural areas was 168, 310, 135 and 115 ng/m^2-year, respectively. The seasonal change of PCDD/Fs in air could be affected by heating and temperature inversion in winter or photodegradation and OH radical reaction in summer. The various homologue profiles of PCDD/Fs probably affect the distribution in environmental sinks, such as sediments or the living organisms in rivers or seas. Additionally, the results of statistical analysis indicated that PCDD I-TEQ dry deposition flux (ng I-TEQ/m^2-month) was negatively correlated with the ambient temperature ($r = -0.843$) for the agricultural rural area, possibly due to vapor pressure (temperature-dependent) effect on the gas/particle PCDD/F partitioning. Furthermore, the PCDD/F concentration of wet deposition flux (ng I-TEQ/m^2-month) was positively correlated with the monthly rainfall ($r = 0.826$–0.988) at the four sampling areas.

Keywords: PCDD/Fs; Dry deposition; Wet deposition; Fluxes; Atmosphere.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) have received a lot of attention in Taiwan, not only for their adverse health effects, but also for the wide distribution of PCDD/Fs in the atmosphere (Kao *et al.*, 2006; Lee *et al.*, 2008; Lee *et al.*, 2009). Municipal solid waste incinerators are widely regarded as the major source of PCDD/F emissions to the atmosphere (H.-Jones *et al.*, 1993; Addink and Altwicker, 2001). However, the situation is different in Taiwan, because sintering plants contribute the largest amounts (Wang *et al.*, 2003; Chen, 2004; Shih *et al.*, 2009). PCDD/Fs exist in the atmosphere as gases and bound to particles. They are emitted into the atmosphere in trace amounts, and can be transported long distances before they are deposited (Lee *et al*, 2003; Zhang *et al.*, 2009). This will result in widespread dispersal through environment and initiate the air-deposition-soil/water-food chain-wildlife human exposure cycle (Lohmann and Jones, 1998; Hu *et al.*, 2009).

For the reference year of 2006 in Taiwan, the total release of PCDD/Fs to air was 85.6 g I-TEQ/yr (EPA, 2008). Atmospheric depositional processes play a key role in the transport and fate of persistent organic pollutants (POPs) at the regional and global scales (Jurado *et al.*, 2004), and PCDD/Fs gas-particle partitioning will influence the deposition processes (Oh *et al.*, 2001). The distribution of PCDD/Fs between the gas-particle phases determines their fate: OH-radical reactions are thought to occur mostly in the gaseous phase, while particulate-bound compounds have, in general, shorter residence times in the atmosphere (Lohmann *et al.*, 2007).

Deposition of PCDD/Fs is divided into dry deposition (gaseous, particulate) and wet deposition. The former is an adsorption at the air-surface interface, when an airborne

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particle comes into contact with a surface. The latter is the removal of atmospheric particles by precipitation (rain and cloud droplets) (Lohmann and Jones, 1998). Dry deposition is generally dominated by higher chlorinated congeners, especially OCDD (Lohmann and Jones, 1998). Furthermore, wet deposition is the major pathway of the higher chlorinated PCDD/Fs to a bare soil (Schröder *et al.*, 1997).

In this study, dry and wet depositions of PCDD/Fs in the ambient air near a commercial suburban area, an industrial area, a coastal rural area, and an agricultural rural area were sampled seasonally to compare the variation of PCDD/F depositions by diverse human activities. Furthermore, the effects of various meteorological parameters on the dry and wet deposition of PCDD/Fs were evaluated by statistical analysis.

METHODs

Sampling Sites

Fig. 1 shows the variation of PCDD/F concentrations among a commercial suburban area (C), an industrial area (I), a coastal rural area (S), and an agricultural rural area (A). Site C is located at the suburb of Taipei, surrounded by hills and trees. Site I is setup at industrial park in Chungli city with population of 380,000. Site S is at the coastal area in Chunan township with population of 70,000 and site A is situated in the agricultural plain of Yongkang city with population of 220,000. Sampling sites around these areas were allocated by the results of the Industrial Source Short Term model (ISCST), and four samples per season in each area were taken in 2006. A total of sixty-four ambient air samples were collected seasonally by using a PS-1 sampler (Graseby Andersen, GA) according to the revised EPA Reference Method T09A. The sampling flow rate was specified at ~0.225 m^3/min, and each sample was collected continuously on three consecutive days. The PS-1 sampler was equipped with a quartz fiber filter for sampling particle-phase PCDD/Fs, followed by a glass cartridge packed with PUF and XAD-2 for sampling the gas-phase PCDD/Fs. A known amount of surrogate standard was spiked in the glass cartridge in the laboratory before sampling. To ensure that the collected samples were free of contamination, one field blank was always taken with each sampling. Once the ambient air sampling was completed, the samples were brought back to the laboratory under refrigeration with temperature less than 10°C. Sampling meteorological information was showed in Table 1.

Analysis of PCDD/Fs

Analysis of ambient air samples for PCDD/Fs was performed according to the US EPA Reference Method TO9A. All chemical analyses were measured in the Super Micro Mass Research and Technology Centre of the Cheng Shiu Institute of Technology. This facility was the first certified lab by the Taiwan EPA to analyze PCDD/Fs in Taiwan, and it has passed international intercalibration standards test on PCDD/Fs in fly ash, sediment, mother's milk, human blood, and cod liver. Each sample was spiked with a known amount of the internal standard. After being extracted for 24 h, the extract was concentrated, treated with concentrated sulfuric acid, and then subjected to a series of sample cleanup and fractionation procedures. Sample cleanup was done using an acidic silica-gel column, an alumina column, and an activated carbon column. Consequently, the elute was concentrated to around 1 mL and then transferred to a vial, further concentrated to near-dryness with a nitrogen stream. Before analyzing PCDD/Fs, the standard solution was added into the sample to ensure
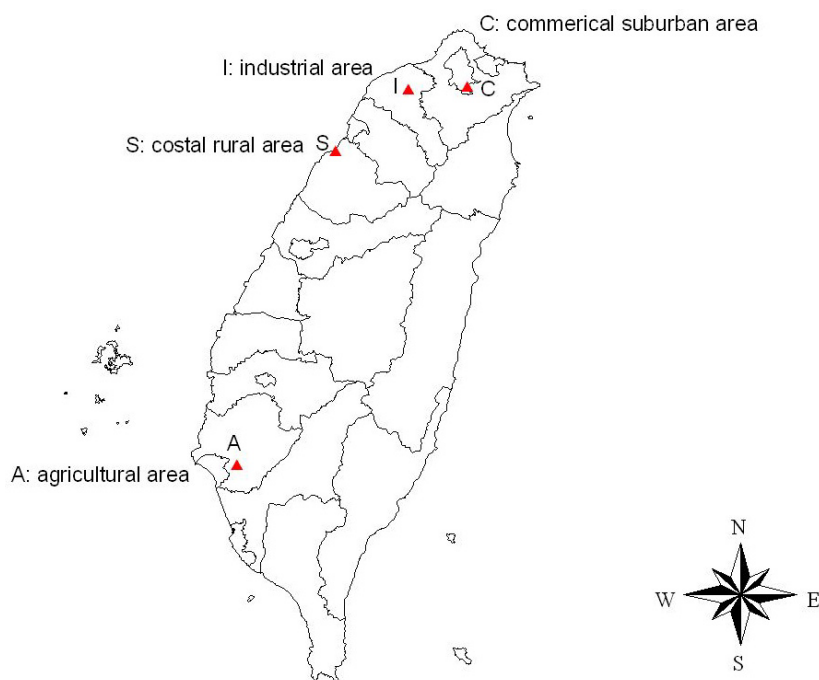

Fig. 1. The sampling sites, C: commercial suburban area, I: industrial area, S: coastal rural area, and A: agricultural area.

Table 1. Meteorological information during PCDD/Fs sampling periods.

District	Season	Sampling date	Temperature (°C)	Wind Direction	PM_{10} (µg/m^3)
Commercial suburban area	Spring	95/4/24–26	21.6	East	48.0
	Summer	95/6/26–28	33.0	South-southeast	37.3
	Autumn	95/8/14–17	31.6	East	49.3
	Winter	95/11/13–16	22.6	East-northeast	52.0
Industrial urban area	Spring	95/4/26–28	21.7	East-northeast	59.0
	Summer	95/6/29–7/1	31.8	West-northwest	53.0
	Autumn	95/8/28–30	29.6	Northeast	61.0
	Winter	95/11/6–9	24.8	East-northeast	88.5
Costal rural area	Spring	95/4/24–26	20.6	Northeast	24.3
	Summer	95/7/10–13	31.4	South-southeast	36.8
	Autumn	95/8/31–9/2	29.5	West-northwest	52.3
	Winter	95/11/6–10	23.1	East-northeast	88.0
Agricultural rural area	Spring	95/4/26–28	28.2	South	48.3
	Summer	95/6/6–9	29.6	East-northeast	36.3
	Autumn	95/8/7–10	28.5	North-northeast	48.3
	Winter	95/11/20–24	26.0	North-northeast	69.4

recovery during the analysis process (Shih *et al.*, 2006a). High-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for PCDD/F analysis. 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, film thickness = 0.25 µm) (J&W Scientific, CA) with a splitless injection, while the HRMS (Micromass Autospec Ultima, Manchester, UK) had a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring was used with the resolving power set at 10,000. The electron energy and source temperature were specified at 35 eV and 250°C, respectively. The oven temperature program was set as follows: initially at 150°C (held for 1 min), then increased by 30°C/min to 220°C (held for 12 min), and finally increased by 1.5 °C/min to 310°C (held for 20 min). Helium was used as the carrier gas. The protocol for quality analysis/quality control was strictly followed (Wang and Lee, 2010). The recovery efficiency of known-addition analysis ranged from 75% to 118%. The MDLs ranged from 0.0001 to 0.0035 ng/Nm^3.

Particle and Gas Partition of PCDD/Fs

Particle and Gas concentration was calculated by gas/particle partitioning multiplying total PCDD/Fs concentrations. Gas/particle partitioning was calculated by partitioning constant as follows (Pankow, 1991, 1994; Pankow and Bidleman, 1992):

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

K_p: partitioning constant (m^3/µg),
TSP: total suspended particle concentration (µg/m^3),
TSP was calculated by TSP/PM_{10} ratio: 1.92(spring), 2.10(summer), 2.57(autumn) and 3.44(winter) in northern Taiwan (Chang *et al.*, 2003); 2.17(spring), 1.38(summer), 1.78(autumn) and 2.30(winter) in central Taiwan (Fang *et al.*, 2006); 1.79(spring),

1.54(summer), 1.37(autumn) and 1.37(winter) in southern Taiwan (Tainan EPB, 2006)
F: particle-associated concentration for PCDD/Fs (pg/m^3),
A: gaseous-associated concentration for PCDD/Fs (pg /m^3).
K_p was calculated as follows:

$$\log K_p = m_r \times \log P_L^0 + b_r \quad (2)$$

P_L^0: subcooled liquid vapor pressure (Torr),
b_r: intercept in a plot of $\log K_p$ vs $\log P_L^0$ referred to our group data by Chao (Chao *et al.*, 2004).
P_L^0 was calculated as follows (Hung *et al.*, 2002):

$$\log P_L^0 = \frac{-1.34(\text{RI})}{T} + 1.67 \times 10^{-3}(\text{RI}) - \frac{1320}{T} + 8.087 \quad (3)$$

RI: gas chromatographic retention indexes, GC-RI, referred to Donnelly and Hale (Hale *et al.*, 1985; Donnelly *et al.*, 1987),
T: ambient temperature (K).

In the previous study, Chao did find subcooled liquid vapor pressure (P_L^0)-based model to be excellent descriptors for the gas–particle partitioning of PCDD/Fs (Chao *et al.*, 2004). This model led to the well estimation of C_p, C_g and increased the reliability of calculation for PCDD/F deposition fluxes.

Dry and Wet Deposition Process of PCDD/Fs

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

$$F_T = F_g + F_{p,} \quad (4)$$
$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \quad (5)$$

F_T: the summation of PCDD/F deposition fluxes from both gas and particle phases,
F_g: the PCDD/F deposition flux contributed by the gas phase,

F_p: the PCDD/F deposition flux contributed by the particle phase,

$$F_T = C_T \times V_{d,T}, \quad F_g = C_g \times V_{d,g} \quad \text{and} \quad F_p = C_p \times V_{d,p} \qquad (6)$$

C_T: the measured concentration of total PCDD/Fs in the ambient air,
$V_{d,T}$: the dry deposition velocity of total PCDD/Fs,
C_g: the calculated concentration of PCDD/Fs in the gas phase,
$V_{d,g}$: the dry deposition velocity of the gas-phase PCDD/Fs,
C_p: the calculated concentration of PCDD/Fs in the particle phase,
$V_{d,p}$: the dry deposition velocity of the particle-phase PCDD/Fs.

The dry deposition velocity of total PCDD/Fs in spring, summer, autumn, and winter were 0.45, 0.52, 0.32 and 0.39 cm/s, respectively (Shih *et al.*, 2006a). Dry deposition velocity of gas-phase PCDD/Fs is mainly by diffusion, and because of a lack of measured data for PCDD/Fs, a selected value (0.010 cm/s) of gas-phase polycyclic aromatic hydrocarbon (PAH) dry deposition velocity, $V_{d,g}$ proposed by Sheu (Sheu and Lee, 1996) and used by Lee (Lee *et al.*, 1996), is also used here to calculate the PCDD/F dry deposition flux contributed by its gas phase.

The wet deposition flux of PCDD/Fs is a combination of both vapor dissolution into rain and the removal of suspended particulates by precipitation. The gas scavenging ratio, S_g, can be estimated by

$$S_g = RT/H, \qquad (7)$$

S_g: the gas scavenging ratio of PCDD/Fs (dimensionless),
R: the universal gas constant (82.06×10^{-6} m^3 atm/mol K),
T: ambient temperature (K),
H: Henry constant (m^3 atm/mol).

$$S_g = C_{rain,dis.}/C_g \qquad (8)$$

$C_{rain,dis}$: the dissolved-phase concentration of PCDD/Fs in the raindrop,
C_g: the concentration of PCDD/Fs in the gas phase.

The particle scavenging ratio, S_p, on the other hand, can be calculated by

$$S_p = C_{rain,particle}/C_p \qquad (9)$$

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

Total scavenging of precipitation (S_{tot}) is the sum of gas and particle scavenging, which can be calculated by

$$S_{tot} = S_g (1-\Phi) + S_p \times \Phi \qquad (10)$$

S_{tot}: the total scavenging ratio of PCDD/Fs (dimensionless),
Φ: the fraction of PCDD/Fs bound to particles.

Due to the lack of real measured data for the particle scavenging ratios of PCDD/Fs, the values used in this study were referenced to those in Eitzer and Hites' work (Eitzer and Hites, 1989).

RESULTS AND DISCUSSION

PCDD/F Concentration in Different Area

The mean PCDD/F concentrations in the atmosphere at different areas are shown in Table 2. The mean TEQ concentration for the commercial suburb, industrial urban, coastal rural and agricultural rural areas was 0.0389, 0.0958, 0.0329 and 0.0369 pg I-TEQ/Nm³, respectively. It is thus obvious that the mean PCDD/F concentration in the industrial urban area was much higher than those in other areas. It was apparently indicated the high concentration was correlated with the industrial activities. As showed in Eduljee and Dyke's study, industrial processes contributed 535–955 g I-TEQ/yr of PCDD/Fs, which was approximately 90% of the total PCDD/Fs inventory in UK. This also demonstrated that the PCDD/F concentration was highly influenced by local industrial activities (Eduljee and Dyke, 1996). These results are comparable to those in the EPA's report which surveyed 80 ambient PCDD/F samples in Taiwan (EPA, 2002a). The mean concentration was 0.105 pg I-TEQ/Nm³ in southern Taiwan, and 0.038 pg I-TEQ/Nm³ in northern Taiwan. The mean TEQ concentrations at each of these areas was much lower than the air quality standard in Japan (0.6 pg I-TEQ/Nm³) (EPA, 2002b). The seasonal distribution in Table 2 shows that the highest and lowest concentrations occurred in winter and spring, respectively, in the commercial, industrial and coastal areas. However, the pattern was a little different for the I-TEQ concentration distribution, for which the highest level occurred in summer in the industrial area. In contrast, the pattern was totally different in the agricultural area, where the PCDD/F and I-TEQ concentrations were both highest in spring and lowest in summer. The data of wind rose shows that the wind direction in spring at agricultural area was mostly from the south, a highly populated commercial urban area, which can be expected to have contributed lots of PCDD/Fs from heavily loaded transportation. This possibly explained the higher PCDD/Fs concentration in the agricultural area in spring.

Fig. 2 shows the seasonal congener profiles of PCDD/Fs in the atmosphere at different areas. The major congeners were OCDD, OCDF, 1,2,3,4,6,7,8-HpCDF and 1,2,3,4,6,7,8-HpCDD and these are all highly chlorinated PCDD/Fs. Furthermore, the chlorine-substituted profiles of PCDD/Fs in the four sampling areas all show that OCDD, HpCDF and OCDF were the three dominant chlorine-substituted PCDD/Fs and account for more than 56% of total PCDD/F concentration. The congener profile for the commercial area in summer is similar to that found in Shih's study in winter (Shih *et al.*, 2006b; Wang *et al.*, 2009), which surveyed the ambient samples around a municipal solid waste incinerator. However, the chlorine-substituted profiles of PCDD/Fs for the four sampling areas are very different to those in Wang's study (Wang *et al.*, 2009), which investigated ambient samples near a power plant. The emission sources at the sampling areas I located at an industrial park were more complex, and resulted in various congener/chlorine-substituted profiles of PCDD/Fs in the atmosphere.

Table 2. Atmospheric concentrations of PCDD/Fs at each area.

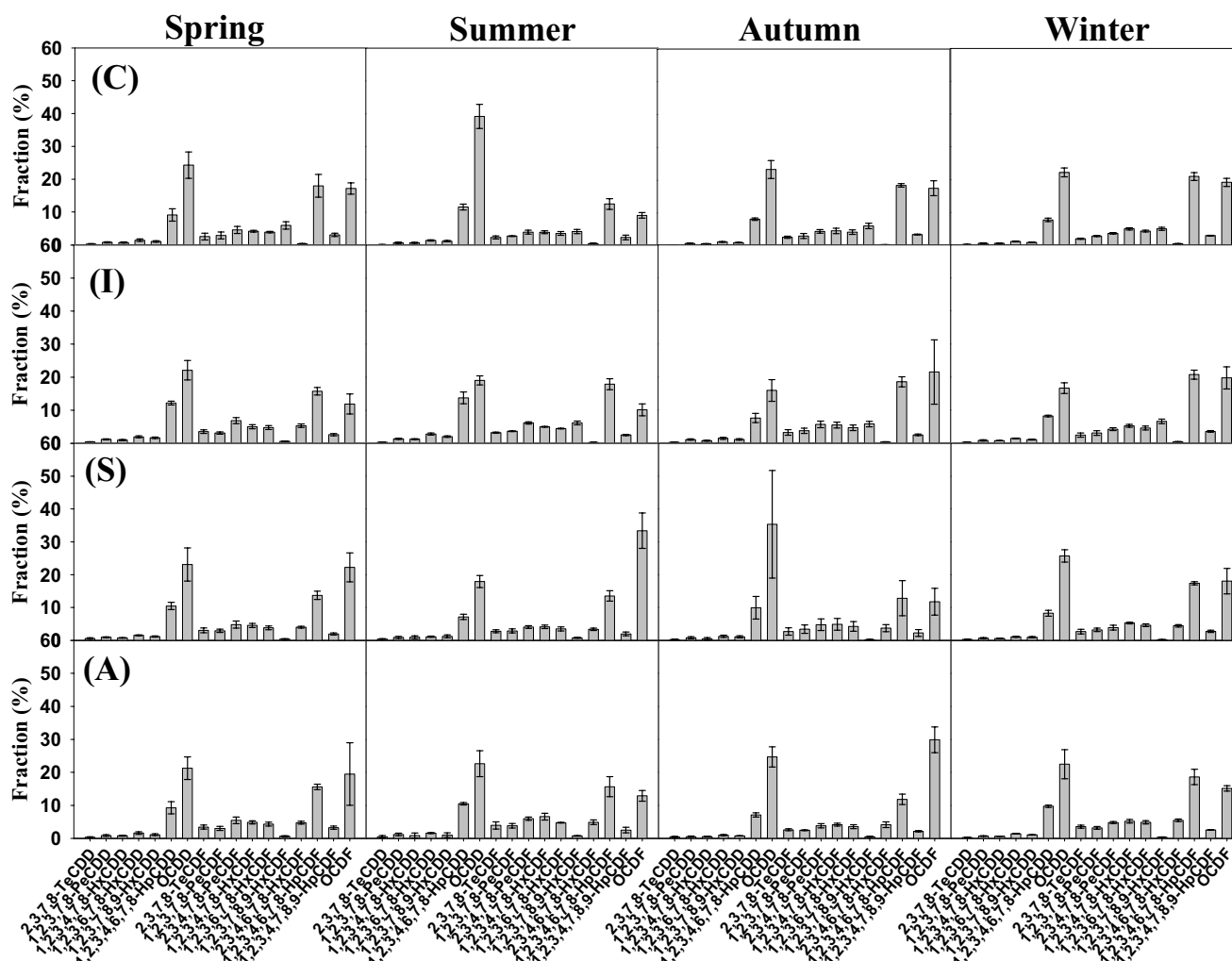| area	commercial area (C)				industrial area (I)				coastal area (S)				agricultural area (A)			
season	spring	summer	autumn	winter	spring	summer	autumn	winter	spring	summer	autumn	winter	spring	summer	autumn	winter
2,3,7,8-TeCDD	0.0015	0.0007	0.0014	0.0029	0.0050	0.0052	0.0036	0.0047	0.0026	0.0027	0.0017	0.0022	0.0027	0.0015	0.0023	0.0021
1,2,3,7,8-PeCDD	0.0046	0.0047	0.0050	0.0069	0.0134	0.0210	0.0150	0.0152	0.0046	0.0058	0.0050	0.0051	0.0072	0.0036	0.0031	0.0055
1,2,3,4,7,8-HxCDD	0.0041	0.0048	0.0043	0.0066	0.0112	0.0197	0.0101	0.0141	0.0036	0.0061	0.0037	0.0042	0.0068	0.0033	0.0033	0.0050
1,2,3,6,7,8-HxCDD	0.0089	0.0100	0.0082	0.0127	0.0222	0.0440	0.0195	0.0271	0.0076	0.0071	0.0082	0.0081	0.0135	0.0052	0.0059	0.0112
1,2,3,7,8,9-HxCDD	0.0064	0.0085	0.0068	0.0097	0.0188	0.0318	0.0149	0.0202	0.0057	0.0080	0.0071	0.0071	0.0093	0.0037	0.0045	0.0082
1,2,3,4,6,7,8-HpCDD	0.0600	0.0826	0.0553	0.0847	0.1370	0.2160	0.1020	0.1550	0.0532	0.0459	0.0732	0.0617	0.0782	0.0344	0.0411	0.0763
OCDD	0.1470	0.2800	0.1580	0.2420	0.2480	0.2980	0.2120	0.3040	0.1200	0.1150	0.2710	0.1920	0.1810	0.0713	0.1420	0.1770
2,3,7,8-TeCDF	0.0139	0.0160	0.0181	0.0220	0.0398	0.0502	0.0419	0.0417	0.0145	0.0179	0.0159	0.0199	0.0292	0.0121	0.0153	0.0275
1,2,3,7,8-PeCDF	0.0161	0.0199	0.0207	0.0308	0.0353	0.0575	0.0496	0.0528	0.0142	0.0185	0.0204	0.0238	0.0254	0.0121	0.0143	0.0247
2,3,4,7,8-PeCDF	0.0269	0.0274	0.0302	0.0398	0.0773	0.0964	0.0757	0.0767	0.0229	0.0260	0.0290	0.0291	0.0463	0.0195	0.0227	0.0374
1,2,3,4,7,8-HxCDF	0.0262	0.0285	0.0314	0.0547	0.0569	0.0782	0.0729	0.0957	0.0223	0.0264	0.0301	0.0395	0.0416	0.0217	0.0246	0.0406
1,2,3,6,7,8-HxCDF	0.0246	0.0267	0.0287	0.0475	0.0540	0.0700	0.0618	0.0829	0.0186	0.0225	0.0261	0.0344	0.0369	0.0156	0.0209	0.0379
1,2,3,7,8,9-HxCDF	0.0399	0.0304	0.0415	0.0550	0.0068	0.0967	0.0776	0.1250	0.0020	0.0049	0.0018	0.0016	0.0057	0.0025	0.0028	0.0029
2,3,4,6,7,8-HxCDF	0.0019	0.0036	0.0024	0.0055	0.0603	0.0041	0.0049	0.0079	0.0199	0.0215	0.0235	0.0332	0.0407	0.0161	0.0246	0.0428
1,2,3,4,6,7,8-HpCDF	0.1230	0.0923	0.1260	0.2290	0.1780	0.2800	0.2610	0.3930	0.0692	0.0871	0.0796	0.1300	0.1350	0.0532	0.0697	0.1450
1,2,3,4,7,8,9-HpCDF	0.0204	0.0181	0.0236	0.0320	0.0296	0.0392	0.0349	0.0680	0.0098	0.0126	0.0135	0.0206	0.0282	0.0088	0.0126	0.0200
OCDF	0.1110	0.0663	0.1190	0.2080	0.1350	0.1580	0.3280	0.3890	0.1110	0.2130	0.0703	0.1350	0.1830	0.0432	0.1750	0.1180
PCDDs	0.2330	0.3910	0.2390	0.3660	0.4550	0.6350	0.3770	0.5400	0.1980	0.1910	0.3690	0.2800	0.2980	0.1230	0.2020	0.2850
PCDFs	0.4040	0.3290	0.4410	0.7240	0.6730	0.9300	1.0100	1.3300	0.3040	0.4500	0.3100	0.4670	0.5720	0.2050	0.3820	0.4970
PCDDs/PCDFs ratio	0.5760	1.1900	0.5410	0.5050	0.6770	0.6820	0.3740	0.4050	0.6500	0.4240	1.1900	0.6000	0.5220	0.6000	0.5290	0.5740
total PCDD/Fs (pg/Nm³)	**0.6370**	**0.7200**	**0.6800**	**1.0900**	**1.1300**	**1.5700**	**1.3800**	**1.8700**	**0.5020**	**0.6410**	**0.6790**	**0.7470**	**0.8700**	**0.3280**	**0.5850**	**0.7820**
PCDDs pg-I-TEQ/Nm³	0.0064	0.0065	0.0066	0.0103	0.0185	0.0277	0.0168	0.0202	0.0072	0.0082	0.0070	0.0075	0.0103	0.0049	0.0058	0.0083
PCDFs pg-I-TEQ/Nm³	0.0264	0.0264	0.0299	0.0427	0.0644	0.0843	0.0695	0.0813	0.0208	0.0245	0.0262	0.0302	0.0417	0.0178	0.0219	0.0368
PCDDs/PCDFs(TEQ)ratio	0.2430	0.2460	0.2200	0.2420	0.2880	0.3290	0.2420	0.2490	0.3480	0.3360	0.2680	0.2480	0.2460	0.2740	0.2650	0.2240
Total I-TEQ (pg I-TEQ/Nm³)	**0.0329**	**0.0329**	**0.0365**	**0.0531**	**0.0829**	**0.1120**	**0.0863**	**0.1020**	**0.0280**	**0.0327**	**0.0333**	**0.0377**	**0.0519**	**0.0227**	**0.0277**	**0.0451**

Fig. 2. Congener profiles of PCDD/F in ambient air.
(C: the commercial suburban area, I: the industrial urban area, S: the coastal rural area, A: the agricultural rural area.)

Gas/Particle Partitioning of PCDD/Fs

The gas-particle partitioning of PCDD/Fs in the ambient air of four areas is illustrated in Fig. 3. It is obvious that Φ (fraction of PCDD/Fs bound to particles) was much lower for total PCDD/Fs and PCDD/F I-TEQ in summer and autumn than it was in spring and winter for the four sampling areas. Most of the PCDD/F congeners that dominated in the particle phase ($\Phi > 0.50$) had a chlorine number more than six in spring and winter for the commercial, industrial and coastal areas. However, only the Φ of 1,2,3,4,6,7,8-HpCDD, OCDD, 1,2,3,4,7,8,9-HpCDF and OCDF was more than 0.50 in summer for these three areas. Accordingly, all PCDD/Fs would probably evaporate from the particle phase to gas phase as the temperature increased. Lee (2008) indicated that temperature had a negative correlation with particle-gas partitioning, while Lohmann (1999) and Ogura (2001) indicated that the seasonal change of PCDD/Fs in air could be affected by heating and temperature inversion in winter or photodegradation and OH radical reaction in summer. The Φ of PCDD and PCDF in this study ranged from 0.72–0.96 and 0.40–0.84, respectively, for commercial,

industrial and coastal areas. Additionally, Φ reached the highest level in winter and the lowest level in summer, for both PCDD and PCDF in these areas. Additionally, the Φ of PCDD-I-TEQ and PCDF-I-TEQ ranged from 0.13–0.57 and 0.099–0.57, respectively. Consequently, total PCDD/Fs was mostly dominated by its particle phase, while total PCDD/F I-TEQ was mostly dominated by its gas phase. However, the distribution was a little different for the agricultural area, for which the Φ of PCDD, PCDF, total PCDD/Fs, PCDD-I-TEQ, PCDF-I-TEQ and PCDD/Fs-I-TEQ were all smaller than those for other areas. The seasonal variation of Φ was thus not so apparent in the agricultural area and this was possibly because the temperature variation did not fluctuate much among the different seasons (26°C–29.6°C) in this area, which was located in southern Taiwan. So did the Φ for individual PCDD/Fs congener (Shih *et al.*, 2006a).

Figs. 4 and 5 illustrate the gas/particle partitioning of dry and wet deposition for four the seasons, respectively. The results show that dry deposition of PCDD/Fs was mainly in the particle phase (99.1%–99.7%) at the four sampling areas in every season. Furthermore, the particle bound
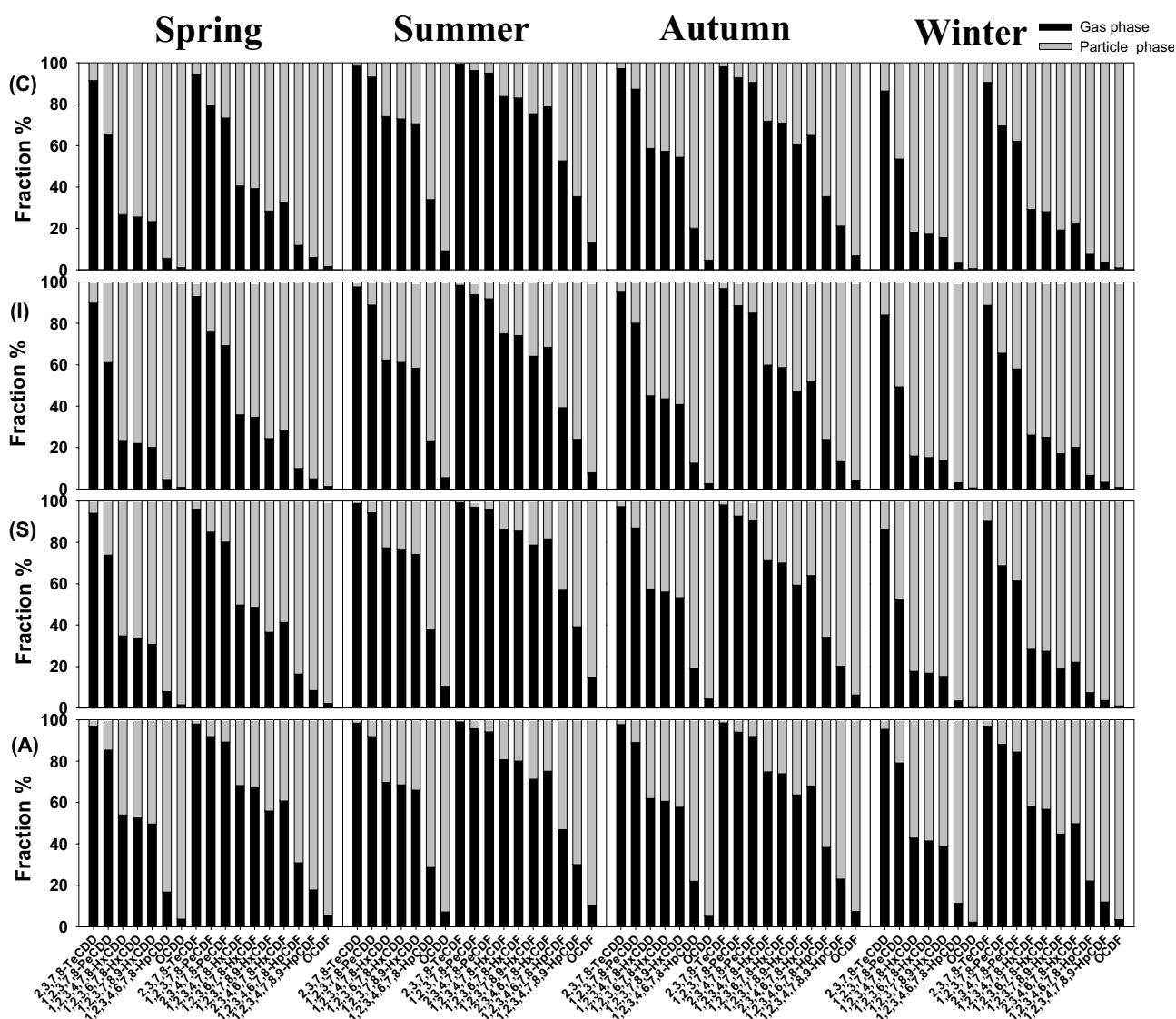
Fig. 3. Gas/particle partitioning of total PCDD/Fs for four seasons.
(C: the commercial suburban area, I: the industrial urban area, S: the coastal rural area, A: the agricultural rural area.)

fraction of wet deposition ranged from 20.2%–99.9%, 29.8%–99.9%, 17.0%–99.9% and 22.7%–99.8% for the commercial, industrial, coastal and agriculture areas, respectively. The results show that TeCDD/Fs and PeCDD/Fs are prone to evaporate as the gas phase with increasing temperature.

Dry and Wet Deposition of PCDD/Fs

Based on the deposition equations (1) and (2), the deposition flux of PCDD/Fs was calculated and is in Fig. 6. C_T was measured, and C_p and C_g were determined according to the gas/particle partitioning shown in Figs. 3–5. The results indicate that most PCDD/Fs of dry deposition fluxes were bound to the particle phase (94.2%–100%), and the dry deposition fluxes of total PCDD/Fs in different seasons ranged from 7.67–18.2, 17.6–40.6, 6.69–11.6, 5.73–15.6 pg I-TEQ/m²-d, with an average of 11.8, 29.1, 9.81 and 10.9 pg I-TEQ/m²-d for commercial, industrial, coastal, and agricultural areas, respectively. The dry deposition flux for

the four sampling areas reached the highest level in winter (except for the industrial area, which was the highest in summer) and the lowest level in autumn. Furthermore, the mean dry deposition flux for the commercial, industrial, coastal, and agricultural areas was 302, 567, 250 and 245 pg/m²-d, respectively. These values were similar to those in Kurz's (1993) and Koester's (1992) studies. Additionally, OCDD was the dominant congener in all the dry deposition fluxes, just as in Wu's study (Wu *et al.*, 2009). The fraction of OCDD to all deposition fluxes reached the highest level in summer (44.8%), spring (28.1%), autumn (41.3%) and summer (26.6%) for the commercial, industrial, coastal, and agricultural areas, respectively. The fraction of OCDD was an average of 29.3%, 20.4%, 28.7% and 24.2% for the commercial, industrial, coastal, and agricultural areas, respectively. These values were similar with Hiester's (1993), Kurz's (1993) and Wallenhorst's (1997) studies, however, they were smaller than those in Wu's (Wu *et al.*, 2009) investigation.
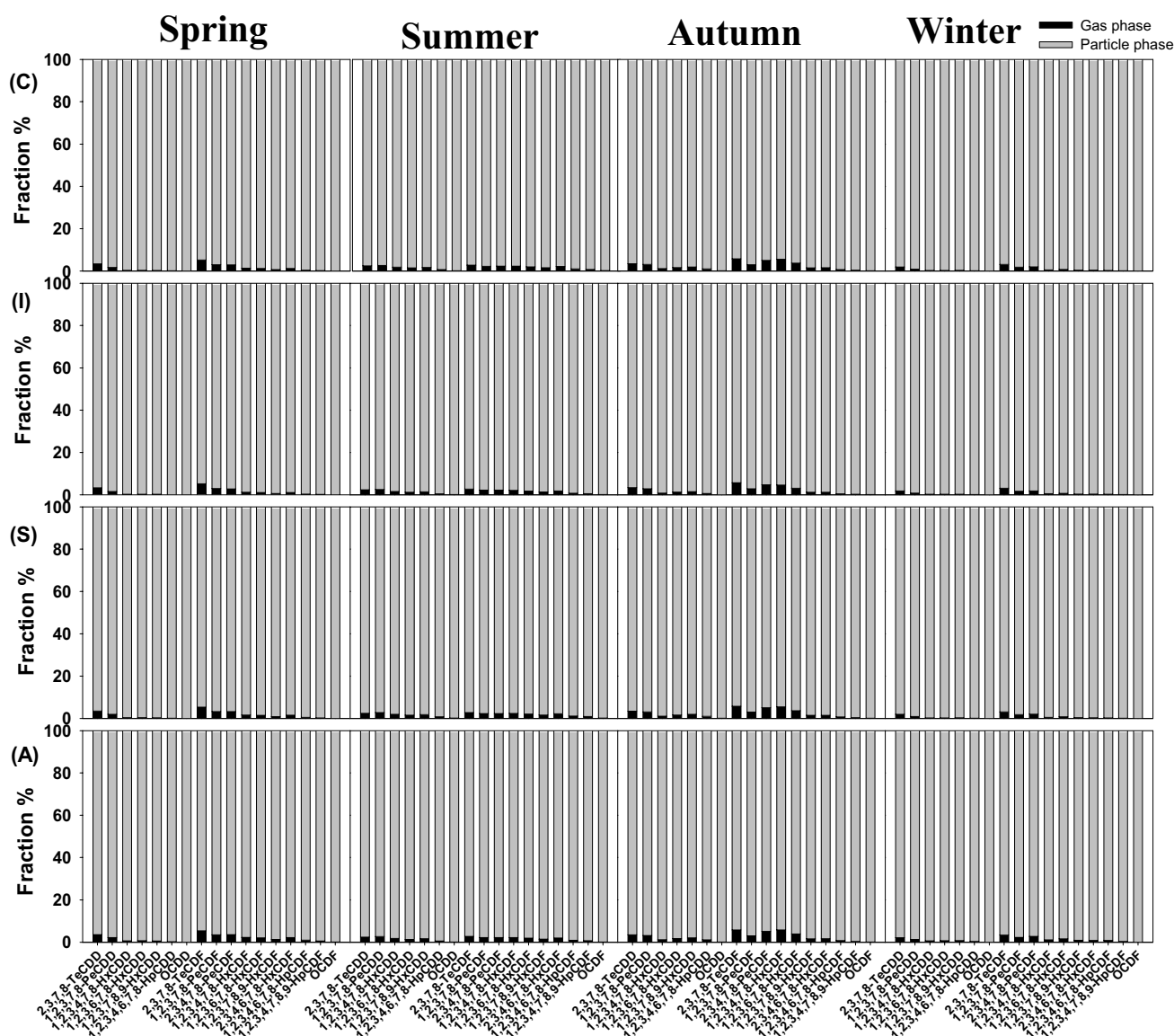
Fig. 4. Gas/particle partitioning of dry deposition for four seasons.
(C: the commercial suburban area, I: the industrial urban area, S: the coastal rural area, A: the agricultural rural area.)

Total PCDD/Fs in the rain's dissolved phase ranged from 19.7–40.3, 38.5–69.2, 17.0–27.4, 8.11–26.0 pg/L, with an average of 25.6, 47.8, 20.7 and 19.2 pg/L for commercial, industrial, coastal, and agricultural areas, respectively. Total PCDD/Fs were 92.1%–98.9% bound to particles. These values were smaller than those in Koester's study (Koester and Hites, 1992). The wet PCDD/F deposition in Indianapolis and Bloomington was 160 and 150 pg/L, respectively. Indianapolis has a population of 800,000 and is an urban location, while Bloomington is a suburb location of 50,000 population. Koester (1992) also indicated that smaller amounts of precipitation were expected to have higher particle-bound concentrations of PCDD/Fs, and found an inversely correlated relationship (r = –0.7) between the amount of rainfall and particle-bound PCDD/F concentrations. However, no such relationship was found in this study. It was probably due to the limited samples would make insufficient databank to get

statistically correlated results. This warrants further investigation in the future. Additionally, the seasonal distribution of wet deposition showed that the highest wet deposition fluxes were in winter for all areas, possibly due to the fact that there was less rainfall in winter than in other seasons. Furthermore, the homologues wet deposition profiles in this study were different to those in Koester's study. The first two dominant homologues bound to particles of wet deposition were octachlorinated and heptachlorinated PCDD in Koester's study, but in this work, they were octachlorinated PCDD and heptachlorinated PCDF for commercial and industrial areas, octachlorinated PCDD and octachlorinated PCDF for coastal and agricultural areas. These varied homologue profiles of PCDD/F probably affect the distribution in environmental sinks, such as the sediments or living organisms in rivers or seas.

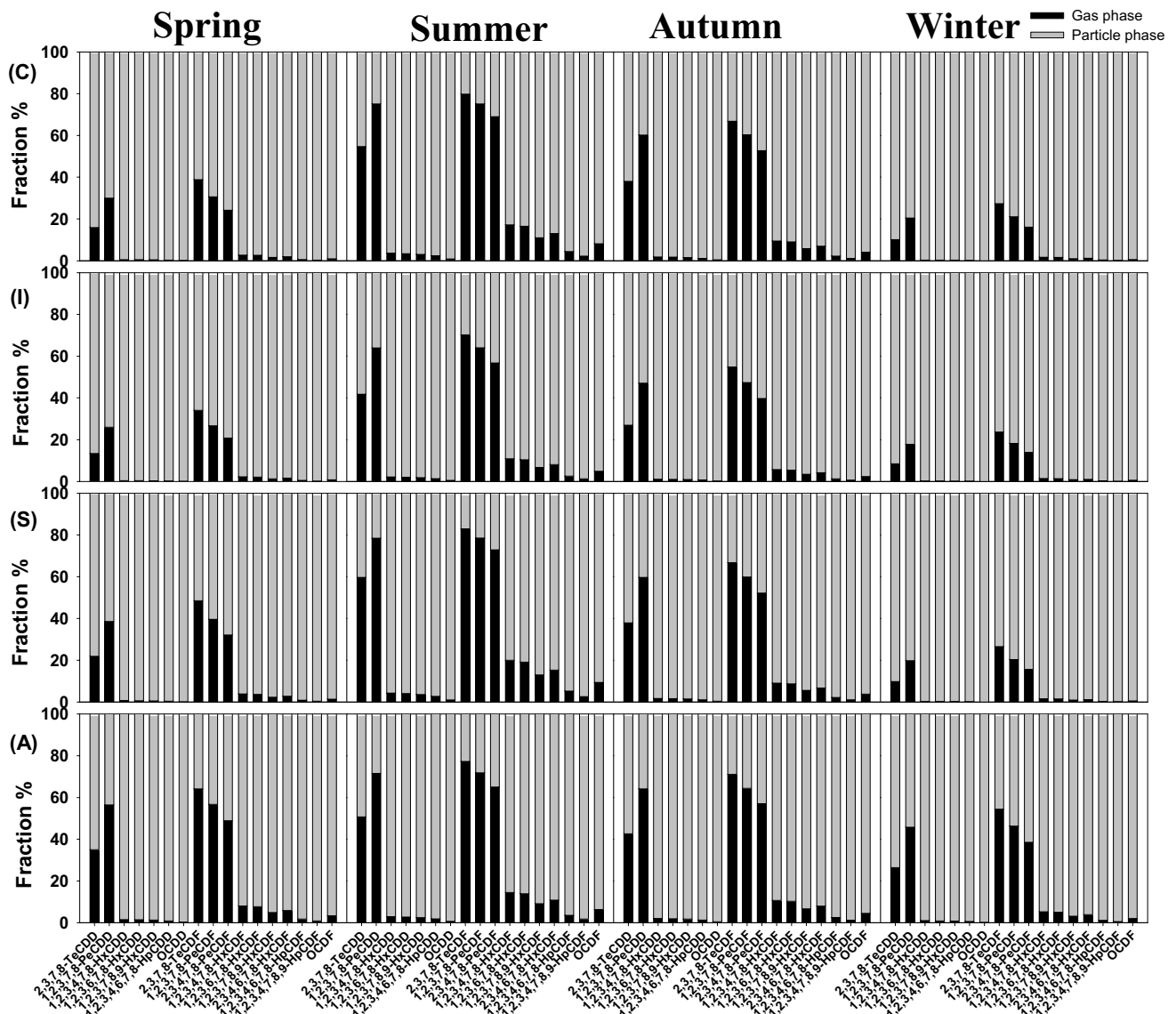The correlated coefficients of PCDD/Fs dry deposition

Fig. 5. Gas/particle partitioning of wet deposition for four seasons.
(C: the commercial suburban area, I: the industrial urban area, S: the coastal rural area, A: the agricultural rural area.)

flux with the ambient meteorological conditions are shown in Table 3. The PCDD/F dry deposition fluxes at the four sampling areas were low/middle-correlated with the ambient meteorological conditions. The PCDD I-TEQ dry deposition flux (ng I-TEQ/m²-month) was highly negative-correlated with the ambient temperature ($r = -0.843$) for the agricultural rural area, possibly due to vapor pressure (temperature-dependent) affecting gas/particle PCDD/F partitioning. The higher particle-bound PCDD/Fs were formed at lower ambient temperature, and the particle-bound PCDD/Fs were the major scavenging mechanism for dry deposition. The results were similar with Bidleman's study (Bidleman *et al.*, 1986). Additionally, the PCDD/F concentration of wet deposition flux (ng I-TEQ/m²-month) was highly positively correlated with the monthly rainfall ($r = 0.826$–0.988) at the four sampling areas. Further investigation is warranted to see if heavy rain leads to higher TEQ in soils or sediments.

Seasonal Deposition of PCDD/Fs

The pattern of dry deposition flux, as shown in Fig. 6, was seasonally and geographically variated. The total deposition flux of PCDD/Fs was highest in summer (49.1–97.6 ng/m²-season), at the commercial, industrial, and coastal areas, however, the total deposition flux was highest in spring at the agricultural area. The dry/wet deposition flux at the industrial area was much higher than that at other areas, especially for I-TEQ wet deposition. The PCDD/F concentration in the atmosphere could be affected by the different seasons, because of variated in domestic heating, photolysis and chemical reactions (Lohmann and Jones, 1998). Hippelein *et al.*, (1996) found that PCDD/F concentrations were higher in the winter than those in the summer in rural Germany, due to the domestic heating. Since wet precipitation is more effective in scavenging particle PCDD/Fs, the fluxes of PCDD/Fs in rainy season are higher than those in dry seasons

Table 3. Correlated coefficient of dry, wet and total deposition fluxes of PCDD/Fs with the ambient meteorological parameters.

Correlated coefficient		PCDD/Fs (ng/m²-month)					PCDD/Fs I-TEQ (μg I-TEQ/m²-month)				
		Average temperature (°C)	Monthly rainfall (mm)	PM₁₀ concentration (μg/m³)	Wind speed (m/s)	Relative humidity (%)	Average temperature (°C)	Monthly rainfall (mm)	PM₁₀ concentration (μg/m³)	Wind speed (m/s)	Relative humidity (%)
Dry deposition	The commercial suburban area	−0.241	0.027	−0.276	−0.568†	0.315†	−0.465†	−0.208	−0.134	−0.579†	0.312†
	The industrial urban area	0.192	0.236	−0.425†	−0.500†	0.087	0.036	0.267	−0.336†	−0.473†	0.252
	The coastal rural area	0.326†	0.593†	−0.546†	0.414†	0.221	−0.144	0.200	−0.256	0.297	0.099
	The agricultural rural area	−0.689†	−0.586†	0.621†	0.409†	−0.610†	−0.843*	−0.464†	0.574†	0.684†	−0.511†
Wet deposition	The commercial suburban area	0.186	0.872*	−0.273	−0.507†	0.236	−0.311†	0.405†	0.078	−0.410†	0.352†
	The industrial urban area	0.020	0.920*	−0.050	−0.421†	0.250	−0.205	0.847*	0.197	−0.307†	0.386†
	The coastal rural area	0.177	0.988*	−0.231	0.225	0.563†	−0.136	0.809*	0.103	0.053	0.582†
	The agricultural rural area	0.662†	0.826*	−0.841*	−0.351†	0.892*	0.594†	0.862*	−0.805*	−0.219	0.856*
Total deposition	The commercial suburban area	−0.006	0.602†	−0.334†	−0.650†	0.330†	−0.471†	−0.021	−0.078	−0.595†	0.366†
	The industrial urban area	0.111	0.751*	−0.250	−0.544†	0.216	−0.059	0.564†	−0.177	−0.496†	0.358†
	The coastal rural area	0.246	0.944*	−0.364†	0.313†	0.497†	−0.172	0.544†	−0.137	0.244	0.358†
	The agricultural rural area	−0.126	0.092	−0.071	0.106	0.120	−0.639†	−0.145	0.280	0.621†	−0.196

Note: † means middle positive-correlated ($0.3 \leq$ the absolute value of correlated coefficient <0.7); * means highly positive-correlated (the absolute value of correlated coefficient ≥ 0.7).

(Kaupp and McLachlan, 1998). The rainy season occurs from May to October in Taiwan and so the wet deposition flux in summer is higher than that in other seasons.

The annual total deposition flux of PCDD/Fs in commercial, industrial, coastal and agricultural areas was 168, 310, 135 and 115 ng/m²-year, respectively. These values were smaller than the total PCDD/F fluxes measured in Indianapolis (540 ng/m²-year) and Bloomington (370 ng/m²-year) (Koester and Hites, 1992). The annual total deposition flux of PCDD/Fs was correlated with the PCDD/F emission concentrations which are deeply influenced by human activities. The annual release for PCDD/Fs in USA (US EPA, 2001) was 2,888 g I-TEQ/yr which was 30 times higher than it in Taiwan. This probably resulted in higher total deposition flux. Additionally, the annual dry deposition flux of PCDD/Fs in the ambient at the industrial area was 207 ng/m²-year, which was very similar with the data in Wu's study (Wu *et al.*, 2009) but much higher than 9 ng/m²-year in Jurado's study (Jurado *et al.*, 2004). The PCDD/F emission concentrations are deeply influenced by human activities and there is considerable scope for reducing them. In this study, dry deposition accounted for 65.6%, 66.8%, 67.7% and 77.8% of total PCDD/F deposition fluxes at commercial, industrial, coastal and agricultural areas, respectively. The monthly rainfall ranged from 0 mm to 499 mm at the four sampling areas in 2006, and the rainfall density has appeared to decrease in recent years. The results of this study imply 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. The impact of dry deposition on the PCDD/F distribution in soil or sediment will probably increase in the future, due to climate change and thus it should pay more attention on it.

The total deposition fluxes of PCDD/Fs were highly positive-correlated (0.602–0.944) with the rainfall at commercial, industrial and coastal areas. Wallenhorst (1997) has also reported a positive relationship between total deposition flux and rainfall. However, the total deposition fluxes of total I-TEQ were only positive-correlated (0.564 and 0.544) with the rainfall at industrial and coastal areas.

CONCLUSION

1. The mean PCDD/F-TEQ concentration for the commercial suburb, industrial urban, coastal rural and agricultural rural areas was 0.0389, 0.0958, 0.0329 and 0.0369 pg I-TEQ/Nm³, respectively, which were all much lower than the air quality standard in Japan (0.6 pg I-TEQ/Nm³).
2. The chlorine-substituted profiles of PCDD/Fs in the four sampling areas all show that OCDD, HpCDF and OCDF were the three dominant chlorine-substituted PCDD/Fs and account for more than 56% of total PCDD/F concentration.
3. The dry deposition of PCDD/Fs was mainly in the particle phase (99.1%–99.7%) at the four sampling areas in every season. The particle bound fraction of wet deposition ranged from 20.2%–99.9%,
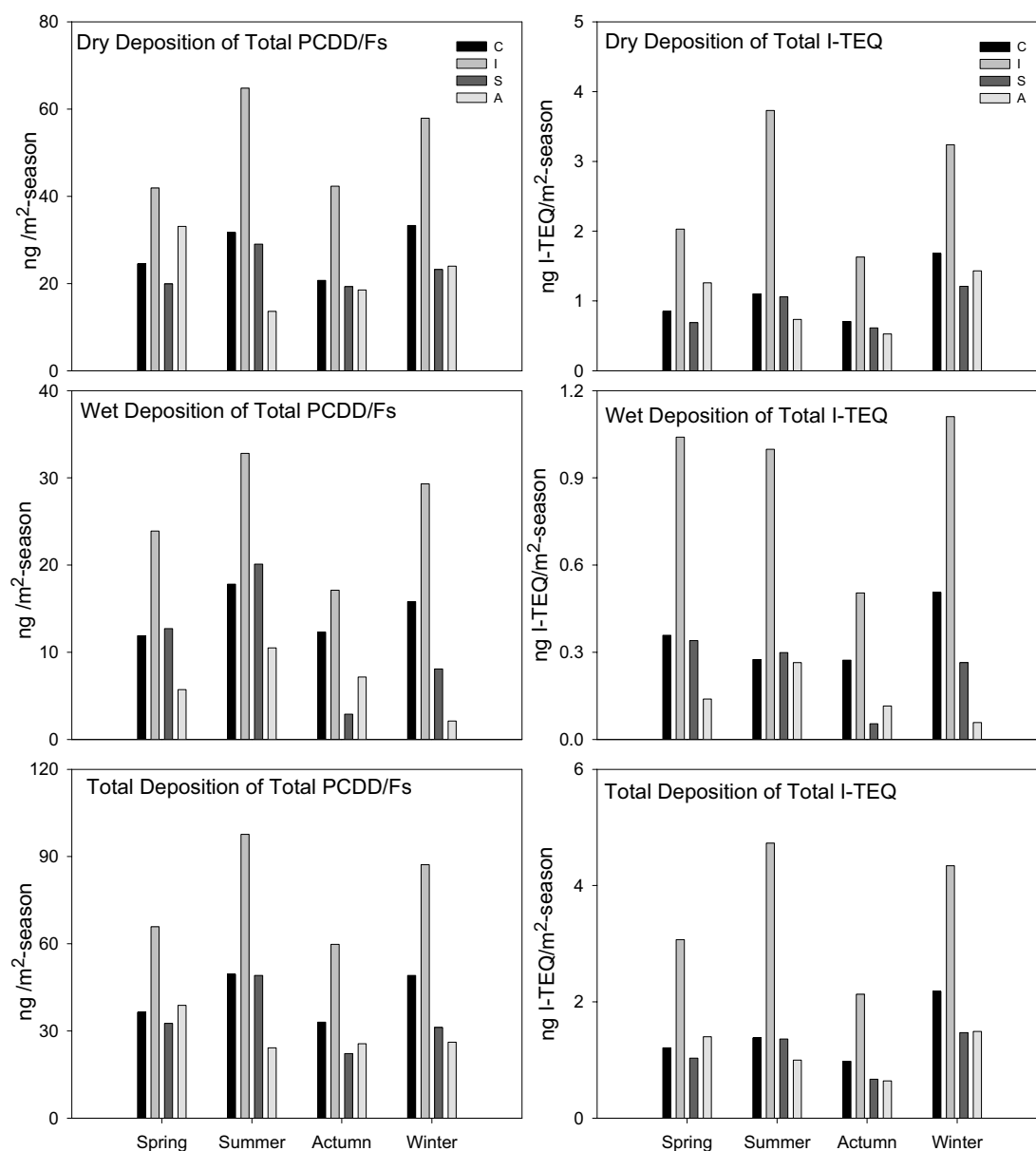
Fig. 6. The dry, wet and total deposition fluxes of PCDD/Fs and I-TEQ.
(C: the commercial suburban area, I: the industrial urban area, S: the coastal rural area, A: the agricultural rural area.)

29.8%–99.9%, 17.0%–99.9% and 22.7%–99.8% for the commercial, industrial, coastal and agriculture areas, respectively.

4. The dry deposition fluxes of the total PCDD/Fs in different seasons ranged from 7.67–18.2, 17.6–40.6, 6.69–11.6, 5.73–15.6 pg I-TEQ/m^2-d, with an average of 11.8, 29.1, 9.81 and 10.9 pg I-TEQ/m^2-d for the commercial, industrial, coastal, and agricultural areas, respectively.

5. Total PCDD/Fs in the rain's dissolved phase ranged from 19.7–40.3, 38.5–69.2, 17.0–27.4, 8.11–26.0 pg/L, with an average of 25.6, 47.8, 20.7 and 19.2 pg/L for the commercial, industrial, coastal, and agricultural areas, respectively.

6. The PCDD I-TEQ dry deposition flux (ng I-TEQ/m^2-month) was highly negative-correlated with the ambient

temperature ($r = -0.843$) for the agricultural rural area, possibly due to vapor pressure (temperature-dependent) affecting gas/particle PCDD/F partitioning.

7. The PCDD/F concentration of wet deposition flux (ng I-TEQ/m^2-month) was highly positively correlated with the monthly rainfall ($r = 0.826$–0.988) at the four sampling areas.

8. The annual dry deposition flux of PCDD/Fs in the ambient air at the industrial area was 207 ng/m^2-year, which was much higher than the 9 ng/m^2-year measured in the Atlantic Ocean.

9. The dominant mechanisms of dry and wet deposition were particle phase deposition and particle scavenging. Besides, PCDD/F emission concentrations are thus deeply influenced by human activities, and much scope remains for their reduction.

REFERENCES

Addink, R. and Altwicker, E.R. (2001). Formation of Polychlorinated Dibenzo-*p*-dioxins/dibenzofurans from Residual Carbon on Municipal Solid Waste Incinerator Fly Ash Using Na37Cl. *Chemosphere* 44: 1361–1367.

Bidleman, T.F., Billings, W.N. and Foreman, W.T. (1986). Vapor-particle Partitioning of Semivolatile Organic Compounds: Estimates from Field Collections. *Environ. Sci. Technol.* 20: 1038–1043.

Chang, M.B., Weng, Y.M., Lee, T.Y., Chen, Y.W., Chang, S.H. and Chi, K.H. (2003). Sampling and Analysis of Ambient Dioxins in Norther Taiwan. *Chemosphere* 51: 1103–1110.

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, C.M. (2004). The Emission Inventory of PCDD/PCDF in Taiwan. *Chemosphere* 54: 1413–1420.

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.

Eduljee, G.H. and Dyke, P. (1996) An Updated Inventory of Potential PCDD and PCDF Emission Sources in the UK. *Sci. Total Environ.* 177: 303–321.

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

EPA, Taiwan (2002a). The PCDD/F Concentrations in Ambient Atmosphere in Taiwan. EPA-91-1601-02-06.

EPA, Japan Ministry of the Environment (2002b). The Environmental Monitoring Report on the Persistent Organic Pollutants (POPs) in Japan.

EPA, Taiwan (2008). PCDD/F Emission from the Exhaust Gases of the Heavy-duty, Diesel Passenger and SUV Vehicles. EPA, EPA-96-FA-12-03-A115.Technical Report.

Fang, G.C., Wu, Y.S., Wen, C.C., Huang, S.H. and Rau, J.Y. (2006). Ambient Air Particulate Concentrations and Metallic Elements Principal Component Analysis at Taichung Harbor (TH) and WuChi Traffic (WT) near Taiwan Strait During 2004-2005. *J. Hazard. Mater.* 137: 314–323.

H.-Jones, P., de Gerlache, J., Marti, E., Mischer, G., Scherrer, M.C., Bontinck, W.J. and Niessen, H.J. (1993). The Global Exposure of Man to Dioxins: a Perspective on Industrial Waste Incineration. *Chemosphere* 26: 1491–1497.

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

Hiester, E., Böhm, R., Eynck, P., Gerlach, A., Mülder, W. and Ristow, H. (1993). Long Term Monitoring of PCDD, PCDF and PCB in Bulk Deposition Samples. *Organohalogen Compd.* 12: 147–150.

Hippelein, M., Kuapp, H., Dörr, G. and McLachlan, M.S. (1996). Testing of a Sampling System and Analytical Method for Determination of Semivolatile Organic Compounds in Ambient Air. *Chemosphere* 32: 1605–1616.

Hu, M.T., Chen, S.J., Lai, Y.C., Huang, K.L., Chang-Chien, G.P. and Tsai, J.H. (2009). Characteristics of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofurans from Joss Paper Burned in Taiwanese Temples. *Aerosol Air Qual. Res.* 9: 369–377.

Hung, H., Blanchard, P., Poole, G., Thibert, B., 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.

Kao, J.H., Chen, K.S., Chang-Chien, G.P. and Chou, I.C. (2006). Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Various Stationary Sources. *Aerosol Air Qual. Res.* 6: 170–179.

Kaupp, H. and McLachlan, M.S. (1998). Atmospheric Particle Size Distributions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) and Polycyclic Aromatic Hydrocarbons (PAHs) and their Implications for Wet and Dry Deposition. *Atmos. Environ.* 33: 85–95.

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

Kurz, R., Boeske, J. and Lahl, U. (1993). Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Soil, Deposition and Airborne Particulate Matter in the Vicinity of a Municipal Solid Waste Incinerator. *Organohalogen Compd.* 12: 151–154.

Lee, S.J., Ale, D., Chang, Y.S., Oh, J.E. and Shin, S.K. (2008). Seasonal and Particle Size-Dependent Variations in Gas/particle Partitioning of PCDD/Fs. *Environ. Pollut.* 153: 215–222.

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. and Chen, C.K. (2003). Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Incinerations of Both Medical and Municipal Solid Wastes. *Aerosol Air Qual. Res.* 3: 1–6.

Lohmann, R., Gioia, R., Eisenreich, S.J. and Jones, K.C. (2007). Assessing the Importance of Ab- and Adsorption

to the Gas-particle Partitioning of PCDD/Fs. *Atmos. Environ.* 41: 7767–7777.

Lohmann, R., Green, N.J.L. and Jones, K.C. (1999). Detailed Studies of the Factors Controlling Atmospheric PCDD/F Concentrations. *Environ. Sci. Technol.* 33: 4440–4447.

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.

Ogura, I., Masunaga, S. and Nakanishi, J. (2001). Congener-specific Characterization of PCDDs/PCDFs in Atmospheric Deposition: Comparison of Profiles among Deposition, Source, and Environmental Sink. *Chemosphere* 45: 173–183.

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.

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. 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.

Pankow, J.F. (1994). An Adsorption Model of Gas/Particle Partitioning of Organic Compounds in the Atmosphere. *Atmos. Environ.* 28: 185–188.

Schröder, J., Welsch-Pausch, K. and McLachlan, M.S. (1997). Measurement of Atmospheric Deposition of Polychlorinated Dibenzo-*p*-dioxins (PCDDs) and Dibenzofurans (PCDFs) to a Soil. *Atmos. Environ.* 31: 2983–2989.

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

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

Shih, S.I., Wang, Y.F., Chang, J.E., Jang, J.S., Kuo, F.L., Wang, L.C. and Chang-Chien, G.P. (2006b). Comparisons of Levels of Polychlorinated Dibenzo-*p*-

dioxins/dibenzofurans in the Surrounding Environment and Workplace of two Municipal Solid Waste Incinerators. *J. Hazard. Mater.* 137: 1817–1830.

Shih, T.S., Shih, M., Lee, W.J., Huang, S.L., Wang, L.C., Chen, Y.C. and Tsai, P.J. (2009). Particle Size Distributions and Health-related Exposures of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) of Sinter Plant Workers. *Chemosphere* 74: 1463–1470.

Tainan EPB of Taiwan, (2006). http://www.tnepb.gov.tw/.

US EPA (2001). Database of Sources of Environmental Releases of Dioxin like Compounds in the United States, EPA/600/C-01/012.

Wallenhorst, T., Krauβ, P. and Hagenmaier, H. (1997). PCDD/F in Ambient Air and Deposition in Baden-württemberg, Germany. *Chemosphere* 34: 1369–1378.

Wang, I.C. and Lee, W.J. (2010). Polychlorinated Dibenzo-*p*-dioxin, Polychlorinated Dibenzofurans and Polychlorinated Biphenyls in Farmed Fish, Water, Sediment, and Feed. *J. Environ. Sci. Health., Part A.* 45: 201–210.

Wang, L.C., Lee, W.J., Tsai, P.J., Lee, W.S. and Chang-Chien, G.P. (2003). Emission of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Stack Flue Gases of Sinter Plants. *Chemosphere* 50: 1123–1129.

Wang, Y.F., Chao, H.R., Wu, C.H., Wang, L.C., Chang-Chien, G.P., Yang, H.H., Lin, D.Y. and Tsou, T.C. (2009). Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from a Heavy Oil-fueled Power Plant in Northern Taiwan. *J. Hazard. Mater.* 163: 266–272.

Wu, Y.L., Lin, L.F., Hsieh, L.T., Wang, L.C., 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.

Zhang, B., Meng, F., Shi, C., Yang, F., Wen, D., Aronsson, J., Gbor, P.K. and Sloan, J.J. (2009). Modeling the Atmospheric Transport and Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in North America. *Atmos. Environ.* 43: 2204–2212.