



Atmospheric Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans at Coastal and High Mountain Areas in Taiwan

Chandra Suryani R.¹, Wen-Jhy Lee^{1*}, Endah Mutiara M.P.^{2†}, John Kennedy Mwangi¹, Lin-Chi Wang³, Neng-Huei Lin⁴, Guo-Ping Chang-Chien⁵

¹ Department of Environmental Engineering, National Cheng Kung University, No. 1, University Road, Tainan 70101, Taiwan

² Department of chemistry, Sepuluh November Institute of Technology, Surabaya 60111, Indonesia

³ Department of Civil Engineering and Geomatics, Cheng Shiu University, 840, Chengching Rd., Kaohsiung 833, Taiwan

⁴ Department of Atmospheric Sciences, National Central University, Chung-Li, Taiwan

⁵ Super Micro Mass Research & Technology Center, Cheng Shiu University, 840, Chengching Rd., Niasong Dist., Kaohsiung 833, Taiwan

ABSTRACT

Atmospheric deposition is of great importance for the sink of air pollutants to the environment, either from local sources as well as coming from long range transport. To further understand the combined impact of both long-range transport from South East Asia and local emission sources of pollutants, the characteristics of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the ambient air of two background sites in Taiwan namely, Hengchun (coastal area) and Lulin (high mountain area), were simulated by regression of PM₁₀ versus total-PCDD/F mass concentration, modeling of gas-particle partitioning, and simulations of both dry and wet deposition. The simulated PCDD/F concentrations at Hengchun, ranged between 0.0039–0.0106 and 0.0054–0.0138 pg WHO₂₀₀₅-TEQ m⁻³ for 2012 and 2013, respectively. For Lulin site, the PCDD/F concentrations ranged between 0.0016–0.0029 and 0.0016–0.0032 pg WHO₂₀₀₅-TEQ m⁻³ for 2012 and 2013, respectively. The WHO₂₀₀₅-TEQ ratios of PCDDs to PCDFs at both sites were less than unity, indicating that PCDF dominated the total toxicity. The results show that higher chlorinated PCDD/Fs primarily present in particulate phase for all seasons at both sampling sites especially in winter. Average dry deposition fluxes at Hengchun (57.1 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) were approximately 5.1 times higher than those at Lulin (11.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). The annual average dry deposition velocities were estimated to be 0.28 and 0.22 cm/s for Hengchun and Lulin, respectively. For the whole period (2012–2013), the mean monthly wet deposition fluxes at Hengchun (averaged 11.7 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) were 1.44 times higher than that at Lulin (averaged 8.11 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). The calculated annual average total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the rain were 0.064 and 0.027 pg WHO₂₀₀₅-TEQ L⁻¹ for Hengchun and Lulin, respectively. The estimated annual average scavenging ratios of total-PCDD/Fs-WHO₂₀₀₅-TEQ were 8015 and 13450 for Hengchun and Lulin, respectively. Similarly, for the entire study period (2012–2013), the average annual total (dry + wet) deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ in the terms of pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ at Hengchun (824.9) was 3.5 times higher than those in Lulin (232.0). On the basis of total PCDD/Fs-WHO₂₀₀₅-TEQ, the mean fraction contributed by dry deposition at Hengchun were 78.2% and 78.9% in 2012 and 2013, respectively, while at Lulin the average fraction contributed by dry deposition at Lulin were 56.5% and 69.1% in 2012 and 2013, respectively. Higher chlorinated congeners OCDD, OCDF, 1,2,3,4,6,7,8-HpCDF and 1,2,3,4,6,7,8-HpCDD dominated in terms of mass fractions of total PCDD/F deposition fluxes for all seasons at both sites. However on the basis of total PCDD/Fs-WHO₂₀₀₅-TEQ deposition fluxes, the most dominant congeners were 2,3,4,7,8-PeCDF, 1,2,3,7,8-PeCDD and 2,3,7,8-TeCDD during the whole sampling period at both sites. The results of this study provide useful information for both environmental impact assessment and control strategies of persistent organic compounds (POPs).

Keywords: PCDD/Fs; POPs; Dry deposition; Wet deposition; Gas-particle partitioning; Atmosphere; Concentration.

* Corresponding author.
Tel.: +886-209-3155; Fax: +886-275-2790
E-mail address: wjlee@mail.ncku.edu.tw

† Corresponding author.
Tel.: +62816505743; Fax: +6231-5981008
E-mail address: emmutri@gmail.com

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) which belong to the dioxin family (Schechter *et al.*, 2006) continue to be accorded increasing concern due to their toxic and ubiquitous and persistence nature in the environment (Lohmann and Jones, 1998; Oh *et al.*, 2001; Chiu *et al.*, 2011; Huang *et al.*, 2011a, b).

The 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) is arguably one of the most toxic chemicals (Van den Berg *et al.*, 1998; Mitrou *et al.*, 2001;) and the rest of dioxin like chemicals including other of PCDD/F congeners and polychlorinated biphenyls (PCBs), which occur as mixtures in commercial and environmental samples (Lee *et al.*, 2003) are assigned toxic equivalent factors (TEFs) relative to the 2,3,7,8-TCDD (Lohmann and Jones, 1998) according to the WHO₁₉₉₈ scheme (Van den Berg *et al.*, 1998), which was revised later to the WHO₂₀₀₅ scheme (Van den Berg *et al.*, 2006). These TEFs are multiplied with corresponding congener concentration to give toxic equivalents.

Exposure to PCDD/Fs occurs mainly through accumulation up the food chain considering that PCDD/Fs are highly bioaccumulative (Correa *et al.*, 2006). Additionally, high exposure incidents such as industrial accidents and spillages are also responsible for human exposure (Kulkarni *et al.*, 2008). These compounds are classified as endocrine disruptors and act as AhR agonists that bind to AhR (Schechter *et al.*, 2006), and cause AhR-mediated biochemical and toxic responses in animals and occur as mixtures of several congeners in the environmental matrices. From occupational and epidemiological studies, the potential health effects of PCDD/Fs have been shown to include mutagenic and carcinogenic potentials, genetic and endocrine disruptions, reproductive and developmental abnormalities (Schechter *et al.*, 2006; Kulkarni *et al.*, 2008; Huang *et al.*, 2011a).

Generally, as with most of the emerging persistent organic pollutants (POPs) in the environment, the PCDD/Fs are mostly emitted from anthropogenic activities (Moon *et al.*, 2005; Kulkarni *et al.*, 2008; Wang *et al.*, 2010; Huang *et al.*, 2011a). Combustion processes (Correa *et al.*, 2006; Shih *et al.*, 2006; Chiu *et al.*, 2011) involving chlorine containing organic matter (Wang *et al.*, 2009) such as municipal solid waste incineration (Lee *et al.*, 2003), laboratory waste incinerators (Liao *et al.*, 2014; Wu *et al.*, 2014), hazardous waste incinerators (Wang *et al.*, 2014), iron ore sintering plants (Fang *et al.*, 2011), steel and cement kilns (Van Thuong *et al.*, 2014), landfill fires (Moussiopoulos *et al.*, 2006), vehicle tailpipe exhausts (Chuang *et al.*, 2010), boilers (Chen *et al.*, 2011), fast food restaurants (Lin *et al.*, 2011), coal fire-power plants (Lin *et al.*, 2010b), bio-mass open burning (Shih *et al.*, 2008; Chang *et al.*, 2013; Chang *et al.*, 2014) and dust storms (Chi *et al.*, 2014) have been pointed out to be among the leading primary sources of PCDD/Fs and other dioxin like compounds. The emission of PCDD/Fs from the mentioned processes can be attributed to either their presence the feedstock, or be formed from chlorine containing precursors compounds as well as be formed through the *de novo* mechanism at certain temperature window (Wang *et al.*, 2010; Fujimori *et al.*, 2014).

PCDD/Fs are also classified as semi-volatile organic compounds (SVOCs) and when they are released into the atmosphere, they are ultimately subjected to atmospheric dispersion and deposition processes (Halsall *et al.*, 1997; Shih *et al.*, 2006) to the environmental compartments or sinks such as water bodies, soil and vegetation (Halsall *et al.*, 1997; Ren *et al.*, 2007). By virtue of being SVOCs, PCDD/Fs will partition between gas and particulate phases in the atmosphere, which governs their subsequent fate (Kaupp *et al.*, 1994; Lohmann *et al.*, 2000). Removal of PCDD/Fs in the atmosphere can occur through both dry and wet deposition processes (Fang *et al.*, 2011; Mi *et al.*, 2012) as well as destruction and degradation of gaseous PCDD/Fs via photolysis and OH radical induced chemical reactions (Lohmann *et al.*, 2000; Moon *et al.*, 2005). This study is focus mainly on atmospheric deposition including dry and wet deposition processes.

Dry deposition is responsible primarily for removal of PCDD/Fs bound to coarse particles (Moon *et al.*, 2005) as well as small fractions bound to fine particles and gaseous phase from the atmosphere during non-precipitation days (Giorgi, 1988). Mechanisms such as turbulent diffusion, sedimentation, Brownian motion, interception, inertial forces, thermophoresis, electrical migration and diffusiphoresis are ways in which dry deposition occurs. The governing factors of dry deposition process include wind, temperature, humidity, size and shape of particles as well as the surface characteristics of both particles and sink media (Giorgi, 1988; Oh *et al.*, 2002; Moon *et al.*, 2005; Wang *et al.*, 2010; Fang *et al.*, 2011; Amodio *et al.*, 2014). According to (Shih *et al.*, 2006) dry deposition process accounts for approximately 15% of the total deposition flux of the higher chlorinated homologues and half of the flux of the most volatile homologues.

Three methods of determining dry deposition fluxes include measuring ambient air concentrations and the associated deposition velocities, using mass balance approach as well as employing surrogate surfaces to directly measure the fluxes (Tasdemir *et al.*, 2004). Artifacts affecting direct measurements include blow off from particles collected on the surfaces, uncertainties of using artificial surfaces in place of natural surfaces such as water, soil and vegetation with different surface characteristics, the appropriate time for exposure, which echoes that in the environment (Halsall *et al.*, 1997; Wu *et al.*, 2009). This presents drawbacks into having a universally accepted method altogether (Tasdemir *et al.*, 2004).

Monitoring dry deposition characteristics of PCDD/Fs in different geographic sites and areas is of great importance in understanding the fate and distribution of these pollutants in the environment as well as the estimating potential exposure to the populations living in these locations. Previously, various studies have been done to elucidate PCDD/F dry deposition scenarios in an industrial coastal city in South Korea (Fang *et al.*, 2011), Manchester and Cardiff cities in the UK (Halsall *et al.*, 1997), in the vicinity of municipal solid waste incinerators in South Taiwan (Wu *et al.*, 2009), commercial and residential districts of Guanzhou city in China (Ren *et al.*, 2007), major water shed in Northern

Taiwan (Lin *et al.*, 2010a) as well as the Atlantic Ocean (Jurado *et al.*, 2004).

Wet deposition is also the major removal mechanism of particles, especially the fine particles (Moon *et al.*, 2005) as well as gaseous phase POPs (Melymuk *et al.*, 2011), in atmosphere via precipitation in form of rainfall and cloud droplets, or snow in the cold weather condition (Lohmann and Jones, 1998) and is majorly responsible for the presence of higher chlorinated homologues in the environmental sinks (Shih *et al.*, 2006; Lin *et al.*, 2010a; Wang *et al.*, 2010). Precipitation scavenging accounts for the majority of removing these SVOCs from atmosphere by wet deposition (Huang *et al.*, 2011a, b). The wet deposition flux of PCDD/Fs can be obtained by combining both vapor dissolution into rain and the removal of suspended particulates by precipitation.

Studies focusing on atmospheric deposition of PCDD/Fs in mountain regions are scarce, therefore, this study aims to address the research gap by providing an insight to the characteristics of PCDD/Fs deposition fluxes in a background mountain area and compare it with a coastal region of Taiwan. This will serve as useful information for both environmental impact assessment and control strategies of persistent organic compounds (POPs).

The objectives of this study were to investigate ambient air concentrations, gas-particle partitioning in addition to dry, wet and total deposition fluxes of atmospheric PCDD/Fs at coastal and high mountain area in Taiwan via modeling and simulation. Two sampling sites, in Hengchun, Southern Taiwan and in Mount Lulin, Central Taiwan, were selected. Collected samples were analyzed for specific 17 PCDD/Fs congeners to establish atmospheric concentration level, and then gaseous and particulate concentration could be calculated. The atmospheric deposition fluxes were simulated by model calculations. Results of this study will provide long-term data of PCDD/Fs in Taiwan.

METHODS

Sampling Sites

Samples used in this experiment were obtained from two astronomical observatory sites in Taiwan, which include the Hengchun coastal area (22°06'N 120°70'W, 6 m elevation above sea level) in southern tip of Taiwan and Lulin Atmospheric Background Station (23°47'N 120°87'E, 2862 m above sea level) in central Taiwan (Fig. 1).

The sampling sites were chosen, since there are few studies focusing on the difference in atmospheric deposition patterns between coastal and high mountain areas of Taiwan. Sampling period was from February 2012 to April 2013.

Meteorological Conditions and PM Concentration during the Sampling Periods

The pollutant transmissions and deposition in the atmosphere are affected by the meteorological conditions, such as wind speed, rainfall intensity, PM₁₀ concentrations and the atmospheric stability. In this study, the pertinent meteorological information and PM₁₀ concentrations for Hengchun and Lulin, respectively, for the year 2012 and 2013 were obtained from the nearby air quality monitoring



Fig. 1. Sampling sites at the Hengchun coastal area (H) and at Lulin mountain background station (L).

stations. The meteorological conditions prevailing in the sampling areas over the whole sampling period of 2012–2013 are summarized in the Tables 1–4.

During 2012, the monthly average atmospheric temperatures at Hengchun were between 21.1°C and 29.1°C and averaged 25.6°C, while those at Lulin were between 4.2°C and 12.9°C and averaged 9.4°C. In 2013, the atmospheric temperatures at Hengchun were between 21.0°C and 28.6°C and averaged 25.4°C, while those at Lulin were between 5.5°C and 14.3°C and averaged 10.9°C. In year 2012 and 2013, the maximum monthly average temperature occurred in July and June at both sites. The monthly average atmospheric temperatures at Hengchun are much higher than that in Lulin, since Lulin is located in a high altitude area (2862 m high) with cooler temperature than coastal area like Hengchun (6 m high). Additionally, Taiwan lies in the sub-tropical regions especially Hengchun, which is in the southern part, thus as reported by Shih *et al.* (2006) and Wang *et al.* (2010) and the temperature values reported herein are similar to those reported by Huang *et al.* (2011) for a rural area in Taiwan.

The Taiwan air quality monitoring network collects hourly data of PM₁₀ using the Weddington beta gauge recommended by the US EPA (Chang and Tsai, 2003; Tsai and Cheng, 1996). In 2012, the monthly average PM₁₀ concentrations at Hengchun were between 15.0 and 34.0 µg m⁻³ and averaged 24.3 µg m⁻³, while those at Lulin were between 5.0 and 18.0 µg m⁻³ and averaged 8.5 µg m⁻³. While in 2013, the monthly average PM₁₀ concentrations at Hengchun were between 19.0 and 43.0 µg m⁻³ and averaged 28.8 µg m⁻³,

while those at Lulin were between 5.4 and 20.2 $\mu\text{g m}^{-3}$ and averaged 9.7 $\mu\text{g m}^{-3}$. Overall, the monthly average highest PM_{10} concentrations at Hengchun occurred in the month of

October (averaged at 38.5 $\mu\text{g m}^{-3}$) for both years. However, at Lulin, the highest monthly average PM_{10} concentrations occurred in March (averaged 19.1 $\mu\text{g m}^{-3}$) in both years.

Table 1. Meteorological data for gas-particle partition simulation at Hengchun.

Year	Month	Temperature ($^{\circ}\text{C}$)	PM_{10} ($\mu\text{g m}^{-3}$)	TSP ($\mu\text{g m}^{-3}$)	Wind speed (m s^{-1})
2012	Jan.	21.0	21.0	26.4	11.9
	Feb.	21.3	15.0	18.6	10.9
	March	23.9	26.0	32.2	12.1
	April	25.8	26.0	32.2	10.2
	May	27.4	20.0	24.8	8.9
	June	28.2	24.0	29.8	8.9
	July	28.6	17.0	21.1	7.8
	Aug.	28.0	33.0	40.9	20.5
	Sep.	28.1	28.0	34.7	11.1
	Oct.	26.0	34.0	42.2	10.3
	Nov.	24.5	26.0	32.2	10.2
	Dec.	22.2	22.0	27.3	11.6
2013	Jan	21.4	24.0	29.8	13.8
	Feb	23.5	24.0	29.8	12.0
	Mar	24.3	30.0	37.2	11.1
	Apr	25.0	32.0	39.7	10.6
	May	27.1	29.0	35.9	8.6
	Jun	29.1	26.0	32.2	7.5
	Jul	28.6	19.0	23.6	10.9
	Aug	28.2	25.0	31.0	10.6
	Sep	28.0	32.0	39.7	19.1
	Oct	26.2	43.0	53.3	10.5
	Nov	24.2	34.0	42.2	11.5
	Dec	21.1	28.0	34.7	13.4

Table 2. Meteorological data for gas-particle partition simulation at Lulin.

Year	Month	Temperature($^{\circ}\text{C}$)	PM_{10} ($\mu\text{g m}^{-3}$)	TSP ($\mu\text{g m}^{-3}$)	Wind Speed (m s^{-1})
2012	Jan	4.2	6.0	6.9	5.0
	Feb	5.4	9.0	11.4	5.3
	Mar	7.9	18.0	22.6	4.5
	Apr	7.9	13.0	15.5	6.1
	May	10.6	9.0	11.5	3.6
	Jun	12.3	5.0	6.5	4.3
	Jul	13.0	6.0	7.2	2.6
	Aug	11.8	6.0	7.5	3.5
	Sep	11.6	8.0	9.3	2.3
	Oct	11.0	10.0	13.0	2.3
	Nov	9.5	6.0	7.0	4.4
	Dec	7.1	6.0	7.4	4.6
2013	Jan	5.5	9.0	10.8	5.3
	Feb	9.7	11.0	14.2	4.2
	Mar	9.1	20.0	25.1	4.7
	Apr	10.1	15.0	18.3	4.8
	May	12.0	8.0	10.1	4.4
	Jun	14.3	8.0	9.8	3.4
	Jul	14.1	8.0	9.7	2.8
	Aug	14.3	7.0	8.5	3.2
	Sep	13.9	10.0	12.9	2.0
	Oct	12.0	10.0	11.9	2.3
	Nov	9.5	6.0	7.0	3.5
	Dec	5.8	5.0	6.7	6.4

Table 3. Prevailing rainfall condition during simulation period at Hengchun.

Year	Month	Rainfall Amount (mm)	Days with Rainfall	Days without Rainfall	Relative Humidity (%)
2012	Jan.	1.6	8	23	74
	Feb.	109	17	11	78
	March	6.2	9	22	75
	April	47.8	11	19	77
	May	300.2	17	14	78
	June	769.4	23	7	83
	July	426.8	18	13	81
	Aug.	723.4	25	6	86
	Sep.	59.4	19	11	76
	Oct.	16	14	17	69
	Nov.	45.2	9	21	74
	Dec.	68.4	18	13	73
2013	Jan.	78	13	18	70
	Feb.	67.6	17	11	73
	March	8.4	7	24	70
	April	79.2	7	23	74
	May	130.8	19	12	81
	June	246.8	16	14	78
	July	251.6	27	4	77
	Aug.	534.8	20	11	80
	Sep.	266.2	20	10	73
	Oct.	39	13	18	68
	Nov.	69	9	21	73
	Dec.	89.2	14	17	69

Table 4. Prevailing rainfall condition during simulation period at Lulin.

	Month	Rainfall Amount (mm)	Days with Rainfall	Days without Rainfall	Relative Humidity (%)
2012	Jan.	106	9	22	71
	Feb.	167	6	22	85
	March	36	9	22	70
	April	288	22	8	96
	May	616	25	6	89
	June	886	17	13	93
	July	306	25	6	82
	Aug.	712	26	5	92
	Sep.	164	19	11	85
	Oct.	16	8	23	60
	Nov.	258	15	15	83
	Dec.	182	12	19	80
2013	Jan.	36	13	18	75
	Feb.	4.4	17	11	65
	March	44	7	24	66
	April	428	7	23	88
	May	732	19	12	93
	June	139	16	14	82
	July	424	27	4	82
	Aug.	1057	20	11	89
	Sep.	373	20	10	84
	Oct.	23	13	18	70
	Nov.	108	9	21	73
	Dec.	136	14	17	80

The annual average PM₁₀ concentration at Hengchun (24.9 $\mu\text{g m}^{-3}$) is approximately 2.7 times higher than Lulin (9.1 $\mu\text{g m}^{-3}$). The lowest monthly average PM₁₀ concentration

occurred in February 2012 (15 $\mu\text{g m}^{-3}$) and July 2013 (19 $\mu\text{g m}^{-3}$) at Hengchun and in June 2012 (5.2 $\mu\text{g m}^{-3}$) and December 2013 (5.4 $\mu\text{g m}^{-3}$) at Lulin, respectively.

For the year 2012, the wind speeds at Hengchun were between 1 and 6 m s⁻¹ and averaged 4 m s⁻¹, while those at Lulin were between 2 and 6 m s⁻¹ and averaged 4 m s⁻¹. As for the year 2013, the wind speeds at Hengchun were between 3 and 8 m s⁻¹ and averaged 6 m s⁻¹, while those at Lulin were between 2 and 6 m s⁻¹ and averaged 4 m s⁻¹. It is clear there was no significant difference in wind speed at both sampling locations.

Tables 2 and 3 illustrate the prevailing rainfall conditions during the simulation period. At both sites August had the highest amount of rainfall as well as highest days with rainfall while March was the driest month for the whole simulation period of 2012–2013. During the year 2012, the monthly rainfall intensities at Hengchun were between 1.6 mm and 769.4 mm with an annual total of 2573.4 mm, while those at Lulin were between 16 mm and 886.2 mm monthly and with an annual total of 3735.8 mm. On the other hand, during 2013, the monthly rainfall intensities at Hengchun were between 8.4 mm and 534.8 mm and with an annual total of 1860.6 mm, while those at Lulin were between 4.4 mm and 1056.6 mm monthly and with an annual total of 3503.2 mm. Overall, the highest rainfall intensities at Hengchun were 769.4 mm in June 2012 and 534.8 mm in August 2013. However, at Lulin, the highest monthly rainfall intensities were 886.2 mm in June 2012 and 1056.6 mm in August 2013, respectively. Generally, Lulin recorded higher total rainfall intensity than Hengchun due to the location in high mountain area.

Sampling Procedures and Analysis

Samples were collected, for a period of five days, using PS-1 sampler (Graseby Andersen, GA) according to the T09A method which was referred by United States Environmental Protection Agency (US-EPA). The PS-1 sampler was used to collect both gas and particle-phase compounds. Particle-phase compounds were collected by quartz fiber filter, whereas the gas-phase was collected using polyurethane foam. To evaluate contamination during sampling, one field blank was taken during the individual sampling events. Field blanks were loaded into the sampling system, but no air was drawn through them. They experienced the same handling, storage, and analysis procedures as the actual samples.

All the chemical analyses in this study were carried out in an accredited laboratory, Super Micro Mass Research and Technology Centre, in Cheng Shiu University which is certified by Taiwan EPA to analyze PCDD/Fs in Taiwan. High-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) (Hewlett Packard 6970 Series, CA, USA) was used for PCDD/F analysis. Detailed analytical procedures and instrumental parameters of PCDD/Fs given in the previous work (Wang *et al.*, 2010).

Gas-Particle Partitioning

Gaseous and particulate concentrations of PCDD/Fs were evaluated by the gas-particle partitioning multiplying the total concentrations of PCDD/Fs. The gas-particle partitioning was simulated by an equation, proposed by several researchers, that successfully describes gas-particle partitioning constant (Yamasaki *et al.*, 1982; Pankow, 1987;

Pankow and Bidleman, 1991, 1992):

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

K_p : gas-particle partitioning constant (m³ μg⁻¹),

TSP: concentration of total suspended particulate material (μg m⁻³),

F: particle phase concentration of PCDD/Fs (pg m⁻³),

A: gaseous phase concentration of PCDD/Fs (pg m⁻³).

Plotting log K_p against the logarithm of the subcooled liquid vapor pressure (P_L^o), gives:

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

P_L^o : subcooled liquid vapor pressure (Torr),

m_r : slope of a plot of log K_p vs log P_L^o ,

b_r : y-intercept in a plot of log K_p vs log P_L^o (Lohmann and Jones, 1998).

Eitzer and Hites (1989) have correlated P_L^o of PCDD/Fs with gas chromatographic retention indexes (GC-RI) on a non-polar (DB-5) GC-column using p,p'-DDT as a reference standard (Eitzer and Hites, 1989), and the correlation has been re-developed by (Hung *et al.*, 2002):

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

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

T: ambient temperature (K) (Hung *et al.*, 2002).

A complete datasets on the gas-particle partitioning of PCDD/Fs in Taiwan have been reported by (Chao *et al.*, 2004). From their data, parameters for Eq. (1) were determined as $m_r = -1.29$ and $b_r = -7.2$ with $R^2 = 0.94$. In this study, those parameters are also used for estimating the partitioning constant (K_p) of PCDD/Fs.

Dry Deposition Fluxes of PCDD/Fs

The dry deposition fluxes 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 (Wang *et al.*, 2010),

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

$$F_T = C_T \times V_{d,T}, \quad (6)$$

$$F_g = C_g \times V_{d,g} \text{ and } F_p = C_p \times V_{d,p} \quad (7)$$

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.

Dry deposition of particle-phase PCDD/Fs occurs mainly via the gravitational settling. The dry deposition velocities of particle-phase PCDD/Fs ($V_{d,p}$) can be simulated by Eqs. (5) and (7).

Theory of Scavenging Ratios

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

$$S_g = RT/H \quad (8)$$

S_g : the gas scavenging ratio of PCDD/Fs (dimensionless),

R : the universal gas constant ($82.06 \times 10^{-6} \text{ m}^3 \text{ atm mol}^{-1} \text{ K}^{-1}$),

T : ambient temperature (K),

H : Henry constant ($\text{m}^3 \text{ atm mol}^{-1}$).

$$S_g = C_{rain,dis}/C_g \quad (9)$$

$C_{rain,dis}$: 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:

$$S_p = C_{rain,particle}/C_p \quad (10)$$

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:

$$S_{tot} = S_g(1 - \Phi) + S_p \times \Phi \quad (11)$$

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 (1989) work.

Determination of Wet Deposition Fluxes of PCDD/Fs

Wet deposition is the removal of particles in atmosphere by precipitation (rainfall and cloud droplets) and precipitation scavenging accounts for the majority of removing SVOCs from atmosphere by wet deposition (Huang *et al.*, 2011b). Wet deposition flux of SVOCs is a combination of both vapor dissolution into rain and removal of suspended particulates by

precipitation (Bidleman, 1988; Koester and Hites, 1992).

The wet deposition flux of SVOCs can be evaluated as follows:

$$F_{w,T} = F_{w,dis} + F_{w,p} \quad (12)$$

$$F_{w,dis} = C_{rain,dis} \times Rainfall \quad (13)$$

$$F_{w,p} = C_{rain,particle} \times Rainfall \quad (14)$$

$F_{w,T}$: the wet deposition flux of SVOCs from both vapor dissolution into rain and removal of suspended particulates by precipitation,

$F_{w,dis}$: the wet deposition flux contributed by vapor dissolution into rain,

$F_{w,p}$: the wet deposition flux contributed by removal of suspended particulates by precipitation,

Rainfall: monthly rainfall (m).

RESULTS AND DISCUSSION

Simulated Ambient Air PCDD/F Concentrations

Fig. 2 shows the regression line obtained for five total PCDD/F mass concentrations values measured in this study versus the corresponding PM_{10} data. There is a strong correlation ($R = 0.99$) between the PM_{10} values and the total-PCDD/F mass concentrations. From the results in the study of Huang *et al.* (2011), it was also shown that there was a good correlation ($R = 0.94$) between PM_{10} concentrations and total-PCDD/F mass concentration. Additionally, in the same study, the PM_{10} concentration and particle phase PCDD/Fs contributed and controlled more than 90% of the atmospheric PCDD/F depositions on a mass basis. Therefore, based on these inferences, this study simulated concentrations data for PCDD/F by regression analysis of PM_{10} versus total PCDD/F mass concentration. Monthly simulated concentrations of total-PCDD/Fs-WHO₂₀₀₅-TEQ are presented in Fig. 3.

The total-PCDD/Fs-WHO₂₀₀₅-TEQ concentration during 2012, at Hengchun, ranged between 0.0039 and 0.0106 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$, while those during 2013 ranged between 0.0054 and 0.0138 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$. The annual mean total-PCDD/Fs-WHO₂₀₀₅-TEQ-concentrations at Hengchun were 0.0072 and 0.0088 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$ in 2012 and 2013, respectively.

However, during 2012, at Lulin, the total-PCDD/Fs-WHO₂₀₀₅-TEQ concentration ranged between 0.0016 and 0.0029 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$, while those during 2013 ranged between 0.0016 and 0.0032 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$. The annual mean total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in atmosphere at Lulin were 0.0019 and 0.0021 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$ in 2012 and 2013, respectively.

The highest monthly average total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations at Hengchun occurred in October (averaged 0.0122 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$) for both years, however, the lowest monthly average concentrations occurred in February (0.0039 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$) 2012 and July (0.0054 $\text{pg WHO}_{2005}\text{-TEQ m}^{-3}$) 2013. However, at Lulin, the highest monthly average total-PCDD/Fs-WHO₂₀₀₅-TEQ

concentrations occurred in March (averaged 0.0031 pg WHO₂₀₀₅-TEQ m⁻³) in both years, while the lowest monthly average concentrations occurred in June (0.0106 pg WHO₂₀₀₅-

TEQ m⁻³) 2012 and December (0.0016 pg WHO₂₀₀₅-TEQ m⁻³) 2013. The mean total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in Lulin are approximately 67.8% lower than that in

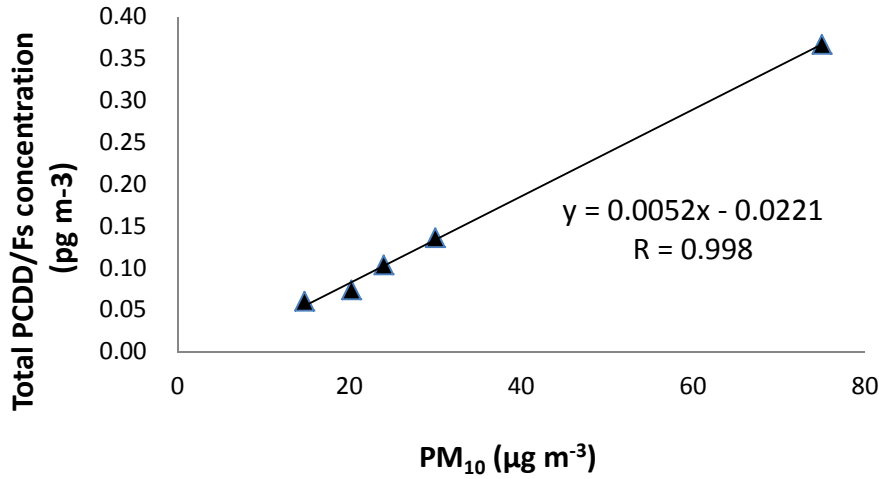


Fig. 2. Regression of PM₁₀ (µg m⁻³) vs. total PCDD/F mass concentration (pg m⁻³).

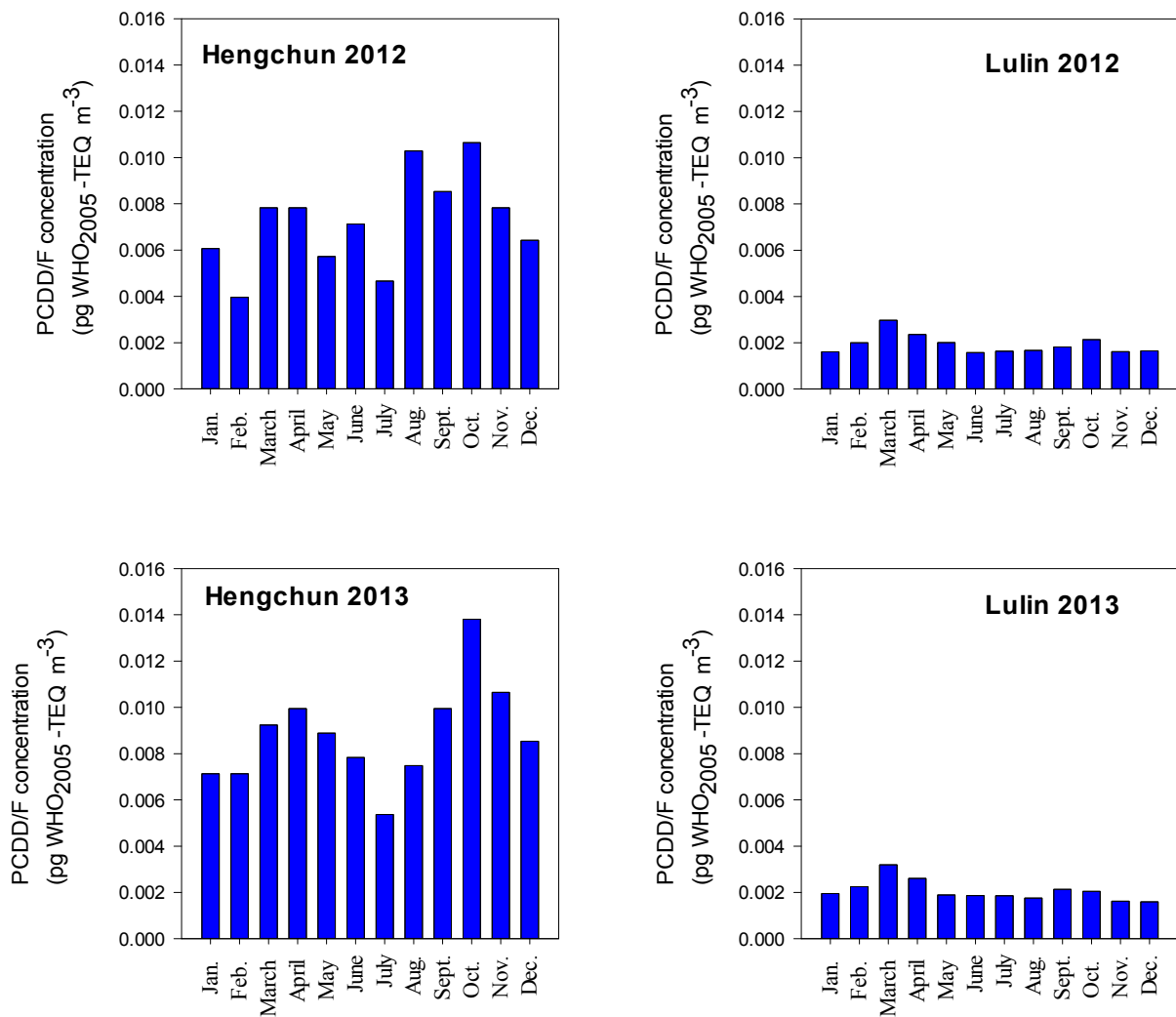


Fig. 3. Simulated total-PCDD/Fs-WHO₂₀₀₅-TEQ concentration in the ambient air of Hengchun and Lulin, respectively.

Hengchun. This is due to the fact that Lulin atmosphere has a lower PM₁₀ concentration.

Hengchun and Lulin are both background sites with no significant anthropogenic sources existing in the vicinity. Particularly, Lulin being a mountain site had lower PM₁₀ and total-PCDD/Fs-TEQ concentrations due to the location above sea level and far distance from the possible South East Asia sources. At a height of 2862 m above sea level, Lulin is above the boundary layer, hence the presence of PCDD/Fs maybe from long range transport. For comparison, Chi *et al.* (2013) reports a range of 0.00187–0.0102 pg I-TEQ m⁻³ for a mountain site in Northern Taiwan and a range of 0.00237–0.00374 pg I-TEQ m⁻³ for the same southern Taiwan coastal site. In another study, Chang *et al.* (2013), reported ambient concentrations ranging from 0.00232–0.00428 I-TEQ m⁻³ for Lulin site. Chang *et al.* (2013) attributed the presence of PCDD/Fs in these background sites to the fact that Taiwan lies in the downstream side of biomass burning plumes originating from South East Asia.

Gas-Particle Partitioning of PCDD/Fs

The distribution of PCDD/Fs between the gas-particle phases determines their fates in the environment. When PCDD/Fs are emitted into the atmosphere, they can be partitioned between the gas and particulate phases based on their concentrations, vapor pressure, the atmospheric temperature, and the ambient air particle concentration (Hoff *et al.*, 1996). Gas-particle partitioning was calculated using Eq. (1), while the Tables 1 and 2 illustrate the meteorological data for gas-particle partition simulation. Total suspended particulate (TSP) concentrations were evaluated using the following relationship TSP: PM₁₀ = 1.24:1 (Huang *et al.*, 2011a) in Taiwan.

Monthly gas-particle partitioning of PCDD/Fs in the ambient air of Hengchun and Lulin in 2012 and 2013 are illustrated in Figs. 4 and 5. According to Figs. 4 and 5, for both Hengchun and Lulin, the highest particle-bound PCDD/Fs occurred in winter and is lowest in summer. The partitioning of PCDD/Fs in gas and particle phase is controlled by three key factors, the atmospheric total suspended particulate (TSP) concentration, the atmospheric temperature and the vapor pressure of PCDD/F congeners. Higher chlorinated PCDD/Fs primarily present in particulate phase for all seasons at both sampling sites similar to the observations of (Wang *et al.*, 2010). Higher chlorinated PCDD/Fs, with higher molecular weight, have a lower vapor pressure. Thus, higher chlorinated PCDD/Fs are mostly present in particle phase than gas phase. The above findings are comparable with those reported in previous studies (Wu *et al.*, 2009; Lin *et al.*, 2010a; Huang *et al.*, 2011a). These results are associated with different vapor pressures; a property strongly related to temperature and the main factor influencing partition of PCDD/Fs (Pankow, 1987).

Additionally, the results show that in winter at both sites, PCDD/Fs bound to particle phase were higher than that in other seasons. This is due to the temperature in winter (averaged 21.8°C at Hengchun and 6.3°C at Lulin) were lower than that in summer (averaged 28.5°C at Hengchun

and 13.3°C at Lulin). As the temperature rose, all PCDD/Fs would probably evaporate from the particle phase to gas phase. Thus, at high temperatures, the PCDD/Fs mostly exist in the gas phase. Therefore, since Hengchun has a higher mean atmospheric temperature than Lulin, it can be explained that PCDD/Fs that is bound to particle phase are lower at this site. These findings are comparable with those reported earlier by Huang *et al.* (2011a) and Mi *et al.* (2012), where the PCDD/Fs bound to particle was found to be increased with decreasing temperature since as temperature increased, the higher chlorinated PCDD/Fs bound to particle phase decrease.

Dry Deposition of PCDD/Fs

Dry deposition velocities of total PCDD/Fs were selected to be 0.45, 0.52, 0.32, and 0.39 cm s⁻¹ in spring, summer, autumn and winter, respectively (Huang *et al.*, 2011a). Due to lack of measured data for PCDD/Fs, a selected value (0.010 cm s⁻¹) is used here to simulate PCDD/Fs dry deposition flux contributed by their gas phase (Mi *et al.*, 2012). The dry deposition velocity contributed by the particle phase was then calculated using Eqs. (5) and (7).

Monthly dry deposition fluxes of total-PCDD/Fs-WHO₂₀₀₅-TEQ at both sites are illustrated in Fig. 6. During 2012, at Hengchun, the monthly dry deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ ranged between 22.7 and 82.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹, while those during 2013 ranged between 33.6 and 104.3 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹. The annual total dry deposition flux of total PCDD/Fs-WHO-TEQ at Hengchun were 602.2 and 767.5 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. In Hengchun, when the dry deposition flux was divided by the ambient air concentration, the resulting dry deposition velocities were estimated as 0.27 and 0.28 cm s⁻¹ for 2012 and 2013, respectively and averaged 0.28 cm s⁻¹.

However, during 2012, at Lulin, the monthly dry deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ ranged between 4.5 and 31.3 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹, while those of 2013 ranged between 3.9 and 27.3 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹. The annual total dry deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ at Lulin were 142.1 and 127.2 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. At Lulin, the estimated dry deposition velocities were 0.24 and 0.19 cm s⁻¹ for 2012 and 2013, respectively and averaged 0.22 cm s⁻¹.

The highest dry deposition fluxes at Hengchun occurred in March 2012 (82.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) and April 2013 (104.3 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). While the lowest occurred in February 2012 (22.7 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) and August 2013 (33.6 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). Highest dry deposition fluxes at Lulin occurred in March (averaged 29.3 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) in both years; while the lowest deposition fluxes were in August (averaged 4.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) in both years. Similar to Huang *et al.* (2011a) the lowest dry deposition fluxes in this study occurred in summer but conversely this study reports the highest dry deposition fluxes at the onset of spring, while Huang *et al.* (2011a) reports the highest occurrence in winter.

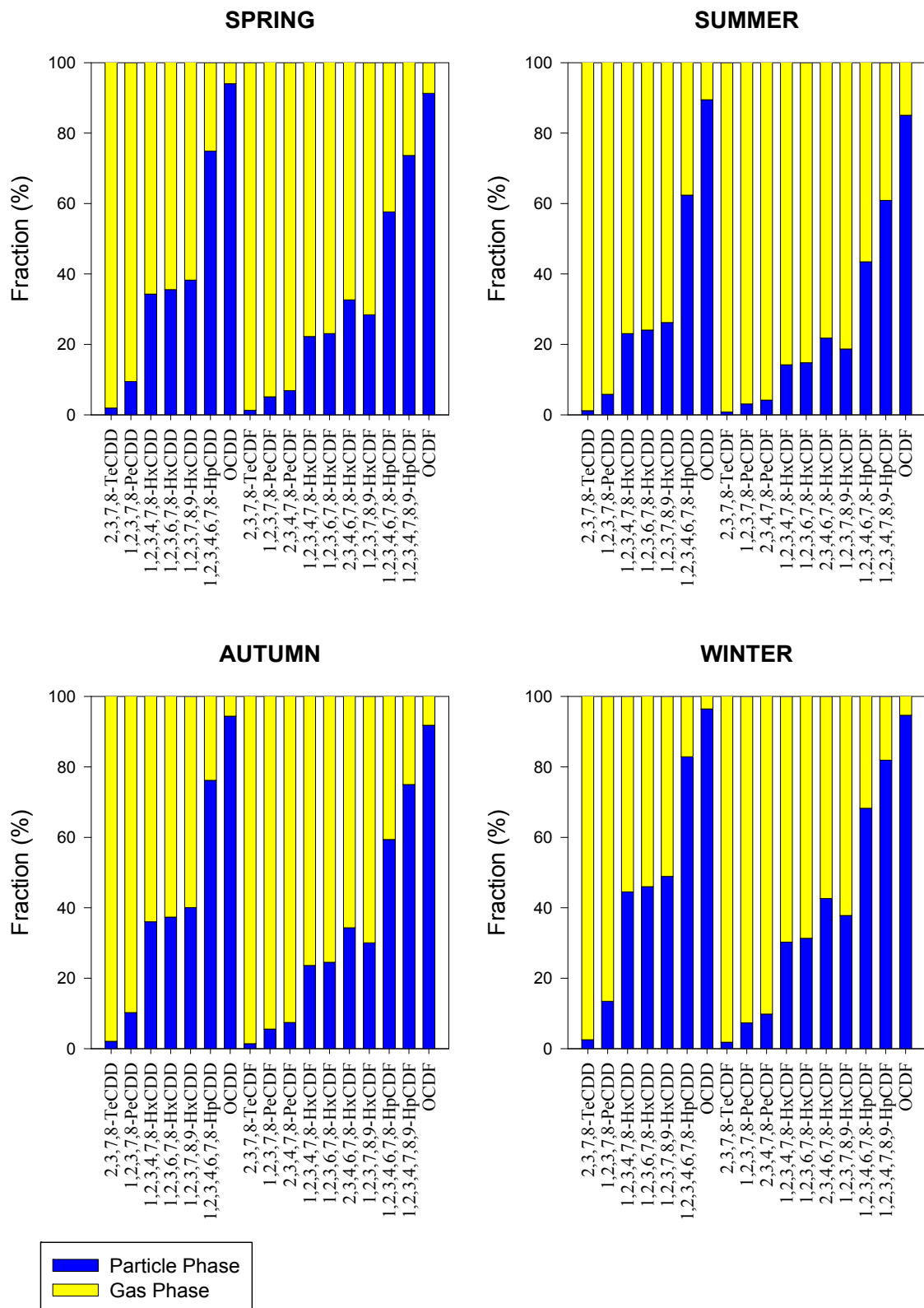


Fig. 4. Seasonal gas-particle partitioning of individual PCDD/F mass concentration at Hengchun.

Dry deposition fluxes at Hengchun averaged 57.1 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) were approximately five times higher than those at Lulin (11.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). This is due to the fact that PM₁₀ concentration at

Hengchun (averaged 26.6 μg m⁻³) was higher than that in Lulin (averaged 11.3 μg m⁻³). Previous studies (Shih *et al.*, 2006; Huang *et al.*, 2011a) reported that the total dry deposition flux was found to decrease as the temperature

increased and indicated that temperature influences the amount of PCDD/Fs that are bound to particles and subsequently are deposited via dry deposition. Huang *et al.* (2011a) reports total dry deposition fluxes of PCDD/Fs

ranging from 0.0274–0.718 ng I-TEQ m⁻² month⁻¹ for a rural area in Taiwan, while Mi *et al.* (2012) reports an average of 8.30–27.5 pg I-TEQ m⁻² year⁻¹ for industrial, rural and urban areas and Wang *et al.* (2010) reported

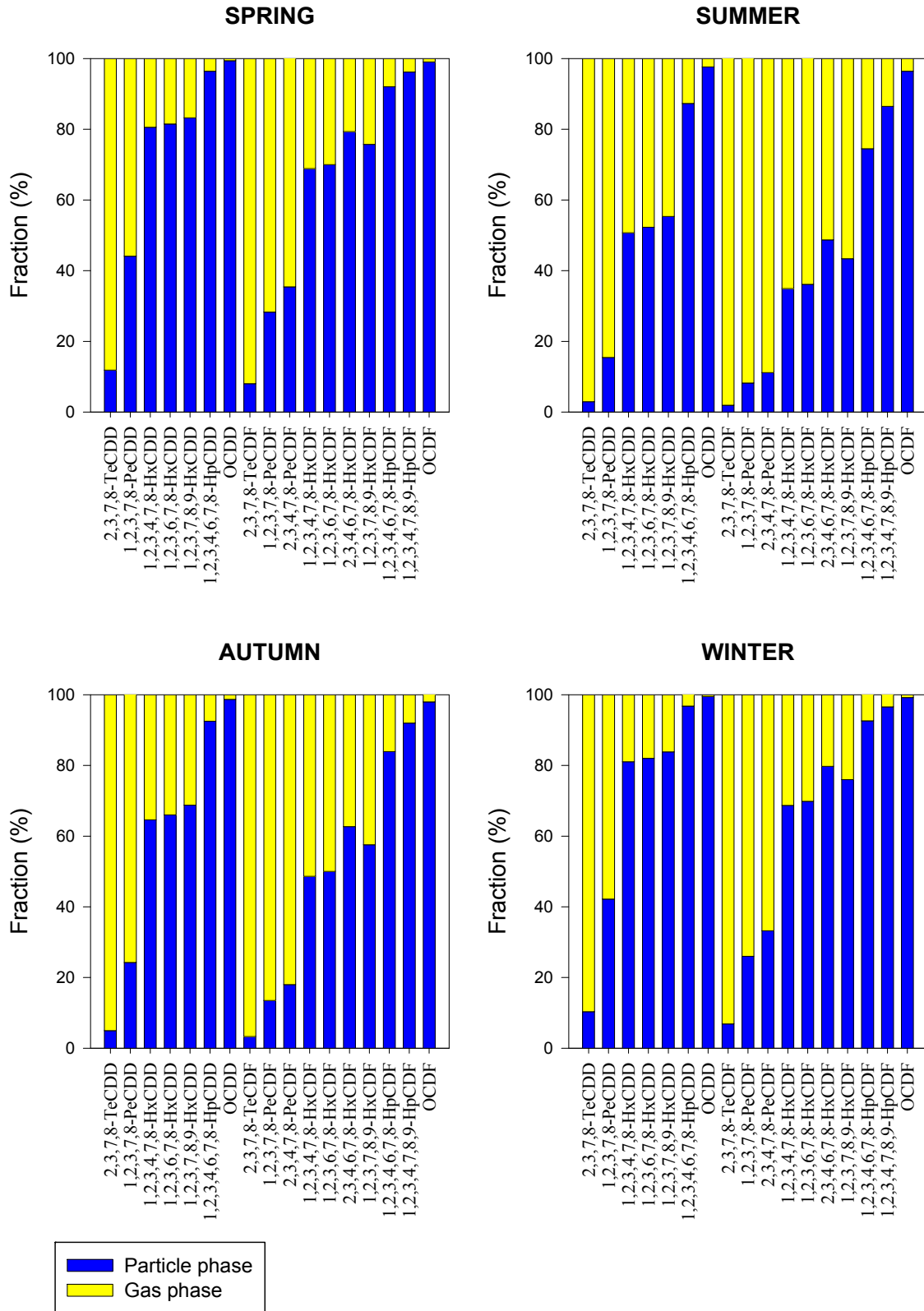


Fig. 5. Seasonal gas-particle partitioning of individual PCDD/F mass concentration at Lulin.

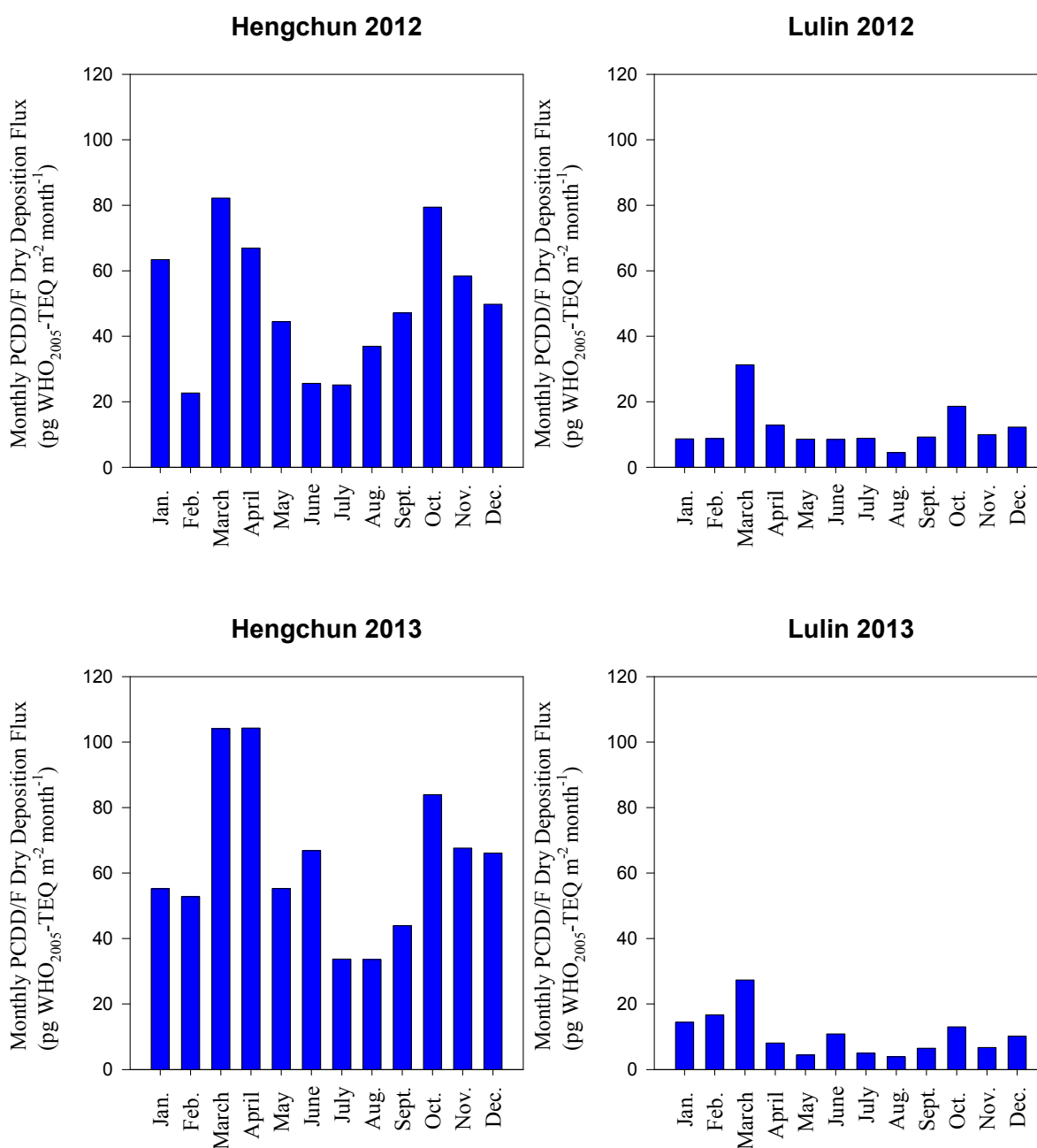


Fig. 6. Monthly dry deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ at Hengchun and Lulin, respectively.

fluxes ranging at 7.67–18.2, 17.6–40.6, 6.69–11.6, 5.73–15.6 pg I-TEQ m⁻² day⁻¹ for commercial, industrial, coastal, and agricultural areas are comparable to 0.49–4.24 pg WHO₂₀₀₅-TEQ m⁻² day⁻¹ reported in this study. Additionally, Shih *et al.* (2006) reports seasonal dry deposition fluxes ranging from 3.07 to 18.91 pg I-TEQ m⁻² day⁻¹ which are significantly higher than other studies mentioned earlier.

Monthly Wet Deposition Flux of Total PCDD/Fs-WHO₂₀₀₅-TEQ

Wet deposition is the combination of both vapor-phase and particle-bound SVOCs that are removed from the atmosphere by rain. The method to evaluate wet deposition fluxes of PCDD/Fs is as described earlier in methodology

part.

Monthly wet deposition fluxes of total-PCDD/Fs-WHO₂₀₀₅-TEQ at both sites can be seen in Fig. 7. During 2012, at Hengchun, the monthly wet deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ ranged between 0.1 and 63.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹, while those during 2013 ranged between 0.8 and 30.1 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹. The annual total wet deposition flux of total-PCDD/Fs-WHO-TEQ concentration at Hengchun were 151.0 and 129.1 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. By using the annual total rainfall and the annual total wet deposition fluxes, the estimated total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the rain were 0.059 and 0.069 pg WHO₂₀₀₅-TEQ L⁻¹ for 2012 and 2013,

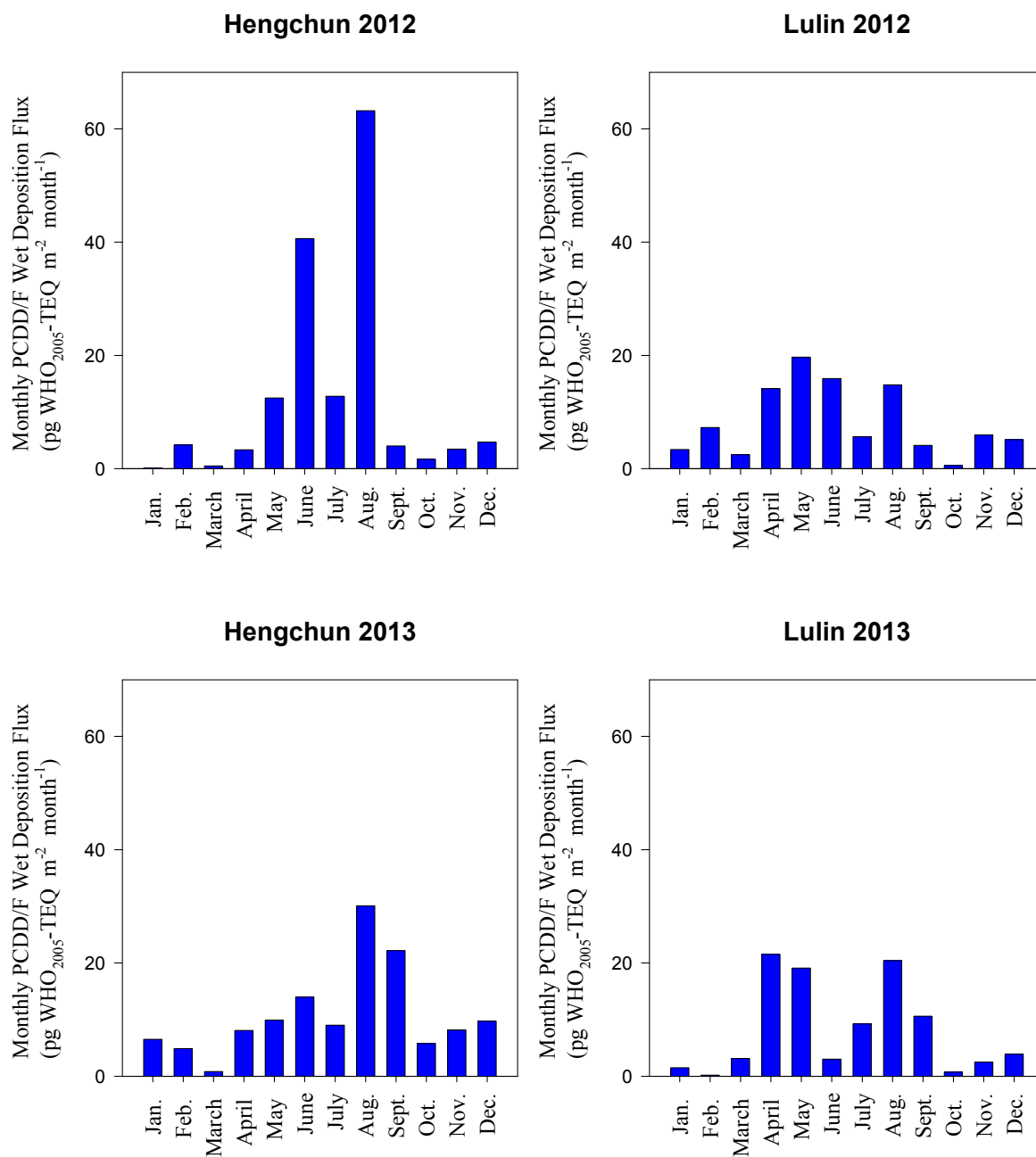


Fig. 7. Monthly wet deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ at Hengchun and Lulin, respectively.

respectively and averaged $0.064 \text{ pg WHO}_{2005}\text{-TEQ L}^{-1}$. By dividing the concentrations in the rainfall by the concentration in the ambient air, the corresponding scavenging ratios of total-PCDD/Fs-WHO₂₀₀₅-TEQ were calculated as 8050 and 7880 for 2012 and 2013 respectively and averaged 8015.

However, during 2012, at Lulin, the monthly wet deposition fluxes of total-PCDD/Fs-WHO₂₀₀₅-TEQ ranged between 0.50 and $19.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$, while those during 2013 ranged between 0.20 and $21.5 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$. The annual total wet deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ at Lulin were 99.0 and $95.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ year}^{-1}$ in 2012 and 2013, respectively. The estimated total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the rain were 0.026 and $0.027 \text{ pg WHO}_{2005}\text{-TEQ L}^{-1}$ for 2012 and 2013, respectively, and averaged $0.027 \text{ pg WHO}_{2005}\text{-TEQ L}^{-1}$.

The estimated scavenging ratios of total-PCDD/Fs-WHO₂₀₀₅-TEQ were calculated as 13900 and 13000 for 2012 and 2013 respectively and averaged 13450. Due to the fact that, the annual average temperature at Lulin is much lower than that in Hengchun and the fraction of PCDD/Fs-WHO₂₀₀₅-TEQ in the particulate phase in Lulin is much higher resulting in a higher scavenging ratio.

Highest wet deposition fluxes at Hengchun were observed in August (averaged $46.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) in both 2012 and 2013. Highest wet deposition flux obtained in these periods was due to tremendous rainfall intensity recorded in August for both years (averaged 629.1 mm). However, those of the lowest wet deposition fluxes occurred

in January ($0.1 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2012 and March ($0.8 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2013. At Hengchun, lowest rainfall intensities were recorded in January 2012 (1.6 mm) and March 2013 (8.4 mm), thus the wet depositions were also lowest in those periods.

On the other hand, at Lulin, the highest wet deposition fluxes were in May 2012 ($19.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) and April 2013 ($21.5 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$). Highest wet deposition flux obtained in these periods was due to high rainfall intensity were occurred in May 2012 (616 mm) and in April 2013 (427.6 mm). The lowest wet deposition flux occurred in October ($0.6 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2012 and February ($0.2 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2013 as a result of low rainfall intensities recoded in October 2012 (16 mm) and in February 2013 (4.4 mm).

The PCDD/Fs scavenging with raindrops, therefore, the wet deposition fluxes fluctuate dramatically from month to month. Wet depositions are strongly influenced by the meteorological factors such as ambient temperature, rainfall intensity and wind speed. The wet deposition fluxes of PCDD/Fs increase primarily with increasing rainfall intensity. The scavenging ratios of both sites can be seen in (Rahenderi, 2014).

Furthermore, particulate matter (PM_{10}) concentration also plays a big role in the wet deposition. Higher PM_{10} concentration means that there are more particulates can be scavenged by wet deposition. For example, if we compare wet deposition at Lulin in 2012, highest rainfall occurred in June 2012 (886.2 mm). But, highest wet deposition occurred in the second highest rainfall period in May 2012 (616 mm). Considering the PM_{10} concentration, in May 2012 mean monthly PM_{10} concentration was higher ($9 \mu\text{g m}^{-3}$) than that in June 2012 ($5 \mu\text{g m}^{-3}$). Thus, more particles were scavenged in May than June, so that the wet deposition in May is higher than in June 2012. During the whole period (2012 and 2013), the mean monthly wet deposition at Hengchun (averaged $11.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) was 1.44 times higher than that at Lulin (averaged $8.11 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$). However, the rainfall intensity at Lulin (monthly average of 301 mm) was 1.64 times higher than at Hengchun (monthly average of 184mm). Therefore, the concentrations of both PM_{10} and PCDD/Fs and gas-particle partitioning of PCDD/Fs in the ambient air are also key factors affecting wet deposition of PCDD/Fs in addition to rainfall intensity. On the other hand, in this study, other parameters effect like snow or fog are not being observed in this simulation.

Monthly Total (Wet + Dry) PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ Deposition Fluxes

Total deposition fluxes were calculated by summation of dry and wet deposition fluxes. Monthly total deposition fluxes of total-PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ at both sites can be seen in Fig. 8. During 2012, at Hengchun, the monthly total deposition flux of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ ranged between 26.9 and $100.2 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$, while those during 2013 ranged between 42.8 and $112.4 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$. However, during 2012, at

Lulin, the monthly total deposition flux of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ ranged between 12.1 and $33.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$, while those during 2013 ranged between 9.2 and $30.5 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$.

Highest total deposition flux at Hengchun was recorded in August ($100.2 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2012 and in March ($99.2 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2013; while the lowest occurred in February ($26.9 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2012 and in July ($42.8 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) 2013. Highest total deposition fluxes at Lulin occurred in March 2012 ($33.7 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) and in March 2013 ($30.5 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$), while the lowest occurred in January 2012 ($12.1 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$) and in November 2013 ($9.2 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$).

The annual total deposition flux of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ at Hengchun were 753.2 and $896.6 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ year}^{-1}$ in 2012 and 2013, respectively. The annual total deposition flux of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ at Lulin were 241.1 and $222.9 \text{ pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ year}^{-1}$ in 2012 and 2013, respectively.

During the whole periods (2012 and 2013), the average annual total (dry + wet) deposition flux of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ in the terms of $\text{pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ year}^{-1}$ at Hengchun (824.9) was 3.5 times higher than those in Lulin (232.0). These values are lower than 5.02–5.11 $\text{ng I-TEQ m}^{-2} \text{ year}^{-1}$ reported near two municipal waste incinerators in southern Taiwan (Huang *et al.*, 2011b) and far much lower than 27.0 $\text{ng I-TEQ m}^{-2} \text{ year}^{-1}$ in a drinking water treatment plant in Taiwan (Lin *et al.*, 2010a).

Comparisons between total depositions simulated in this study and in other previous studies are illustrated in Table 5. It can be seen from the table that total deposition simulated in this study are much lower compared to other previous studies.

Contribution Fractions of Dry Deposition to the Total (Dry + Wet) Deposition Fluxes of Total-PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$

Monthly fraction contributed by dry deposition in monthly total (dry + wet) deposition fluxes of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ can be seen in Fig. 9. At Hengchun, during 2012, the fraction contributed by dry deposition ranged between 30.5% and 99.8%, while during 2013 ranged between 51.7% and 99.1%. On the basis of monthly average, the mean fraction of total (dry + wet) deposition fluxes of total PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ contributed by dry deposition at Hengchun were 78.2% and 78.9% in 2012 and 2013, respectively; however, at Lulin, during 2012, the fraction contributed by dry deposition ranged between 19.2% and 97.0%, while during 2013 ranged between 23.6% and 98.6%. Mean fraction of total (dry + wet) deposition fluxes of total-PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ contributed by dry deposition at Lulin were 56.5% and 69.1% in 2012 and 2013, respectively.

The highest fraction of total (dry + wet) deposition fluxes of total-PCDD/Fs- $\text{WHO}_{2005}\text{-TEQ}$ contributed by dry deposition at Hengchun occurred in January 2012 (99.8%) and March 2013 (99.1%); in January 2012, the dry deposition

contributed 99.8% in the total deposition flux, since the rainfall intensity in that period is the lowest (1.6 mm).

Thus, fewer particles were scavenged by wet deposition. March 2013 at Hengchun also have the lowest rainfall

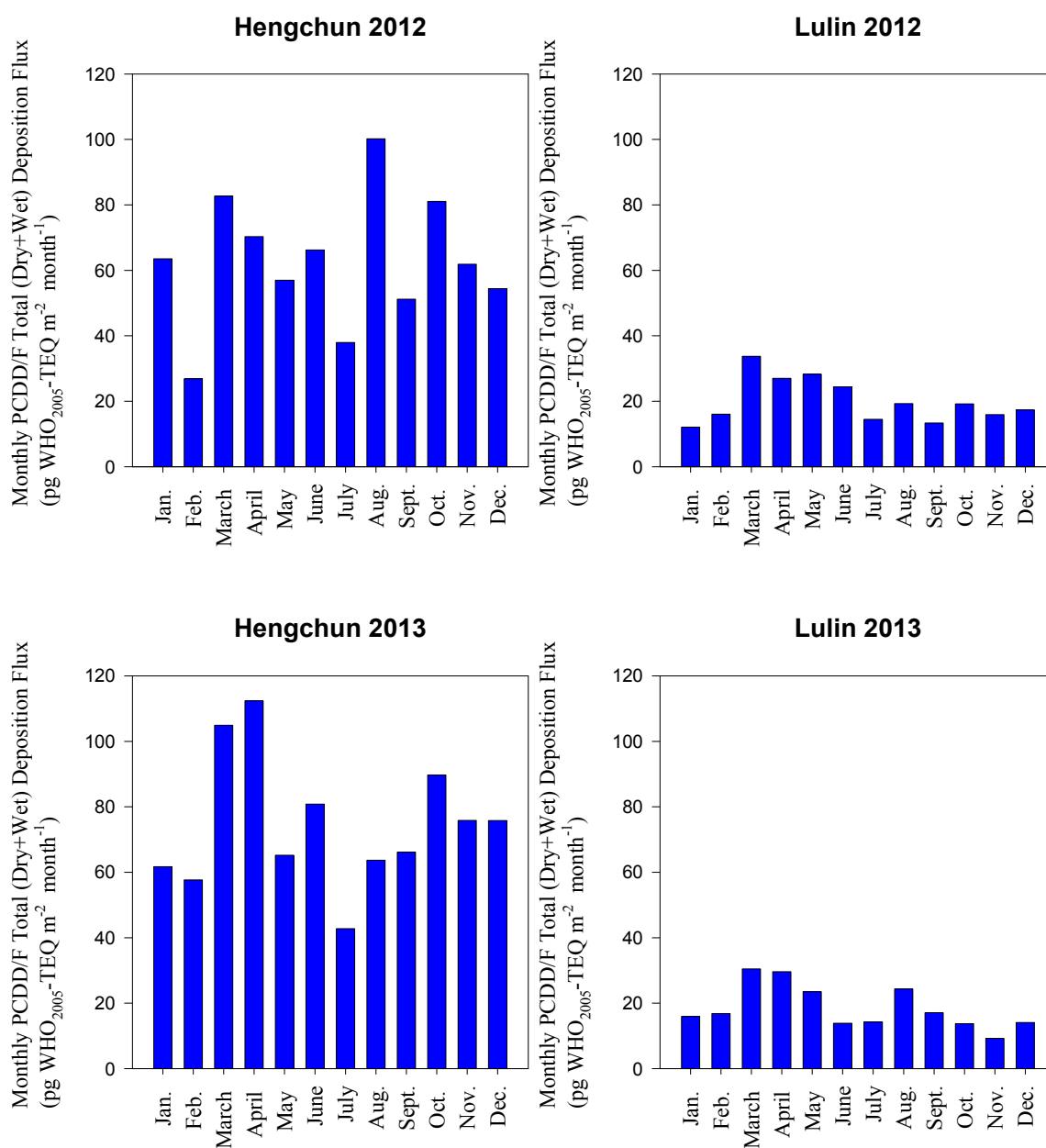


Fig. 8. Monthly total (dry + wet) Dry deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ at Hengchun and Lulin, respectively.

Table 5. Comparison of total (dry + wet) deposition fluxes of total PCDD/Fs-TEQ between previous studies and this study.

Country	Location	Sampling Period	Total TEQ flux	Reference
Taiwan	Industrial	2009–2010	5020–5110 pg I-TEQ m ⁻² year ⁻¹	Huang <i>et al.</i> , 2011
Taiwan	Coastal	2013	657 pg WHO ₂₀₀₅ -TEQ m ⁻² year ⁻¹	This study
	Mountain		249 pg WHO ₂₀₀₅ -TEQ m ⁻² year ⁻¹	
Tokyo	Urban		17000 pg I-TEQ m ⁻² year ⁻¹	Ogura <i>et al.</i> , 2001
Yokohama	Urban	2001	11000 pg I-TEQ m ⁻² year ⁻¹	
Tanzawa	Mountain		5700 pg I-TEQ m ⁻² year ⁻¹	Guerzoni <i>et al.</i> , 2004
France	Lagoon	2011	4280 pg I-TEQ m ⁻² year ⁻¹	
Italy	Lake	2005	140 pg WHO-TEQ m ⁻² week ⁻¹ (only one week registered precipitation)	Castro-Jiménez <i>et al.</i> , 2008

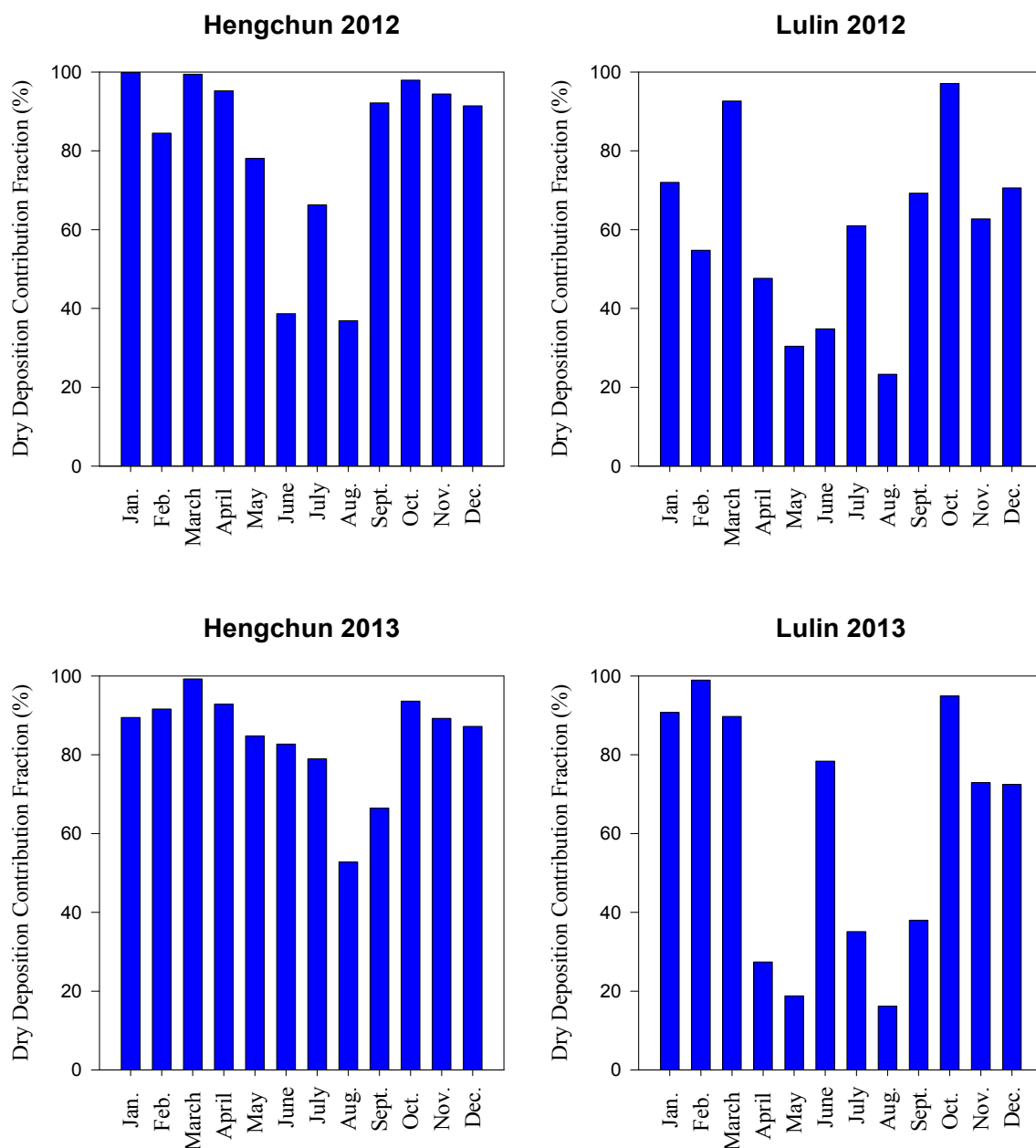


Fig. 9. Contribution fractions (%) of dry deposition to the total PCDD/F (dry + wet) deposition fluxes in the terms of WHO₂₀₀₅-TEQ at Hengchun and Lulin, respectively.

intensity (8.4 mm). Lowest fraction contributed by dry deposition occurred in August 2012 (30.5%) and July 2013 (51.7%). Lowest fraction contributed by dry deposition occurred when the rainfall occurrence is high (723.4 mm) in August 2012 and in July 2013 (251.6 mm).

The highest fraction contributed by dry deposition at Lulin was in October 2012 (97.0%) and February 2013 (98.6%). In October 2012, the dry deposition contributed about 97.0% in the total-PCDD/Fs-WHO₂₀₀₅-TEQ deposition flux, since the rainfall intensity in that period is low (16 mm). February 2013 at Lulin also have the lowest rainfall intensity (4.4 mm). Lowest fraction contributed by dry deposition occurred in May 2012 (19.2%) and in August 2013 (23.6%), due to tremendous rainfall intensity occurred

in May 2012 (616 mm) and August 2013 (1056.6 mm), thus, the particles were scavenged more by rainfall via wet deposition than the dry deposition.

Congener Profile for PCDD/Fs in Total Deposition Fluxes

With regard to the relative mass contribution (%) of individual PCDD/Fs, the high chlorinated congeners were predominant (Fig. 10). At Hengchun the OCDD, OCDF and 1,2,3,4,6,7,8-HpCDF were the major, while at Lulin the OCDD, OCDF followed by 1,2,3,4,6,7,8-HpCDD were predominant in total deposition flux of PCDD/F mass ($\mu\text{g m}^{-2} \text{ month}^{-1}$). This was also the case in studies done by Ren *et al.* (2007), Castro-Jiménez *et al.* (2008) and Wang *et al.* (2010).

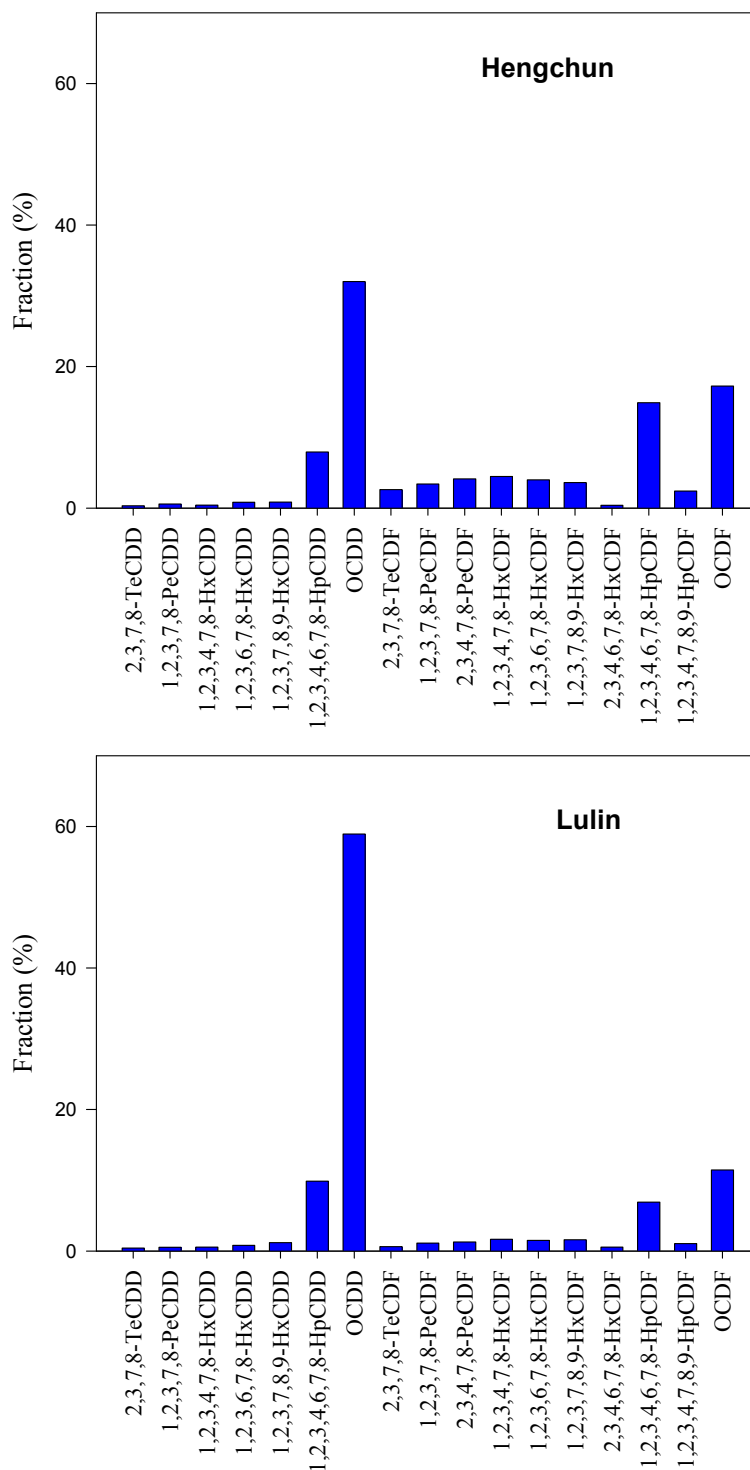


Fig. 10. Congener profiles for total mass deposition fluxes at Hengchun and Lulin, respectively.

However, on the basis of WHO₂₀₀₅-TEQ, at Hengchun 2,3,4,7,8-PeCDF followed by 1,2,3,7,8-PeCDD were predominant, however at Lulin 1,2,3,7,8-PeCDD followed by 2,3,4,7,8-PeCDF contributed the highest fractions to the total-PCDD/Fs-WHO₂₀₀₅-TEQ deposition fluxes (Fig. 11). Similarly, (Moon *et al.*, 2005) and (Ren *et al.*, 2007) reported that 2,3,4,7,8-PeCDF having greater abundance in the WHO₂₀₀₅-TEQs. The results from both background

sites mirror the observations by (Huang *et al.*, 2011a), who notes that total toxicity (TEQ) basis is dominated by gas phase, while the total mass basis is dominated by particle phase. This is particularly due to the fact that lower congeners are more toxic especially the TCDD and they exist mostly in the gas phase owing to their higher vapor pressures, thereby contributing more to the total-PCDD/Fs-WHO₂₀₀₅-TEQ.

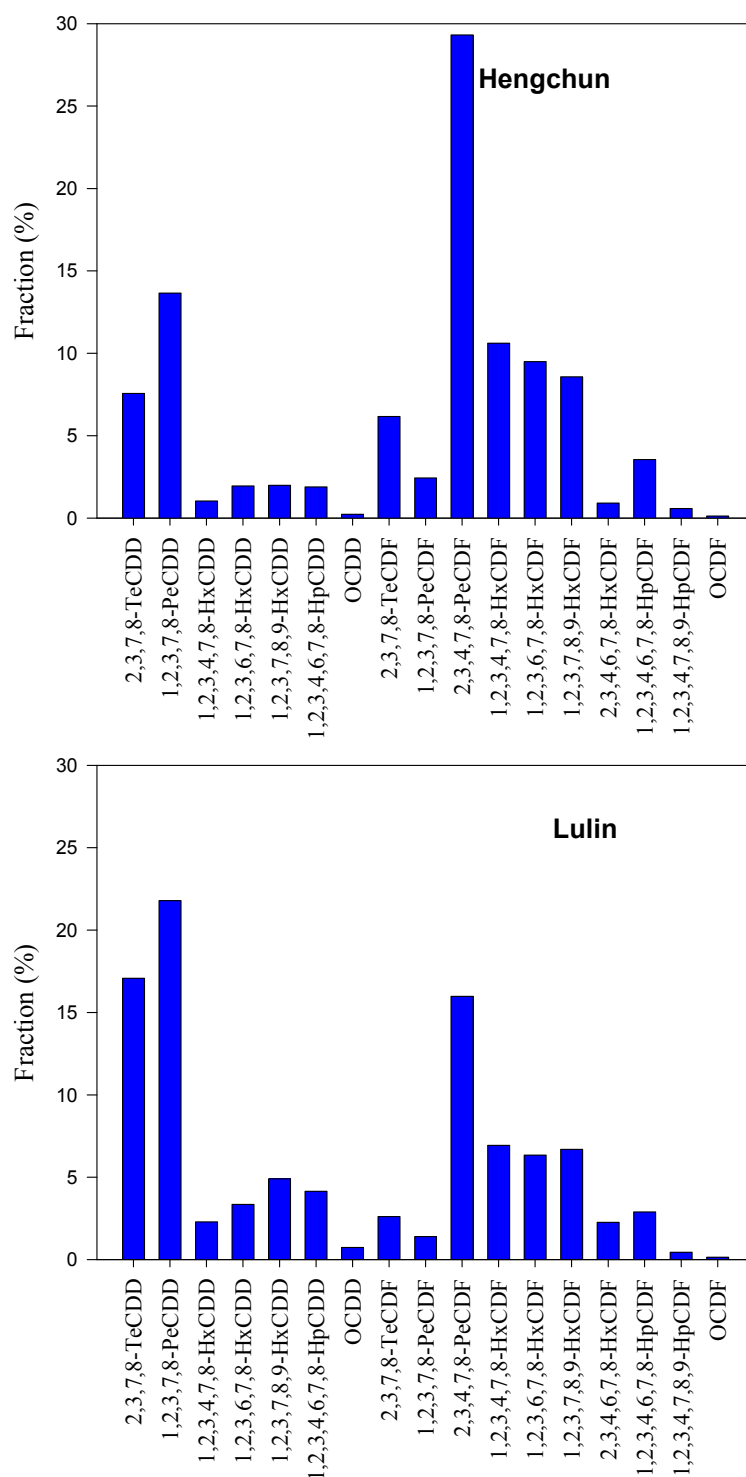


Fig. 11. Congener profiles for PCDD/Fs-WHO₂₀₀₅-TEQ deposition fluxes at Hengchun and Lulin, respectively.

CONCLUSIONS

The results of this study can be summarized as follows:

1. The simulated total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations at Hengchun (coastal site) which had more anthropogenic activities, ranged between 0.0039–0.0106 pg WHO₂₀₀₅-TEQ m⁻³ and 0.0054–0.0138 pg WHO₂₀₀₅-TEQ m⁻³ for 2012 to 2013, respectively. For

Lulin, a background (mountain) site, the PCDD/F concentrations ranged between 0.0016–0.0029 pg WHO₂₀₀₅-TEQ m⁻³ and 0.0016–0.0032 pg WHO₂₀₀₅-TEQ m⁻³ for 2012 and 2013, respectively.

2. The annual total dry deposition flux of total-PCDD/Fs-WHO-TEQ at Hengchun were 602.2 and 767.5 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. The annual total dry deposition flux of total-PCDD/Fs-

- WHO₂₀₀₅-TEQ at Lulin were 142.1 and 127.2 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. Dry deposition fluxes at Hengchun (averaged 57.1 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) were approximately five times higher than those at Lulin (11.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). The lack of anthropogenic activities around Lulin site and the altitude of location may influence the results, which resulted in lower than those recorded at Hengchun.
- In Hengchun, the resulting dry deposition velocities were estimated as 0.27 and 0.28 cm s⁻¹ for 2012 and 2013, respectively, and averaged 0.28 cm s⁻¹. At Lulin, the estimated dry deposition velocities were 0.24 and 0.19 cm s⁻¹ for 2012 and 2013, respectively, and averaged 0.22 cm s⁻¹.
 - In this study, the annual total wet deposition fluxes of total-PCDD/Fs-WHO₂₀₀₅-TEQ at Hengchun were 151.0 and 129.1 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively, while the annual total wet deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ at Lulin were 99.0 and 95.7 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. For the whole period (2012 and 2013), the mean monthly wet deposition at Hengchun (averaged 11.7 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) was 1.44 times higher than that at Lulin (averaged 8.11 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). However, the rainfall intensity at Lulin (monthly average of 301 mm) was 1.64 times higher than at Hengchun (monthly average of 184 mm). Therefore, the concentrations of both PM₁₀ and PCDD/Fs in the ambient air are two key factors influencing wet deposition of PCDD/Fs in addition to rainfall intensity.
 - At Hengchun, the estimated total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the rain were 0.059 and 0.069 pg WHO₂₀₀₅-TEQ/L for 2012 and 2013, respectively and averaged 0.064 pg WHO₂₀₀₅-TEQ L⁻¹. For Lulin, the estimated total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the rain were 0.026 and 0.027 pg WHO₂₀₀₅-TEQ L⁻¹ for 2012 and 2013, respectively, and averaged 0.027 pg WHO₂₀₀₅-TEQ L⁻¹. The corresponding scavenging ratios of total-PCDD/Fs-WHO₂₀₀₅-TEQ for Hengchun area were 8050 and 7880 for 2012 and 2013 respectively and averaged 8015, while for Lulin, the estimated scavenging ratios of total-PCDD/Fs-WHO₂₀₀₅-TEQ were calculated as 13900 and 13000 for 2012 and 2013 respectively and averaged 13450.
 - Highest wet deposition fluxes of total-PCDD/Fs-WHO₂₀₀₅-TEQ at Hengchun were observed in August (averaged 46.7 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) in both 2012 and 2013. Highest wet deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ obtained in these periods was due to tremendous rainfall intensity recorded in August for both years (averaged 629.1 mm). However, those of the lowest wet deposition fluxes occurred in January (0.1 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) 2012 and March (0.8 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) 2013. At Hengchun, lowest rainfall intensities were recorded in January 2012 (1.6 mm) and March 2013 (8.4 mm), thus the wet depositions were also lowest in those periods.
 - On the other hand, at Lulin, the highest wet deposition fluxes of total-PCDD/Fs-WHO₂₀₀₅-TEQ were in May 2012 (19.7 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) and April 2013 (21.5 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹). Highest wet deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ obtained in these periods was due to high rainfall intensity were occurred in May 2012 (616 mm) and in April 2013 (427.6 mm). The lowest wet deposition flux occurred in October (0.6 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) 2012 and February (0.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹) 2013 as a result of low rainfall intensities recorded in October 2012 (16 mm) and in February 2013 (4.4 mm).
 - At Hengchun, the monthly total (dry + wet) deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ ranged between 26.9 and 100.2 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹ in 2012, while those during 2013 ranged between 42.8 and 112.4 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹. However, during 2012, at Lulin, the monthly total (dry + wet) deposition flux of total-PCDD/Fs-WHO₂₀₀₅-TEQ ranged between 12.1 and 33.7 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹, while those during 2013 ranged between 9.2 and 30.5 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹. The annual total (dry + wet) deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ at Hengchun were 753.2 and 896.6 pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ in 2012 and 2013, respectively. The annual total (dry + wet) deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ at Lulin were 241.1 and 222.9 pg WHO₂₀₀₅-TEQ m⁻² month⁻¹ in 2012 and 2013, respectively. During the whole period (2012 and 2013), the average annual total (dry + wet) deposition flux of total PCDD/Fs-WHO₂₀₀₅-TEQ in the terms of pg WHO₂₀₀₅-TEQ m⁻² year⁻¹ at Hengchun (824.9) was 3.5 times higher than those in Lulin (232.0).
 - Mean fraction contributed by dry deposition at Hengchun were 78.2% and 78.9% in 2012 and 2013, respectively while at Lulin the average fraction contributed by dry deposition at Lulin were 56.5% and 69.1% in 2012 and 2013, respectively. Thus, for the whole sampling period dry deposition was majorly responsible for removal of PCDD/Fs from the atmosphere for both coastal and mountain areas.
 - The highest fraction contributed by dry deposition at Lulin was in October 2012 (97.0%) and February 2013 (98.6%). In October 2012, the dry deposition contributed about 97.0% in the total deposition flux, since the rainfall intensity in that period is low (16 mm). February 2013 at Lulin also have the lowest rainfall intensity (4.4 mm). Lowest fraction contributed by dry deposition occurred in May 2012 (19.2%) and in August 2013 (23.6%), due to tremendous rainfall intensity occurred in May 2012 (616 mm) and August 2013 (1056.6 mm), thus, the particulates were scavenged more by rainfall via wet deposition than the dry deposition.
 - With regard to the relative mass contribution (%) of individual PCDD/Fs, the high chlorinated congeners were predominant. At Hengchun the OCDD, OCDF and 1,2,3,4,6,7,8-HpCDF were the major while at Lulin the OCDD OCDF followed by 1,2,3,4,6,7,8-HpCDD were predominant in total deposition flux of PCDD/F mass (pg/m²-month).

12. On toxicity basis, WHO₂₀₀₅-TEQ, at Hengchun 2,3,4,7,8-PeCDF followed by 1,2,3,7,8-PeCDD were predominant, however at Lulin 1,2,3,7,8-PeCDD followed by 2,3,4,7,8-PeCDF contributed the highest fractions to the total deposition fluxes.

ACKNOWLEDGMENTS

The authors wish to acknowledge the Super Micro Mass Research and Technology Centre, in Cheng Shiu University for helping with analysis of PCDD/Fs.

REFERENCES

- Amodio, M., Catino, S., Dambruoso, P., de Gennaro, G., Di Gilio, A., Giungato, P., Laiola, E., Marzocca, A., Mazzone, A. and Sardaro, A. (2014). Atmospheric Deposition: Sampling Procedures, Analytical Methods, and Main Recent Findings from the Scientific Literature. *Adv. Meteorol.* 2014: 161730.
- Bidleman, T.F. (1988). Atmospheric Processes. *Environ. Sci. Technol.* 22: 361–367.
- Castro-Jiménez, J., Mariani, G., Eisenreich, S., Christoph, E., Hanke, G., Canuti, E., Skejo, H. and Umlauf, G. (2008). Atmospheric Input of POPs into Lake Maggiore (Northern Italy): PCDD/F and Dioxin-like PCB Profiles and Fluxes in the Atmosphere and Aquatic System. *Chemosphere* 73: S122–S130.
- Chang, C. and Tsai, C. (2003). A Model for the Relative Humidity Effect on the Readings of the PM₁₀ Beta-gauge Monitor. *J. Aerosol Sci.* 34: 1685–1697.
- Chang, S.S., Lee, W.J., Wang, L.C., Lin, N.H. and Chang-Chien, G.P. (2013). Influence of the Southeast Asian Biomass Burnings on the Atmospheric Persistent Organic Pollutants Observed at near Sources and Receptor Site. *Atmos. Environ.* 78: 184–194.
- Chang, S.S., Lee, W.J., Holsen, T.M., Li, H.W., Wang, L.C. and Chang-Chien, G.P. (2014). Emissions of Polychlorinated-*p*-dibenzo Dioxin, Dibenzofurans (PCDD/Fs) and Polybrominated Diphenyl Ethers (PBDEs) from Rice Straw Biomass Burning. *Atmos. Environ.* 94: 573–581.
- 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., Lin, C.Y., Wang, S.H., Lin, N.H., Sheu, G.R. and Lee, C.T. (2013). Evaluation of the Distributions of ambient PCDD/Fs at Remote Locations in and around Taiwan. *Atmos. Environ.* 78: 203–210.
- Chi, K.H., Chou, C.C., Peng, C.M., Chang, M.B., Lin, C.Y. and Li, C.T. (2014). Increase of Ambient PCDD/F Concentrations in Northern Taiwan during Asian Dust Storm and Winter Monsoon Episodes. *Aerosol Air Qual. Res.* 14: 1279–1291.
- 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., Chang-Chien, G.P., Wang, L.C. and Huang, Y.C. (2010). Emissions of Polychlorinated Dibenzo-*p*-dioxin and Polychlorinated Dibenzofuran from Motorcycles. *Aerosol Air Qual. Res.* 10: 533–539.
- Correa, O., Raun, L., Rifai, H., Suarez, M., Holsen, T. and Koenig, L. (2006). Depositional Flux of Polychlorinated Dibenzo-*p*-dioxins and Polychlorinated Dibenzofurans in an urban setting. *Chemosphere* 64: 1550–1561.
- Donnelly, J., Munslow, W., Mitchum, R. and Sovocool, G. (1987). Correlation of Structure with Retention Index for Chlorinated Dibenzo-*p*-dioxins. *J. Chromatogr. A* 392: 51–63.
- 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.
- Fang, M., Choi, S.D., Baek, S.Y., Park, H. and Chang, Y.S. (2011). Atmospheric Bulk Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in the Vicinity of an Iron and Steel Making Plant. *Chemosphere* 84: 894–899.
- Fujimori, T., Nishimura, K., Oshita, K., Takeda, N. and Takaoka, M. (2014). Influence of the Properties of Macromolecular Carbon on *de Novo* Synthesis of PCDDs, PCDFs, PCBs, and Chlorobenzenes. *Aerosol Air Qual. Res.* 14: 1131–1141.
- Giorgi, F. (1988). Dry Deposition Velocities of Atmospheric Aerosols as Inferred by Applying a Particle Dry Deposition Parameterization to a General Circulation Model. *Tellus Ser. B* 40: 23–41.
- 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.
- Halsall, C.J., Coleman, P. and Jones, K.C. (1997). Atmospheric Deposition of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofurans (PCDD/Fs) and Polycyclic Aromatic Hydrocarbons (PAHs) in Two UK Cities. *Chemosphere* 35: 1919–1931.
- Hoff, R., Strachan, W., Sweet, C., Chan, C., Shackleton, M., Bidleman, T., Brice, K., Burniston, D., Cussion, S. and Gatz, D. (1996). Atmospheric Deposition of Toxic Chemicals to the Great Lakes: A Review of Data through 1994. *Atmos. Environ.* 30: 3505–3527.
- Huang, C.J., Chen, K.S., Lai, Y.C., Wang, L.C. and Chang-Chien, G.P. (2011a). Characterization of Atmospheric Dry Deposition of Polychlorinated Dibenzop-dioxins/Dibenzofuran in a Rural Area of Taiwan. *Aerosol Air Qual. Res.* 11: 448–459.

- Huang, C.J., Chen, K.S., Lai, Y.C., Wang, L.C. and Chang-Chien, G.P. (2011b). Wet Deposition of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofuran in a Rural Area of Taiwan. *Aerosol Air Qual. Res.* 11: 732–748.
- 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., Simó, 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.
- Kaupp, H., Towara, J. and McLachlan, M. (1994). Distribution of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Atmospheric Particulate Matter with Respect to Particle Size. *Atmos. Environ.* 28: 585–593.
- Koester, C.J. and Hites, R.A. (1992). Wet and Dry Deposition of Chlorinated Dioxins and Furans. *Environ. Sci. Technol.* 26: 1375–1382.
- Kulkarni, P.S., Crespo, J.G. and Afonso, C.A. (2008). Dioxins Sources and Current Remediation Technologies—A Review. *Environ. Int.* 34: 139–153.
- 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 the Incinerations of Both Medical and Municipal Solid Wastes. *Aerosol Air Qual. Res.* 3: 1–6.
- Liao, W.T., Wang, Y.F., Tsai, C.H., Tsai, Y.I., Wu, Z.L. and Kuo, Y.M. (2014). Polychlorinated Dibenzo-*p*-dioxin and Dibenzofuran (PCDD/F) Emission Behavior during Incineration of Laboratory Wastes. Part 2: PCDD/F Profiles and Characteristics of Output Materials. *Aerosol Air Qual. Res.* 14: 1206–1214.
- 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: 231–244.
- 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.
- Lin, Y.C., Wu, T.Y., Chen, Y.M., Hsieh, Y.P. and Hu, M.T. (2011). PCDD/F Levels in the Stacks and Food of Fast Food Restaurants. *Aerosol Air Qual. Res.* 11: 437–447.
- Lohmann, R. and Jones, K.C. (1998). Dioxins and Furans in Air and Deposition: A Review of Levels, Behaviour and Processes. *Sci. Total Environ.* 219: 53–81.
- Lohmann, R., Lee, R.G., Green, N.J. and Jones, K.C. (2000). Gas-particle Partitioning of PCDD/Fs in Daily Air Samples. *Atmos. Environ.* 34: 2529–2537.
- Melymuk, L., Robson, M., Diamond, M.L., Bradley, L.E. and Backus, S. (2011). Wet Deposition Loadings of Organic Contaminants to Lake Ontario: Assessing the Influence of Precipitation from Urban and Rural Sites. *Atmos. Environ.* 45: 5042–5049.
- Mi, H.H., Wu, Z.S., Lin, L.F., Lai, Y.C., Lee, Y.Y., Wang, L.C. and Chang-Chien, G.P. (2012). Atmospheric Dry Deposition of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofurans (PCDD/Fs) and Polychlorinated Biphenyls (PCBs) in Southern Taiwan. *Aerosol Air Qual. Res.* 12: 1016–1029.
- Mitrou, P., Dimitriadis, G. and Raptis, S. (2001). Toxic Effects of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin and Related Compounds. *Eur. J. Inter. Med.* 12: 406–411.
- 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: 1525–1534.
- 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., Chang, Y.S., Kim, E.J. and Lee, D.W. (2002). Distribution of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in Different Sizes of Airborne Particles. *Atmos. Environ.* 36: 5109–5117.
- 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. and Bidleman, T.F. (1991). Effects of Temperature, TSP and Per Cent Non-exchangeable Material in Determining the Gas-particle Partitioning of Organic Compounds. *Atmos. Environ.* 25: 2241–2249.
- 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.
- Ren, M., Peng, P.a., Zhang, S., Yu, L., Zhang, G., Mai, B., Sheng, G. and Fu, J. (2007). Atmospheric Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in Guangzhou, China. *Atmos. Environ.* 41: 592–605.
- Schechter, A., Birnbaum, L., Ryan, J.J. and Constable, J.D. (2006). Dioxins: An Overview. *Environ. Res.* 101: 419–428.
- 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.
- Shih, S.I., Lee, W.J., Lin, L.F., Huang, J.Y., Su, J.W. and Chang-Chien, G.P. (2008). Significance of Biomass Open Burning on the Levels of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in the Ambient Air. *J. Hazard. Mater.* 153: 276–284.
- Tasdemir, Y., Odabasi, M., Vardar, N., Sofuoglu, A., Murphy, T.J. and Holsen, T.M. (2004). Dry Deposition Fluxes and Velocities of Polychlorinated Biphenyls (PCBs) Associated with Particles. *Atmos. Environ.* 38: 2447–2456.
- Tsai, C. and Cheng, Y.H. (1996). Comparison of Two Ambient Beta Gauge PM₁₀ Samplers. *J. Air Waste*

- Manage. Assoc.* 46: 142–147.
- Van den Berg, M., Birnbaum, L., Bosveld, A.T.C., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X.R., Liem, A.K.D., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Waern, F. and Zacharewski, T. (1998). Toxic Equivalency Factors (TEFs) for PCBs, PCDDs, PCDFs for Humans and Wildlife. *Environ. Health Perspect.* 106: 775–792.
- Van den Berg, M., Birnbaum, L.S., Denison, M., De Vito, M., Farland, W., Feeley, M., Fiedler, H., Hakansson, H., Hanberg, A. and Haws, L. (2006). The 2005 World Health Organization Reevaluation of Human and Mammalian Toxic Equivalency Factors for Dioxins and Dioxin-like Compounds. *Toxicol. Sci.* 93: 223–241.
- Van Thuong, N., Nam, V.D., Hue, N.T.M., Son, L.K., Van Thuy, N., Tung, H.D., Tuan, N.A., Minh, T.B., Huy, D.Q. and Minh, N.H. (2014). The Emission of Polychlorinated Dibenzo-*p*-dioxins and Polychlorinated Dibenzofurans from Steel and Cement-Kiln Plants in Vietnam. *Aerosol Air Qual. Res.* 14: 1189–1198.
- Wang, J.B., Chang-Chien, G.P., Lin, W.Y., Yeh, J.H. and Hung, C.H. (2009). A Seasonality Study of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Ambient Air in Kaohsiung (Taiwan) Clustered with Metallurgical Industries. *J. Hazard. Mater.* 162: 103–110.
- Wang, Q., Jin, Y., Li, X., Chen, J., Lu, S., Chen, T., Yan, J., Zhou, M. and Wang, H. (2014). PCDD/F Emissions from Hazardous Waste Incinerators in China. *Aerosol Air Qual. Res.* 14: 1152–1159.
- 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, J.L., Lin, T.C., Wang, L.C. and Chang-Chien, G.P. (2014). Memory Effects of Polychlorinated Dibenzo-*p*-dioxin and Furan Emissions in a Laboratory Waste Incinerator. *Aerosol Air Qual. Res.* 14: 1168–1178.
- 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.
- Yamasaki, H., Kuwata, K. and Miyamoto, H. (1982). Effects of Ambient Temperature on Aspects of Airborne Polycyclic Aromatic Hydrocarbons. *Environ. Sci. Technol.* 16: 189–194.

Received for review, April 20, 2015

Revised, June 3, 2015

Accepted, June 7, 2015