



## Atmospheric (Dry + Wet) Deposition of PCDD/Fs in Taiwan

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### ABSTRACT

Air pollution is becoming increasingly worse with economic development, and atmospheric deposition is an important mechanism for the removal of air pollutants. In 2017, the average dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan ranged between 57 (Lienchiang County in autumn) and 589 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City in winter), with an average of 221 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. The average total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan ranged between 65 (Lienchiang County in autumn) and 681 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City in winter), with an average of 263 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. The fractions of dry deposition fluxes contributing to the total deposition fluxes ranged between 37.8% (Yilan County in winter) and 99.9% (Kaohsiung City in winter), with an average of 82.1%. This study provided valuable information for the control strategies of PCDD/Fs.

**Keywords:** PM<sub>2.5</sub>; PCDD/Fs; Dry deposition; Total deposition.

### INTRODUCTION

Air pollution has been considered a danger to human health (morbidity, lung cancer, cardiovascular and cardiopulmonary diseases, etc.) for centuries (e.g., Dockery *et al.*, 1993; McDonnell *et al.*, 2000; Pope and Dockery, 2006; Pope *et al.*, 2009). The World Health Organization (WHO) estimates that ambient particulate matter (PM) is the world's 13th leading cause of death and contributes to approximately 800,000 premature deaths each year. The major effect of ambient PM on the pulmonary system is the exacerbation of inflammation, especially in susceptible people. Among particles of different sizes, the diameters of PM range from a few micrometres (µm) (PM<sub>2.5</sub> ranging from ~0 to 2.5 µm, PM<sub>10</sub> ranging from ~0 to 10 µm) to around 100 micrometres (TSP ranging from ~0 to 100 µm) (Chow *et al.*, 2015; Lu *et al.*, 2016).

Increased vehicular traffic and other combustion processes have resulted in a significant increase in ambient persistent organic pollutants (POPs) over the past two decades. Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are typical POPs that are not formed as a result of industrial activities, but

almost invariably as unwanted byproducts in all thermal systems and are distributed ubiquitously (U.S. EPA, 2003). They are generally formed unintentionally during a variety of anthropogenic combustion activities and industrial processes such as the manufacture and use of organochlorine chemicals (Hashimoto *et al.*, 1990; Lin *et al.*, 2014; Cheruiyot *et al.*, 2015, 2016). In addition, natural combustion processes such as forest fires, lightning and volcanic eruptions are thought to produce PCDD/Fs (Prange *et al.*, 2002; Kim *et al.*, 2003). Human exposure to high level PCDD/Fs may result in liver damage and chloracne in the short term (Marinković *et al.*, 2010), and may affect the endocrine, immune, reproductive, and nervous system in the long term (Srogi, 2008; Marinković *et al.*, 2010). Different PCDD/F sources are characterized by specific congeners. 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) was regulated as the most toxic congener, and the 2,3,7,8-substituted congeners (including 10 PCDDs and 7 PCDFs) are usually measured as molecular markers of specific sources to the environment (Alcock and Jones, 1996; Watanabe *et al.*, 1998; U.S. EPA, 2000). The atmosphere is an important pathway for the transport and global distribution of air pollutants. Once emitted to the receiving environment, atmospheric transport moves them away from their emission sources to where they pollute water and the soil and eventually enter to the food chain (Lohmann and Jones, 1998; Hu *et al.*, 2009).

Ambient pollutants are mainly removed by dry and wet deposition (Cheruiyot *et al.*, 2015, 2016). Atmospheric deposition is an important pathway for the air pollutants loading to the soil and water systems (Brzuzy and Hites,

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1996; Jurado *et al.*, 2005). PCDD/Fs partition between atmospheric particles and the corresponding gas phase in the atmosphere is an important factor in their transformation and transport (Pankow and Bidleman, 1991; Chang *et al.*, 2004; Lee *et al.*, 2008). The gas-particle partition is affected according to the ambient temperature, compound properties, humidity, vapor pressure and the particle surface available for sorption (Pankow, 1987; Lohmann *et al.*, 1999; Chang *et al.*, 2004). This process has a decisive influence on transport, deposition, and degradation processes (Bidleman *et al.*, 1988; Pankow *et al.*, 1994). The dry deposition fluxes of many substances appear to be dominated by large particles, even in the case of compounds with extremely low loading in this particle range (Shih *et al.*, 2006; Huang *et al.*, 2011), and the wet deposition fluxes are affected by the rainfall intensity and the levels of ambient pollutants (Kaupp and McLachlan, 1998).

Based on previous studies (Lee *et al.*, 2018), the objective of this study was to characterize the meteorological conditions in Taiwan, including the ambient temperature and rainfall, to estimate the seasonal variations in the monthly dry deposition fluxes in Taiwan; to measure the seasonal variations in the monthly total (dry and wet) deposition fluxes in Taiwan, and to compare the fraction of dry and wet deposition fluxes contributing to the total deposition fluxes.

## MATERIALS AND METHODS

### Sample Collection

In this study, the meteorological data, including ambient temperature and rainfall are from 48 local air quality stations associated with 9 cities and 13 counties during the year 2017 in Taiwan. The PCDD/Fs concentrations and gas-particle partitions were collected from previous study (Lee *et al.*, 2018a, b). The detailed information for the air quality stations include Dongshan and Yilan in Yilan County, Hualien in Hualien County, Kinmen in Kinmen County, Chushan, Nantou and Puli in Nantou County, Pingtung, Hungchun and Chaozhou in Pingtung County, Sani, Miaoli and Toufen in Miaoli County, Dayuan, Zhongli, Pingchen, Taoyuan, Longtan and Guanyin in Taoyuan City, Daliao, Siaogang, Renwu, Tsoying, Linyuan, Qianqiu, Meinung, Fuxing, Nanzih, Fongshan and Ciaotou in Kaohsiung City, Keelung in Keelung City, Matsu in Lienchiang County, Douliou, Lunbei and Mailiao in Yunlin County, Sanchong, Tucheng, Yonho, Xizhi, Banqiao, Linkou, Tamsui, Cailiao, Xindian, Xinzhuang and Wanli in New Taipei City, Hsinchu in Hsinchu City, Zhudong and Hukou in Hsinchu County, Chiayi in Chiayi City, Puzi and Xingang in Chiayi County, Erlin, Changhua and Siansi in Changhua County, Dali, Xitun, Shalu, Zhongming and Fengyuan in Taichung City, Shilin, Tatung, Zhongshan, Guting, Songshan, Yangming and Wanhua in Taipei City, Taitung and Guanshan in Taitung County, Annan, Shanhu, Xinying and Tainan in Tainan City, Magong in Penghu County.

### Atmospheric Dry Deposition of PCDD/Fs

The atmospheric dry deposition flux of PCDD/Fs is a

combination of both gas- and particle-phase fluxes, which are given by:

$$F_{d,T} = F_{d,g} + F_{d,p} \quad (1)$$

$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \quad (2)$$

$F_{d,T}$ : the total PCDD/F deposition flux contributed by the summation of both gas- and particle-phase fluxes;

$F_{d,g}$ : the PCDD/F deposition flux contributed by the gas phase;

$F_{d,p}$ : the PCDD/F deposition flux contributed by the particle phase;

$C_T$ : 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.

In this study, the mean dry deposition velocity of total PCDD/Fs ( $V_{d,T} = 0.42 \text{ cm s}^{-1}$ ) as proposed by Shih *et al.* (2006) is used. The dry deposition of gas-phase PCDD/Fs occurs mainly by diffusion. Due to a lack of measured data for PCDD/Fs, a selected value ( $0.010 \text{ cm s}^{-1}$ ) of gas-phase PAH dry deposition velocity,  $V_{d,g}$ , proposed by Sheu *et al.* (1996) and used by Lee *et al.* (1996) is also used here to calculate the PCDD/F dry deposition flux contributed by its gas phase. Dry deposition of particle-phase PCDD/Fs is mainly achieved through gravitational settling, and the dry deposition velocity of particle-phase PCDD/Fs,  $V_{d,p}$ , can be calculated using an equation.

## RESULTS AND DISCUSSION

### Dry Deposition Fluxes

Ambient temperature plays an important role in PCDD/Fs dry deposition fluxes. The seasonal variations of average ambient temperature in Taiwan are presented in Table 1. During 2017, the average ambient temperature ranged between  $17.82^\circ\text{C}$  (Lienchiang County) and  $25.42^\circ\text{C}$  (Kaohsiung City), with an average of  $22.67^\circ\text{C}$  in spring. In summer, the average ambient temperature ranged between  $27.10^\circ\text{C}$  (Nantou County) and  $29.86^\circ\text{C}$  (New Taipei City), with an average of  $29.12^\circ\text{C}$ . In autumn, the average ambient temperature ranged between  $25.04^\circ\text{C}$  (Yilan County) and  $27.52^\circ\text{C}$  (Kaohsiung City), with an average of  $25.95^\circ\text{C}$ . In winter, the average ambient temperature ranged between  $12.40^\circ\text{C}$  (Lienchiang County) and  $21.08^\circ\text{C}$  (Pingtung County), with an average of  $17.75^\circ\text{C}$ . In whole, the average ambient temperature in 2017 ranged between  $20.19^\circ\text{C}$  (Lienchiang County) and  $25.86^\circ\text{C}$  (Kaohsiung City), with an average of  $23.87^\circ\text{C}$ . The ambient temperature shows distinct seasonal variations, but the temperature difference is relatively small.

Based on the PCDD/F concentrations and ambient temperature, the dry deposition fluxes were calculated by

**Table 1.** The seasonal variations of average ambient temperature in Taiwan during 2017 (Unit: °C).

Season	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Spring	21.72	22.58	20.72	23.06	25.22	21.69	21.57	25.42
Summer	29.36	29.65	29.57	27.10	29.24	28.51	29.28	29.76
Autumn	25.04	26.12	25.84	26.21	27.40	25.07	25.39	27.52
Winter	17.38	19.16	14.95	18.21	21.08	16.46	16.73	20.74
Average	23.37	24.37	22.77	23.64	25.73	22.93	23.24	25.86
Season	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	Chiayi County
Spring	20.93	17.82	24.04	22.10	22.00	22.23	24.32	24.39
Summer	29.26	27.27	29.68	29.86	29.56	29.06	29.13	29.64
Autumn	25.26	23.28	26.87	25.86	25.85	25.29	26.19	26.76
Winter	16.71	12.40	18.67	17.56	17.07	17.15	18.85	18.52
Average	23.04	20.19	24.81	23.85	23.62	23.43	24.62	24.82
Season	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	Average	
Spring	23.44	23.28	21.40	22.91	24.73	23.22	22.67	
Summer	29.51	28.58	28.99	28.97	29.65	29.02	29.12	
Autumn	26.62	25.98	25.06	26.08	27.05	26.12	25.95	
Winter	18.01	18.14	16.84	19.36	19.17	17.31	17.75	
Average	24.39	24.00	23.07	24.33	25.15	23.92	23.87	

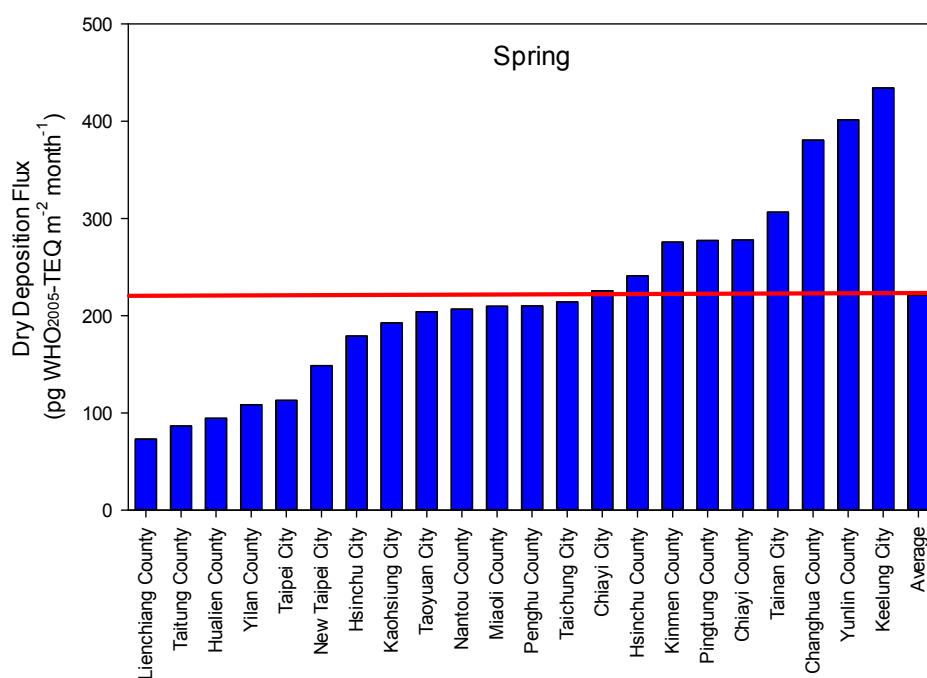
using the equations given above. Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in spring 2017 are presented in Fig. 1(A). The dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged between 73 (Lienchiang County) and 434 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Yunlin County), with an average of 221 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in summer 2017 are presented in Fig. 1(B). The dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged between 83 (Chiayi City) and 531 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 190 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in autumn 2017 are presented in Fig. 1(C). The dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged between 57 (Lienchiang County) and 418 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Yunlin County), with an average of 203 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in winter 2017 are presented in Fig. 1(D). The dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged between 66 (Lienchiang County) and 588 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 269 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. In whole, the average dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan ranged between 57 (Lienchiang County) and 588 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 221 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. There are distinct seasonal variations suggesting that the dry deposition fluxes are higher in cool seasons (spring and winter) than that in warm seasons (summer and autumn). This is probably due to the fact that a lower fraction of PCDD/Fs is adsorbed to particles when temperatures are warmer (Koester and Hites, 1992).

The dry deposition fluxes were similar to those for

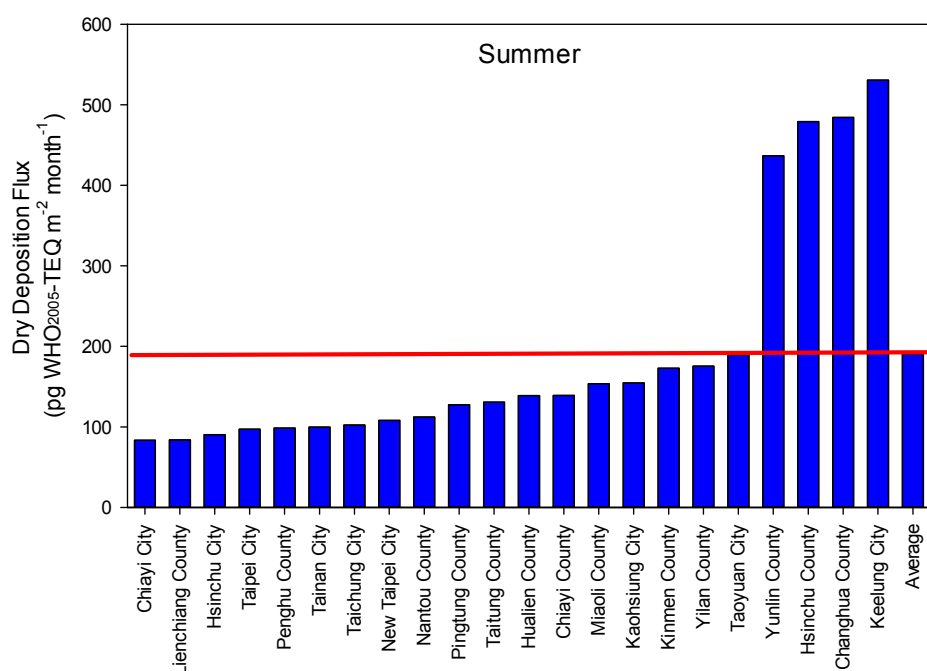
industrial (22.1–632 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) and rural areas (7.8–418 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in Taiwan in 2014 and 2015 (Lee *et al.*, 2016), Guangzhou (60.6–558 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in China during 2014 (Zhu *et al.*, 2017a), commercial (7.67–18.2 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>), coastal (6.69–11.6 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>), and those for agricultural areas (5.73–15.6 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) in Taiwan in 2006 (Wang *et al.*, 2010). They were lower than those in industrial areas (17.6–40.6 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) in Taiwan in 2006 (Wang *et al.*, 2010), urban areas (3.72–56.8 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) in Taiwan in 2009 (Wu *et al.*, 2009), Nanjing (104–1160 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>), and in Shijiazhuang (699–2230 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) and Harbin (208–1290 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in China in 2014 (Zhu *et al.*, 2017a, b). However they were higher than those in coastal (33.6–104.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) and mountain areas (3.9–27.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in Taiwan in 2013 (Chandra Suryani *et al.*, 2015), urban (2.7–10.1 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) and suburban areas (3.0–12.6 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) in Korea in 2002 (Moon *et al.*, 2005), urban areas (1.5 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) in USA during 2003 and 2004 (Correa *et al.*, 2006), industrial area (0–9.2 pg I-TEQ m<sup>-2</sup> day<sup>-1</sup>) in Italy in 1998 and 1999 (Guerzoni *et al.*, 2004). A comparison of atmospheric dry deposition flux in different areas is shown in Table 3.

#### Total Deposition Fluxes

Ambient temperature and rainfall are important factors that affect atmospheric dry and wet deposition fluxes. The monthly seasonal variations in the number of rainy days in Taiwan during 2017 are presented in Table 2. During 2017, the average monthly rainy days ranged between 5.7 (Kaohsiung City) and 18.3 (Keelung City), with an average of 12.0 in spring. In summer, the average monthly



**Fig. 1(A).** Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in spring 2017.

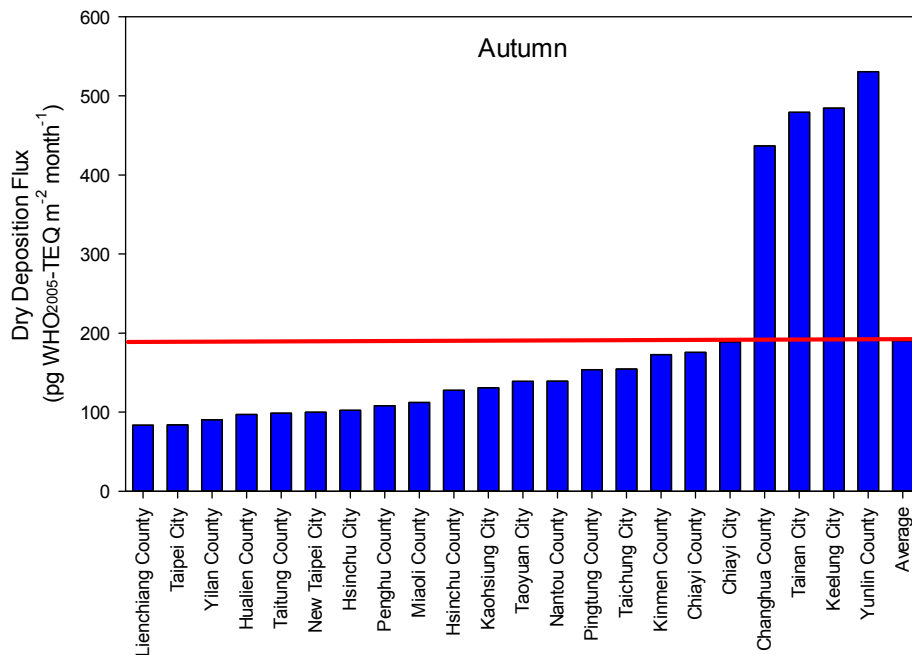


**Fig. 1(B).** Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in summer 2017.

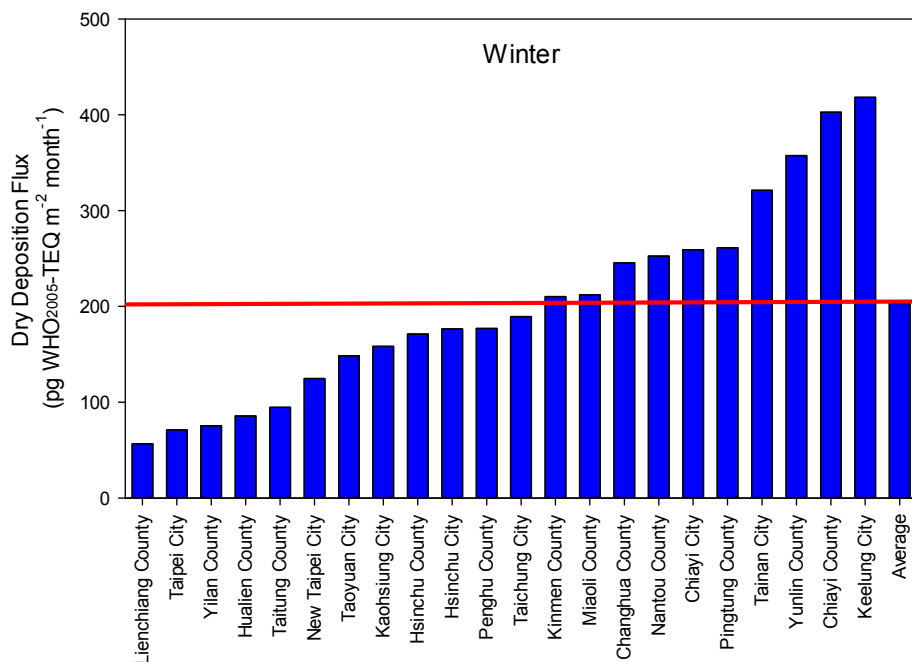
rainy days ranged between 6.7 (Taoyuan County) and 16.9 (Chiayi City and Chiayi County), with an average of 11.7. In autumn, the average monthly rainy days ranged between 3.3 (Tainan City) and 19.7 (Keelung City), with an average of 9.4. In winter, the average monthly rainy days ranged between 0.3 (Kaohsiung City) and 19.2 (Yilan County), with an average of 7.3. In whole, the average monthly rainfall in various areas in Taiwan ranged between 0.3 (Kaohsiung City) and 19.7 (Keelung City), with an average of 10.1.

Atmospheric total deposition fluxes of total-PCDD/Fs-

WHO<sub>2005</sub>-TEQ are a combination of dry and wet deposition. The wet deposition data used here is from previous study (Lee *et al.*, 2018a). The atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in spring of 2017 are presented in Fig. 2(A). The total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged between 102 (Taitung County) and 485 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 274 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in



**Fig. 1(C).** Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in autumn 2017.



**Fig. 1(D).** Atmospheric dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in winter 2017.

Taiwan in summer of 2017 are presented in Fig. 2(B). They ranged between 111 (Hsinchu City) and 667 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Changhua County), with an average of 253 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in autumn of 2017 are presented in Fig. 2(C). They ranged between 65 (Lienchiang County) and 453 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 230 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in

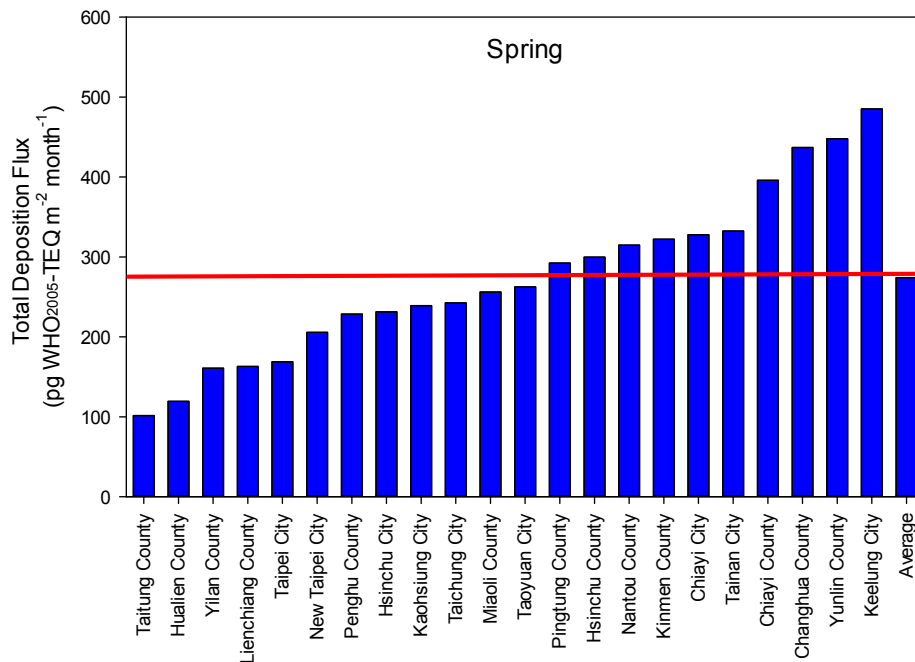
various areas in Taiwan in winter of 2017 are presented in Fig. 2(D). They ranged between 81 (Lienchiang County) and 681 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 296 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. In whole, the average total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan ranged between 65 (Lienchiang County) and 681 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> (Keelung City), with an average of 263 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. The total deposition fluxes detected in this study are similar to those measured in mountainous and in

**Table 2.** The monthly seasonal variations of rainy days in Taiwan during 2017.

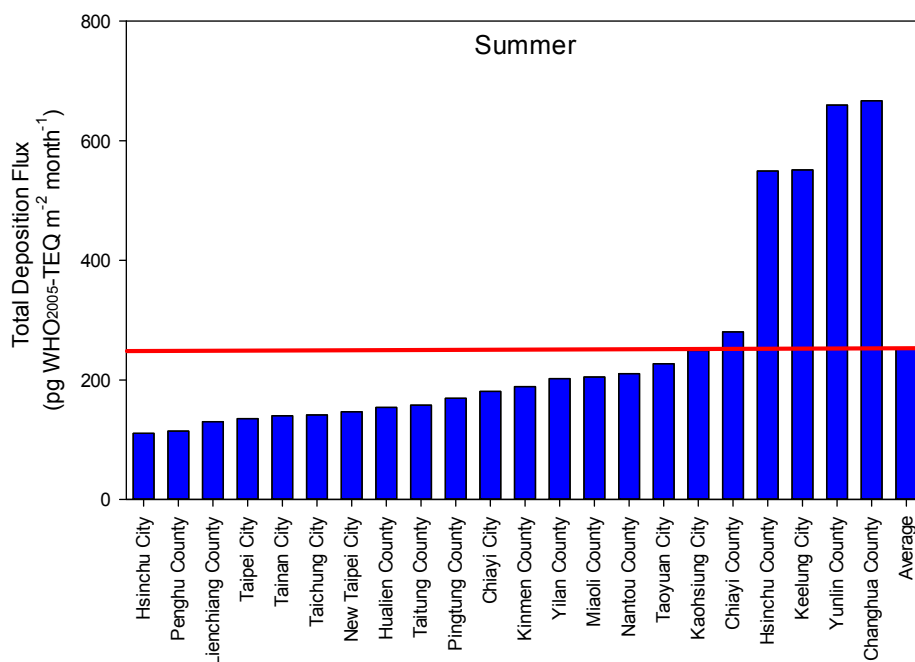
Area	Spring	Summer	Autumn	Winter
Keelung City	18.3	9.3	19.7	18.3
Taipei City	16.6	11.9	19.3	17.7
New Taipei City	14.3	11.2	14.0	8.7
Taoyuan City	12.3	6.7	11.0	8.7
Hsinchu City	11.7	9.3	10.0	8.0
Hsinchu County	11.7	9.3	10.0	8.0
Miaoli County	11.3	11.3	6.7	4.3
Taichung City	9.2	11.3	3.8	3.7
Nantou County	14.0	16.7	7.0	5.7
Changhua County	7.3	12.3	3.7	3.3
Yunlin County	8.0	15.3	5.0	2.3
Chiayi City	12.9	16.9	9.9	5.2
Chiayi County	12.9	16.9	9.9	5.2
Tainan City	6.0	13.0	3.3	1.7
Kaohsiung City	5.7	11.3	6.3	0.3
Pingtung County	6.7	15.3	9.2	2.3
Yilan County	17.8	10.5	17.5	19.2
Hualien County	17.7	9.0	12.0	13.3
Taitung County	16.6	11.9	12.7	12.6
Lienchiang County	13.3	9.7	6.7	6.3
Kinmen County	12.0	9.0	4.3	3.3
Penghu Country	7.3	10.2	4.3	2.7
Average	12.0	11.7	9.4	7.3

**Table 3.** The comparisons of atmospheric dry deposition flux in different area.

Area	Location	Period	Range (pg WHO <sub>2005</sub> -TEQ m <sup>-2</sup> month <sup>-1</sup> )	Average (pg WHO <sub>2005</sub> -TEQ m <sup>-2</sup> month <sup>-1</sup> )	Reference
Taiwan	Hengchun	2012	22.7–82.2	50.2	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Hengchun	2013	33.6–104.3	64.0	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Lulin	2012	4.50–31.3	11.8	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Lulin	2013	3.90–27.3	10.6	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Kaohsiung	2014	22.6–555.0	210.7	Lee <i>et al.</i> , 2016
Taiwan	Kaohsiung	2015	32.1–404.0	182	Lee <i>et al.</i> , 2016
Taiwan	Meinong	2014	9.7–418.0	148	Lee <i>et al.</i> , 2016
Taiwan	Meinong	2015	7.80–373.0	129	Lee <i>et al.</i> , 2016
Taiwan	Xiaogang	2014	22.1–632	234	Lee <i>et al.</i> , 2016
Taiwan	Xiaogang	2015	31.1–469	190.8	Lee <i>et al.</i> , 2016
China	Guangzhou	2014	60.6–558	205.8	Zhu <i>et al.</i> , 2017a
China	Nanjing	2014	104–1160	540.0	Zhu <i>et al.</i> , 2017a
China	Harbin	2014	308–1290	686.7	Zhu <i>et al.</i> , 2017b
China	Shijiazhuang	2014	699–2230	1283.3	Zhu <i>et al.</i> , 2017b
Taiwan	Mailiao	2014	197.4–847.2	472.3	Chen <i>et al.</i> , 2017
Taiwan	Mailiao	2015	238.1–645.6	456.7	Chen <i>et al.</i> , 2017
Taiwan	Mailiao	2016	239.5–604.4	413.8	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2014	163.6–627.0	398.5	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2015	156.1–498.5	342.1	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2016	178.3–433.9	319.8	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2014	137.7–636.8	343.4	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2015	151.3–434.0	301.3	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2016	165.9–385.9	287.7	Chen <i>et al.</i> , 2017
China	Handan	2016	467–1959	965.8	Zhao <i>et al.</i> , 2018
China	Handan	2017	489–1765	1001.7	Zhao <i>et al.</i> , 2018
China	Kaifeng	2016	354–1213	802.5	Zhao <i>et al.</i> , 2018
China	Kaifeng	2017	377–1140	757.8	Zhao <i>et al.</i> , 2018



**Fig. 2(A).** Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in spring 2017.



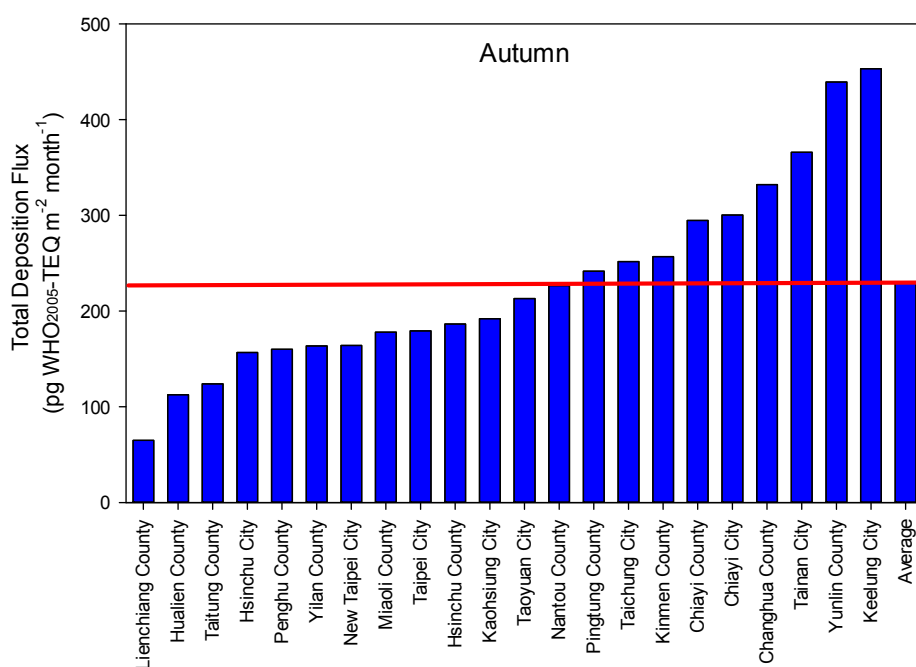
**Fig. 2(B).** Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in summer 2017.

rural areas (5020–5110 pg I-TEQ m<sup>-2</sup> year<sup>-1</sup>) in Taiwan in 2009 and 2010 (Huang *et al.*, 2011), Tanzawa (5700 pg I-TEQ m<sup>-2</sup> year<sup>-1</sup>) in Japan in 2001 (Ogura *et al.*, 2001), Lagoon (4280 pg I-TEQ m<sup>-2</sup> year<sup>-1</sup>) in France in 2001 (Guerzoni *et al.*, 2004), Guangzhou (3043 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> year<sup>-1</sup>) and Nanjing (7610 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> year<sup>-1</sup>) in China in 2014 (Zhu *et al.*, 2017a). These values were higher than those in coastal (657 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> year<sup>-1</sup>) and mountainous areas (249 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> year<sup>-1</sup>) in Taiwan in 2013 (Chandra Suryani *et al.*,

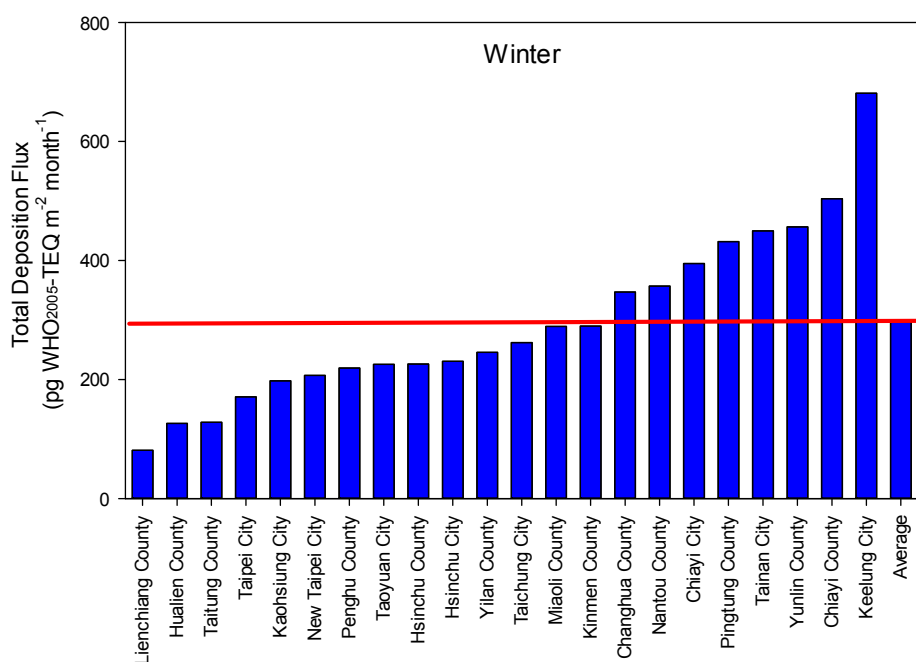
2015). But lower than that in urban area in Tokyo (17000 pg I-TEQ m<sup>-2</sup> year<sup>-1</sup>) and Yokohama (11000 pg I-TEQ m<sup>-2</sup> year<sup>-1</sup>) in Japan in 2001 (Ogura *et al.*, 2001), Shijiazhuang (16100 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> year<sup>-1</sup>) in China in 2014 (Zhu *et al.*, 2017b). A comparisons of atmospheric total deposition flux in different areas is provided in Table 4.

#### **Fractions of Dry and Wet Deposition**

The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition



**Fig. 2(C).** Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in autumn 2017.



**Fig. 2(D).** Atmospheric total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in various areas in Taiwan in winter 2017.

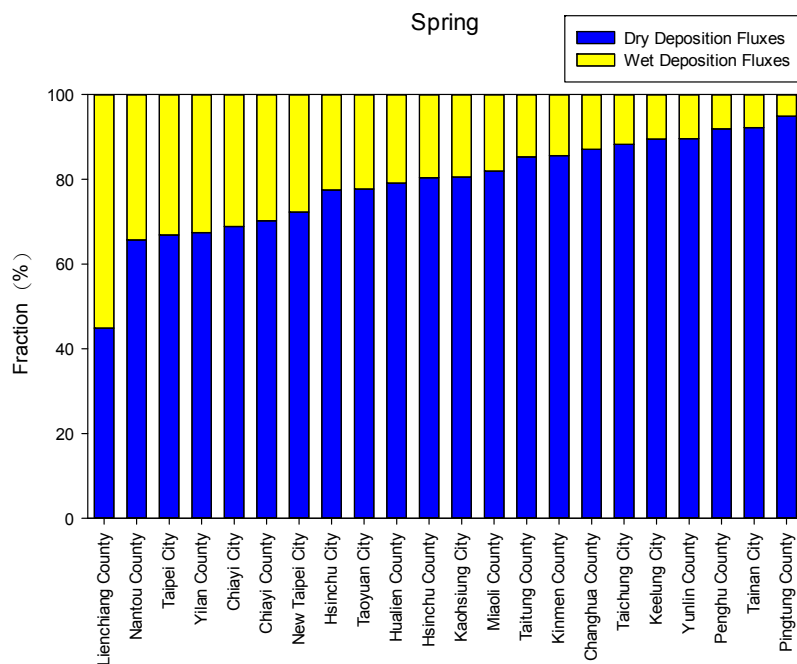
in spring are present in Fig. 3(A), the fractions of dry deposition fluxes contribute to the total deposition fluxes range between 44.9% (Lienchiang County) and 94.9% (Pingtung County), with an average of 79.0%. The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in summer are present in Fig. 3(B), the fractions of dry deposition fluxes contribute to the total deposition fluxes range between 46.0% (Chiayi City) and 96.3% (Keelung City), with an average of 74.5%. The fraction of total deposition flux in

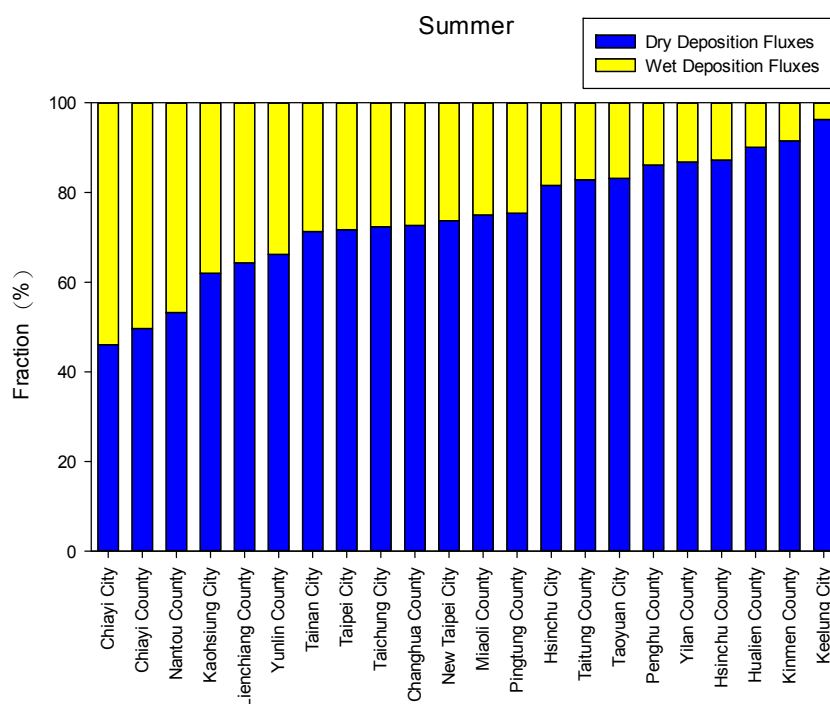
total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in autumn are present in Fig. 3(C), the fractions of dry deposition fluxes contribute to the total deposition fluxes range between 39.7% (Taipei City) and 98.8% (Penghu County), with an average of 86.2%. The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in winter are present in Fig. 3(D), the fractions of dry deposition fluxes contribute to the total deposition fluxes range between 37.8% (Yilan County) and 99.9% (Kaohsiung City), with an



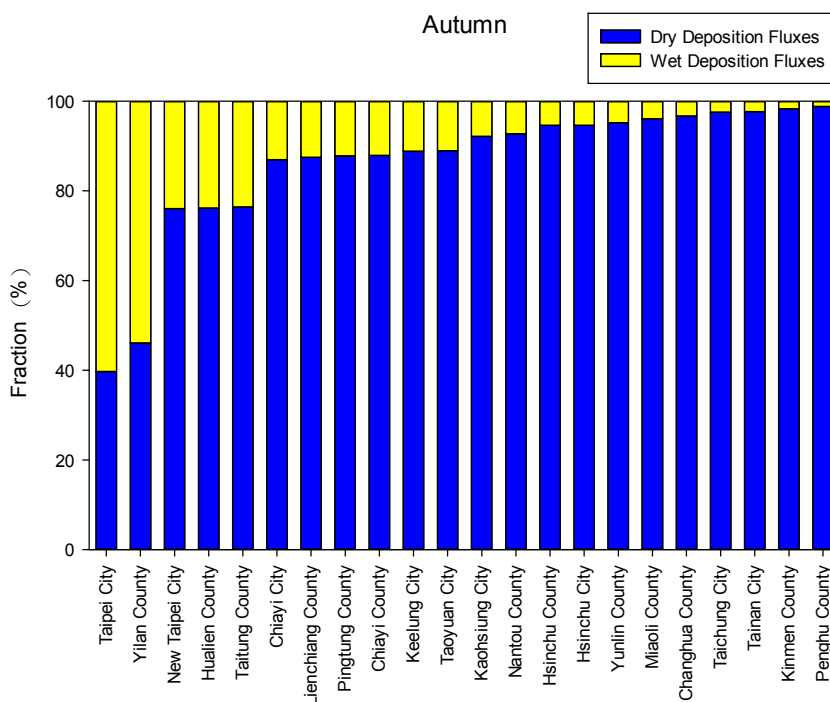
**Table 4.** The Comparisons of Atmospheric Total (wet + dry) Deposition of Total WHO<sub>2005</sub>-TEQ Flux in Different Area.

Area	Location	Period	Range (pg WHO <sub>2005</sub> -TEQ m <sup>-2</sup> month <sup>-1</sup> )	Average (pg WHO <sub>2005</sub> -TEQ m <sup>-2</sup> month <sup>-1</sup> )	Reference
Taiwan	Hengchun	2012	26.9–100.2	62.8	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Hengchun	2013	42.8–112.4	74.7	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Lulin	2012	12.1–33.7	20.1	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Lulin	2013	9.20–30.5	18.6	Chandra Suryani <i>et al.</i> , 2015
Taiwan	South	2012	100.3–408.4	–	Tseng <i>et al.</i> , 2014
Taiwan	South	2013	83.9–428.3	–	Tseng <i>et al.</i> , 2014
Taiwan	Kaohsiung	2014	75.0–555	254	Lee <i>et al.</i> , 2016
Taiwan	Kaohsiung	2015	46.4–400	220	Lee <i>et al.</i> , 2016
Taiwan	Meinong	2014	44.0–420	172	Lee <i>et al.</i> , 2016
Taiwan	Meinong	2015	36.3–376	155	Lee <i>et al.</i> , 2016
Taiwan	Xiaogang	2014	33.3–640	280	Lee <i>et al.</i> , 2016
Taiwan	Xiaogang	2015	47.7–480	230	Lee <i>et al.</i> , 2016
China	Guangzhou	2014	97.7–559	253.6	Zhu <i>et al.</i> , 2017a
China	Nanjing	2014	135–1250	643	Zhu <i>et al.</i> , 2017a
China	Harbin	2014	343–1290	718	Zhu <i>et al.</i> , 2017b
China	Shijiazhuang	2014	713–2230	1340	Zhu <i>et al.</i> , 2017b
Taiwan	Mailiao	2014	223.1–847.2	493.8	Chen <i>et al.</i> , 2017
Taiwan	Mailiao	2015	242.5–660.8	497.8	Chen <i>et al.</i> , 2017
Taiwan	Mailiao	2016	274.5–633.4	466.8	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2014	185.5–627.6	421.3	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2015	161.0–499.3	366.2	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2016	196.8–523.0	354.3	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2014	151.1–637.8	365.2	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2015	157.4–454.5	329.2	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2016	533.7–190.8	335.5	Chen <i>et al.</i> , 2017
China	Handan	2016	545–2046	1048.3	Zhao <i>et al.</i> , 2018
China	Handan	2017	534–1813	1052.5	Zhao <i>et al.</i> , 2018
China	Kaifeng	2016	391–1227	867.5	Zhao <i>et al.</i> , 2018
China	Kaifeng	2017	412–1395	825.8	Zhao <i>et al.</i> , 2018

**Fig. 3(A).** The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in spring, respectively.



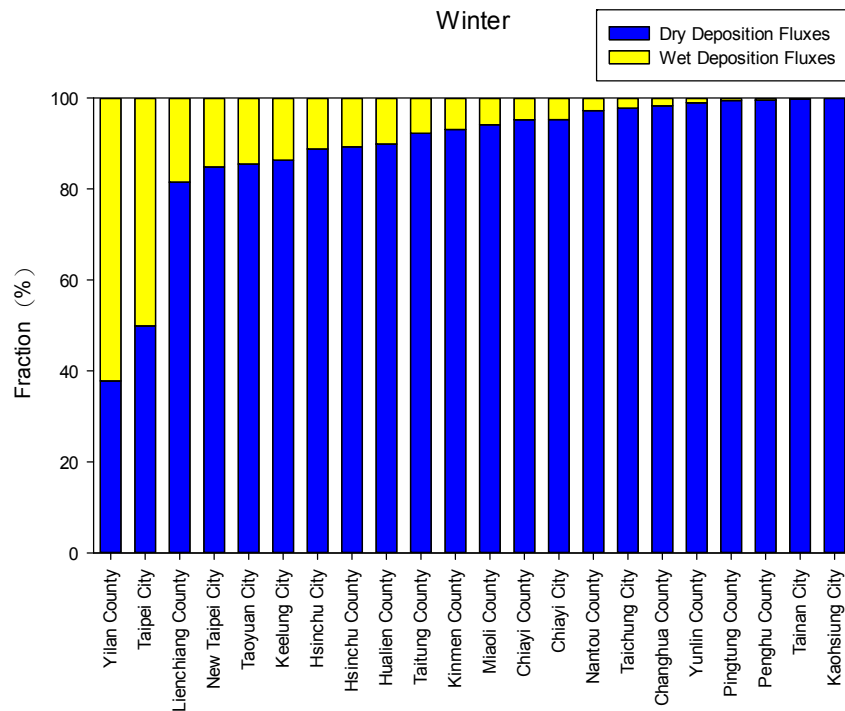
**Fig. 3(B).** The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in summer, respectively.



**Fig. 3(C).** The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in autumn, respectively.

average of 88.8%. In whole, the fractions of dry deposition fluxes contribute to the total deposition fluxes range between 37.8% (Yilan County) and 99.9% (Kaohsiung City), with an average of 82.1%. The fractions of total deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ are major contributed by

dry deposition fluxes like previous studies (Wang *et al.*, 2010; Chandra Suryani *et al.*, 2015; Zhu *et al.*, 2017a, b). The contribution fractions of dry deposition on the atmospheric total (Wet + Dry) WHO<sub>2005</sub>-TEQ deposition published on the previous documents were shown in Table 5.



**Fig. 3(D).** The fraction of total deposition flux in total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by the dry and wet deposition in winter, respectively.

**Table 5.** The contribution fractions of dry deposition on the atmospheric total (dry + wet) WHO<sub>2005</sub>-TEQ deposition.

Area	Location	Period	Range (%)	Average (%)	Reference
Taiwan	Hengchun	2012	30.5–99.8	78.2	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Hengchun	2013	51.7–99.1	78.9	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Lulin	2012	19.2–97.0	56.5	Chandra Suryani <i>et al.</i> , 2015
Taiwan	Lulin	2013	23.6–98.6	69.1	Chandra Suryani <i>et al.</i> , 2015
Taiwan	South	2012	12.1–98.7	67.7	Tseng <i>et al.</i> , 2014
Taiwan	South	2013	12.4–100	73.7	Tseng <i>et al.</i> , 2014
Taiwan	Kaohsiung	2014	-	83.0	Lee <i>et al.</i> , 2016
Taiwan	Kaohsiung	2015	-	82.7	Lee <i>et al.</i> , 2016
Taiwan	Meinong	2014	-	86.0	Lee <i>et al.</i> , 2016
Taiwan	Meinong	2015	-	83.2	Lee <i>et al.</i> , 2016
Taiwan	Xiaogang	2014	-	83.6	Lee <i>et al.</i> , 2016
Taiwan	Xiaogang	2015	-	83.0	Lee <i>et al.</i> , 2016
China	Guangzhou	2014	50.6–99.8	78.6	Zhu <i>et al.</i> , 2017a
China	Nanjing	2014	66.5–98.6	82.4	Zhu <i>et al.</i> , 2017a
China	Harbin	2014	99.7–81.7	-	Zhu <i>et al.</i> , 2017b
China	Shijiazhuang	2014	84.3–100	-	Zhu <i>et al.</i> , 2017b
Taiwan	Mailiao	2014	76.2–100	93.9	Chen <i>et al.</i> , 2017
Taiwan	Mailiao	2015	68.8–99.8	91.3	Chen <i>et al.</i> , 2017
Taiwan	Mailiao	2016	74.9–98.4	88.6	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2014	72.0–100	93.3	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2015	73.5–99.8	92.5	Chen <i>et al.</i> , 2017
Taiwan	Lunbei	2016	77.1–98.6	90.8	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2014	75.0–99.9	92.7	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2015	71.7–99.8	90.4	Chen <i>et al.</i> , 2017
Taiwan	Taisi	2016	69.6–97.7	86.6	Chen <i>et al.</i> , 2017
China	Handan	2016	60.8–100	-	Zhao <i>et al.</i> , 2018
China	Handan	2017	88.0–99.9	-	Zhao <i>et al.</i> , 2018
China	Kaifeng	2016	78.5–99.9	90.9	Zhao <i>et al.</i> , 2018
China	Kaifeng	2017	84.5–99.7	91.8	Zhao <i>et al.</i> , 2018

## CONCLUSION

1. The average dry deposition fluxes of total-PCDD/Fs- $\text{WHO}_{2005}$ -TEQ in various areas in Taiwan range between 57 (Lienchiang County) and 588  $\text{pg WHO}_{2005}\text{-TEQ m}^{-2}\text{ month}^{-1}$  (Keelung City), with an average of 221  $\text{pg WHO}_{2005}\text{-TEQ m}^{-2}\text{ month}^{-1}$ .
2. The average total deposition fluxes of total-PCDD/Fs- $\text{WHO}_{2005}$ -TEQ in various areas in Taiwan range between 65 (Lienchiang County) and 681  $\text{pg WHO}_{2005}\text{-TEQ m}^{-2}\text{ month}^{-1}$  (Keelung City), with an average of 263  $\text{pg WHO}_{2005}\text{-TEQ m}^{-2}\text{ month}^{-1}$ .
3. The fractions of dry deposition fluxes contribute to the total deposition fluxes range between 37.8% (Yilan County) and 99.9% (Kaohsiung City), with an average of 82.1%.

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