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Wet Deposition of PCDD/Fs in Taiwan

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ABSTRACT

In 2017, the seasonal variations in the wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in ambient air were evaluated in Taiwan. The results showed the annual wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ to be 42.5 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the seasonal distributions were 53.3, 62.9, 26.7 and 27.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, autumn and winter, respectively. The average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ was 12300. There were obvious seasonal variations in S_{tot}, for which the values were 13840, 6540, 8280 and 20540 in spring, summer, autumn, and winter, respectively. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain were 0.453, 0.176, 0.218 and 0.649 pg WHO_{2005}-TEQ L^{-1} in spring, summer, autumn and winter, respectively. Atmospheric deposition is the major removal pathway for PCDD/Fs. The results of this study provide an evaluation of the adverse effects of PCDD/Fs exposure on human health, and provide a reason for the government to be concerned and to enact better control on air pollution.

Keywords: $PM_{2.5}$; PCDD/Fs; Wet deposition; Scavenging ratio.

INTRODUCTION

Particulate matter (PM) is a complex mixture of solid and liquid particles suspended in the air (Ghosh *et al.*, 2014). According to previous studies, the concentration of PM is affected by both its physical characteristics, including size morphology, composition and porosity, and its chemical characteristics, such as the composition of ionic components, trace metals, and organic carbon (Schmid *et al.*, 2007; Hu *et al.*, 2012). Depending on its aerodynamic diameters, PM is classified into four categories: $PM_{2.5}$ (less than 2.5 µm), PM_{10} (less than 10 µm), and TSP (total suspended particles, less than 100 µm). In recent decades, several epidemiological studies have interlinked a positive correlation between exposure and health impact. Moreover, due to its health impact, especially in the case of fine particulates ($PM_{2.5}$), PM has become one of the most notorious forms of air pollution (Saldarriaga-Norena *et al.*, 2009; Wang *et al.*, 2014; Liao *et al.*, 2015; Lu *et al.*, 2016). The major sources of PM can be divided into two major categories: naturally formed and anthropogenic. Natural sources include volcanic eruptions, wood burning, sea salts, and windblown particulates. On the other hand, vehicular emissions, industrial exhaust gases, power plants, and open burning are major anthropogenic sources (Pipalatkar *et al.*, 2014; Liu *et al.*, 2015; Wang *et al.*, 2015; Tseng *et al.*, 2016; Hsu *et al.*, 2017). According to Frank *et al.* (2006), the weather condition, including temperature, humidity, rain scavenging, and wind speed, and the local geography, such as topography and surface soil, can be important factors contributiong to ambient PM concentrations. Besides primary sources, directly emitted pollutants, the secondary sources, which are usually formed through photochemical reaction should also be taken into consideration. Therefore, environmental conditions and saturation ratios are important factors related to the ambient concentration of PM from both primary sources and secondary sources (Harrison *et al.*, 1997; Marcazzan *et al.*, 2001; Frank *et al.*, 2006).

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) have the similar structures and are defined as persistent organic pollutants (POPs). The emission sources of PCDD/Fs are very multivariate. Similar to PM, the sources of PCDD/Fs can be divided into anthropogenic sources and natural sources. According to previous research, anthropogenic activities account for the major emissions, including municipal solid waste incinerators, medical waste incinerators, traffic emission (Hashimoto *et al.*, 1990; Ni *et al.*, 1999; Oh *et al.*, 1999; Prange *et al.*, 2003; Chen *et al.*, 2017b). POPs are semi-volatile and hydrophobic compounds. These characteristics increase their resistance to environmental

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degradation and their ability to accumulate in soil, water, and food (Hu *et al.*, 2009; Cheruiyot *et al.*, 2015, 2016; Chi *et al.*, 2016; Redfern *et al.*, 2017). Moreover, PCDD/Fs tend to be adsorbed on the surfaces of particulate matter (Wang *et al.*, 2010; Huang *et al.*, 2011a; Lee *et al.*, 2016). According to Wang *et al.* (2010), ambient transport is the major PCDD/Fs disperse route. Besides their persistency, the toxicity of PCDD/Fs is also a major concern to scientists. The health impact of PCDD/Fs has been proven by several previous studies (Liem *et al.*, 2000; Montesano and Hall, 2001). Long-term exposure to PCDD/Fs can cause damage to the immune system, retardation in the development of the endocrine system, and destruction of the reproductive functions (Bock and Köhle, 2006). Since PCDD/Fs are semi-volatile compounds, gas-particle partitioning plays an important role. The partitions of PCDD/Fs include between surface soil and water and between gas and particle phases in the atmosphere, which usually differ due to different concentrations, pollutant properties, vapor pressures, and the atmospheric temperature (Pankow, 1987; Chang *et al.*, 2004; Lee *et al.*, 2016; Zhu *et al.*, 2017a, b). Both dry and wet deposition are important pathways for removing PCDD/Fs from the air to the soil and water system (Welsch-Pausch *et al.*, 1995; Horstmann and McLachlan, 1997; Lohmann and Jones, 1998; Ren *et al.*, 2007). The dry deposition is an important route for PCDD/Fs to transfer from air to land or water. While the wet deposition is a combination removal pathway of PCDD/Fs from vapor to rain and precipitation (Lohmann and Jones, 1998). Therefore, due to different weather conditions, the concentrations will vary from area to area. This study focuses on seasonal variations in atmospheric wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in different areas in Taiwan. In addition, seasonal variations in the scavenging ratio and the total-PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain in different areas in Taiwan are compared and discussed.

MATERIALS AND METHOD

Sample Collection

In this study, the meteorological data, including ambient temperature and rainfall, were collected from 48 local air quality stations in 9 cities and 13 counties in 2017 in Taiwan. The PCDD/F concentration and gas-particle partition data were retrieved from our previous study (Lee *et al.*, 2018). The name and the location of the 48 air quality stations are as follows: Dongshan and Yilan in Yilan County, Hualien in Hualien County, Kinmen in Kinmen County, Chushan, Nantou and Puli in Nantou County, Pingtung, Hungchun and Chaojhou 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, Shanhua, Xinying and Tainan in Tainan City, Magong in Penghu County.

Scavenging Ratio

Due to the slightly solubility, the flux of PCDD/F wet deposition in the form of take vapor dissolution into rain and the removal of suspended particulates by precipitation must be taken into consideration.

The gas scavenging ratio (S_g) can be estimated by:

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

S_g: the gas scavenging ratio of PCDD/Fs (dimensionless);
R: the universal gas constant (82.06×10^{-6} m^3 atm mol^{-1} K^{-1});
T: ambient temperature (K);
H: the Henry constant (m^3 atm mol^{-1}).

Moreover, meteorological factors and particle characteristics are both important influences of particle scavenging. The concentrations of the gas phase in the air (S_g) should also be taken into consideration. Therefore, the gas scavenging ratio can be calculated by:

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

S_g: the gas scavenging ratio of PCDD/Fs (dimensionless);
$C_{rain,dis}$: 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 is defined as the ratio of the concentration of the particle phase in a raindrop divided by the concentrations of the particle phase in the air, where S_p, can be calculated by:

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

where 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.

The total precipitation scavenging is the sum of gas and particle scavenging (S_{tot}) can be calculated by:

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

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

Due to a lack of real measured data or the particle scavenging ratios of PCDD/Fs, the S_p (S_p was 42000) values of OCDD and OCDF measured by Eitzer and Hites (1989) were averaged and used here.

Wet Deposition

Wet deposition is the removal of particles in the atmosphere by precipitation (rainfall and cloud droplets). Precipitation scavenging accounts for the majority of PCDD/Fs removed from the atmosphere by wet deposition (Huang, 2011b). Wet deposition flux of PCDD/Fs is a combination of both vapor dissolution into rain and removal of suspended particulates by precipitation (Bidleman, 1988; Koester and Hltes, 1992).

The wet deposition fluxes of PCDD/Fs can be evaluated by:

$$F_{w,T} = F_{w,dis} + F_{w,p} \quad (5)$$

$$F_{w,dis} = C_{rain,dis} \times Rainfall \quad (6)$$

$$F_{w,p} = C_{rain,particle} \times Rainfall \quad (7)$$

$F_{w,T}$: the wet deposition flux of PCDD/Fs from both vapor dissolution into rain and the 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

Wet Deposition Fluxes

Based on the PCDD/F concentrations, ambient temperature and rainfall, the wet deposition was calculated using Eq. (5). The monthly seasonal variations of average rainfall in Taiwan in 2017 are presented in Table 1. In 2017, the seasonal rainfall ranged between 0.2 (Kaohsiung City in winter) and 662.1 mm (both Chiayi County and Chiayi City in winter), and averaged 168.2 mm. In spring, the rainfall ranged between 41.8 (Pingtung County) and 220.2 mm (Nantou County), and averaged 119.7 mm. In summer, the rainfall ranged between 103.3 (Kinmen County) and 662.1 mm (Chiayi County and Chiayi City), averaged 331.0 mm. In autumn, the rainfall ranged between 17.7 (Penghu County) and 642.8 mm (Yilan County), averaged 159.4 mm. In winter, the rainfall ranged between 0.2 (Kaohsiung City) and 350.1 mm (Yilan County), averaged 62.5 mm.

Among the 22 areas in Taiwan, the seasonal average wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air range between 0.24 (Kaohsiung City in winter) and 222.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ (Yunlin County in summer), and with an average of 42.5 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in 2017. The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in spring 2017 are presented in Fig. 1(A), it can be seen that the wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ range between 14.9 (Taitung County) and 117.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ (Chiayi County), and with an average of 53.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring 2017. The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in summer 2017 are presented in Fig. 1(B), it can be seen that the wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ range between 15.3 (Hualien County) and 222.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ (Yunlin County), and with an average of 2.06 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in summer 2017. The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in autumn 2017 are presented in Fig. 1(C), it can be seen that the wet deposition fluxes of

Table 1. The monthly seasonal variations of average rainfall in Taiwan in 2017 (Unit: mm).

Area	Spring	Summer	Autumn	Winter
Keelung City	219.9	304.3	435.6	326.0
Taipei City	161.7	360.7	641.5	227.6
New Taipei City	145.3	341.7	204.4	68.7
Taoyuan City	117.0	258.3	89.6	58.0
Hsinchu City	126.0	241.8	50.5	46.3
Hsinchu County	126.0	241.8	50.5	46.3
Miaoli County	104.3	341.2	41.0	27.3
Taichung City	70.8	325.7	27.4	10.7
Nantou County	220.2	540.3	83.7	11.8
Changhua County	92.0	473.3	39.3	7.7
Yunlin County	70.5	523.7	53.7	4.7
Chiayi City	189.9	662.1	113.0	19.3
Chiayi County	189.9	662.1	113.0	19.3
Tainan City	52.3	318.3	26.8	1.0
Kaohsiung City	82.0	250.7	42.0	0.2
Pingtung County	41.8	280.4	154.7	3.8
Yilan County	167.8	188.1	642.8	350.1
Hualien County	108.9	167.5	326.0	47.0
Taitung County	87.9	264.2	300.4	37.6
Lienchiang County	134.2	221.6	33.9	27.2
Kinmen County	61.7	103.3	19.8	31.6
Penghu Country	63.5	210.9	17.7	2.7
Average	119.7	331.0	159.4	62.5

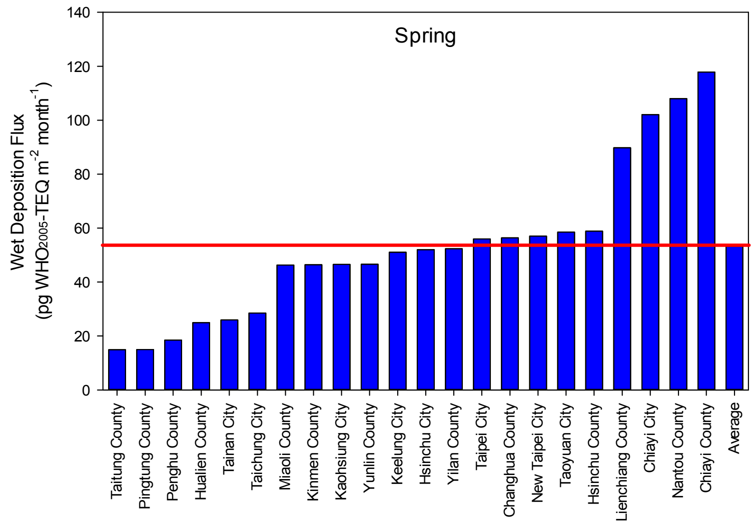

Fig. 1(A). Atmospheric wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in spring 2017.

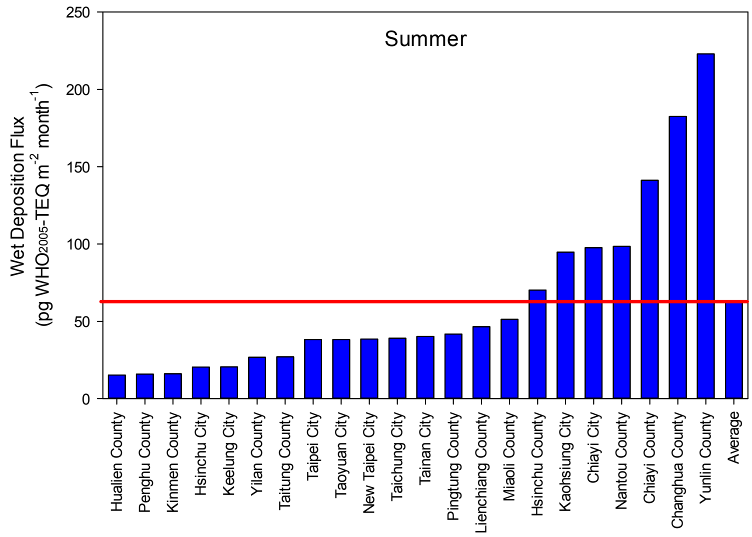

Fig. 1(B). Atmospheric wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in summer 2017.

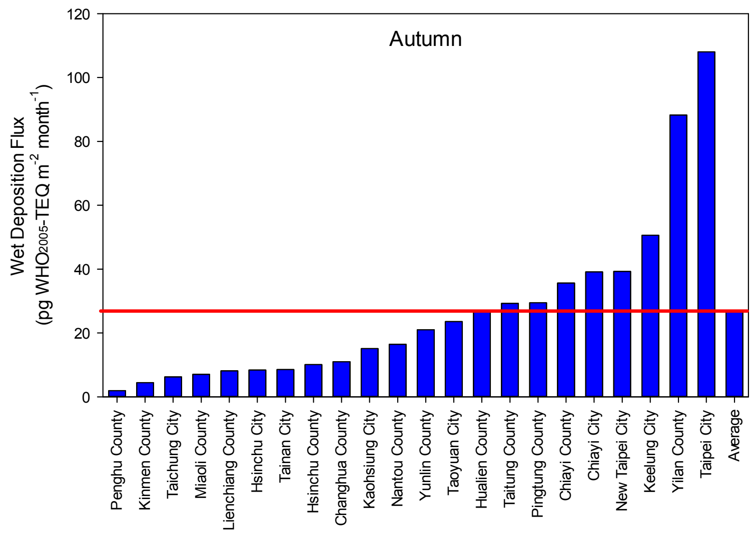

Fig. 1(C). Atmospheric wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in autumn 2017.

total-PCDD/Fs-WHO_{2005}-TEQ range between 1.90 (Penghu County) and 108.0 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ (Taipei City), and with an average of 26.7 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in autumn 2017. The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in winter 2017 are presented in Fig. 1(D), it can be seen that the wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ range between 0.24 (Kaohsiung City) and 152.8 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ (Yilan County), and with an average of 27.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in winter 2017.

On the whole, the wet deposition fluxes are higher in spring (averaged 53.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) and summer (averaged 62.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) than that in winter (averaged 27.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) and autumn (averaged 26.7 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). The wet deposition detected in this study are similar with previous studies in Taiwan, and the average wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ detected in previous studis are presented in Table 2. The wet deposition range of 0.0–69.7 and 1.8–71.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in rural areas (Meinong) in 2014 and 2015, respectively; while in industrial area (Xiaogang) the wet deposition range of 0.0–183 and 0.5–140 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in 2014 and 2015, respectively (Lee *et al.*, 2016). In the rural area of Taiwan, the wet deposition range of 5.4–92.2 and 5.5-120 pg I-TEQ m^{-2} $month^{-1}$ from 2009 to 2010 (Huang *et al.*, 2011b). In the coastal and mountain areas of Taiwan, the wet deposition was 0.8–30.1 (Hengchun) and 0.2–21.5 (Lulin) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in 2013, respectively (Chandra Suryani *et al.*, 2015).

Scavenging Ratio

Based on the ambient temperature and the PCDD/F concentrations in the rain, the scavenging ratio was calculated using Eqs. (1), (2), (3) and (4). The ambient temperature is an important factor for scavenging ratio. In 2017, the average ambient temperature range between 20.19 (Lienchiang County) and 25.86°C (Kaohsiung City), and with an average of 23.87°C. In spring, The average ambient temperature range between 17.82 (Lienchiang County) and 25.42°C (Kaohsiung City), and with an average of 22.67°C. In summer, the average ambient temperature range between 27.10 (Nantou County) and 29.86°C (New Taipei City), and with an average of 29.12°C. In autumn, the average ambient temperature range between 23.28 (Lienchiang County) and 27.52°C (Kaohsiung City), and with an average of 25.95°C. In winter, the average ambient temperature range between 12.40 (Lienchiang County) and 21.08°C (Pingtung County), and with an average of 17.75°C.

Among the 9 cities and 13 counties in Taiwan, the seasonal average scavenging ratio (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ in ambient air range between 4550 (Hsinchu County in summer) and 24630 (Lienchiang County in winter), and with an average of 12300 in 2017. The average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ in spring 2017 are presented in Fig. 2(A), where it can be seen that the average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ range between 9550 (Taitung County) and 19830 (Lienchiang County), and with an average of 13840 in spring 2017. The average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ in summer 2017 are presented in Fig. 2(B), where it can be seen that the average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ range between 4550 (Hsinchu County) and 8410 (Chiayi City), and with an average of 6540 in summer 2017. The average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ in autumn 2017 are presented in Fig. 2(C), where it can be seen that the average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ range between 6270 (Hualien County) and 10740 (Lienchiang County), and with an average of 8280 in autumn 2017. The average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ in winter 2017 are presented in Fig. 2(D), where it can be seen that the average S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ range between 14250 (Taitung County) and 24630 (Lienchiang County), and with an average of 20540 in winter 2017.

There are distinctive seasonal variations of S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ, which the S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ are higher in cool season (averaged 20540 and 13840 in winter and spring, respectively) than that in warm season (averaged 6540 and 8280 in summer and

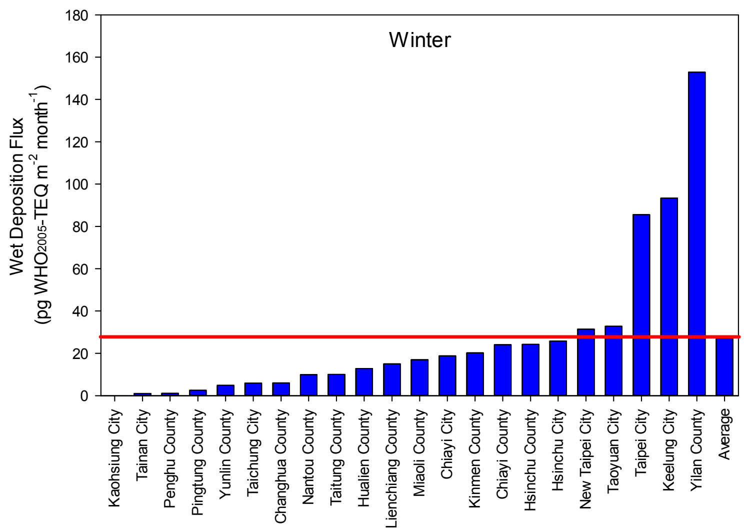

Fig. 1(D). Atmospheric wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in winter 2017.

Table 2. The wet weposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in previous studies.

Area	Location	Period	Range (pg WHO_{2005}-TEQ m^{-2} $month^{-1}$)	Average (pg WHO_{2005}-TEQ m^{-2} $month^{-1}$)	Reference
Taiwan	Hengchun	2012	0.100–63.2	12.6	Chandra Suryani *et al.*, 2015
Taiwan	Hengchun	2013	0.800–30.1	10.8	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2012	0.500–19.7	8.25	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2013	0.200–21.5	7.98	Chandra Suryani *et al.*, 2015
Taiwan	Kaohsiung	2014	0–149	42.6	Lee *et al.*, 2016
Taiwan	Kaohsiung	2015	0–119	34.4	Lee *et al.*, 2016
Taiwan	Meinong	2014	0–69.7	24.1	Lee *et al.*, 2016
Taiwan	Meinong	2015	1.80–71.3	26.5	Lee *et al.*, 2016
Taiwan	Xiaogang	2014	0–183	47.0	Lee *et al.*, 2016
Taiwan	Xiaogang	2015	0.500–140	42.9	Lee *et al.*, 2016
China	Guangzhou	2014	0.580–229	47.5	Zhu *et al.*, 2017a
China	Nanjing	2014	10.7–266	103	Zhu *et al.*, 2017a
China	Harbin	2014	3.00–79.9	30.8	Zhu *et al.*, 2017b
China	Shijiazhuang	2014	0–140	51.8	Zhu *et al.*, 2017b
Taiwan	Mailiao	2014	0–89.9	21.5	Chen *et al.*, 2017
Taiwan	Mailiao	2015	1.00–159	41.0	Chen *et al.*, 2017
Taiwan	Mailiao	2016	8.40–145	53.0	Chen *et al.*, 2017
Taiwan	Lunbei	2014	0–97.1	22.8	Chen *et al.*, 2017
Taiwan	Lunbei	2015	0.800–86.7	24.1	Chen *et al.*, 2017
Taiwan	Lunbei	2016	5.30–112.8	34.5	Chen *et al.*, 2017
Taiwan	Taisi	2014	0.400–75.9	21.8	Chen *et al.*, 2017
Taiwan	Taisi	2015	0.900–76.8	27.9	Chen *et al.*, 2017
Taiwan	Taisi	2016	8.30–155	47.8	Chen *et al.*, 2017
China	Wuhu	2015	46.2–374	128	Wang *et al.*, 2018
China	Bengbu	2015	13.6–278	127	Wang *et al.*, 2018

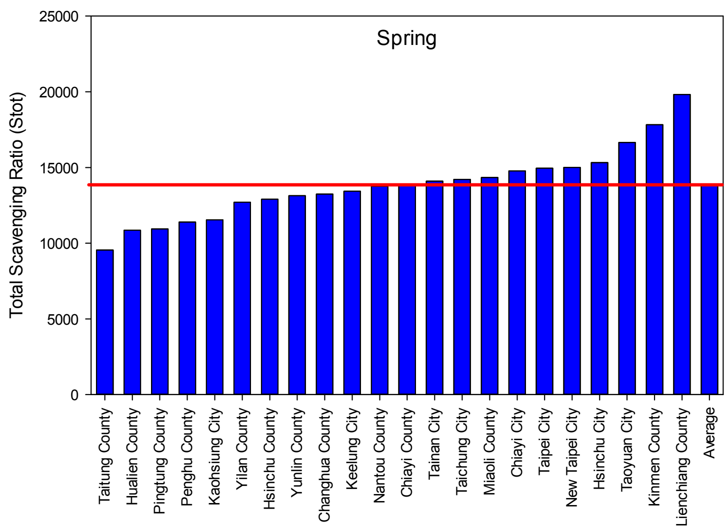

Fig. 2(A). The scavenging ratio (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in spring 2017.

autumn, respectively). The S_{tot} values show a negative correlation with the ambient temperature (Bidleman, 1988; Tseng *et al.*, 2014). It is due to the higher temperature, the greater fraction in the gas phase, and the fact that the S_{tot} in gas phase is less than that in particle phase. Comparisions of the S_{tot} of total-PCDD/Fs-WHO_{2005}-TEQ with previous works are shown in Table 3, where it can be seen that the values detected in this study are similar to those in Lulin (13450) and Yunlin (13900), but higher than those in Xiaogang (6840), Hengchung (8015) and Meinong (4700) (Chandra Suryani *et al.*, 2015; Lee *et al.*, 2016; Chen *et al.*, 2017a).

PCDD/F Concentration in the Rain

According to Eqs. (2) and (3), the total-PCDD/Fs-WHO_{2005}-TEQ concentrations of dissolved and particle in

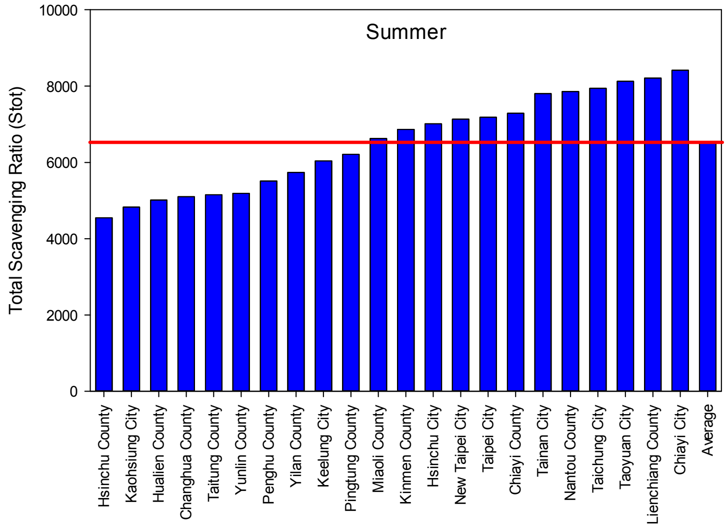

Fig. 2(B). The scavenging ratio (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in summer 2017.

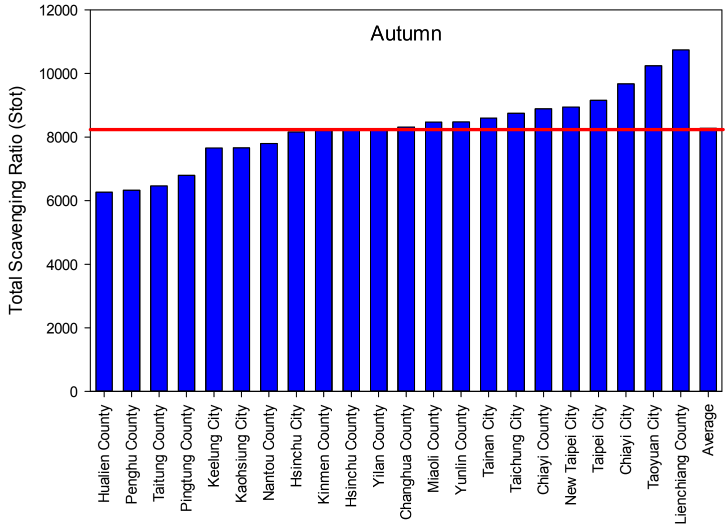

Fig. 2(C). The scavenging ratio (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in autumn 2017.

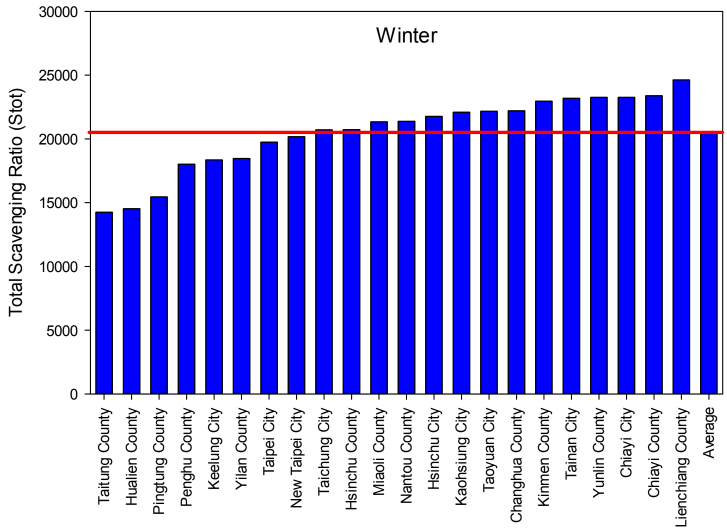

Fig. 2(D). The scavenging ratio (S_{tot}) of total-PCDD/Fs-WHO_{2005}-TEQ in various areas of Taiwan in winter 2017.

Table 3. The scavenging ratios of total PCDD/Fs-WHO_{2005}-TEQ reported in previous studies.

Area	Location	Period	Range	Average	Reference
Taiwan	Hengchun	2012	-	8050	Chandra Suryani *et al.*, 2015
Taiwan	Hengchun	2013	-	7800	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2012	-	13900	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2013	-	13000	Chandra Suryani *et al.*, 2015
Taiwan	Kaohsiung	2014	-	5490	Lee *et al.*, 2016
Taiwan	Kaohsiung	2015	-	6980	Lee *et al.*, 2016
Taiwan	Meinong	2014	-	4490	Lee *et al.*, 2016
Taiwan	Meinong	2015	-	4910	Lee *et al.*, 2016
Taiwan	Xiaogang	2014	-	5820	Lee *et al.*, 2016
Taiwan	Xiaogang	2015	-	7860	Lee *et al.*, 2016
China	Guangzhou	2014	16350–34970	24080	Zhu *et al.*, 2017a
China	Nanjing	2014	19960–39990	30950	Zhu *et al.*, 2017a
China	Harbin	2014	16600–42000	31900	Zhu *et al.*, 2017b
China	Shijiazhuang	2014	17100–41400	30700	Zhu *et al.*, 2017b
Taiwan	Mailiao	2014	7391–28900	16915	Chen *et al.*, 2017
Taiwan	Mailiao	2015	8409–27822	16563	Chen *et al.*, 2017
Taiwan	Mailiao	2016	8160–25956	15512	Chen *et al.*, 2017
Taiwan	Lunbei	2014	6706–27817	16025	Chen *et al.*, 2017
Taiwan	Lunbei	2015	6497–26295	14497	Chen *et al.*, 2017
Taiwan	Lunbei	2016	6635–24593	14495	Chen *et al.*, 2017
Taiwan	Taisi	2014	5763–27772	14574	Chen *et al.*, 2017
Taiwan	Taisi	2015	6398–25270	13973	Chen *et al.*, 2017
Taiwan	Taisi	2016	6592–24017	13603	Chen *et al.*, 2017
China	Wuhu	2015	19580–39370	30140	Wang *et al.*, 2018
China	Bengbu	2015	17130–39400	31270	Wang *et al.*, 2018

the rain were calculated. Among the 9 cities and 13 counties in Taiwan, the seasonal average concentrations of total-PCDD/Fs-WHO_{2005}-TEQ in the rain range between 0.068 (Keelung City in summer) and 1.248 pg WHO_{2005}-TEQ L^{-1} (Chiayi County in winter), and with an average of 0.374 pg WHO_{2005}-TEQ L^{-1} in 2017. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain in spring 2017 are presented in Fig. 3(A), where it can be seen that the average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain range between 0.170 (Taitung County) and 0.753 pg WHO_{2005}-TEQ L^{-1} (Lienchiang County), and with an average of 0.453 pg WHO_{2005}-TEQ L^{-1} in spring 2017. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain in summer 2017 are presented in Fig. 3(B), where it can be seen that the average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain range between 0.068 (Keelung City) and 0.426 pg WHO_{2005}-TEQ L^{-1} (Yunlin County), and with an average of 0.176 pg WHO_{2005}-TEQ L^{-1} in summer 2017. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain in autumn 2017 are presented in Fig. 3(C), where it can be seen that the average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain range between 0.082 (Hualien County) and 0.391 pg WHO_{2005}-TEQ L^{-1} (Yunlin County), and with an average of 0.218 pg WHO_{2005}-TEQ L^{-1} in autumn 2017. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain in winter 2017 are presented in Fig. 3(D), where it can be seen that the average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain range between 0.266 (Taitung County) and 1.248 pg WHO_{2005}-TEQ L^{-1} (Chiayi County), and with an average of 0.649 pg WHO_{2005}-TEQ L^{-1} in winter 2017.

As the results shown, the average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain are higher in the cool season (averaged 0.649 pg WHO_{2005}-TEQ L^{-1} in winter) than that in the warm season (averaged 0.176 pg WHO_{2005}-TEQ L^{-1} in summer). It is mainly affected by the atmospheric PCDD/F concentration and the gas-particle partition. The high concentration of PCDD/Fs may be due to the transport of pollutant will be hindered by the vertical current of the cold air. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain detected in this and other studis are presented in Table 4, where it can be seen that the levels found in this study (averaged 0.374 pg WHO_{2005}-TEQ L^{-1}) are similar to those in Kaohsiung (averaged 0.263 and 0.307 pg WHO_{2005}-TEQ L^{-1} in 2014 and 2015, respectively), Meinong (averaged 0.166 and 0.167 pg WHO_{2005}-TEQ L^{-1} in 2014 and 2015, respectively), Xiaogang (averaged 0.308 and 0.369 pg WHO_{2005}-TEQ L^{-1} in 2014 and 2015, respectively), but higher than those in Hengchun (averaged 0.064 pg WHO_{2005}-TEQ L^{-1} during 2012 and 2013) and Lulin (averaged 0.027 pg WHO_{2005}-TEQ L^{-1} during 2012 and 2013) (Chandra Suryani *et al.*, 2015; Lee *et al.*, 2016).

CONCLUSION

1. In 2017, the rainfall ranged between 0.2 (Kaohsiung City in winter) and 662.1 mm (both Chiayi County and Chiayi City in summer), averaged 168.2 mm. In 2017, the average wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 53.3, 62.9, 26.7 and 27.1

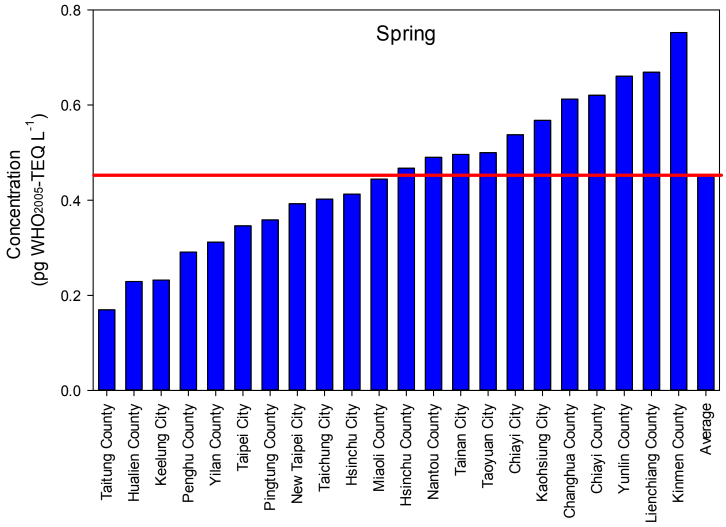

Fig. 3(A). The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in the rain in various areas of Taiwan in spring 2017.

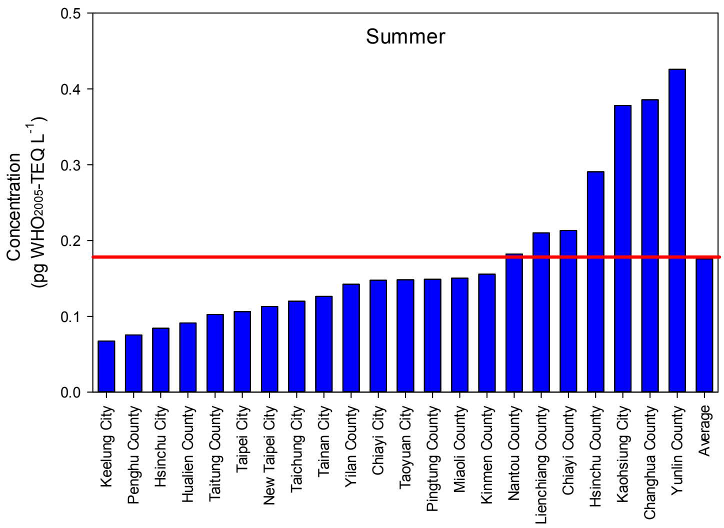

Fig. 3(B). The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in the rain in various areas of Taiwan in summer 2017.

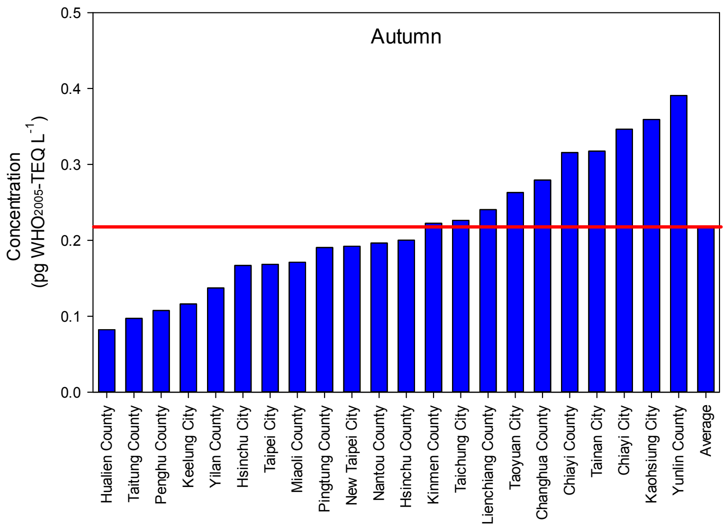

Fig. 3(C). The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in the rain in various areas of Taiwan in autumn 2017.

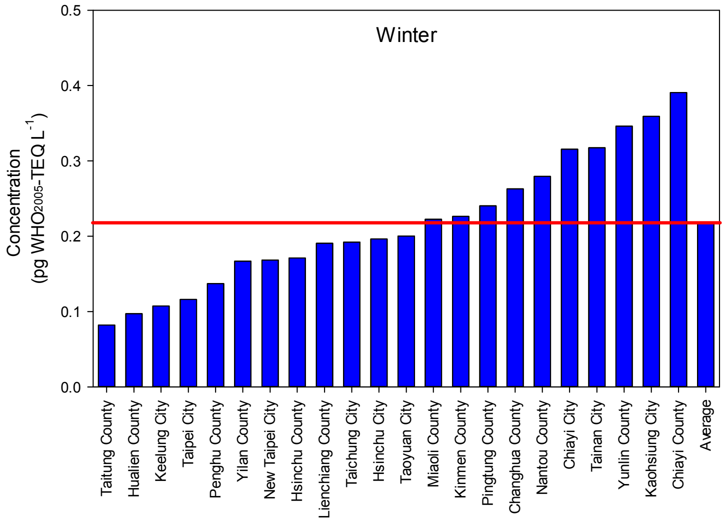

Fig. 3(D). The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in the rain in various areas of Taiwan in winter 2017.

Table 4. The concentration of total PCDD/Fs-WHO$_{2005}$-TEQ in the rain reported in previous studies.

Area	Location	Period	Range (pg WHO$_{2005}$-TEQ L^{-2})	Average (pg WHO$_{2005}$-TEQ L^{-2})	Reference
Taiwan	Hengchun	2012	-	0.0590	Chandra Suryani *et al.*, 2015
Taiwan	Hengchun	2013	-	0.0690	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2012	-	0.0260	Chandra Suryani *et al.*, 2015
Taiwan	Lulin	2013	-	0.0270	Chandra Suryani *et al.*, 2015
Taiwan	Kaohsiung	2014	-	0.263	Lee *et al.*, 2016
Taiwan	Kaohsiung	2015	-	0.307	Lee *et al.*, 2016
Taiwan	Meinong	2014	-	0.166	Lee *et al.*, 2016
Taiwan	Meinong	2015	-	0.167	Lee *et al.*, 2016
Taiwan	Xiaogang	2014	-	0.308	Lee *et al.*, 2016
Taiwan	Xiaogang	2015	-	0.369	Lee *et al.*, 2016
China	Guangzhou	2014	0.0920–1.79	0.490	Zhu *et al.*, 2017a
China	Nanjing	2014	1.91–4.26	1.70	Zhu *et al.*, 2017a
China	Harbin	2014	0.478–4.89	2.28	Zhu *et al.*, 2017b
China	Shijiazhuang	2014	1.12–8.49	3.95	Zhu *et al.*, 2017b
Taiwan	Mailiao	2014	0–1.59	0.577	Chen *et al.*, 2017
Taiwan	Mailiao	2015	0.180–1.65	0.763	Chen *et al.*, 2017
Taiwan	Mailiao	2016	0.190–1.25	0.636	Chen *et al.*, 2017
Taiwan	Lunbei	2014	0–1.60	0.604	Chen *et al.*, 2017
Taiwan	Lunbei	2015	0.0900–1.17	0.515	Chen *et al.*, 2017
Taiwan	Lunbei	2016	0.110–0.940	0.471	Chen *et al.*, 2017
Taiwan	Taisi	2014	0.0700–1.62	0.555	Chen *et al.*, 2017
Taiwan	Taisi	2015	0.0900–1.01	0.436	Chen *et al.*, 2017
Taiwan	Taisi	2016	0.100–0.750	0.393	Chen *et al.*, 2017
China	Wuhu	2015	0.360–3.29	1.41	Wang *et al.*, 2018
China	Bengbu	2015	0.380–2.53	1.61	Wang *et al.*, 2018

pg WHO$_{2005}$-TEQ m^{-2} month^{-1} in spring, summer, autumn and winter, respectively.

2. In 2017, the average ambient temperature ranged between 20.19 (Lienchiang County) and 25.86°C (Kaohsiung City), and with an average of 23.87°C. Obvious seasonal variations were detected in S_{tot}, and the average S_{tot} values for total-PCDD/Fs-WHO$_{2005}$-TEQ were 13840, 6540, 8280 and 20540 in spring, summer, autumn and winter, respectively.
3. The average concentration of total-PCDD/Fs-WHO$_{2005}$-TEQ in the rain were 0.453, 0.176, 0.218 and 0.649 pg WHO$_{2005}$-TEQ L^{-1} in spring, summer, autumn and winter, respectively.
4. Atmospheric PCDD/Fs are accumulated through deposition into soil, water system and eventually into food, so it is important to control the air quality to decrease the effects of air pollutants on human health and ecosystems.

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