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Atmospheric $PM_{2.5}$ and Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Taiwan

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ABSTRACT

In this study, the atmospheric $PM_{2.5}$, increases/decreases of the $PM_{2.5}$, the $PM_{2.5}/PM_{10}$ ratio, total PCDD/Fs-TEQ concentrations, $PM_{2.5}$-bound total PCDD/Fs-TEQ content, and PCDD/F gas-particle partition in Taiwan were investigated for the period 2013 to 2017. In Taiwan, the annual average $PM_{2.5}$ concentrations were found to be 28.9, 24.1, 21.4, 20.2, and 19.9 μg m^{-3} in 2013, 2014, 2015, 2016, and 2017, respectively, which indicated that the annual variations in $PM_{2.5}$ levels were decreasing during the study period. The average increases (+)/decreases (–) of $PM_{2.5}$ concentrations were –16.7%, –11.1%, –5.75%, and –1.73% from 2013 to 2014, from 2014 to 2015, from 2015 to 2016, and from 2016 to 2017, respectively. Based to the relationship between PM_{10} values and total PCDD/F concentrations obtained from previous studies, we estimated that in 2017, the annual average total PCDD/Fs-TEQ concentrations ranged between 0.0148 (Lienchiang County) and 0.0573 pg WHO_{2005}-TEQ m^{-3} (Keelung City), and averaged 0.0296 pg WHO_{2005}-TEQ m^{-3}, while the $PM_{2.5}$-bound total PCDD/Fs-TEQ content ranged from 0.302 (Kaohsiung City) to 0.911 ng WHO_{2005}-TEQ g^{-1} (Keelung City), at an average of 0.572 ng WHO_{2005}-TEQ g^{-1}. These values are suggested to be validated in the future study because the sources and formations of PM_{10} and $PM_{2.5}$ in different areas were diverse, and may be not closely related to combustion sources. The seasonal variations in the gas fraction of total PCDD/Fs-WHO_{2005}-TEQ concentrations were 68.6%, 86.6%, 82.3%, and 52.3% in the spring, summer, autumn, and winter, respectively. Due to the fact that Taiwan is located mostly in the sub-tropical zone, which had annual average temperatures between 23.0 and 24.4°C, averaging 23.8°C during the study period, the majority of PCDD/Fs-TEQ were dominant in the gas phase.

Keywords: $PM_{2.5}$; PCDD/Fs; PM_{10}; Particle-bound PCDD/F content; Gas-particle partition.

INTRODUCTION

Previous epidemiological studies have suggested that there are health effects at unexpectedly low concentrations of particulate air pollutants (Pope and Dockery, 2006). Particulate matter (PM) or aerosol, is a mixture of solid and liquid particulate matter in the ambient air (Ghosh *et al.*, 2014). PM generally reduces visibility and induces respiratory diseases, eventually affecting both air quality and health (Krewski *et al.*, 2000). One pollutant of growing global concern is $PM_{2.5}$, which is a type of PM with aerodynamic diameters less than or equal to 2.5 μm. Increased exposure to $PM_{2.5}$ may cause lung and respiratory diseases and even premature death. According to the National Ambient Air Quality Standard (NAAQS), the health standard for 24-hour averaged $PM_{2.5}$ should be less than 65.6 μg m^{-3}. However, the World Health Organization suggests that the annual $PM_{2.5}$ concentration be no more than 10 μg m^{-3}. The level of $PM_{2.5}$ in ambient air is usually used as an indicator of the level of anthropogenic air pollution. Although many studies have suggested that $PM_{2.5}$ exposure has adverse effects on human health, these effects vary in different locations because of variations in the chemical components of $PM_{2.5}$ (Boldo *et al.*, 2006; Bell *et al.*, 2007; Jahn *et al.*, 2011; Atkinson *et al.*, 2014; Fisk and Chan, 2017).

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) and other persistent organic pollutants (POPs), are stable in the environment

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and accumulate in the soil, water, and food (Lohmann and Jones, 1998; Hu *et al.*, 2009; Cheruiyot *et al.*, 2015, 2016; Redfern *et al.*, 2017). The major sources of PCDD/Fs are anthropogenic rather than natural (Oh *et al.*, 1999; Addink and Altwicker, 2001; Prange *et al.*, 2002, 2003; Kim *et al.*, 2003; Neuer-Etscheidt *et al.*, 2006; Lin *et al.*, 2014; Cheruiyot *et al.*, 2016). In fact, PCDD/Fs are unintentionally formed during combustion and thermal operations. The chemical, physical, and toxicological properties of PCDD/Fs mostly depend on the number and position of chlorine atoms. There are 210 possible congeners of PCDD/Fs (75 PCDDs and 135 PCDFs) with the different numbers and positions of chlorine atoms. Studies have shown that the 2,3,7,8-TCDD comprises the most toxic congener. Both the International Agency for Research on Cancer (IARC) and the US Department of Health and Human Services (US DHHS) determined that it can cause cancer in humans (IARC, 1997; US EPA, 2017). The atmosphere is the major pathway for PCDD/Fs to transport to other areas. Generally, there are particle and gas phases in the ambient air, and the gas-particle partitions will affect the transport route, deposition (dry/wet deposition) processes, and degradation mechanisms (Kouimtzis *et al.*, 2002).

In Taiwan, the control of the air pollution began in 1955 and started as control of coal burning in Taipei. Because of high population density and a rapid increase in the number of incinerators and motor vehicles, several cities in Taiwan developed worsening air quality. From 1988 to 1991, the pollutant standards index (PSI) was often in the range of 50–100 in Taiwan, but some areas were more than 100 (unhealthy air quality) (Bureau of Air Quality Protection and Noise Control, 1991; Fang and Chen, 1996). Previous studies have indicated that meteorological and geographic conditions, emission sources, and land use contribute to this problem, but meteorological and geographic conditions are the most important factors affecting air quality (Harrison *et al.*, 1997; Tsuang and Chao, 1999; Lin, 2001; Marcazzan *et al.*, 2001; Ho *et al.*, 2003; Frank *et al.*, 2006).

The aim of this study was to examine the level of $PM_{2.5}$ and PCDD/Fs in the ambient air in Taiwan. The $PM_{2.5}$ concentrations and $PM_{2.5}/PM_{10}$ ratios from 2013 to 2017 were investigated, as well as the total PCDD/Fs-TEQ concentrations and the $PM_{2.5}$-bound total PCDD/Fs-TEQ contents in 2017. Furthermore, seasonal variations in the PCDD/Fs-TEQ gas-particle partitions in 2017 were also compared and discussed.

MATERIALS AND METHODS

Data Collection

In this study, the meteorological data from 48 air quality stations in 9 cities and 13 counties in Taiwan were collected for 2013 and 2017. Detailed information was obtained for 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. The ambient temperature, $PM_{2.5}$, and PM_{10} from 2013 to 2017 were obtained from local air quality stations.

PCDD/F Concentration

The total PCDD/F concentrations were estimated by averaging the values obtained from two formulas that demonstrate the relationship between PM_{10} values and total PCDD/F concentrations, for which the correlations were presented by Huang *et al.* (2011) and Lee *et al.* (2016) and are shown as Eq. (1) and Eq. (2), respectively as follows:

$$y = 0.0472 + 0.0138x \quad (1)$$

$$y = 0.0117x - 0.021 \quad (2)$$

where y: total PCDD/F concentration (pg m^{-3})
x: PM_{10} concentration (μg m^{-3})

The differences between the estimated total PCDD/F concentrations from the two equations range from 0.0892 pg m^{-3} to 0.383 pg m^{-3} when the PM_{10} concentrations vary from 10 μg m^{-3} to 150 μg m^{-3}. Also apparently, these two equations can not cover all scenarios that address the relationship between PM_{10} and PCDD/Fs. Therefore, efforts are required to develop more universal and compact relationship equations between atmospheric PM and PCDD/F concentrations to moderate variances between field and simulation results.

To obtain the toxicity equivalent (TEQ), the concentrations of PCDD/Fs congeners are multiplied by their respective toxic equivalency factor (TEF) values. There are two TEF schemes, including the International Toxicity Equivalent (I-TEF) and the World Health Organization TEF (WHO-TEF). The WHO has established and regularly re-evaluated TEFs for dioxins and related compounds through expert consultations. WHO-TEF values have been established, which apply to humans, mammals, birds, and fish (van den Berg *et al.*, 2006). In this study, we used the WHO_{2005}-TEF as revised in 2005 to calculate TEQ concentrations.

Gas-Particle Partitioning

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

Pankow, 1987; Pankow and Bidleman, 1991, 1992):

$$K_p = \frac{F/TSP}{A} \quad (3)$$

K_p: temperature-dependent partitioning constant (m^3 μg^{-1})
TSP: concentration of total suspended particulate matter, multiplied by PM_{10} concentration with 1.24 (μg m^{-3})
F: concentration of the compounds of interest bound to particles (pg m^{-3})
A: gaseous concentration of the compound of interest (pg m^{-3}).

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

$$\log K_p = m_r \times \log P_L^o + b_r \quad (4)$$

P_L^o: subcooled liquid vapor pressure (Pa),
m_r: cited slope,
b_r: cited y-intercept.

Complete datasets on the gas-particle partitioning of PCDD/Fs in Taiwan has been reported (Chao *et al.*, 2004), with the values m_r = –1.29 and b_r = –7.2, with R^2 = 0.94. These values were used in this study for establishing the partitioning constant (K_p) of PCDD/Fs.

A previous study correlated the 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. The correlation has been redeveloped as follows (Hung *et al.*, 2002):

$$\log P_L^o = \frac{-1.34\ (RI)}{T} + 1.67\times10^{-3}(RI) - \frac{1320}{T} + 8.087 \quad (5)$$

RI: gas chromatographic retention indexes developed by Donnelly *et al.* (1987) and Hale *et al.* (1985),
T: ambient temperature (K).

RESULTS AND DISCUSSION

$PM_{2.5}$ Concentration

The concentrations of $PM_{2.5}$ in the ambient air in Taiwan from 2013 to 2017 are presented in Figs. 1(a), 1(b), 1(c), 1(d) and 1(e), respectively. Among the 22 cities and counties in Taiwan, the annual average concentration of $PM_{2.5}$ in the ambient air ranged between 12.5 (Taitung County) and 40.3 μg m^{-3} (Chiayi City), with an average of 28.9 μg m^{-3} in 2013. In 2014, the annual average concentration of $PM_{2.5}$ in the ambient air ranged between 10.8 (Taitung County) and 32.6 μg m^{-3} (Chiayi City), with an average of 24.1 μg m^{-3}. In 2015, the annual average concentration of $PM_{2.5}$ in the ambient air ranged between 9.6 (Taitung County) and 29.7 μg m^{-3} (Nantou County), with an average of 21.4 μg m^{-3}. In 2016, the annual average concentration of $PM_{2.5}$ in the ambient air range between 9.0 (Taitung County) and 27.3 μg m^{-3} (Chiayi City), with an average of 20.2 μg m^{-3}. In 2017, the annual average concentration of $PM_{2.5}$ in the ambient air range between 9.1 (Taitung County) and 27.6 μg m^{-3} (Kaohsiung City), with an average of 19.9 μg m^{-3}. With the exception of Taitung County in 2015, 2016, and 2017, the $PM_{2.5}$ levels in the other areas were higher than the WHO standard (10 μg m^{-3}). Among the 22 cities and counties in Taiwan, the lowest $PM_{2.5}$ concentrations occurred in Taitung County from 2013 to 2017, and the values decreased annually (from 12.5 to 9.1 μg m^{-3}). The highest $PM_{2.5}$ concentrations occurred in Chiayi City in 2013, 2014, and 2016, Nantou County in 2015, and Kaohsiung City in

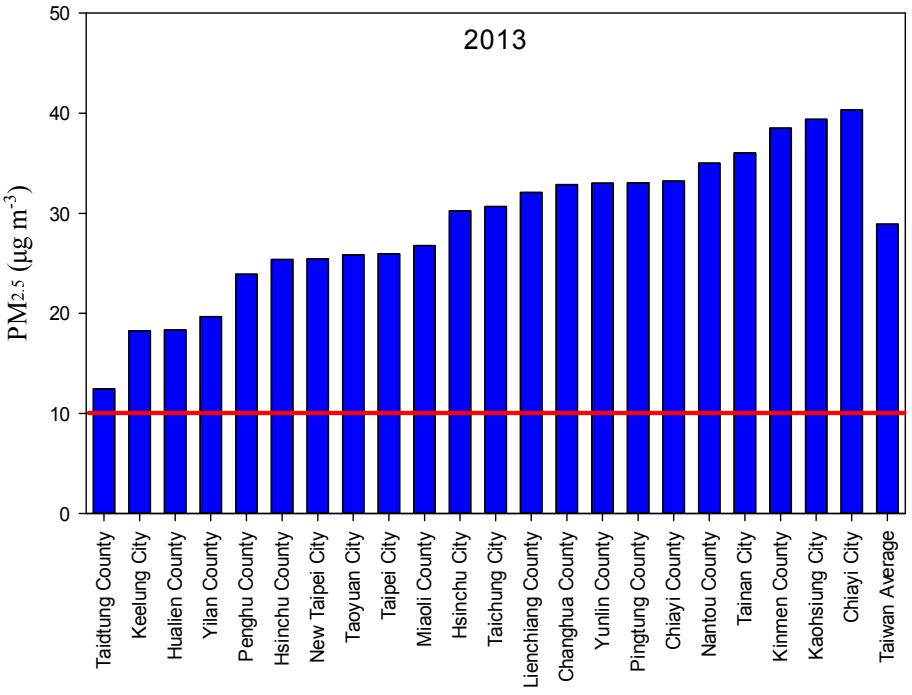

Fig. 1(a). Atmospheric $PM_{2.5}$ concentrations in various areas in Taiwan in 2013.

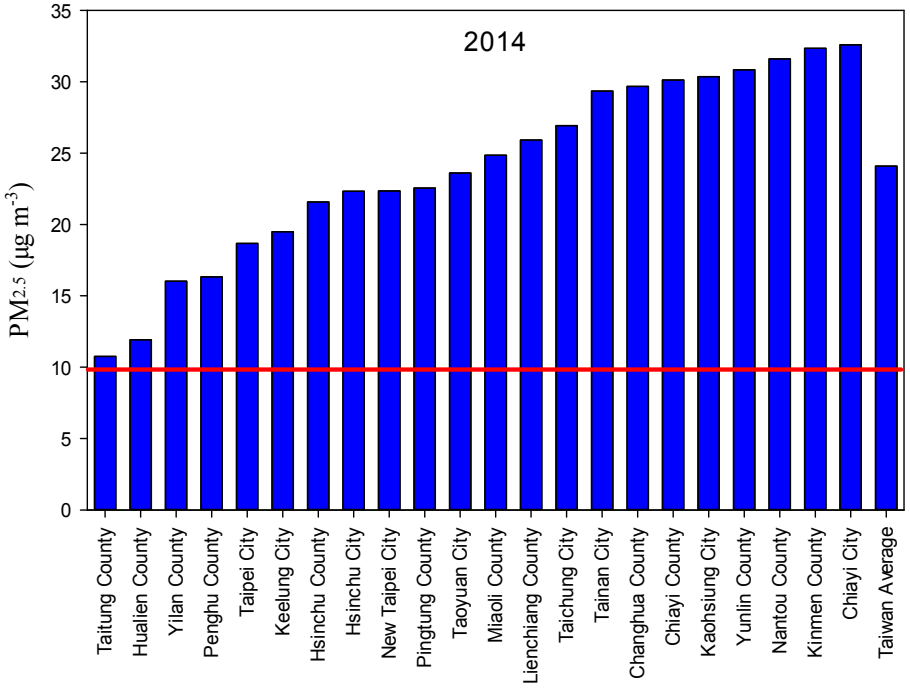

Fig. 1(b). Atmospheric $PM_{2.5}$ concentrations in various areas in Taiwan in 2014.

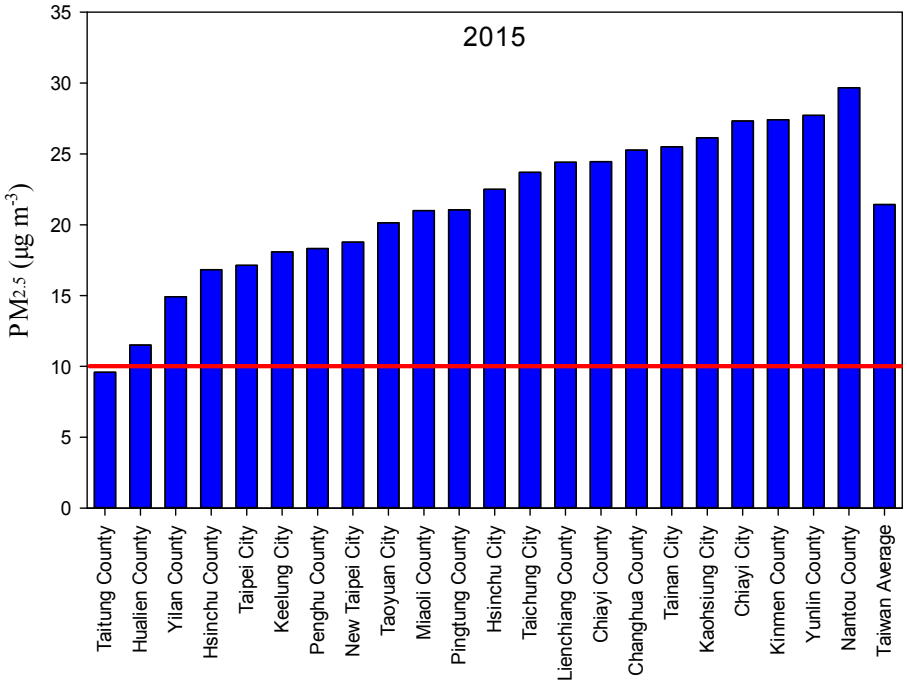

Fig. 1(c). Atmospheric $PM_{2.5}$ concentrations in various areas in Taiwan in 2015.

2017, for which the values decreased annually (from 40.3 to 27.6 μg m^{-3}). Overall, in Taiwan, the annual average $PM_{2.5}$ concentrations decreased annually (from 28.9 to 19.9 μg m^{-3}). As the results show, the air quality of Taiwan improved significantly, but the annual average $PM_{2.5}$ concentration was still far above the WHO air quality standard (10 μg m^{-3}). The above results indicate that even though it occurred slowly, the $PM_{2.5}$ concentrations decreased annually during the study period in Taiwan.

As to seasonal variations, the $PM_{2.5}$ in the spring ranged between 10.5 (Taitung County) and 33.7 μg m^{-3} (Kinmen County), with an average of 23.2 μg m^{-3}. In summer, the $PM_{2.5}$ ranged between 7.2 (Taitung County) and 15.8 μg m^{-3} (Yunlin County), with an average of 12.1 μg m^{-3}. In autumn, the $PM_{2.5}$ ranged between 8.7 (Taitung County) and 29.2 μg m^{-3} (Kaohsiung City), with an average of 19.1 μg m^{-3}.

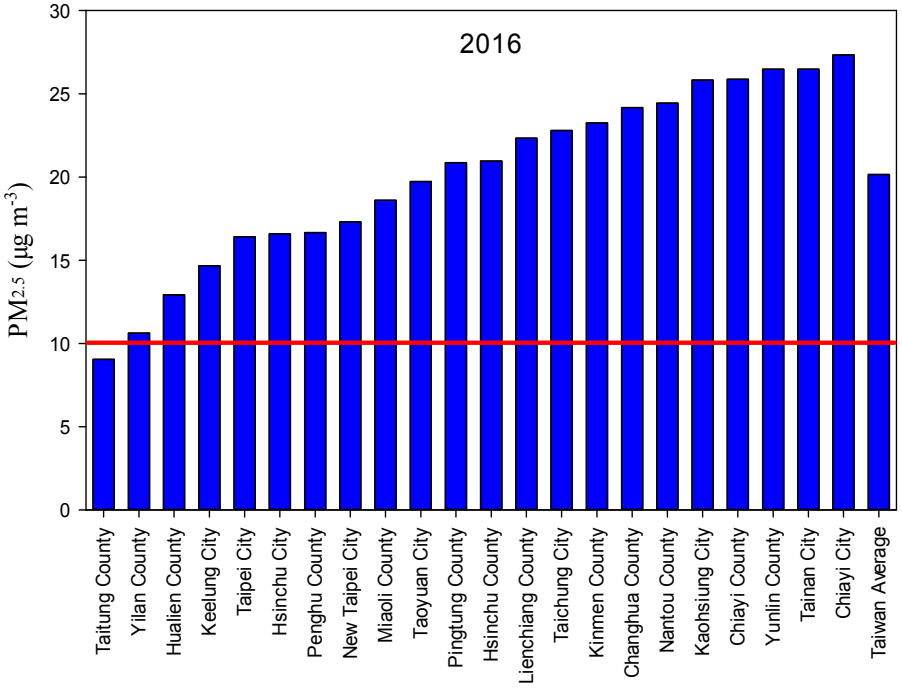

Fig. 1(d). Atmospheric $PM_{2.5}$ concentration in various areas in Taiwan during 2016.

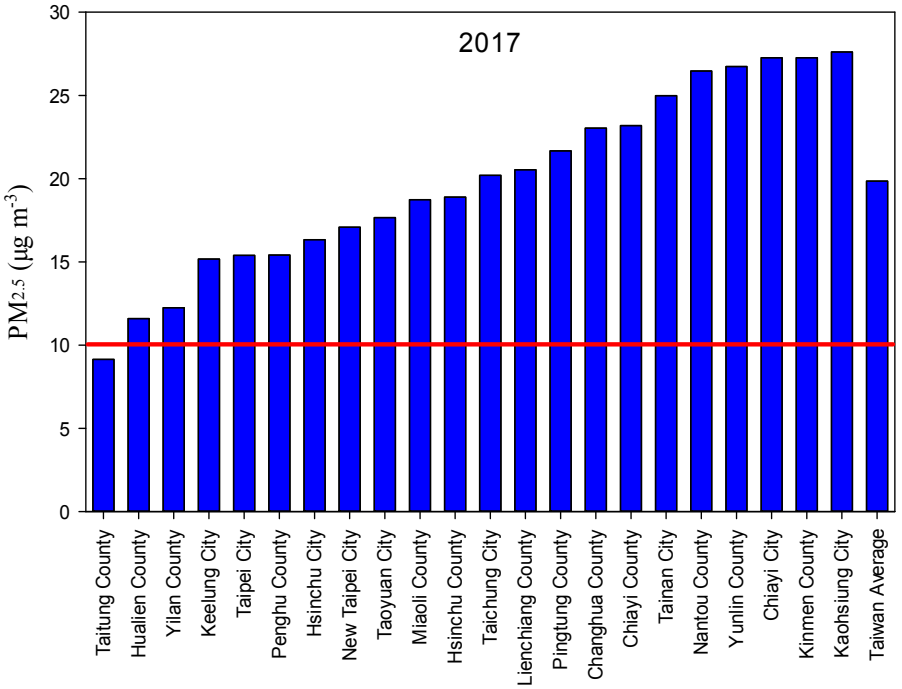

Fig. 1(e). Atmospheric $PM_{2.5}$ concentration in various areas in Taiwan during 2017.

In winter, the $PM_{2.5}$ ranged between 10.2 (Taitung County) and 41.0 μg m^{-3} (Kaohsiung City), with an average of 25.0 μg m^{-3}. The atmospheric $PM_{2.5}$ concentrations were high in spring (23.2 μg m^{-3}) and winter (25.0 μg m^{-3}), and low in summer (12.1 μg m^{-3}) and autumn (19.1 μg m^{-3}). This may have been because low temperatures will result in a lower mixing period, hindering the vertical transport of atmospheric pollutants and elevating the $PM_{2.5}$ concentration in the ambient air.

To compare the increases/decreases for the 22 cities and counties in Taiwan, we further investigated the increases/decreases of $PM_{2.5}$ concentrations from 2013 to 2017, for which the results for the period 2013 to 2014 are presented in Fig. 2(a). As the results show, with the exception of Keelung City, which increased by approximately 6.85%, all of the other cities/counties showed decreasing trends of

$PM_{2.5}$ levels, ranging from –6.62% (Yunlin County) to –35.0% (Hualien County), at an average of –16.7%. The increases/decreases of $PM_{2.5}$ concentrations for the period 2014 to 2015 are presented in Fig. 2(b). The results show that increases in the $PM_{2.5}$ concentration only occurred in Penghu County and Hsinchu City, with increased percentages of 12.2% and 0.75%, respectively, while, those of the other 20 cities or counties exhibited decreases in $PM_{2.5}$ concentrations, for which the percentages ranged between –3.50% (Hualien County) and –22.01% (Hsinchu County) and averaged –11.08%. The increases/decreases of $PM_{2.5}$ concentration for the period 2015 to 2016 are presented in Fig. 2(c). There was almost no change in Chiayi City, while four counties/cities showed increases in $PM_{2.5}$ concentrations:

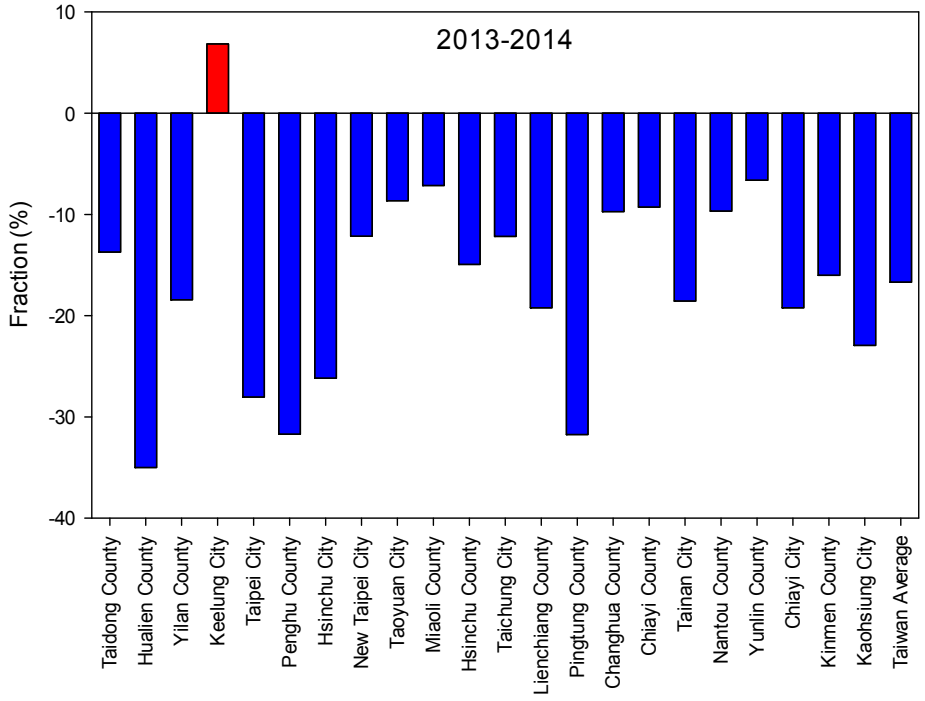

Fig. 2(a). Increases/decreases of $PM_{2.5}$ concentrations in Taiwan from 2013–2014.

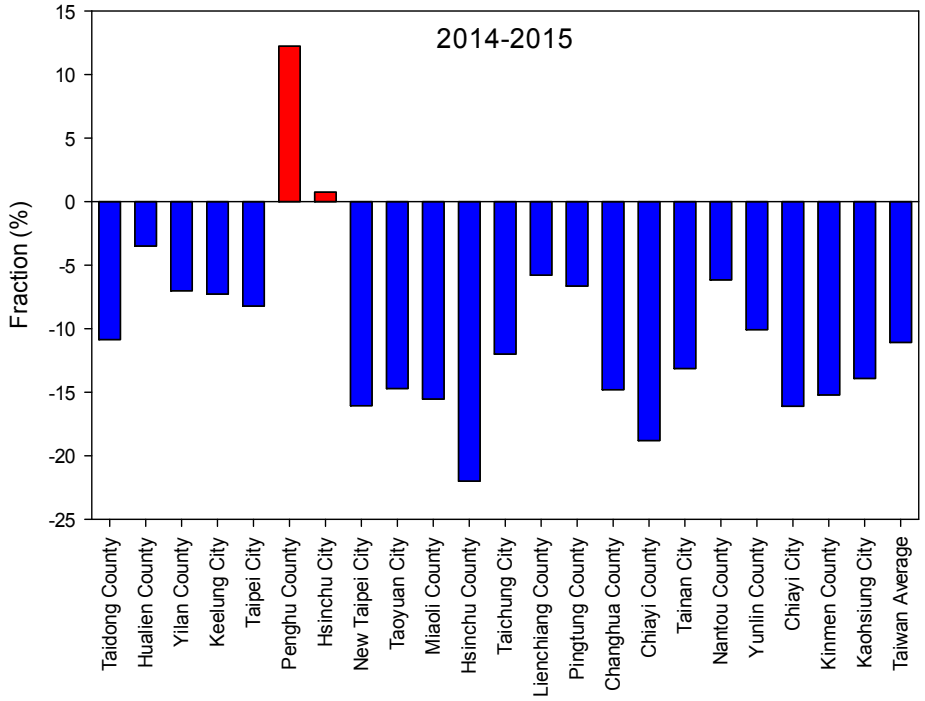

Fig. 2(b). Increases/decreases of $PM_{2.5}$ concentrations in Taiwan from 2014–2015.

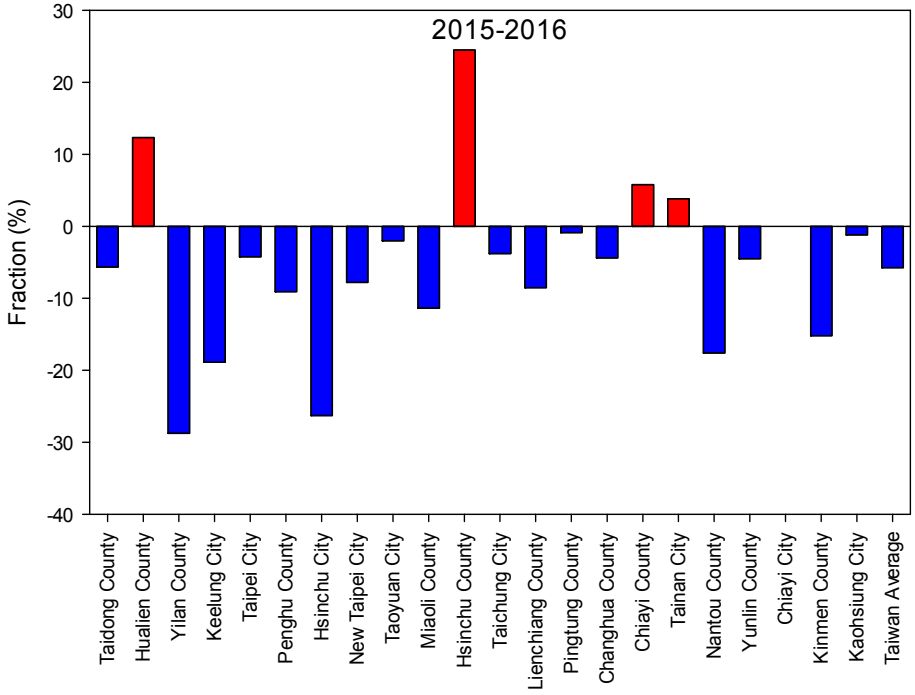

Fig. 2(c). Increases/decreases of $PM_{2.5}$ concentrations in Taiwan from 2015–2016.

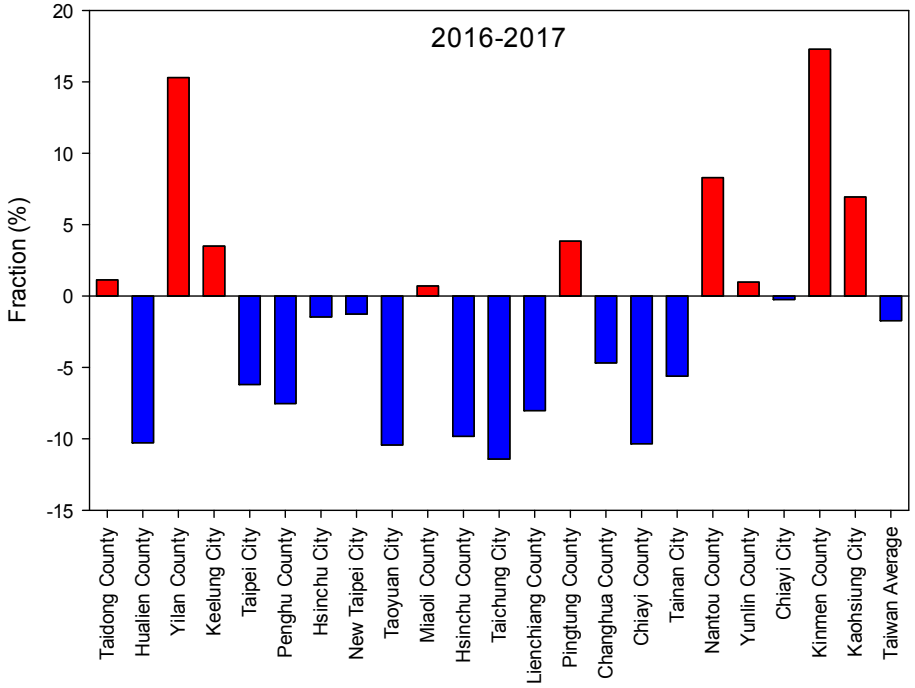

Fig. 2(d). Increases/decreases of $PM_{2.5}$ concentrations in Taiwan from 2016–2017.

Hualien County, Hsinchu County, Chiayi County, and Tainan City, with increased percentages of 12.3%, 24.5%, 5.79% and 3.84%, respectively. The other 17 cities (or counties) showed decreases in $PM_{2.5}$ concentrations, for which the decreases ranged between –1.19% (Kaohsiung City) and –28.8% (Yilan County), and averaged –8.8%; Overall, from 2015 to 2016, the average decreased percentages of $PM_{2.5}$ concentrations was –5.75%. The increases/decreases of $PM_{2.5}$ concentrations for the period 2016 to 2017 are presented in Fig. 2(d). There were 9 cities (or counties) in which the $PM_{2.5}$ concentrations were increased, for which the increased percentages ranged between 0.71% (Miaoli County) and 17.30% (Kinmen County), while the other 13 cities (or counties) showed decreases in $PM_{2.5}$ concentrations,

for which the decreased percentages ranged between –0.25% (Chiayi City) and –11.4% (Taichung City). Overall, the average decreased percentages of $PM_{2.5}$ concentrations from 2016 to 2017 was –1.73%. These results suggest that the $PM_{2.5}$ concentration was decreasing slowly from 2013 to 2017. However, some of the cities (or counties) experienced increases and deserve additional attention.

$PM_{2.5}/PM_{10}$ Ratio

The $PM_{2.5}/PM_{10}$ ratio is dependent on the type of site and varies depending on the area under consideration. The chemical characteristics and emission sources are different for $PM_{2.5}$ and PM_{10}; therefore, the $PM_{2.5}/PM_{10}$ ratio reflects the proportion of PM and the importance of $PM_{2.5}$. A higher $PM_{2.5}/PM_{10}$ ratio usually means worse air quality because $PM_{2.5}$ remains in the air for long periods and significant propagation distance. The annual average $PM_{2.5}/PM_{10}$ ratio in the cities (or counties) in Taiwan are presented in Tables 1, 2, 3, 4, and 5.

As Table 1 shows, among the 22 cities (or counties) in Taiwan, the annual average $PM_{2.5}/PM_{10}$ ratio ranged from 0.33 in October (Chiayi County) to 0.85 in May (Pingtung

Table 1. The atmospheric $PM_{2.5}/PM_{10}$ ratios in Taiwan in 2013.

	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Jan.	0.59	0.54	0.55	0.73	0.62	0.61	0.52	0.59
Feb.	0.57	0.55	0.62	0.68	0.58	0.57	0.50	0.55
Mar.	0.59	0.60	0.52	0.68	0.60	0.57	0.48	0.54
Apr.	0.64	0.74	0.59	0.73	0.66	0.64	0.53	0.63
May.	0.67	0.70	0.55	0.72	0.85	0.58	0.47	0.58
Jun.	0.58	0.56	0.63	0.65	0.63	0.55	0.47	0.56
Jul.	0.54	0.55	0.50	0.72	0.57	0.59	0.49	0.54
Aug.	0.55	0.62	0.51	0.59	0.58	0.66	0.51	0.55
Sep.	0.51	0.73	0.41	0.63	0.64	0.61	0.45	0.64
Oct.	0.53	0.70	0.43	0.56	0.52	0.57	0.43	0.51
Nov.	0.47	0.62	0.46	0.63	0.54	0.59	0.42	0.54
Dec.	0.65	0.81	0.57	0.67	0.65	0.66	0.53	0.62
Ave.	**0.57**	**0.65**	**0.53**	**0.66**	**0.60**	**0.60**	**0.48**	**0.57**
	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	
Jan.	0.69	0.65	0.59	0.60	0.67	0.69	0.63	
Feb.	0.69	0.62	0.60	0.62	0.67	0.66	0.63	
Mar.	0.62	0.52	0.57	0.56	0.62	0.58	0.59	
Apr.	0.65	0.57	0.65	0.63	0.69	0.63	0.61	
May.	0.64	0.59	0.61	0.62	0.66	0.58	0.58	
Jun.	0.65	0.59	0.56	0.63	0.62	0.55	0.59	
Jul.	0.65	0.41	0.54	0.61	0.70	0.65	0.57	
Aug.	0.63	0.40	0.55	0.61	0.75	0.67	0.59	
Sep.	0.48	0.46	0.54	0.50	0.71	0.65	0.60	
Oct.	0.49	0.52	0.39	0.49	0.60	0.57	0.48	
Nov.	0.56	0.51	0.47	0.53	0.63	0.57	0.56	
Dec.	0.69	0.69	0.61	0.64	0.75	0.70	0.67	
Ave.	**0.62**	**0.56**	**0.55**	**0.58**	**0.67**	**0.62**	**0.59**	
	Chiayi County	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	
Jan.	0.45	0.61	0.64	0.60	0.46	0.54	0.54	
Feb.	0.45	0.55	0.64	0.59	0.38	0.51	0.55	
Mar.	0.44	0.56	0.60	0.58	0.46	0.46	0.46	
Apr.	0.50	0.60	0.61	0.64	0.48	0.54	0.51	
May.	0.45	0.57	0.54	0.65	0.38	0.48	0.44	
Jun.	0.43	0.54	0.55	0.70	0.42	0.47	0.50	
Jul.	0.45	0.57	0.61	0.66	0.34	0.48	0.44	
Aug.	0.47	0.60	0.63	0.66	0.37	0.48	0.52	
Sep.	0.46	0.54	0.59	0.58	0.42	0.53	0.54	
Oct.	0.33	0.50	0.56	0.56	0.39	0.40	0.51	
Nov.	0.45	0.54	0.58	0.49	0.42	0.45	0.46	
Dec.	0.50	0.63	0.71	0.62	0.52	0.56	0.56	
Ave.	**0.44**	**0.57**	**0.59**	**0.61**	**0.43**	**0.49**	**0.51**	

Table 2. The atmospheric $PM_{2.5}/PM_{10}$ ration in Taiwan during 2014.

	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Jan.	0.45	0.55	0.50	0.58	0.50	0.57	0.47	0.51
Feb.	0.42	0.41	0.57	0.58	0.47	0.56	0.44	0.50
Mar.	0.46	0.39	0.53	0.64	0.44	0.58	0.44	0.47
Apr.	0.49	0.41	0.53	0.62	0.46	0.59	0.45	0.46
May.	0.47	0.46	0.53	0.50	0.38	0.54	0.45	0.43
Jun.	0.44	0.34	0.45	0.41	0.27	0.45	0.40	0.33
Jul.	0.27	0.31	0.38	0.50	0.35	0.51	0.46	0.36
Aug.	0.33	0.30	0.39	0.43	0.30	0.56	0.48	0.34
Sep.	0.31	0.38	0.39	0.53	0.40	0.58	0.45	0.42
Oct.	0.39	0.38	0.41	0.55	0.47	0.57	0.43	0.47
Nov.	0.39	0.32	0.46	0.60	0.46	0.57	0.40	0.48
Dec.	0.44	0.36	0.40	0.59	0.49	0.52	0.41	0.50
Ave.	**0.41**	**0.39**	**0.47**	**0.56**	**0.44**	**0.56**	**0.44**	**0.47**
	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	
Jan.	0.68	0.57	0.48	0.50	0.58	0.59	0.53	
Feb.	0.71	0.60	0.55	0.53	0.53	0.61	0.56	
Mar.	0.67	0.53	0.56	0.54	0.52	0.58	0.52	
Apr.	0.67	0.53	0.56	0.53	0.50	0.63	0.54	
May.	0.64	0.49	0.51	0.53	0.46	0.58	0.46	
Jun.	0.63	0.43	0.36	0.52	0.35	0.51	0.39	
Jul.	0.57	0.32	0.44	0.48	0.38	0.55	0.49	
Aug.	0.67	0.45	0.52	0.52	0.46	0.57	0.38	
Sep.	0.65	0.48	0.48	0.50	0.59	0.33	0.48	
Oct.	0.58	0.49	0.44	0.45	0.60	0.35	0.45	
Nov.	0.63	0.54	0.46	0.49	0.52	0.34	0.51	
Dec.	0.61	0.52	0.44	0.47	0.51	0.45	0.53	
Ave.	**0.64**	**0.51**	**0.47**	**0.50**	**0.52**	**0.50**	**0.50**	
	Chiayi County	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	
Jan.	0.43	0.55	0.58	0.44	0.42	0.47	0.47	
Feb.	0.46	0.53	0.54	0.43	0.40	0.46	0.47	
Mar.	0.48	0.54	0.52	0.45	0.47	0.45	0.50	
Apr.	0.47	0.52	0.53	0.45	0.46	0.42	0.50	
May.	0.42	0.49	0.47	0.48	0.52	0.39	0.48	
Jun.	0.33	0.52	0.42	0.44	0.34	0.30	0.33	
Jul.	0.40	0.54	0.52	0.40	0.29	0.40	0.32	
Aug.	0.36	0.59	0.50	0.43	0.31	0.39	0.29	
Sep.	0.40	0.56	0.53	0.44	0.36	0.47	0.41	
Oct.	0.37	0.57	0.51	0.42	0.38	0.45	0.41	
Nov.	0.40	0.59	0.53	0.40	0.36	0.46	0.39	
Dec.	0.41	0.57	0.50	0.41	0.48	0.47	0.43	
Ave.	**0.42**	**0.55**	**0.52**	**0.43**	**0.39**	**0.43**	**0.43**	

County) and averaged 0.57 in 2013. The annual average $PM_{2.5}/PM_{10}$ ratio ranged between 0.27 in July (Yilan County) and 0.71 in February (Keelung City), and averaged 0.48 in 2014. The annual average $PM_{2.5}/PM_{10}$ ratio ranged from 0.21 in April (Hualien County) to 0.75 in September (Penghu County) and averaged 0.47 in 2015. The annual average $PM_{2.5}/PM_{10}$ ratio ranged between 0.16 in October (Yilan County) to 0.70 in January (Lienchiang County) and averaged 0.49 in 2016. The annual average $PM_{2.5}/PM_{10}$ ratio ranged between 0.27 in July (Taitung County) to 0.68 in April (Keelung City) and averaged 0.47 in 2017. In Taiwan, the annual average $PM_{2.5}/PM_{10}$ ratios in 2013 (0.5) were higher than those from 2014 to 2017, which had similar $PM_{2.5}/PM_{10}$ ratios (ranging from 0.47 to 0.49). These results were slightly lower than those found for Lunbei and Taisi (0.553 and 0.536, respectively) and higher than those in Mailiao (0.363) (Chen *et al.*, 2017); however, the ratios were lower than those in northern China (ranging between 0.48 and 0.59) (Chen *et al.*, 2014; Xing *et al.*, 2017; Wang *et al.*, 2018). Among the 22 cities (or counties) in Taiwan, the $PM_{2.5}/PM_{10}$ ratios were always low in specific areas like Taitung and Chiayi counties. Some areas such as Kaohsiung

Table 3. The atmospheric $PM_{2.5}/PM_{10}$ ration in Taiwan during 2015.

	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Jan.	0.48	0.40	0.48	0.59	0.48	0.53	0.43	0.48
Feb.	0.49	0.32	0.51	0.61	0.48	0.52	0.43	0.49
Mar.	0.51	0.26	0.52	0.63	0.43	0.52	0.42	0.46
Apr.	0.48	0.21	0.47	0.53	0.40	0.42	0.41	0.38
May.	0.43	0.35	0.50	0.49	0.36	0.40	0.43	0.35
Jun.	0.44	0.46	0.44	0.43	0.32	0.34	0.39	0.27
Jul.	0.45	0.46	0.43	0.45	0.37	0.40	0.43	0.33
Aug.	0.39	0.46	0.38	0.40	0.37	0.44	0.42	0.31
Sep.	0.42	0.48	0.50	0.55	0.44	0.50	0.45	0.42
Oct.	0.38	0.48	0.48	0.55	0.49	0.52	0.41	0.45
Nov.	0.33	0.48	0.50	0.55	0.47	0.51	0.44	0.44
Dec.	0.31	0.52	0.45	0.58	0.49	0.45	0.40	0.47
Ave.	**0.43**	**0.40**	**0.48**	**0.55**	**0.44**	**0.47**	**0.42**	**0.43**
	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	
Jan.	0.66	0.56	0.52	0.49	0.55	0.50	0.49	
Feb.	0.64	0.54	0.51	0.49	0.58	0.50	0.52	
Mar.	0.63	0.56	0.54	0.49	0.61	0.47	0.49	
Apr.	0.61	0.53	0.47	0.48	0.60	0.44	0.44	
May.	0.60	0.52	0.48	0.48	0.61	0.38	0.47	
Jun.	0.60	0.41	0.36	0.44	0.59	0.26	0.37	
Jul.	0.59	0.49	0.41	0.43	0.62	0.32	0.45	
Aug.	0.50	0.52	0.38	0.38	0.67	0.22	0.45	
Sep.	0.56	0.68	0.49	0.40	0.66	0.35	0.40	
Oct.	0.53	0.55	0.49	0.42	0.55	0.42	0.45	
Nov.	0.58	0.60	0.48	0.46	0.56	0.46	0.47	
Dec.	0.59	0.65	0.44	0.45	0.47	0.49	0.41	
Ave.	**0.59**	**0.55**	**0.48**	**0.46**	**0.58**	**0.41**	**0.46**	
	Chiayi County	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	
Jan.	0.42	0.55	0.52	0.46	0.51	0.51	0.48	
Feb.	0.43	0.55	0.51	0.45	0.48	0.48	0.48	
Mar.	0.41	0.54	0.50	0.45	0.38	0.47	0.49	
Apr.	0.34	0.48	0.48	0.47	0.43	0.41	0.51	
May.	0.39	0.51	0.50	0.49	0.45	0.40	0.54	
Jun.	0.28	0.42	0.44	0.44	0.34	0.29	0.69	
Jul.	0.32	0.44	0.49	0.44	0.35	0.39	0.72	
Aug.	0.32	0.47	0.48	0.40	0.31	0.38	NA	
Sep.	0.40	0.51	0.53	0.44	0.41	0.51	0.75	
Oct.	0.40	0.50	0.55	0.42	0.37	0.50	0.58	
Nov.	0.38	0.53	0.51	0.43	0.33	0.45	0.67	
Dec.	0.39	0.52	0.50	0.41	0.40	0.47	0.61	
Ave.	**0.38**	**0.51**	**0.51**	**0.44**	**0.40**	**0.46**	**0.54**	

and New Taipei City were usually shown in the middle, and some areas such as Keelung City and Nantou County were usually high in terms of $PM_{2.5}/PM_{10}$ ratios. However, some areas fluctuated significantly, for example, the $PM_{2.5}/PM_{10}$ ratio in Hsinchu City was 0.58 in 2015 but 0.43 in 2016.

In the case of seasonal variations, the $PM_{2.5}/PM_{10}$ ratio in spring ranged between 0.39 (Yilan County) and 0.65 (Keelung City) and averaged 0.49. In summer, the $PM_{2.5}/PM_{10}$ ratio ranged between 0.30 (Taitung County) and 0.63 (Keelung City) and averaged 0.44. In autumn, the $PM_{2.5}/PM_{10}$ ratio ranged between 0.31 (Taitung County) and 0.56 (Keelung City and averaged 0.45. In winter, the $PM_{2.5}/PM_{10}$ ratio ranged between 0.37 (Taitung County) and 0.88 (Kinmen County) and averaged 0.52. The $PM_{2.5}/PM_{10}$ ratio was high in winter (0.52) and spring (0.49) and low in summer (0.44) and autumn (0.45).

Total PCDD/Fs-TEQ Concentrations

The concentrations of total PCDD/Fs-TEQ in 2017 are presented in Fig. 3. As the results show, the annual average total PCDD/Fs-TEQ concentrations ranged between 0.0148 (Lienchiang County) and 0.0573 pg WHO_{2005}-TEQ m^{-3}

Table 4. The atmospheric $PM_{2.5}/PM_{10}$ ration in Taiwan during 2016.

	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Jan.	0.33	0.50	0.56	0.54	0.55	0.45	0.47	0.55
Feb.	0.35	0.48	0.52	0.57	0.55	0.45	0.47	0.55
Mar.	0.37	0.52	NA	0.49	0.54	NA	NA	0.55
Apr.	0.46	0.49	0.56	0.57	0.52	0.56	0.51	0.49
May.	0.39	0.41	0.46	0.49	0.44	0.52	0.46	0.40
Jun.	0.30	0.50	0.24	0.42	0.30	0.41	0.36	0.27
Jul.	0.30	0.44	0.26	0.49	0.35	0.41	0.41	0.33
Aug.	0.31	0.48	0.39	0.55	0.39	0.37	0.44	0.42
Sep.	0.27	0.41	0.33	0.51	0.51	0.31	0.40	0.42
Oct.	0.16	0.42	0.33	0.57	0.47	0.44	0.39	0.44
Nov.	0.21	0.41	0.41	0.55	0.49	0.44	0.37	0.50
Dec.	0.27	0.44	0.45	0.55	0.51	0.45	0.41	0.48
Ave.	**0.32**	**0.46**	**0.46**	**0.55**	**0.48**	**0.46**	**0.45**	**0.48**
	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	
Jan.	0.62	0.70	0.56	0.53	0.48	0.61	0.56	
Feb.	0.55	0.56	0.51	0.46	0.44	0.59	0.51	
Mar.	0.61	NA	0.63	0.51	NA	NA	0.58	
Apr.	0.66	0.63	0.58	0.55	0.56	0.67	0.52	
May.	0.56	0.56	0.50	0.49	0.47	0.63	0.44	
Jun.	0.52	0.48	0.36	0.44	0.31	0.45	0.31	
Jul.	0.62	0.41	0.44	0.44	0.36	0.51	0.39	
Aug.	0.67	0.44	0.51	0.45	0.35	0.50	0.53	
Sep.	0.48	0.49	0.46	0.44	0.32	0.53	0.41	
Oct.	0.43	0.41	0.50	0.40	0.36	0.54	0.55	
Nov.	0.46	0.48	0.50	0.39	0.39	0.54	0.52	
Dec.	0.54	0.54	0.45	0.39	0.35	0.55	0.52	
Ave.	**0.56**	**0.55**	**0.51**	**0.46**	**0.43**	**0.58**	**0.50**	
	Chiayi County	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	
Jan.	0.47	0.54	0.55	0.50	0.55	0.58	0.69	
Feb.	0.44	0.56	0.56	0.46	0.50	0.54	0.52	
Mar.	0.52	0.59	0.56	0.56	0.54	0.62	NA	
Apr.	0.49	0.58	0.60	0.57	0.47	0.60	0.63	
May.	0.43	0.52	0.53	0.50	0.40	0.49	0.61	
Jun.	0.35	0.45	0.44	0.45	0.32	0.37	0.58	
Jul.	0.39	0.49	0.51	0.49	0.31	0.44	0.53	
Aug.	0.47	0.49	0.54	0.48	0.38	0.51	0.56	
Sep.	0.41	0.42	0.49	0.46	0.28	0.47	0.52	
Oct.	0.43	0.51	0.57	0.45	0.24	0.52	0.50	
Nov.	0.43	0.47	0.52	0.43	0.28	0.50	0.51	
Dec.	0.40	0.46	0.52	0.44	0.34	0.48	0.49	
Ave.	**0.44**	**0.51**	**0.55**	**0.49**	**0.38**	**0.52**	**0.57**	

(Keelung City) and averaged 0.0296 pg WHO_{2005}-TEQ m^{-3}. Compared with previous studies, the values reported in this study were lower than those found in southern Taiwan (averaging 0.048 and 0.044 pg WHO_{2005}-TEQ m^{-3} in 2014 and 2015, respectively), coastal areas (averaging 0.0380, 0.0346, and 0.0324 pg WHO_{2005}-TEQ m^{-3} in 2014, 2015, and 2016, respectively), central Taiwan, and most areas in northern China (averaging 0.075 pg WHO_{2005}-TEQ m^{-3} in 2014) and southern China (0.0665, 0.0633, 0.0625, 0.0600, 0.0528 and 0.0526 pg-WHO_{2005}-TEQ m^{-3} in Chengdu, Wuhan, Nanjing, Hefei, Luzhou, and Hangzhou, respectively) (Lee *et al.*, 2016; Chen *et al.*, 2017; Tang *et al.*, 2017; Xing *et al.*, 2017; Wang *et al.*, 2018). The concentrations of total PCDD/Fs-TEQ in Chiayi County (0.0407 pg WHO_{2005}-TEQ m^{-3}), Changhua County (0.0477 pg WHO_{2005}-TEQ m^{-3}), Yunlin County (0.0559 pg WHO_{2005}-TEQ m^{-3}), and Keelung City (0.0573 pg WHO_{2005}-TEQ m^{-3}) were significantly higher than those in the other cities (or counties) in Taiwan. The higher concentrations of total PCDD/Fs-TEQ in Changhua and Yunlin counties may have been a result of the fact that both are industrial areas with many petrochemical and metal industries. As for Keelung City, the reasons need to

Table 5. The atmospheric $PM_{2.5}/PM_{10}$ ration in Taiwan during 2017.

	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Jan.	0.36	0.46	0.56	0.60	0.50	0.47	0.44	0.48
Feb.	0.40	0.47	0.58	0.60	0.49	0.47	0.46	0.49
Mar.	0.41	0.45	0.51	0.62	0.48	0.46	0.42	0.50
Apr.	0.41	0.49	0.54	0.57	0.43	0.49	0.45	0.48
May.	0.32	0.50	0.59	0.54	0.41	0.47	0.40	0.46
Jun.	0.30	0.39	0.64	0.42	0.34	0.41	0.37	0.31
Jul.	0.31	0.41	0.48	0.51	0.40	0.42	0.40	0.38
Aug.	0.41	0.46	0.43	0.53	0.38	0.47	0.41	0.45
Sep.	0.40	0.42	0.51	0.58	0.42	0.54	0.44	0.50
Oct.	0.35	0.39	0.44	0.54	0.39	0.53	0.37	0.41
Nov.	0.33	0.35	0.51	0.55	0.46	0.52	0.36	0.46
Dec.	0.38	0.47	NA	0.56	0.47	0.53	0.41	0.46
Ave.	**0.37**	**0.44**	**0.55**	**0.57**	**0.44**	**0.48**	**0.41**	**0.46**
	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	
Jan.	0.62	0.51	0.50	0.46	0.48	0.50	0.53	
Feb.	0.64	0.54	0.49	0.48	0.48	0.57	0.53	
Mar.	0.63	0.49	0.56	0.48	0.46	0.52	0.53	
Apr.	0.68	0.45	0.57	0.51	0.43	0.57	0.54	
May.	0.63	0.38	0.51	0.47	0.38	0.55	0.50	
Jun.	0.64	0.29	0.44	0.47	0.38	0.61	0.34	
Jul.	0.58	0.27	0.49	0.46	0.41	0.54	0.40	
Aug.	0.67	0.39	0.47	0.47	0.42	0.53	0.40	
Sep.	0.67	0.53	0.51	0.48	0.53	0.54	0.52	
Oct.	0.50	0.47	0.35	0.39	0.46	0.52	0.43	
Nov.	0.50	0.55	0.36	0.43	0.48	0.47	0.44	
Dec.	0.52	NA	0.42	0.43	0.46	0.53	0.47	
Ave.	**0.61**	**0.48**	**0.47**	**0.46**	**0.45**	**0.54**	**0.48**	
	Chiayi County	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	
Jan.	0.38	0.47	0.51	0.49	0.35	0.47	0.40	
Feb.	0.39	0.46	0.51	0.52	0.43	0.48	0.43	
Mar.	0.43	0.49	0.52	0.49	0.43	0.51	0.49	
Apr.	0.44	0.52	0.55	0.52	0.42	0.50	0.46	
May.	0.39	0.48	0.50	0.47	0.37	0.47	0.44	
Jun.	0.30	0.41	0.45	0.45	0.30	0.33	0.44	
Jul.	0.36	0.53	0.50	0.44	0.27	0.42	0.60	
Aug.	0.38	0.53	0.49	0.48	0.31	0.39	0.56	
Sep.	0.42	0.55	0.51	0.50	0.33	0.50	0.58	
Oct.	0.30	0.46	0.45	0.44	0.31	0.38	0.46	
Nov.	0.31	0.46	0.41	0.44	0.30	0.38	0.45	
Dec.	0.34	0.53	0.47	0.45	0.33	0.40	NA	
Ave.	**0.37**	**0.49**	**0.49**	**0.48**	**0.35**	**0.44**	**0.48**	

be further investigated. Because its air quality monitoring station is close to Keelung harbor, the PM emissions from ships and vessels could have direct impact on the PM_{10} concentrations measured in the Keelung air quality monitoring station. Besides this, its PM_{10} concentrations can be elevated due to sea spray aerosol. However, these factors are not reflected on the relationship equations between PM_{10} and PCDD/Fs, which were obtained in previous studies that their atmospheric environments were rural and metallurgical complex areas. Therefore, these values are suggested to be validated in the future study because the sources and formations of PM_{10} and $PM_{2.5}$ in different areas were diverse, and may be not closely related to combustion sources. The total PCDD/Fs-TEQ concentrations of the above four areas in Taiwan were similar to those in Bengbu (averaging 0.054, 0.054 and 0.061 pg WHO_{2005}-TEQ m^{-3} in 2015, 2016, and 2017, respectively) and Wuhu (averaging 0.050, 0.047 and 0.050 pg WHO_{2005}-TEQ m^{-3} in 2015, 2016 and 2017, respectively) (Wang *et al.*, 2018). Overall, the total PCDD/Fs-TEQ concentrations in Taiwan were lower than the regulated standard in Japan (0.6 pg WHO_{2005}-TEQ m^{-3}).

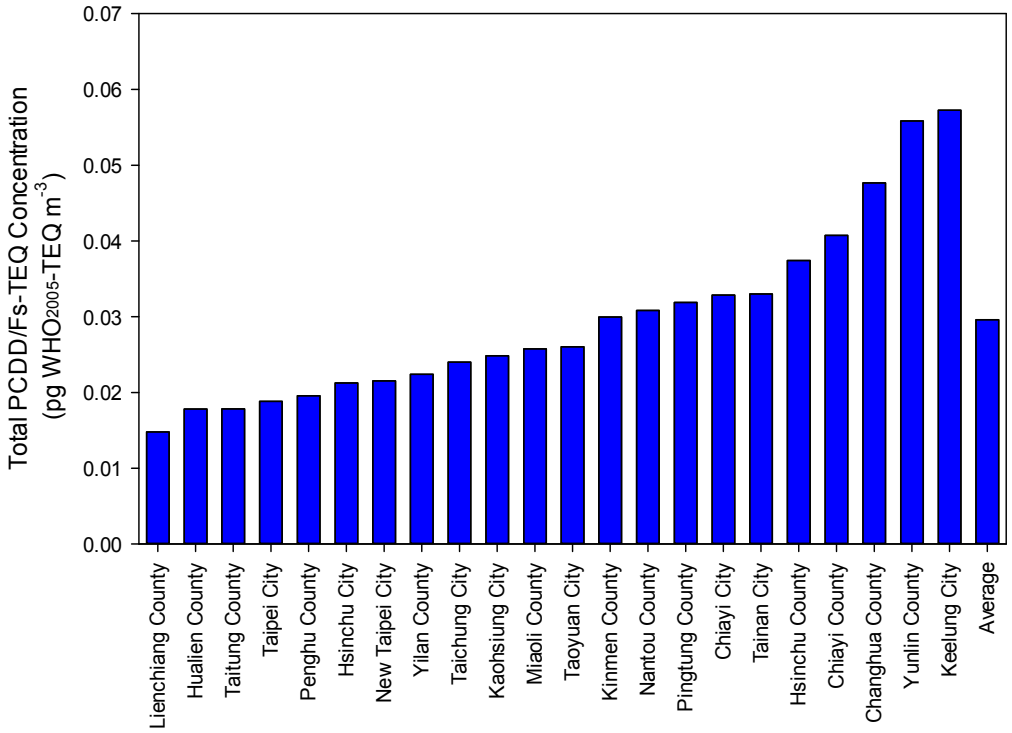

Fig. 3. The total PCDD/Fs-TEQ concentrations in Taiwan in 2017 (Unit: pg WHO_{2005}-TEQ m^{-3}).

In the case of seasonal variations, the concentrations of total PCDD/Fs-TEQ in spring ranged between 0.0173 (Lienchiang County) and 0.0503 pg WHO_{2005}-TEQ m^{-3} (Yunlin County) and averaged 0.0325 pg WHO_{2005}-TEQ m^{-3}. In summer, the concentrations of total PCDD/Fs-TEQ ranged between 0.0112 (Lienchiang County) and 0.0821 pg WHO_{2005}-TEQ m^{-3} (Yunlin County) and averaged 0.0293 pg WHO_{2005}-TEQ m^{-3}. In autumn, the concentrations of total PCDD/Fs-TEQ ranged between 0.0131 (Hualien County) and 0.0469 pg WHO_{2005}-TEQ m^{-3} (Keelung City) and averaged 0.0259 pg WHO_{2005}-TEQ m^{-3}. In winter, the concentrations of total PCDD/Fs-TEQ ranged between 0.0156 (Lienchiang County) and 0.0546 pg WHO_{2005}-TEQ m^{-3} (Keelung City) and averaged 0.0309 pg WHO_{2005}-TEQ m^{-3}. The above results indicate that the total PCDD/Fs-WHO_{2005}-TEQ concentrations were lower in summer (0.0293 pg WHO_{2005}-TEQ m^{-3}) and autumn (0.0259 pg WHO_{2005}-TEQ m^{-3}) and were higher in spring (0.0325 pg WHO_{2005}-TEQ m^{-3}) and winter (0.0309 pg WHO_{2005}-TEQ m^{-3}). These trends are similar to those for the $PM_{2.5}$ concentrations and $PM_{2.5}/PM_{10}$ ratios. The vertical transport of air current is much better in hot periods and more stable in cold seasons.

$PM_{2.5}$-Bound PCDD/Fs-TEQ Content

The results for the total $PM_{2.5}$-bound PCDD/Fs-TEQ content are presented in Fig. 4. As the results show, the total $PM_{2.5}$-bound PCDD/Fs-TEQ content ranged between 0.302 (Kaohsiung City) and 0.911 ng WHO_{2005}-TEQ g^{-1} (Keelung City) and averaged 0.572 ng WHO_{2005}-TEQ g^{-1}. There are four areas for which the $PM_{2.5}$-bound total PCDD/Fs-TEQ content was much higher than the other areas: Changhua County (0.802 ng WHO_{2005}-TEQ g^{-1}), Yunlin County (0.846 ng WHO_{2005}-TEQ g^{-1}), Hsinchu County (0.894 ng WHO_{2005}-TEQ g^{-1}) and Keelung City (0.911 ng WHO_{2005}-TEQ g^{-1}). Generally, metal industries, incineration, petrochemical plants, and other factories are the major emission sources of PCDD/Fs and other POPs (Oh *et al.*, 1999; Cheruiyot *et al.*, 2015, 2016; Redfern *et al.*, 2017). Three of these four areas are industrial areas: Yunlin County, Changhua County, and Hsinchu County. Except for the above four areas, the other 18 cities (or counties) exhibited similar values for $PM_{2.5}$-bound total PCDD/Fs-TEQ content and ranged between 0.302 and 0.575 ng WHO_{2005}-TEQ g^{-1} and averaged 0.484 ng WHO_{2005}-TEQ g^{-1}. The $PM_{2.5}$-bound total PCDD/Fs-TEQ content in Taiwan was similar to that of Wuhu and Bengbu in China, for which the annual average values were 0.419 and 0.435 ng WHO_{2005}-TEQ g^{-1} in 2015, 0.404 and 0.457 ng WHO_{2005}-TEQ g^{-1} in 2016, and 0.498 and 0.511 ng WHO_{2005}-TEQ g^{-1} in 2017, respectively (Wang *et al.*, 2018). Compared with the $PM_{2.5}$ concentrations in the ambient air, there was a negative but insignificant correlation between the $PM_{2.5}$-bound total PCDD/Fs-TEQ content and $PM_{2.5}$ concentrations. For example, the highest values of $PM_{2.5}$ concentration occurred in Kaohsiung City in 2017 (27.6 μg m^{-3}), but the $PM_{2.5}$-bound total PCDD/Fs-TEQ content was the lowest (0.302 ng WHO_{2005}-TEQ g^{-1}); the lowest $PM_{2.5}$ concentration occurred in Taitung County (9.14 μg m^{-3}), but the $PM_{2.5}$-bound total PCDD/Fs-TEQ content was at the middle level (0.575 ng WHO_{2005}-TEQ g^{-1}). The results were similar to those found in previous studies (Xing *et al.*, 2017; Wang *et al.*, 2018).

In the case of seasonal variations, the $PM_{2.5}$-bound total PCDD/Fs-TEQ content in spring ranged between 0.364 (Kaohsiung City) and 0.776 ng WHO_{2005}-TEQ g^{-1} (Keelung

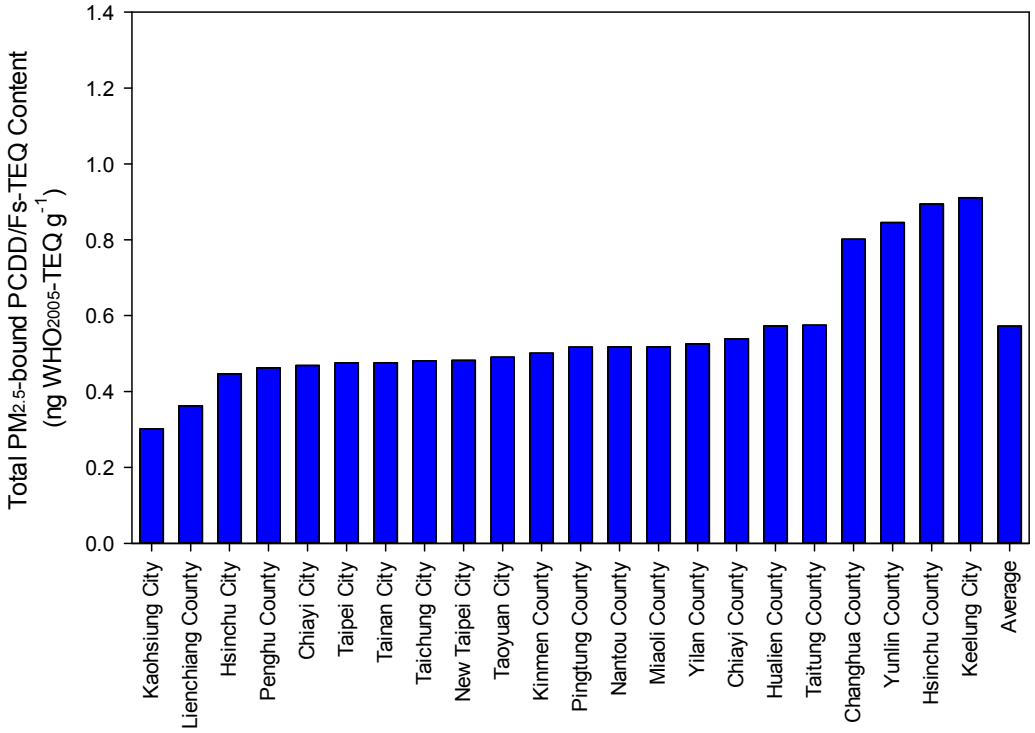

Fig. 4. The total $PM_{2.5}$-bound PCDD/Fs-TEQ content in Taiwan in 2017 (Unit: ng WHO_{2005}-TEQ g^{-1}).

City) and averaged 0.545 ng WHO_{2005}-TEQ g^{-1}. In summer, the $PM_{2.5}$-bound total PCDD/Fs-TEQ content ranged between 0.426 (Hsinchu City) and 2.123 ng WHO_{2005}-TEQ g^{-1} (Yunlin County) and averaged 0.821 ng WHO_{2005}-TEQ g^{-1}. In autumn, the $PM_{2.5}$-bound total PCDD/Fs-TEQ content ranged between 0.278 (Kaohsiung City) and 0.78 ng WHO_{2005}-TEQ g^{-1} (Keelung City) and averaged 0.474 ng WHO_{2005}-TEQ g^{-1}. In winter, the $PM_{2.5}$-bound total PCDD/Fs-TEQ content ranged between 0.218 (Kaohsiung City) and 0.8 ng WHO_{2005}-TEQ g^{-1} (Keelung City) and averaged 0.478 ng WHO_{2005}-TEQ g^{-1}. The $PM_{2.5}$-bound total PCDD/Fs-TEQ content was in the following order: summer (0.821 ng WHO_{2005}-TEQ g^{-1}) > spring (0.545 ng WHO_{2005}-TEQ g^{-1}) > winter (0.478 ng WHO_{2005}-TEQ g^{-1}) > autumn (0.474 ng WHO_{2005}-TEQ g^{-1}). The sensitivity analysis indicated that atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content was the most sensitive to total PCDD/F mass concentration, followed by PM_{10} concentration, and then the air temperature and $PM_{2.5}$ concentration (Wang *et al.*, 2018).

Gas-Particle Partitions

The gas-particle partition is an important factor related to the fate, transport, and transformation of PCDD/Fs. The ambient temperature, total suspended particle concentration, and vapor pressure all affect gas-particle partitions (Pankow, 1994). The ambient temperature in the 22 cities/counties) are presented in Table 6. The annual temperature in 2017 ranged between 20.2 (Lienchiang County) and 25.9°C (Kaohsiung City), with an average of 23.9°C in Taiwan.

The results for the seasonal gas-particle partitioning of total PCDD/Fs-TEQ in 2017 are presented in Fig. 5. In spring, the fraction of total PCDD/Fs-TEQ contributed by the gas phase ranged between 54.0% (Lienchiang County) and 79.1% (Taitung County) and averaged 68.6%; those in summer ranged from 82.2% (Chiayi City) to 90.8% (Hsinchu County) and averaged 86.6%; those in autumn ranged between 76.2% (Lienchiang County) and 87.3% (Hualien County) and averaged 82.3%, which were closed to those in summer. As for winter, the fraction of total PCDD/Fs-TEQ contributed by the gas phase ranged between 42.3% (Lienchiang County) and 67.7% (Taitung County) and averaged 52.3%. The results suggest that due to temperature fluctuations, the seasonal variations in the PCDD/F partitions vary significantly. Because the atmospheric temperatures fluctuate from spring to winter, a higher fraction was found in the gas phase in warm seasons (averaging 86.6% and 82.3% in summer and autumn, respectively) as compared to that in cold seasons (averaging 31.4% and 47.7% in spring and winter, respectively), for which the mechanisms were similar to those found in previous studies (Xu *et al.*, 2009; Wang *et al.*, 2010; Lee *et al.*, 2016; Zhu *et al.*, 2017). Unlike the results of total PCDD/Fs mass concentration (Huang *et al.*, 2011; Xing *et al.*, 2017), the gas phase was predominant in the ambient air of the total PCDD/Fs-TEQ concentrations for all four seasons. This may have been due to the fact that PCDD/F congeners (like 2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, 2,3,7,8-TeCDF), which have a relative high TEF, usually show a higher fraction in the gas phase than in the particle phase. Otherwise, the annual temperature in 2017 ranged between 20.2 (Lienchiang County) and 25.9°C (Kaohsiung City) and averaged 23.9°C in Taiwan, where the higher temperature resulted in a higher fraction of gas phase PCDD/Fs. The gas phases of PCDD/Fs increased with increases in ambient temperature,

Table 6. The atmospheric temperatures in Taiwan in 2017 (Unit: °C).

	Yilan County	Hualien County	Kinmen County	Nantou County	Pingtung County	Miaoli County	Taoyuan City	Kaohsiung City
Spring	21.7	22.6	20.7	23.1	25.2	21.7	21.6	25.4
Summer	29.4	29.7	29.6	27.1	29.2	28.5	29.3	29.8
Autumn	25.0	26.1	25.9	26.2	27.4	25.1	25.4	27.5
Winter	17.4	19.2	15.0	18.2	21.1	16.5	16.7	20.7
Average	**23.4**	**24.4**	**22.8**	**23.6**	**25.7**	**22.9**	**23.2**	**25.9**
	Keelung City	Lienchiang County	Yunlin County	New Taipei City	Hsinchu City	Hsinchu County	Chiayi City	
Spring	20.9	17.8	24.0	22.1	22.0	22.2	24.3	
Summer	29.3	27.3	29.7	29.9	29.6	29.1	29.1	
Autumn	25.3	23.3	26.9	25.9	25.9	25.3	26.2	
Winter	16.7	12.4	18.7	17.6	17.1	17.2	18.9	
Average	**23.0**	**20.2**	**24.8**	**23.9**	**23.6**	**23.4**	**24.6**	
	Chiayi County	Changhua County	Taichung City	Taipei City	Taitung County	Tainan City	Penghu County	
Spring	24.4	23.4	23.3	21.4	22.9	24.7	23.2	
Summer	29.6	29.5	28.6	29.0	29.0	29.7	29.0	
Autumn	26.8	26.6	26.0	25.1	26.1	27.1	26.1	
Winter	18.5	18.0	18.1	16.8	19.4	19.2	17.3	
Average	**24.8**	**24.4**	**24.0**	**23.1**	**24.3**	**25.2**	**24.0**	

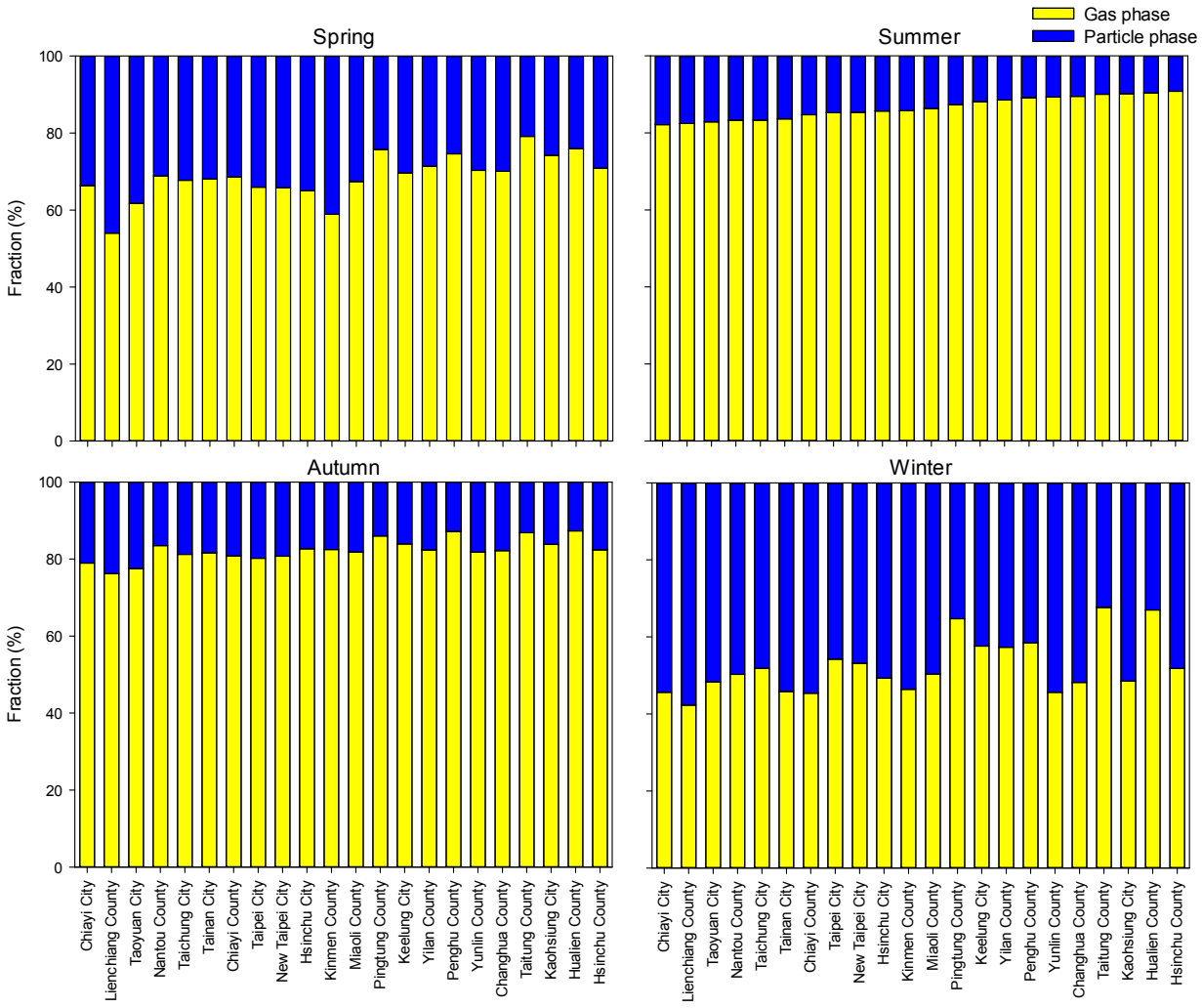

Fig. 5. Seasonal gas-particle partitioning of PCDD/Fs-TEQ in Taiwan in 2017.

and as temperature increased, some of the particle phase PCDD/Fs evaporated and existed in the gas phase.

CONCLUSION

1. The annual average $PM_{2.5}$ concentrations were 28.9, 24.1, 21.4, 20.2, and 19.9 μg m^{-3} in 2013, 2014, 2015, 2016, and 2017 respectively. The concentration of $PM_{2.5}$ decreased annually from 2013 to 2017, but the $PM_{2.5}$ concentrations were still far above the WHO air quality standard (10 μg m^{-3}).
2. The average increases/decreases of $PM_{2.5}$ concentrations in Taiwan were –16.7%, –11.1%, –5.75%, and –1.73% from 2013 to 2014, from 2014 to 2015, from 2015 to 2016, and from 2016 to 2017, respectively, showing the $PM_{2.5}$ concentration decreased steadily.
3. In Taiwan, the average $PM_{2.5}/PM_{10}$ ratios were 0.57, 0.48, 0.47, 0.49, and 0.47 in 2013, 2014, 2015, 2016, and 2017, respectively.
   The annual average total PCDD/Fs-TEQ concentrations in Taiwan ranged between 0.0148 (Lienchiang County) and 0.0573 pg WHO_{2005}-TEQ m^{-3} (Keelung City) and averaged 0.0296 pg WHO_{2005}-TEQ m^{-3}.
4. The $PM_{2.5}$-bound total PCDD/Fs-TEQ content ranged between 0.302 (Kaohsiung City) and 0.911 ng WHO_{2005}-TEQ g^{-1} (Keelung City) and averaged 0.572 ng WHO_{2005}-TEQ g^{-1}.
5. In spring, the fraction of total PCDD/Fs-TEQ contributed by the gas phase ranged between 54.0% (Lienchiang County) and 79.1% (Taitung County) and averaged 68.6%; in summer, it ranged from 82.2% (Chiayi City) to 90.8% (Hsinchu County) and averaged 86.6%; in autumn, it ranged between 76.2% (Lienchiang County) and 87.3% (Hualien County) and averaged 82.3%, which was closed to that in summer. In the winter, the fraction of total PCDD/Fs-TEQ contributed by the gas phase ranged between 42.3% (Lienchiang County) and 67.7% (Taitung County) and averaged 52.3%. The results suggest that due to temperature fluctuations, the seasonal variations in PCDD/F partitions were very significant.

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