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Part I: $PM_{2.5}$ and Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in the Ambient Air of Southern China

Haiyan Tang[1], Kangping Cui[1*], Jin Xing[1], Jinning Zhu[1*], Wen-Jhy Lee[1,2], John Kennedy Mwangi[2], Yu-Cheng Lee[3]**

[1] *School of Resources and Environmental Engineering, Hefei University of Technology, Hefei 230009, China*
[2] *Department of Environmental Engineering, National Cheng Kung University, Tainan 70101, Taiwan*
[3] *School of International business, Jinan University, Guangzhou 510632, China*

ABSTRACT

The atmospheric $PM_{2.5}$, $PM_{2.5}/PM_{10}$, PCDD/Fs-WHO_{2005}-TEQ, and PCDD/F (polychlorinated dibenzo-*p*-dioxins and dibenzofuran) phase distributions of 23 cities in southern China, during 2014–2016, were investigated in this study. In general, the cities with higher latitudes had higher $PM_{2.5}$ concentrations than those with lower latitudes. During 2014–2016, the lowest three-year average concentrations of $PM_{2.5}$ occurred at Sanya and Haikou and were 16.4 and 21.7 µg m^{-3}, respectively; while the highest concentrations of $PM_{2.5}$ was occurred at Wuhan and Luzhou and were 68.8 and 63.1 µg m^{-3}, respectively. During 2015–2016, the $PM_{2.5}$ concentrations of most of cities decreased, but those of five cities (Chengdu, Luzhou, Nanchang, Qujing and Quanzhou) increased, indicting that the air quality of these five cities was still not well controlled. The average R_M values of the 23 cities were 5.20, 4.49 and 4.13 in 2014, 2015 and 2016, respectively, which revealed that the $PM_{2.5}$ concentrations in the cities of southern China slowly decreased, although they were still far above the WHO air quality regulated standard (10 µg m^{-3}). In general, a city with a higher $PM_{2.5}$ concentration was also had a higher $PM_{2.5}/PM_{10}$ ratio. Among the 23 cities, the six highest three-year averages of total-PCDD/Fs-WHO_{2005}-TEQ concentrations were 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. During 2014, the six cities (Hefei, Nanjing, Wuhan, Guiyang, Shanghai and Chengdu) with the lowest temperatures in winter (an average of 5.4°C), their average particle phase fractions of total-PCDD/Fs-WHO_{2005}-TEQ that were approximately 76%, 53%, 71% and 93% in the spring, summer, fall and winter, respectively; while, the six cities (Haikou, Fuzhou, Guangzhou, Nanning, Nanchang and Changsha) with the highest temperatures in summer (an average of 16.5°C), had average particle phase fractions of total-PCDD/Fs-WHO_{2005}-TEQ that were approximately 61%, 42%, 57% and 81% in the spring, summer, fall and winter, respectively. The results of this study provide information showing the trends of both atmospheric $PM_{2.5}$ and PCDD/Fs in the cities of southern China. In addition, this study provided the overview relating to the $PM_{2.5}$ and PCDD/Fs in ambient air of southern China, which was not reported in previous studies. The results of this study were of great importance to present the trends of air quality in China. It is also useful for the establishment of control strategies in the future.

Keywords: $PM_{2.5}$; PCDD/Fs; $PM_{2.5}/PM_{10}$ ratio; Phase distribution; Particle-bound; Southern China.

INTRODUCTION

Particulate matter is an aerosol of solid or liquid particulate matter in the atmosphere (Ghosh *et al.*, 2014). According to the aerodynamic equivalent diameter, particulate matter can be divided into $PM_{2.5}$, PM_{10} and TSP (Lu *et al.*, 2016). The aerodynamic equivalent diameter of $PM_{2.5}$ is 0 to 2.5 µm (Chow *et al.*, 2015). $PM_{2.5}$ is an inhalable particulate matter, which can lead to reduced visibility, poor air quality and health hazards, and thus it is a widespread public concern. Aerosols can be classified as the primary and secondary. Primary aerosols are released directly into the atmosphere by natural and artificial emission sources (Kong *et al.*, 2014). Natural aerosols are mainly emitted from forest fires, wood burning, volcanic eruptions, and marine spraying, while artificial aerosols are primarily from automobiles, factories, construction activities and artificial open burning. Secondary

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[*] Corresponding author.
Tel.: +86-186-5595-3355
E-mail address: cuikangping@163.com (K. Cui)
Tel.: +86-159-7217-0421
E-mail address: zhujinning@whu.edu.cn (J. Zhu)
[**] Corresponding author.
Tel.: +86-151-5596-2356; +886-913-027-189
E-mail address: wjlee@mail.ncku.edu.tw

aerosols are produced by the photochemical oxidation or catalytic oxidation reactions of nitrogen oxides (NO, NO_2, N_2O) and/or volatiles. Studies have shown that the formation of secondary aerosols depends on their saturation ratio and environmental conditions (Lee *et al.*, 2016). Moreover, aerosols in the ambient air can be removed from the atmosphere by dry or wet deposition (Cheruiyot *et al.*, 2015; Cheruiyot *et al.*, 2016; Li *et al.*, 2016b).

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are combined as PCDD/Fs, which belong to the dioxin family (Schecter *et al.*, 2006), and these substance have drawn increasing attention due to the ubiquitous nature of their toxicity, as well as the long-distance transport and their overall ability to disperse due to their stability, persistence, lipophilicity, bioaccumulation and other properties (Chen *et al.*, 2010; Lee *et al.*, 2016). PCDD/Fs are semi-volatile organic compounds (SVOCs) and persistent organic pollutants (POPs) that are toxic to human health and ecosystem (Cheruiyot *et al.*, 2015, 2016; Li *et al.*, 2016a). There are 210 possible congeners of PCDD/Fs, and of these only 17 with chlorine atoms attached to the 2, 3, 7 and 8 positions have been shown to especially toxic. In particular, the 2,3,7,8-tetrachloro-dibenzo-*p*-dioxin (2,3,7,8-TCDD) is one of the most toxic chemicals (Van den Berg *et al.*, 1998; Li *et al.*, 2016a). And these is given a toxicity equivalence factor (TEF) of 1, and the other 16 congeners have TEF values set relative to this. Furthermore, these TEFs are multiplied by the corresponding concentrations of individual congeners, and this then produces the toxic equivalent quantities (TEQs) (Lohmann and Jones, 1998; Van den Berg *et al.*, 1998; Cheruiyot *et al.*, 2016).

PCDD/Fs are mainly derived from natural combustion processes and human activities. The natural processes include volcanic eruptions and forest fires, while the human include manufacturing processes, mostly those associated with waste combustion, emissions from chemical plants, and metal smelting processes, as well as vehicle exhaust emissions (Wang *et al.*, 2003; Lin *et al.*, 2007; Hsieh *et al.*, 2009; Chuang *et al.*, 2011). Since PCDD/Fs are SVOCs, their distribution in the atmosphere between the gas- and the particle-phase determines their subsequent fate (Lohmann *et al.*, 2000; Suryani R. *et al.*, 2015). PCDD/Fs can be degraded by chemically reactive OH radicals and photochemical reactions (Chi *et al.*, 2009), and the removal of PCDD/Fs in the ambient air can occur by both dry and wet depositions (Huang *et al.*, 2011a; Mi *et al.*, 2012).

The PCDD/Fs have been shown to accumulate in the food chain due to their properties of being chemically stable and low dissolved properties (Shih *et al.*, 2006). This can then lead to enzyme induction, reproductive and developmental toxicity, immunotoxicity, adverse endocrine effects, chloramphenicol and tumor promotion and other hazards to human health (King-Heiden *et al.*, 2012; Tsai *et al.*, 2014).

Due to the rapid economic and industrial growth in China over the past twenty years, the $PM_{2.5}$ concentration in the ambient air is a significant environmental issue (Pui *et al.*, 2014). Guo *et al.* (2014) showed that the $PM_{2.5}$ concentrations in Beijing were extremely high, which were above 500 $\mu g\ m^{-3}$ for several months in the recent years. Several studies suggest the carbonaceous species, including organic carbon (OC) and elemental carbon (EC), are major components of $PM_{2.5}$ in China (Dan *et al.*, 2004; Zhou *et al.*, 2012; Tian *et al.*, 2013). Among these, OC usually includes mutagenic or carcinogenic compounds, such as PAHs, PCBs, PCDD/Fs and PBDEs. EC involves salt formation from SO_2, NO_x, O_3 and other gaseous compounds, which may also intervene in some important gas-to-particle heterogeneous reactions (Novakov, 1984; Yang *et al.*, 2005; Gu *et al.*, 2010). It is thus important to examine both $PM_{2.5}$ and PCDD/F levels and distributions in China in order to establish control strategies to effectively reduce these above two air pollutants in the atmosphere.

As such, the objectives of this study were to investigate the (1) $PM_{2.5}$ concentrations, (2) R_M ratio, (3) $PM_{2.5}/PM_{10}$ ratio, (4) total-PCDD/F-WHO_{2005}-TEQ concentrations, and (5) phase distributions of PCDD/Fs. The results of this study provide useful background information for the control of both $PM_{2.5}$ and persistent organic pollutants (POPs) in China and in other countries.

METHODS

In this study, a total of 23 cities belonging to 13 provinces and two municipalities in southern China were examined for the period 2014–2016 (Fig. 1). These cities were Nanjing and Suzhou in Jiangsu province, Hangzhou and Wenzhou in Zhejiang province, Hefei in Anhui province, Wuhan in Hubei province, Changsha in Hunan province, Guiyang in Guizhou province, Nanchang in Jiangxi province, Guangzhou and Shenzhen in Guangdong province, Chengdu and Luzhou in Sichuan province, Kunming and Qujing in Yunnan province, Nanning and Guilin in Guangxi province, Fuzhou and Quanzhou in Fujian province, Haikou and Sanya in Hainan province, Shanghai in Shanghai municipality and Chongqing in Chongqing municipality. Both $PM_{2.5}$ and PM_{10} concentrations for each city were obtained from the Air Quality Monitoring Stations (The Real Atmosphere Network) website. The main emission sources of these 23 cities in southern China are industrial sources and motor vehicle exhaust emissions. For example, Xu reported that in Guangzhou City, the industrial sources ranked first among all emission sources, accounting for 32.1% of the total, of which 20.6% was from coal-fired burners, 11.5% from industrial technology, followed by motor vehicle exhaust gas, accounting for 21.7% (Xu, 2015). In addition, the total-PCDD/F-WHO_{2005}-TEQ concentrations in each month for each city were simulated using the regression analyses (Wang *et al.*, 2010; Huang *et al.*, 2011a; Lee *et al.*, 2016). Taiwan is also located off the coast of China, and thus the data for China is compared with that for Taiwan. In addition, to reduce system error, two regression equations were selected for these comparison, and the results were averaged. The differences between these two regression equations were between 20% and 30%, which was acceptable for the trace amount PCDD/Fs. The regression equations were as follows: $Y_1 = 0.0138X + 0.0472$ and $Y_2 = 0.0117X - 0.021$; Y_1 and Y_2 is total-PCDD/F mass concentration, while X is PM_{10} concentration in ambient air, respectively. The final total-PCDD/F mass concentration is the average of Y_1 and Y_2.

Fig. 1. A Map of China.

Finally, by using the meteorological data, PM_{10} concentration and gas-particle partitioning model, the phase distribution of PCDD/Fs in the cities of Guangzhou and Nanjing city, respectively, were also calculated and discussed.

Gas-Particle Partitioning

Gaseous and particulate concentrations of PCDD/Fs were evaluated by the gas-particle partitioning multiplied by the total concentrations of PCDD/Fs. The gas-particle partitioning was simulated by 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):

$$K_p = \frac{F/TSP}{A} \tag{1}$$

where K_p: temperature-dependent partitioning constant ($m^3\ \mu g^{-1}$)
TSP: concentration of total suspended particulate matter, which was multiplied by PM_{10} concentration with 1.24 ($\mu g\ m^{-3}$)
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

$$\mathrm{Log}\ K_p = m_r \times \log P_L^o + b_r \tag{2}$$

where P_L^o: subcooled liquid vapor pressure (Pa)
m_r: cited slope, –1.29 (Chao *et al.*, 2004)
b_r: cited y-intercept, –7.2 (Chao *et al.*, 2004).

Chao *et al.* (2004) reported the gas-particle partitioning of PCDD/Fs in Taiwan and gave values for $m_r = -1.29$ 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.

Previous research has 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).

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

where RIs are the gas chromatographic retention indexes developed by Donnelly *et al.* (1987) and Hale *et al.* (1985), and T is ambient temperature (K), which was obtained from China's Meteorological Yearbook. The total PCDD/Fs-TEQ of mixture is obtained by summing up the individual TEQ, and the results are shown in Tables 5–8. The seventeen

2,3,7,8 substituted PCDD/F congener profiles in spring, summer, fall and winter, respectively, were cited from Wang *et al.* (2010).

RESULTS AND DISCUSSION

PM$_{2.5}$ Concentration

The PM$_{2.5}$ concentration can influence both the level and the phase distributions of PCDD/Fs in the atmosphere. During the periods between 2014 and 2016, the corresponding PM$_{2.5}$ concentrations in various cities of southern China are as shown in Tables 1–4. Among the 23 cities examined, the lowest PM$_{2.5}$ concentrations during 2014–2016 were all found to be at Sanya and Haikou both in Hainan Province, with the concentrations of 18.5 and 22.3 µg m^{-3}, 17.1 and 21.5 µg m^{-3} and 13.6 and 21.2 µg m^{-3}, in the 2014, 2015, and 2016, respectively. This is because Hainan Province located at the southeast site of China with both less traffic vehicles and industries. While during 2014 and 2015, the highest PM$_{2.5}$ concentrations were found to be at Wuhan City in Hubei Province with the PM$_{2.5}$ concentration of 80.5 µg m^{-3} and 68.8 µg m^{-3}, respectively; however in 2016, the highest PM$_{2.5}$ concentration was found to be 64.0 µg m^{-3} for Luzhou City in Sichuan Province. During 2014–2016, the

Table 1. Atmospheric PM$_{2.5}$ Concentration in Various Cities (2014–2016) (µg m^{-3}).

Province		Shanghai	Jiangsu		Zhejiang		Anhui
City	Month	Shanghai	Nanjing	Suzhou	Hangzhou	Wenzhou	Hefei
2014	Jan.	76.5	129	99.5	103	76.4	154.8
	Feb.	52.3	78.2	67.0	60.0	38.0	84.8
	Mar.	57.0	74.8	70.6	59.6	55.5	77.5
	Apr.	52.9	60.0	61.8	60.2	49.7	66.5
	May	62.8	84.7	76.1	59.0	53.4	76.1
	June	44.1	89.6	61.1	53.8	37.0	94.5
	July	45.0	64.8	55.6	43.9	28.3	58.5
	Aug.	38.0	42.2	51.7	45.3	30.8	54.1
	Sep.	35.2	50.7	41.7	45.5	31.6	54.5
	Oct.	40.0	67.0	54.4	58.8	46.6	74.8
	Nov.	52.2	81.8	71.0	75.1	47.1	95.4
	Dec.	71.7	63.3	80.6	71.8	58.9	68.2
	Average	52.3	73.8	65.9	61.4	46.1	79.9
	Range	35.2–76.5	42.2–129	41.7–99.5	43.9–103.4	28.3–76.4	54.1–154.8
2015	Jan.	83.0	95.8	96.2	88.5	73.1	101.5
	Feb.	64.3	73.0	72.0	64.8	51.3	83.6
	Mar.	54.0	56.0	60.1	52.9	44.7	63.4
	Apr.	55.8	50.2	62.6	53.8	54.9	57.8
	May	43.6	51.7	45.7	53.8	46.2	62.2
	June	43.3	45.8	43.2	40.0	40.1	45.1
	July	39.4	36.1	39.1	31.2	26.7	40.0
	Aug.	40.0	32.8	41.6	38.5	29.9	39.1
	Sep.	33.5	30.1	38.6	42.9	34.0	41.3
	Oct.	47.4	56.3	53.4	55.6	39.5	70.5
	Nov.	57.5	57.0	54.4	52.1	39.0	65.3
	Dec.	81.7	94.2	86.9	90.5	45.0	112.9
	Average	53.6	56.6	57.8	55.4	43.7	65.2
	Range	33.5–83.0	30.1–95.8	38.6–96.2	31.2–88.5	26.7–73.1	39.1–101.5
2016	Jan.	69.8	79.2	72.4	80.2	50.9	87.9
	Feb.	54.4	69.5	62.3	63.8	44.6	79.7
	Mar.	52.1	77.4	62.9	71.5	52.5	86.9
	Apr.	55.6	44.9	45.7	45.9	48.2	50.5
	May	50.8	42.9	42.6	46.4	43.3	48.5
	June	39.6	36.6	32.2	35.5	32.1	41.5
	July	38.3	30.4	30.8	32.0	28.4	36.0
	Aug.	18.9	25.7	23.0	25.0	22.6	36.6
	Sep.	32.2	33.4	32.8	33.3	27.4	45.5
	Oct.	21.8	24.5	26.7	31.9	26.1	34.4
	Nov.	44.0	43.4	51.2	51.6	40.3	54.8
	Dec.	57.7	65.7	73.8	69.4	47.0	86.0
	Average	44.6	47.8	46.4	48.9	38.6	57.4
	Range	18.9–69.8	24.5–79.2	23.0–73.8	25.0–80.2	22.6–50.9	34.4–87.9

Table 2. Atmospheric $PM_{2.5}$ Concentration in Various Cities (2014–2016) ($\mu g\ m^{-3}$).

Province		Hubei	Hunan	Jiangxi	Guizhou	Guangdong	
City	Month	Wuhan	Changsha	Nanchang	Guiyang	Guangzhou	Shenzhen
2014	Jan.	182.9	160.8	103.9	93.1	90.5	59.9
	Feb.	93.2	76.6	36.5	50.0	45.9	31.1
	Mar.	83.0	65.1	42.4	44.0	55.4	36.6
	Apr.	62.9	61.1	39.7	42.7	48.6	32.0
	May	74.8	65.4	56.2	44.3	32.6	16.6
	June	83.2	72.6	46.7	37.9	34.1	19.6
	July	44.2	47.8	33.2	24.5	35.5	19.9
	Aug.	43.6	46.6	30.8	25.7	31.3	14.7
	Sep.	46.1	51.5	44.0	29.8	35.5	26.0
	Oct.	90.7	98.8	76.2	52.6	58.7	45.1
	Nov.	77.5	73.5	55.1	38.3	49.6	43.1
	Dec.	83.7	78.0	47.6	63.2	55.5	50.1
	Average	80.5	74.8	51.0	45.5	47.8	32.9
	Range	43.6–182.9	46.6–160.8	30.8–103.9	24.5–93.1	31.3–90.5	14.7–59.9
2015	Jan.	123.6	114.5	67.3	66.8	59.8	49.4
	Feb.	107.7	90.6	52.0	53.4	54.9	47.3
	Mar.	68.2	55.0	34.0	44.3	33.1	31.2
	Apr.	58.6	49.4	33.3	40.4	38.2	27.1
	May	65.5	59.5	45.3	30.5	26.6	16.0
	June	40.8	31.3	24.1	20.1	20.5	11.0
	July	35.8	37.6	27.1	28.8	27.8	19.1
	Aug.	41.3	39.8	30.4	27.1	38.5	24.6
	Sep.	47.4	50.9	32.1	28.8	41.5	29.6
	Oct.	71.2	69.5	53.3	42.6	44.7	37.6
	Nov.	57.2	43.6	32.4	33.1	38.1	31.7
	Dec.	108.6	77.5	68.3	42.2	41.5	31.8
	Average	68.8	59.9	41.6	38.2	38.9	29.7
	Range	35.8–123.6	31.3–114.5	24.1–68.3	20.1–66.8	20.5–59.8	11.0–49.4
2016	Jan.	106.7	81.8	59.4	40.7	34.9	29.8
	Feb.	79.9	70.6	66.5	56.4	36.4	30.4
	Mar.	83.9	69.6	69.3	42.4	47.5	34.9
	Apr.	47.1	44.8	34.4	36.1	37.9	25.9
	May	43.6	39.6	29.6	30.5	30.6	22.4
	June	31.0	32.8	21.6	22.3	23.4	12.8
	July	24.3	31.1	23.5	17.1	26.6	17.1
	Aug.	33.7	36.8	22.8	30.1	36.7	24.0
	Sep.	44.5	52.7	35.2	42.1	35.6	27.0
	Oct.	35.2	43.4	28.9	31.1	30.1	24.4
	Nov.	60.7	55.1	43.4	34.6	40.9	30.8
	Dec.	92.8	83.9	78.6	57.9	50.9	44.5
	Average	57.0	53.5	42.8	36.8	36.0	27.0
	Range	24.3–106.7	31.1–83.9	21.6–78.6	17.1–57.9	23.4–50.9	12.8–44.5

lowest three-year average concentrations of $PM_{2.5}$ occurred at Sanya and Haikou and were 16.4 and 21.7 $\mu g\ m^{-3}$, respectively; while the highest concentrations of $PM_{2.5}$ occurred at Wuhan and Luzhou and were 68.8 and 63.1 $\mu g\ m^{-3}$, respectively.

Among 23 cities, the top six with the highest $PM_{2.5}$ concentrations were Wuhan, Hefei, Chengdu, Luzhou, Changsha and Nanjing with $PM_{2.5}$ concentrations between 47.8 and 80.5 $\mu g\ m^{-3}$ and an average of 64.4 $\mu g\ m^{-3}$. In contrast, the six cities with the lowest $PM_{2.5}$ concentration were Sanya, Haikou, Quanzhou, Fuzhou, Kunming and Shenzhen, with $PM_{2.5}$ concentrations between 13.6 and 32.9 $\mu g\ m^{-3}$ and an average of 26.0 $\mu g\ m^{-3}$. In general, we can see that is the lower the latitude of the city, the lower its $PM_{2.5}$ concentration. Moreover, the high results for both Wuhan and Hefei were probably due to them being more industrialized and with higher populations, which thus require more coal and other fossil fuel combustion, with more air pollutant emissions from both industry and vehicles, while both are also at relatively high latitudes. During the winter time, this is a very low ground temperature in higher latitudes, which makes it easier for a more stable state of vertical atmospheric convection to occur, which causes an atmospheric temperature inversion. This will then readily

Table 3. Atmospheric $PM_{2.5}$ Concentration in Various Cities (2014–2016) (µg m^{-3}).

Province		Sichuan		Yunnan		Guangxi	
City	Month	Chengdu	Luzhou	Kunming	Qujing	Nanning	Guilin
2014	Jan.	179	168	44.4	47.4	112	145
	Feb.	113	100	28.1	34.4	52.1	55.2
	Mar.	83.7	63.8	46.1	41.4	48.9	61.2
	Apr.	59.9	50.4	51.3	47.4	38.3	52.1
	May	75.7	64.4	29.3	32.0	32.5	55.3
	June	43.8	43.8	21.8	25.9	29.6	54.7
	July	39.9	35.5	21.8	26.0	21.1	34.0
	Aug.	37.9	38.4	21.6	26.0	21.5	34.8
	Sep.	39.9	39.4	21.9	27.5	32.9	45.6
	Oct.	64.0	57.5	35.6	40.8	70.1	85.1
	Nov.	53.1	38.7	27.4	28.0	49.2	63.3
	Dec.	83.8	74.0	40.1	41.9	70.6	61.6
	Average	72.8	64.5	32.5	34.9	48.2	62.3
	Range	37.9–179	35.5–168	21.6–51.3	25.9–47.4	21.1–112	34.0–145
2015	Jan.	126	127	34.3	33.8	71.3	81.7
	Feb.	85.6	95.8	31.4	30.5	72.9	82.4
	Mar.	67.6	66.5	38.5	36.5	39.0	46.9
	Apr.	53.9	53.7	32.9	31.4	43.2	49.3
	May	49.0	54.1	29.7	32.0	27.8	38.0
	June	38.1	47.0	18.7	20.2	18.7	25.2
	July	45.4	37.1	29.9	32.8	29.2	32.8
	Aug.	44.1	46.8	22.4	26.2	33.6	35.5
	Sep.	35.1	33.6	23.4	25.7	29.4	50.7
	Oct.	63.1	57.1	28.5	32.4	52.9	58.5
	Nov.	50.5	52.4	25.8	24.2	34.1	39.0
	Dec.	90.2	57.5	33.0	34.0	41.3	50.6
	Average	59.9	60.7	29.0	30.0	41.1	49.2
	Range	35.1–126	33.6–127	18.7–38.5	20.2–36.5	18.7–72.9	25.2–82.4
2016	Jan.	80.8	72.3	31.1	28.1	42.9	55.1
	Feb.	66.7	83.8	39.2	40.8	60.4	68.6
	Mar.	74.6	79.3	36.6	39.5	46.8	65.7
	Apr.	64.6	51.1	31.9	33.8	38.1	43.6
	May	56.7	59.6	25.9	27.6	27.4	43.2
	June	43.6	50.7	15.0	17.9	17.1	25.7
	July	29.8	38.8	16.6	20.0	18.7	24.4
	Aug.	41.0	55.4	22.5	30.8	27.2	36.0
	Sep.	45.4	67.0	28.2	37.7	38.8	52.3
	Oct.	46.5	53.1	26.8	30.0	35.8	38.9
	Nov.	85.8	69.5	25.5	26.1	29.1	39.2
	Dec.	116.5	87.4	35.6	39.3	56.6	69.9
	Average	62.7	64.0	27.9	31.0	36.6	46.9
	Range	29.8–116.5	38.8–87.4	15.0–39.2	17.9–40.8	17.1–56.6	24.4–69.9

lead to an accumulation of $PM_{2.5}$ within the human activity zone, resulting in the $PM_{2.5}$ concentrations for the central cities of China that were much higher than those of southern cities. Moreover, the air current blowing from the north parts of China with very high $PM_{2.5}$ concentration levels may also increase the $PM_{2.5}$ concentrations in the cities of central China, like Wuhan and Hefei.

A report in the Chengdu Daily Newspaper (2010) stated that due to the influences of El Niño and the La Nina, every year, just before and after November, the meteorological conditions in the Sichuan basin (including Chengdu and Luzhou) become significantly getting worse, coupled with weak cold air activity. This basin has thus experienced three regional episodes of air pollution, causing a rise in $PM_{2.5}$ concentrations in both Luzhou and Chengdu.

Comparing the annual average of $PM_{2.5}$ concentrations in the 23 southern cities during 2014–2016, it can be concluded that the concentrations are highest in 2014, followed by 2015, with the lowest in 2016. Among the 23 cities, Nanjing saw the highest reduction in $PM_{2.5}$ concentration between 2014 and 2015, falling from 73.8 to 56.6 µg m^{-3}, with a reduction rate of 26.4%, and this continued to fall to 47.8 µg m^{-3} in 2016, down by 15.3%. It can be seen that in 2014 the annual average $PM_{2.5}$ concentration of the 23 cities in

Table 4. Atmospheric $PM_{2.5}$ Concentration in Various Cities (2014–2016) (μg m^{-3}).

Province		Fujian		Hainan		Chongqing
City	Month	Fuzhou	Quanzhou	Haikou	Sanya	Chongqing
2014	Jan.	56.2	57.1	54.7	46.9	131
	Feb.	25.9	26.6	20.6	17.1	85.9
	Mar.	39.1	38.5	19.6	17.7	55.5
	Apr.	35.1	32.6	16.8	17.9	42.9
	May	33.1	33.5	13.3	12.7	47.9
	June	26.1	25.5	13.7	11.1	35.7
	July	20.3	23.5	10.4	9.20	31.5
	Aug.	24.3	31.1	10.9	8.30	32.4
	Sep.	23.5	25.6	15.7	13.3	39.1
	Oct.	31.9	29.1	33.8	26.6	70.5
	Nov.	31.0	28.5	23.2	18.5	86.9
	Dec.	36.7	32.8	34.9	23.1	95.6
	Average	31.9	32.0	22.3	18.5	62.9
	Range	20.3–56.2	23.5–57.1	10.4–54.7	8.30–46.9	31.5–131
2015	Jan.	49.3	42.4	38.2	30.5	122
	Feb.	37.9	35.1	38.4	28.0	81.6
	Mar.	31.6	28.7	16.8	16.7	50.7
	Apr.	35.4	28.4	20.4	16.4	46.1
	May	30.3	29.5	13.4	13.0	40.1
	June	21.1	21.5	9.50	9.40	35.8
	July	18.5	21.3	15.0	13.6	46.1
	Aug.	21.4	18.9	16.7	11.1	42.7
	Sep.	21.6	23.7	16.5	12.8	34.1
	Oct.	26.2	25.5	33.6	24.5	48.2
	Nov.	22.6	22.5	16.6	13.1	46.5
	Dec.	28.1	25.1	22.5	16.1	65.4
	Average	28.7	26.9	21.5	17.1	54.9
	Range	18.5–49.3	18.9–42.4	9.50–38.4	9.40–30.5	34.1–122
2016	Jan.	36.1	28.1	21.4	15.4	70.7
	Feb.	33.3	38.3	31.3	19.9	71.0
	Mar.	40.5	37.7	24.4	16.3	57.4
	Apr.	30.0	31.8	24.3	16.4	44.7
	May	29.3	26.5	18.2	11.5	48.4
	June	20.0	20.9	11.8	8.00	42.1
	July	20.2	22.7	12.4	8.60	34.0
	Aug.	19.4	25.4	14.1	10.5	41.4
	Sep.	19.9	22.1	20.0	11.8	70.0
	Oct.	17.7	24.0	22.7	13.3	39.9
	Nov.	26.2	29.9	18.3	11.6	53.4
	Dec.	35.0	31.1	35.6	19.9	74.4
	Average	27.3	28.2	21.2	13.6	54.0
	Range	17.7–40.5	20.9–38.3	11.8–35.6	8.60–19.9	34.0–74.4

this study was 49.8 μg m^{-3}, and the relative standard deviation (RSD%) was 39.0%; while for 2015, the annual average in $PM_{2.5}$ concentration was 42.9 μg m^{-3}, and the relative standard deviation (RSD%) was 37.0%; and for 2016, the annual average in $PM_{2.5}$ concentration was 41.3 μg m^{-3}, and the relative standard deviation (RSD%) was 29.7%. The annual average concentration of $PM_{2.5}$ in 2015 decreased by 6.9 μg m^{-3} (14.9%) compared to 2014, while for 2016 it fell by 1.6 μg m^{-3} (3.7%) compared to 2015. Thus, during the last three years, even though the air quality of most cities was improved, the $PM_{2.5}$ concentrations were still very high, and thus more efforts need to be made to address this issue.

Comparing the figures for 2015 and 2016, the $PM_{2.5}$ concentrations of the cities were all decreased. For example, for Wuhan it decreased from 68.8 to 57.0 μg m^{-3}, for Hefei from 65.2 to 57.4 μg m^{-3}, the Changsha deceased from 59.9 to 53.5 μg m^{-3}, the Shanghai deceased from 53.6 to 44.6 μg m^{-3}, the Nanjing deceased from 56.6 to 47.8 μg m^{-3}, and the Hangzhou deceased from 55.4 to 48.9 μg m^{-3}. However, the $PM_{2.5}$ concentration of some cities were increased from 2015 to 2016. For example, for Chengdu it increased from 59.9 to 62.7 μg m^{-3}, for Luzhou increased from 60.7 to 64.0 μg m^{-3}, the Nanchang increased from 41.6 to 42.8 μg m^{-3}, the Qujing increased from 30.0 to 31.0 μg m^{-3} and the Quanzhou increased from 26.9 to 28.2 µg m^{-3}. During 2015 and 2016, these five cities with higher $PM_{2.5}$ concentrations all had poorly controlled air quality, and this is probably because the amount of emissions increased, for examples, due to more vehicles and factories, as well as less air dispersion.

The monthly averages of the $PM_{2.5}$ concentrations in the 23 cities in 2014 and 2015 show that December the concentration of $PM_{2.5}$ in Nanjing increased from 63.3 to 94.2 µg m^{-3}, that for Hangzhou increased from 71.8 to 90.5 µg m^{-3}, Hefei from 68.2 to 112.9 µg m^{-3}, Wuhan from 83.7 to 108.6 µg m^{-3}, and Chengdu from 83.8 to 90.2 µg m^{-3}, and in the other months the contractions all fell. The reason for this is probably because the temperature in December 2015 was lower than that in December 2014, and thus the vertical atmospheric convection was hindered and slower. Therefore, the cumulative effect of air pollutants resulted in greater $PM_{2.5}$ concentrations. However, in December 2016, there are several cities with a higher $PM_{2.5}$ concentration than seen in December 2015. This is because of climate change, and the heights of the atmospheric temperature inversion layer during December were approximately 2000, 1000 and 500 meters in 2014, 2015 and 2016, respectively. This meteorological condition in 2016 was increasingly detrimental to the dispersion of air pollutants.

From the point of view of seasonal variations, the $PM_{2.5}$ concentration was the highest in winter and lowest in summer. For example, during 2016, in Wuhan, the average $PM_{2.5}$ concentrations during winter (January, February and December), spring (March, April, May), summer (June, July, August) and fall (September, October, November) were 93.1, 58.2, 29.7 and 46.8 µg m^{-3}, respectively; in Hefei, the average $PM_{2.5}$ concentrations in winter, spring, summer and fall were 84.5, 62.0, 38.0 and 44.9 µg m^{-3}, respectively; in Nanjing, the average $PM_{2.5}$ concentrations in winter, spring, summer and fall were 71.5, 55.1, 30.9 and 33.8 µg m^{-3}, respectively; in Chengdu, the average $PM_{2.5}$ concentrations in winter, spring, summer and fall were 88.0, 65.3, 38.1 and 59.2 µg m^{-3}, respectively. This is because the winter temperature was lower and the vertical transport of air current was thus hindered, and making the $PM_{2.5}$ accumulate. Moreover, in winter both rainfall and wind speed were weaker, reducing the effects of both rainfall scavenging and wind blowing with regard to removing the PM.

The R_M of Atmospheric $PM_{2.5}$

R_M refers to the ratio of the annual average of $PM_{2.5}$ concentration over the air quality standard of 10 µg m^{-3}, as regulated by the World Health Organization (WHO). From Figs. 2(A)–2(C), it can be seen that in 2014 and 2015 the highest R_M values were in Wuhan, with 8.10 and 6.90, respectively, however for 2016 Wuhan was in fourth place (R_M of 5.70), while Luzhou and Chengdu rose to the first and second places (6.40 and 6.27), respectively. During 2014–2016 Sanya always had the lowest R_M values, with 1.89, 1.70 and 1.36, respectively. The R_M values showed that, in the 23 cities of southern China examined in this work, during 2014–2016 the top six R_M for each year were between 5.4 and 8.1 and averaged 6.5. This means that the average $PM_{2.5}$ concentrations were up to 6.5 times the WHO air quality regulated standard. However, during 2014–2016, the six lowest R_M were in the range between 1.36 and 3.5, and averaged 2.78. In addition, when we compared the values among 2014–2016, the average R_M values were 5.20, 4.49 and 4.13 in 2014, 2015 and 2016, respectively. These results indicate that, during the last three years the $PM_{2.5}$ concentrations in these cities in southern China slowly decreased, but were still high and well above the WHO air quality regulated standard. The emissions sources thus need better control strategies, and action and the $PM_{2.5}$ levels in the ambient air need to be significantly reduced.

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

As for the atmospheric $PM_{2.5}/PM_{10}$ ratios, during 2014 the six highest annual average of $PM_{2.5}/PM_{10}$ concentration ratios were 0.84 in Changsha, 0.77 in Guilin, 0.74 in Suzhou, 0.72 in Shanghai, 0.72 in Hefei and 0.71 in Wuhan. In 2015, the six highest annual average $PM_{2.5}/PM_{10}$ concentration ratios were 0.75 in Changsha, 0.73 in Shanghai, 0.71 in Guilin, 0.71 in Suzhou, 0.68 in Luzhou and 0.67 in Hefei. While in 2016, the six highest annual averages $PM_{2.5}/PM_{10}$ concentration ratios were 0.73 in Luzhou, 0.71 in Guilin, 0.69 in Chongqing, 0.69 in Shanghai, 0.68 in Changsha

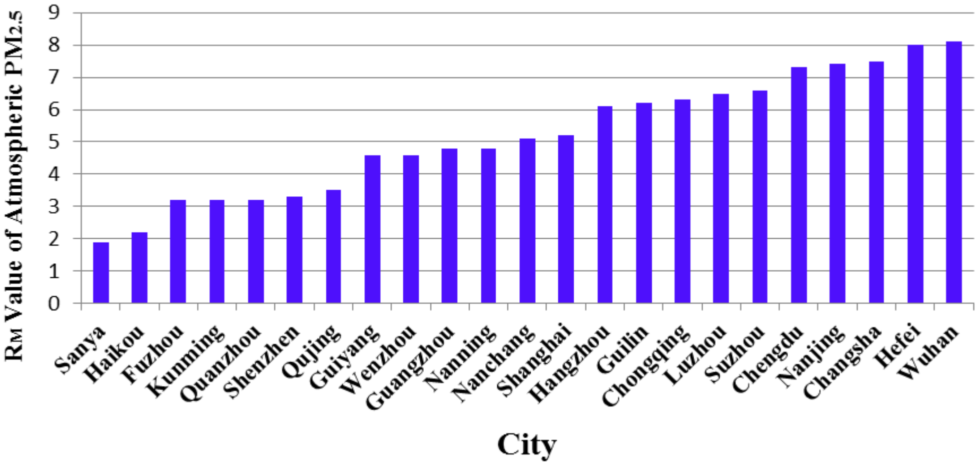

Fig. 2(A). The R_M Value of Atmospheric $PM_{2.5}$ in Various Cities in Southern China (2014).

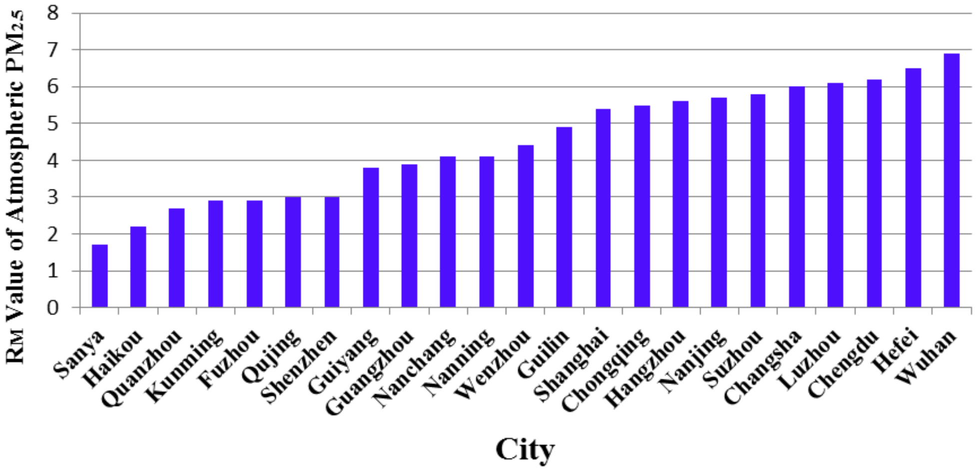

Fig. 2(B). The R_M Value of Atmospheric $PM_{2.5}$ in Various Cities in Southern China (2015).

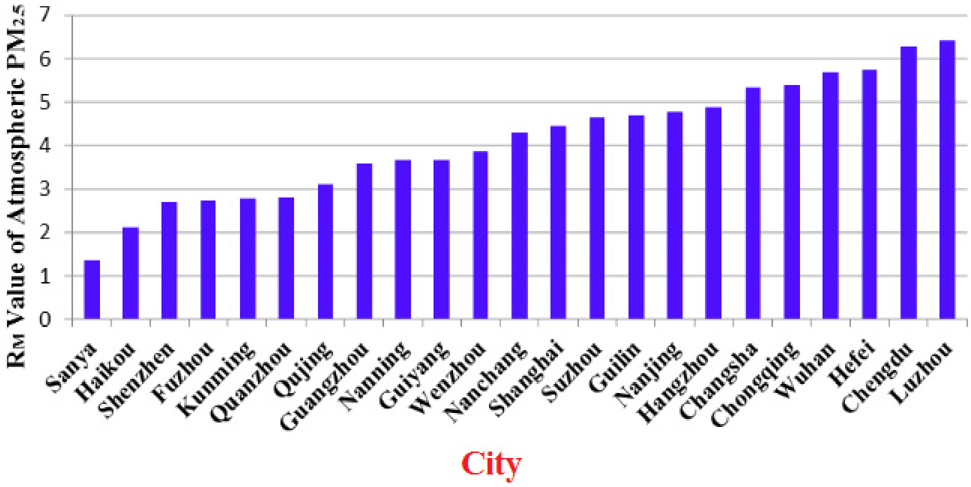

Fig. 2(C). The R_M Value of Atmospheric $PM_{2.5}$ in Various Cities in Southern China (2016).

and 0.64 in Hefei. However, in 2014, the six lowest annual averages of $PM_{2.5}/PM_{10}$ concentration ratios were 0.50 in Fuzhou, 0.50 in Quanzhou, 0.51 in Sanya, 0.51 in Kunming, 0.53 in Haikou and 0.57 in Nanning. In 2015, the six lowest annual averages of $PM_{2.5}/PM_{10}$ concentration ratios were 0.52 in Fuzhou, 0.52 in Quanzhou, 0.53 in Haikou, 0.53 in Sanya, 0.53 in Kunming and 0.55 in Nanchang. Finally, in 2016 they were 0.48 in Sanya, 0.49 in Kunming, 0.52 in Fuzhou, 0.54 in Nanchang, 0.54 in Haikou and 0.55 in Qujing. In the atmosphere of the southern China region, the $PM_{2.5}/PM_{10}$ concentration ratios were in the range of 0.48 and 0.84 and averaged 0.62.

In general, there was a higher $PM_{2.5}$ concentration with a higher $PM_{2.5}/PM_{10}$ ratio. For examples, during 2014–2016, among the 23 cities examined in this work, the top six with the highest $PM_{2.5}$ concentrations were Wuhan, Hefei, Chengdu, Luzhou, Changsha and Nanjing, and the $PM_{2.5}$ concentrations averaged 64.4 $\mu g\ m^{-3}$ while the $PM_{2.5}/PM_{10}$ ratio averaged 0.66. However, during 2014–2016, the six cities with lowest $PM_{2.5}$ concentrations were Sanya, Haikou, Quanzhou, Fuzhou, Kunming and Shenzhen, which had an average $PM_{2.5}$ concentration of 26.0 $\mu g\ m^{-3}$ and with an average $PM_{2.5}/PM_{10}$ ratio of 0.53.

The results indicated that $PM_{2.5}$ is the major portion of atmospheric particles. They also revealed that the atmospheric particles in the southern China were mostly resulted from the gas-particle transformation, going through the condensation and flocculation processes, and finally in the accumulation mode of $PM_{2.5}$. Moreover, some of particles due to the resuspension of road dust or entrainment of naked lands may also contribute certain fractions of the $PM_{2.5}$ concentration in the ambient air. A higher $PM_{2.5}/PM_{10}$ concentration ratio means the air pollutant is more detrimental to human health, and so needs more attention.

Simulated Total-PCDD/F-WHO$_{2005}$-TEQ Concentrations in Ambient Air

Lee *et al.* (2016) showed that there is a strong correlation (R = 0.99) between the PM_{10} level and the total-PCDD/F mass concentration. Therefore, this study calculated the concentrations of PCDD/Fs by regression analysis. The modeled concentrations of total-PCDD/Fs-WHO$_{2005}$-TEQ are shown in Tables 5–8.

From Tables 5–8, it can be seen that the total-PCDD/Fs-

WHO$_{2005}$-TEQ concentration in the 23 southern cities for 2014 was between 0.0140 and 0.1573 pg-WHO$_{2005}$-TEQ m^{-3}. The highest annual average of the total-PCDD/Fs-WHO$_{2005}$-TEQ concentration was 0.0772 pg-WHO$_{2005}$-TEQ m^{-3} in Nanjing, and the lowest was 0.0223 pg-WHO$_{2005}$-TEQ m^{-3} in Sanya. The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in Sanya was 71.1% less than in Nanjing. While for 2015, the total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in the 23 cities ranged between 0.0136 and 0.1199 pg-WHO$_{2005}$-TEQ m^{-3}. The highest annual average of the total-PCDD/Fs-WHO$_{2005}$-TEQ concentration was 0.0658 pg-WHO$_{2005}$-TEQ m^{-3} in Wuhan, and the lowest was 0.0204 pg-WHO$_{2005}$-TEQ m^{-3} in Sanya.The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in Sanya was 69.0% less than in Wuhan. For 2016, the total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in the 23 cities ranged between 0.0109 and 0.1079 pg-WHO$_{2005}$-TEQ m^{-3}. The highest annual average of the total-PCDD/Fs-WHO$_{2005}$-TEQ concentration was 0.0605 pg-WHO$_{2005}$-TEQ m^{-3} in Chengdu, and the lowest was 0.0166 pg-WHO$_{2005}$-TEQ m^{-3} in Sanya. The total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in Sanya was 72.6% less than in Chengdu.

Moreover, Lee *et al.* (2016) reported that in the Kaohsiung

Table 5. Atmospheric Total-PCDD/F-WHO$_{2005}$-TEQ Concentration in Various Cities (2014–2016) (Unit: pg-WHO$_{2005}$-TEQ m^{-3}).

Province		Shanghai	Jiangsu		Zhejiang		Anhui
City	Month	Shanghai	Nanjing	Suzhou	Hangzhou	Wenzhou	Hefei
2014	Jan.	0.0557	0.1237	0.0628	0.0873	0.0735	0.1008
	Feb.	0.0369	0.0672	0.0470	0.0441	0.0326	0.0547
	Mar.	0.0547	0.0916	0.0658	0.0617	0.0592	0.0698
	Apr.	0.0440	0.0703	0.0541	0.0627	0.0567	0.0531
	May	0.0648	0.1118	0.0780	0.0749	0.0586	0.0961
	June	0.0327	0.0723	0.0470	0.0501	0.0340	0.0710
	July	0.0357	0.0519	0.0440	0.0406	0.0287	0.0540
	Aug.	0.0296	0.0336	0.0406	0.0397	0.0274	0.0466
	Sep.	0.0349	0.0542	0.0420	0.0466	0.0332	0.0553
	Oct.	0.0451	0.0789	0.0583	0.0655	0.0516	0.0854
	Nov.	0.0471	0.0870	0.0594	0.0707	0.0501	0.0798
	Dec.	0.0696	0.0840	0.0753	0.0719	0.0619	0.0619
	Average	0.0459	0.0772	0.0562	0.0597	0.0473	0.0691
	Range	0.0296–0.0696	0.0336–0.1137	0.0406–0.0753	0.0397–0.0873	0.0274–0.0735	0.0466–0.1008
2015	Jan.	0.0666	0.0950	0.0691	0.0739	0.0719	0.0749
	Feb.	0.0528	0.0710	0.0557	0.0537	0.0466	0.0648
	Mar.	0.0430	0.0627	0.0485	0.0531	0.0466	0.0576
	Apr.	0.0531	0.0627	0.0602	0.0582	0.0627	0.0658
	May	0.0415	0.0586	0.0450	0.0536	0.0496	0.0658
	June	0.0323	0.0431	0.0366	0.0344	0.0401	0.0419
	July	0.0323	0.0362	0.0336	0.0279	0.0261	0.0344
	Aug.	0.0327	0.0366	0.0379	0.0344	0.0274	0.0419
	Sep.	0.0327	0.0410	0.0410	0.0445	0.0358	0.0481
	Oct.	0.0481	0.0737	0.0614	0.0594	0.0451	0.0758
	Nov.	0.0492	0.0583	0.0496	0.0507	0.0430	0.0548
	Dec.	0.0642	0.0883	0.0696	0.0830	0.0446	0.0893
	Average	0.0457	0.0606	0.0507	0.0522	0.0450	0.0596
	Range	0.0323–0.0666	0.0362–0.0950	0.0336–0.0696	0.0279–0.0830	0.0261–0.0719	0.0344–0.0893
2016	Jan.	0.0511	0.0695	0.0559	0.0673	0.0459	0.0612
	Feb.	0.0437	0.0655	0.0516	0.0568	0.0406	0.0603
	Mar.	0.0452	0.0788	0.0548	0.0733	0.0567	0.0710
	Apr.	0.0484	0.0530	0.0484	0.0488	0.0530	0.0544
	May	0.0456	0.0521	0.0479	0.0461	0.0479	0.0558
	June	0.0284	0.0328	0.0300	0.0304	0.0328	0.0336
	July	0.0312	0.0296	0.0300	0.0288	0.0275	0.0332
	Aug.	0.0194	0.0288	0.0255	0.0215	0.0235	0.0401
	Sep.	0.0281	0.0409	0.0371	0.0309	0.0290	0.0557
	Oct.	0.0233	0.0295	0.0290	0.0338	0.0319	0.0328
	Nov.	0.0385	0.0519	0.0495	0.0523	0.0442	0.0523
	Dec.	0.0437	0.0634	0.0546	0.0616	0.0498	0.0647
	Average	0.0372	0.0497	0.0429	0.0460	0.0402	0.0513
	Range	0.0194–0.0511	0.0288–0.0788	0.0255–0.0559	0.0215–0.0733	0.0235–0.0567	0.0328–0.071

Table 6. Atmospheric Total-PCDD/F-WHO_{2005}-TEQ Concentration in Various Cities (2014–2016) (Unit: pg-WHO_{2005}-TEQ m^{-3}).

Province		Hubei	Hunan	Jiangxi	Guizhou	Guangdong	
City	Month	Wuhan	Changsha	Nanchang	Guiyang	Guangzhou	Shenzhen
2014	Jan.	0.1199	0.0796	0.0926	0.0672	0.0719	0.0557
	Feb.	0.0508	0.0360	0.0336	0.0360	0.0379	0.0283
	Mar.	0.0794	0.0557	0.0485	0.0440	0.0516	0.0400
	Apr.	0.0627	0.0547	0.0445	0.0485	0.0415	0.0349
	May	0.0931	0.0688	0.0638	0.0511	0.0328	0.0213
	June	0.0710	0.0527	0.0427	0.0340	0.0300	0.0240
	July	0.0462	0.0444	0.0362	0.0257	0.0310	0.0244
	Aug.	0.0410	0.0387	0.0340	0.0279	0.0279	0.0204
	Sep.	0.0501	0.0481	0.0496	0.0338	0.0373	0.0338
	Oct.	0.0921	0.0824	0.0711	0.0522	0.0563	0.0501
	Nov.	0.0620	0.0527	0.0522	0.0384	0.0445	0.0435
	Dec.	0.0777	0.0561	0.0369	0.0581	0.0508	0.0475
	Average	0.0705	0.0558	0.0505	0.0431	0.0428	0.0353
	Range	0.0410–0.1199	0.0387–0.0796	0.0340–0.0926	0.0257–0.0672	0.0279–0.0719	0.0204–0.0557
2015	Jan.	0.0849	0.0642	0.0628	0.0561	0.0533	0.0494
	Feb.	0.0719	0.0575	0.0508	0.0422	0.0466	0.0446
	Mar.	0.0638	0.0450	0.0394	0.0415	0.0324	0.0344
	Apr.	0.0718	0.0485	0.0471	0.0491	0.0394	0.0344
	May	0.0729	0.0511	0.0531	0.0344	0.0278	0.0217
	June	0.0440	0.0287	0.0287	0.0218	0.0204	0.0148
	July	0.0383	0.0318	0.0336	0.0296	0.0257	0.0200
	Aug.	0.0480	0.0344	0.0423	0.0266	0.0340	0.0244
	Sep.	0.0583	0.0507	0.0410	0.0302	0.0430	0.0307
	Oct.	0.0860	0.0707	0.0620	0.0455	0.0507	0.0394
	Nov.	0.0527	0.0410	0.0353	0.0379	0.0445	0.0349
	Dec.	0.0964	0.0642	0.0614	0.0407	0.0399	0.0312
	Average	0.0658	0.0490	0.0465	0.0380	0.0381	0.0317
	Range	0.0383–0.0964	0.0287–0.0707	0.0287–0.0628	0.0218–0.0561	0.0204–0.0533	0.0148–0.0494
2016	Jan.	0.0795	0.0568	0.0511	0.0358	0.0310	0.0253
	Feb.	0.0690	0.0572	0.0599	0.0472	0.0301	0.0262
	Mar.	0.0806	0.0608	0.0705	0.0433	0.0424	0.0323
	Apr.	0.0530	0.0415	0.0405	0.0401	0.0327	0.0244
	May	0.0511	0.0392	0.0373	0.0382	0.0304	0.0221
	June	0.0336	0.0279	0.0259	0.0263	0.0219	0.0134
	July	0.0300	0.0292	0.0292	0.0215	0.0239	0.0162
	Aug.	0.0409	0.0336	0.0271	0.0288	0.0296	0.0203
	Sep.	0.0604	0.0519	0.0461	0.0442	0.0352	0.0262
	Oct.	0.0366	0.0376	0.0366	0.0338	0.0300	0.0238
	Nov.	0.0385	0.0519	0.0495	0.0523	0.0442	0.0523
	Dec.	0.0690	0.0612	0.0721	0.0516	0.0468	0.0402
	Average	0.0535	0.0457	0.0455	0.0386	0.0332	0.0269
	Range	0.0300–0.0806	0.0279–0.0612	0.0259–0.0721	0.0215–0.0523	0.0219–0.0468	0.0134–0.0523

area, southern Taiwan, the total-PCDD/F-WHO_{2005}-TEQ concentrations were in the ranges of 0.021–0.077 and 0.021–0.072 pg WHO_{2005}-TEQ m^{-3}, respectively, and averaged 0.048 and 0.044 pg WHO_{2005}-TEQ m^{-3}, respectively for 2014 and 2015 (Lee *et al.*, 2016), which was much lower than those in both Nanjing (0.0772 pg-WHO_{2005}-TEQ m^{-3}) in 2014 and Wuhan (0.0658 pg-WHO_{2005}-TEQ m^{-3}) in 2015.

During 2014-2016, the six highest three-year averages of total-PCDD/Fs-TEQ concentrations were 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. However, during 2014-2016, the six lowest three-year averages of total-PCDD/Fs-TEQ concentrations were 0.0201, 0.0245, 0.0313, 0.0335, 0.0342 and 0.0349 pg-WHO_{2005}-TEQ m^{-3} in Sanya, Haikou, Shenzhen, Qujing, Quanzhou and Fuzhou, respectively. The reasons for this difference were that the southern cities with higher latitudes are located in downstream of the air pollution current from the northern cities, and may be affected by the long-range transport of the pollutants from resulting in higher $PM_{2.5}$ and total-PCDD/Fs-WHO_{2005}-TEQ concentrations. These results can be compared with those in Chi *et al.* (2013),

Table 7. Atmospheric Total-PCDD/F-WHO_{2005}-TEQ Concentration in Various Cities (2014–2016) (Unit: pg-WHO_{2005}-TEQ m^{-3}).

Province		Sichuan		Yunnan		Guangxi	
City	Month	Chengdu	Luzhou	Kunming	Qujing	Nanning	Guilin
2014	Jan.	0.1573	0.1290	0.0470	0.0571	0.0998	0.1118
	Feb.	0.0926	0.0758	0.0330	0.0393	0.0446	0.0316
	Mar.	0.0890	0.0638	0.0602	0.0511	0.0511	0.0466
	Apr.	0.0652	0.0496	0.0632	0.0536	0.0460	0.0445
	May	0.0850	0.0652	0.0400	0.0355	0.0419	0.0536
	June	0.0406	0.0349	0.0244	0.0223	0.0323	0.0410
	July	0.0401	0.0310	0.0257	0.0236	0.0266	0.0306
	Aug.	0.0362	0.0323	0.0249	0.0218	0.0300	0.0296
	Sep.	0.0451	0.0379	0.0312	0.0266	0.0420	0.0410
	Oct.	0.0685	0.0553	0.0471	0.0394	0.0742	0.0737
	Nov.	0.0635	0.0384	0.0405	0.0420	0.0553	0.0548
	Dec.	0.0960	0.0125	0.0451	0.0427	0.0739	0.0528
	Average	0.0733	0.0521	0.0402	0.0379	0.0515	0.0510
	Range	0.0362–0.1573	0.0310–0.1290	0.0244–0.0632	0.0218–0.0571	0.0266–0.0998	0.0296–0.1118
2015	Jan.	0.1199	0.1075	0.0374	0.0336	0.0691	0.0666
	Feb.	0.0777	0.0844	0.0330	0.0302	0.0600	0.0648
	Mar.	0.0743	0.0652	0.0440	0.0390	0.0390	0.0415
	Apr.	0.0658	0.0541	0.0405	0.0324	0.0536	0.0496
	May	0.0638	0.0541	0.0394	0.0318	0.0369	0.0359
	June	0.0397	0.0357	0.0204	0.0174	0.0244	0.0223
	July	0.0462	0.0323	0.0310	0.0244	0.0323	0.0279
	Aug.	0.0440	0.0366	0.0244	0.0204	0.0383	0.0306
	Sep.	0.0399	0.0353	0.0307	0.0241	0.0353	0.0455
	Oct.	0.0717	0.0589	0.0399	0.0323	0.0691	0.0516
	Nov.	0.0553	0.0501	0.0408	0.0382	0.0449	0.0449
	Dec.	0.0883	0.0514	0.0345	0.0323	0.0380	0.0380
	Average	0.0656	0.0555	0.0347	0.0297	0.0451	0.0433
	Range	0.0397–0.1199	0.0323–0.1075	0.0204–0.0440	0.0174–0.0382	0.0244–0.0691	0.0223–0.0666
2016	Jan.	0.0712	0.0572	0.0332	0.0310	0.0398	0.0428
	Feb.	0.0625	0.0642	0.0341	0.0358	0.0498	0.0537
	Mar.	0.0779	0.0631	0.0405	0.0433	0.0475	0.0553
	Apr.	0.0617	0.0405	0.0378	0.0429	0.0396	0.0373
	May	0.0581	0.0479	0.0304	0.0327	0.0323	0.0382
	June	0.0397	0.0328	0.0182	0.0194	0.0198	0.0211
	July	0.0267	0.0271	0.0203	0.0198	0.0219	0.0215
	Aug.	0.0356	0.0340	0.0235	0.0259	0.0255	0.0288
	Sep.	0.0428	0.0490	0.0319	0.0366	0.0404	0.0419
	Oct.	0.0500	0.0552	0.0366	0.0328	0.0381	0.0328
	Nov.	0.0913	0.0676	0.0414	0.0324	0.0319	0.0357
	Dec.	0.1079	0.0708	0.0463	0.0433	0.0511	0.0520
	Average	0.0605	0.0508	0.0329	0.0330	0.0365	0.0384
	Range	0.0267–0.1079	0.0271–0.0708	0.0182–0.0463	0.0194–0.0433	0.0198–0.0511	0.0211–0.0553

which proposed an atmospheric PCDD/F concentration in the range of 0.00187–0.0102 pg I-TEQ m^{-3} in the northern mountainous area of Taiwan, while that for a coastal area in southern Taiwan ranged between 0.00237 and 0.00374 pg I-TEQ m^{-3}.

The highest monthly average total-PCDD/Fs-WHO_{2005}-TEQ concentrations occurred in October in Chengdu for 2014 and 2015 (averaged 0.1573 and 0.1199 pg-WHO_{2005}-TEQ m^{-3}, respectively), while for 2016, it occurred in December in Chengdu, with a total-PCDD/Fs-WHO_{2005}-TEQ concentration of 0.1097 pg-WHO_{2005}-TEQ m^{-3}. However, the lowest monthly average concentrations occurred in July (0.0140 pg-WHO_{2005}-TEQ m^{-3}) during 2014 in Sanya, while for 2015 and 2016, it occurred in June at Sanya (0.0136 and 0.0109 pg-WHO_{2005}-TEQ m^{-3}, respectively). With regard to total-PCDD/Fs-WHO_{2005}-TEQ concentration during the whole of 2014–2016, it was found that the concentration in winter was higher than in summer. Comparing the monthly total-PCDD/Fs-WHO_{2005}-TEQ concentrations with the atmospheric $PM_{2.5}$ and PM_{10} levels showed that the concentrations were closely related to the levels of particulate matter in the ambient air. The higher the levels

Table 8. Atmospheric Total-PCDD/F-WHO$_{2005}$-TEQ Concentration in Various Cities (2014–2016) (Unit: pg-WHO$_{2005}$-TEQ m^{-3}).

Province		Fujian		Hainan		Chongqing
City	Month	Fuzhou	Quanzhou	Haikou	Sanya	Chongqing
2014	Jan.	0.0551	0.0619	0.0490	0.0437	0.1118
	Feb.	0.0288	0.0288	0.0221	0.0192	0.0729
	Mar.	0.0496	0.0547	0.0264	0.0233	0.0586
	Apr.	0.0445	0.0501	0.0233	0.0202	0.0450
	May	0.0440	0.0450	0.0192	0.0157	0.0557
	June	0.0318	0.0327	0.0174	0.0144	0.0327
	July	0.0279	0.0349	0.0144	0.0140	0.0349
	Aug.	0.0327	0.0397	0.0144	0.0140	0.0336
	Sep.	0.0353	0.0332	0.0204	0.0204	0.0445
	Oct.	0.0435	0.0373	0.0379	0.0327	0.0685
	Nov.	0.0405	0.0327	0.0282	0.0245	0.0813
	Dec.	0.0403	0.0360	0.0350	0.0249	0.0733
	Average	0.0395	0.0406	0.0256	0.0223	0.0594
	Range	0.0279–0.0551	0.0288–0.0619	0.0144–0.0490	0.0140–0.0437	0.0327–0.1118
2015	Jan.	0.0475	0.0407	0.0369	0.0316	0.1051
	Feb.	0.0379	0.0330	0.0355	0.0269	0.0710
	Mar.	0.0390	0.0359	0.0227	0.0213	0.0561
	Apr.	0.0485	0.0374	0.0258	0.0217	0.0522
	May	0.0400	0.0405	0.0202	0.0182	0.0485
	June	0.0283	0.0318	0.0144	0.0136	0.0340
	July	0.0240	0.0283	0.0179	0.0166	0.0440
	Aug.	0.0249	0.0240	0.0196	0.0140	0.0387
	Sep.	0.0292	0.0323	0.0221	0.0164	0.0358
	Oct.	0.0338	0.0332	0.0358	0.0266	0.0507
	Nov.	0.0307	0.0266	0.0236	0.0184	0.0466
	Dec.	0.0316	0.0273	0.0259	0.0192	0.0551
	Average	0.0346	0.0326	0.0250	0.0204	0.0532
	Range	0.0240–0.0475	0.0240–0.0407	0.0144–0.0369	0.0136–0.0316	0.0340–0.1051
2016	Jan.	0.0315	0.0258	0.0214	0.0162	0.0516
	Feb.	0.0306	0.0354	0.0271	0.0192	0.0511
	Mar.	0.0387	0.0369	0.0267	0.0198	0.0507
	Apr.	0.0359	0.0355	0.0276	0.0198	0.0396
	May	0.0364	0.0304	0.0217	0.0147	0.0465
	June	0.0235	0.0231	0.0154	0.0109	0.0332
	July	0.0251	0.0259	0.0162	0.0117	0.0304
	Aug.	0.0235	0.0247	0.0158	0.0146	0.0336
	Sep.	0.0247	0.0243	0.0228	0.0162	0.0571
	Oct.	0.0247	0.0243	0.0238	0.0176	0.0381
	Nov.	0.0338	0.0338	0.0214	0.0167	0.0504
	Dec.	0.0384	0.0315	0.0341	0.0223	0.0594
	Average	0.0306	0.0293	0.0228	0.0166	0.0451
	Range	0.0235–0.0387	0.0231–0.0369	0.0154–0.0341	0.0109–0.0223	0.0304–0.0594

of particulate matter, the higher the total-PCDD/Fs-WHO$_{2005}$-TEQ concentrations. Therefore, reductions in ambient dioxin levels should control the concentration of particulates in the atmosphere from the source.

Gas-Particle Partitioning of PCDD/Fs

The gas-particle partitioning of PCDD/Fs is an important factor that influences the efficiency of atmospheric removal by wet and dry deposition (Bidleman and Harner, 2000). When PCDD/Fs are emitted into the atmosphere, they have a certain distribution between the gas and particle phases based on their concentrations, the atmospheric temperature, vapor pressure and the ambient air particulate concentration (Hoff *et al.*, 1996). The gas-particle partitioning of PCDD/Fs in the ambient air of the 23 cities examined in this work is similar. The results for 12 of these cities are further compared and discussed, below.

The seasonal gas-particle partitioning of PCDD/Fs in the ambient air of 12 cities in 2014 is illustrated in Tables 9–14. During summer 2014 (June, July and August), the top six cities with the average highest temperatures were Haikou (28.9°C), Fuzhou (28.6°C), Guangzhou (28.4°C), Nanning

Table 9. Seasonal Gas-Particle Partitioning of Individual PCDD/F Mass Concentrations in Various Cities during 2014.

PCDD/Fs	Shanghai								Nanjing							
	Spring		Summer		Fall		Winter		Spring		Summer		Fall		Winter	
	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)
2,3,7,8-TeCDD	21	79	4	96	11	89	56	44	28	72	6	94	22	78	73	27
1,2,3,7,8-PeCDD	57	43	17	83	39	61	89	11	66	34	25	75	56	44	95	5
1,2,3,4,7,8-HxCDD	89	11	51	49	73	27	98	2	91	9	61	39	84	16	99	1
1,2,3,6,7,8-HxCDD	87	13	52	48	74	26	98	2	91	9	63	37	85	15	99	1
1,2,3,7,8,9-HxCDD	89	11	55	45	76	24	98	2	92	8	65	35	86	14	99	1
1,2,3,4,6,7,8-HpCDD	98	2	85	15	94	6	100	0	98	2	90	10	97	3	100	0
OCDD	100	0	97	3	99	1	100	0	100	0	98	2	100	0	100	0
2,3,7,8-TeCDF	15	85	3	97	8	92	45	55	21	79	4	96	16	84	63	37
1,2,3,7,8-PeCDF	41	59	10	90	26	74	79	21	51	49	15	85	41	59	89	11
2,3,4,7,8-PeCDF	49	51	13	87	31	69	84	16	58	42	19	81	48	52	92	8
1,2,3,4,7,8-HxCDF	78	22	36	64	61	39	96	4	84	16	47	53	75	25	98	2
1,2,3,6,7,8-HxCDF	79	21	38	62	62	38	96	4	85	15	48	52	76	24	98	2
1,2,3,7,8,9-HxCDF	85	15	49	51	72	28	98	2	90	10	59	41	83	17	99	1
2,3,4,6,7,8-HxCDF	83	17	44	56	68	32	97	3	88	12	55	45	81	19	99	1
1,2,3,4,6,7,8-HpCDF	94	6	73	27	87	13	99	1	96	4	80	20	93	7	100	0
1,2,3,4,7,8,9-HpCDF	97	3	85	15	93	7	100	0	98	2	89	11	97	3	100	0
OCDF	99	1	96	4	98	2	100	0	100	0	97	3	99	1	100	0

Table 10. Seasonal Gas-Particle Partitioning of Individual PCDD/F Mass Concentrations in Various Cities during 2014.

PCDD/Fs	Hefei								Wuhan							
	Spring		Summer		Fall		Winter		Spring		Summer		Fall		Winter	
	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)
2,3,7,8-TeCDD	23	77	6	94	22	78	70	30	24	76	5	95	21	79	70	30
1,2,3,7,8-PeCDD	60	40	25	75	57	43	94	6	61	39	23	77	55	45	94	6
1,2,3,4,7,8-HxCDD	88	12	62	38	85	15	99	1	89	11	58	42	83	17	99	1
1,2,3,6,7,8-HxCDD	89	11	63	37	85	15	99	1	90	10	60	40	84	16	99	1
1,2,3,7,8,9-HxCDD	90	10	66	34	87	13	99	1	90	10	63	37	86	14	99	1
1,2,3,4,6,7,8-HpCDD	98	2	90	10	97	3	100	0	98	2	89	11	96	4	100	0
OCDD	97	3	95	5	100	0	100	0	100	0	98	2	99	1	100	0
2,3,7,8-TeCDF	17	83	4	96	17	83	59	41	17	83	4	96	15	85	60	40
1,2,3,7,8-PeCDF	44	56	14	86	42	58	87	13	45	55	13	87	41	59	87	13
2,3,4,7,8-PeCDF	52	48	19	81	49	51	91	9	53	47	17	83	48	52	91	9
1,2,3,4,7,8-HxCDF	80	20	48	52	76	24	98	2	81	19	44	56	75	25	98	2
1,2,3,6,7,8-HxCDF	81	19	49	51	77	23	98	2	82	18	45	55	76	24	98	2
1,2,3,7,8,9-HxCDF	87	13	60	40	84	16	99	1	88	12	57	43	83	17	99	1
2,3,4,6,7,8-HxCDF	85	15	56	44	81	19	98	2	86	14	52	48	80	20	99	1
1,2,3,4,6,7,8-HpCDF	95	5	81	19	93	7	100	0	95	5	78	22	93	7	100	0
1,2,3,4,7,8,9-HpCDF	98	2	90	10	97	3	100	0	98	2	88	12	96	4	100	0
OCDF	99	1	97	3	99	1	100	0	99	1	97	3	99	1	100	0

Table 11. Seasonal Gas-Particle Partitioning of Individual PCDD/F Mass Concentrations in Various Cities during 2014.

PCDD/Fs	Guiyang								Chengdu							
	Spring		Summer		Fall		Winter		Spring		Summer		Fall		Winter	
	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)
2,3,7,8-TeCDD	22	78	6	94	17	83	60	40	26	74	6	94	21	79	73	27
1,2,3,7,8-PeCDD	60	40	24	76	51	49	91	9	64	36	24	76	56	44	94	6
1,2,3,4,7,8-HxCDD	89	11	62	38	82	18	98	2	90	10	62	38	85	15	99	1
1,2,3,6,7,8-HxCDD	89	11	64	36	83	17	99	1	90	10	63	37	85	15	99	1
1,2,3,7,8,9-HxCDD	90	10	66	34	84	16	99	1	91	9	66	34	87	13	99	1
1,2,3,4,6,7,8-HpCDD	98	2	91	9	96	4	100	0	98	2	91	9	97	3	100	0
OCDD	100	0	98	2	100	0	100	0	100	0	98	2	100	0	100	0
2,3,7,8-TeCDF	16	84	4	96	12	88	49	51	19	81	4	96	15	85	64	36
1,2,3,7,8-PeCDF	43	57	14	86	36	64	82	18	48	52	14	86	41	59	89	11
2,3,4,7,8-PeCDF	51	49	18	82	42	58	87	13	56	44	18	82	48	52	92	8
1,2,3,4,7,8-HxCDF	81	19	47	53	72	28	97	3	83	17	47	53	76	24	98	2
1,2,3,6,7,8-HxCDF	82	18	48	52	73	27	97	3	83	17	48	52	77	23	98	2
1,2,3,7,8,9-HxCDF	88	12	60	40	81	19	98	2	89	11	60	40	84	16	99	1
2,3,4,6,7,8-HxCDF	85	15	55	45	78	22	98	2	87	13	55	45	81	19	99	1
1,2,3,4,6,7,8-HpCDF	96	4	81	19	92	8	99	1	96	4	81	19	93	7	100	0
1,2,3,4,7,8,9-HpCDF	98	2	89	11	96	4	100	0	98	2	90	10	97	3	100	0
OCDF	99	1	97	3	99	1	100	0	99	1	97	3	99	1	100	0

Table 12. Seasonal Gas-Particle Partitioning of Individual PCDD/F Mass Concentrations in Various Cities during 2014.

PCDD/Fs	Changsha								Nanchang							
	Spring		Summer		Fall		Winter		Spring		Summer		Fall		Winter	
	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)
2,3,7,8-TeCDD	17	83	4	96	14	86	50	50	15	85	3	97	12	88	47	53
1,2,3,7,8-PeCDD	52	48	18	82	44	56	86	14	47	53	15	85	41	59	85	15
1,2,3,4,7,8-HxCDD	84	16	51	49	73	23	97	3	82	18	46	54	74	26	97	3
1,2,3,6,7,8-HxCDD	85	15	53	47	78	22	97	3	82	18	47	53	75	25	97	3
1,2,3,7,8,9-HxCDD	86	14	55	45	80	20	98	2	84	16	50	50	77	23	98	2
1,2,3,4,6,7,8-HpCDD	97	3	85	15	95	5	100	0	97	3	83	17	94	6	99	1
OCDD	99	1	97	3	99	1	100	0	99	1	96	4	99	1	100	0
2,3,7,8-TeCDF	12	88	3	97	10	90	39	61	10	90	2	98	9	91	37	63
1,2,3,7,8-PeCDF	36	64	10	90	30	70	75	25	32	68	8	92	28	72	72	28
2,3,4,7,8-PeCDF	43	57	13	87	36	64	80	20	39	61	11	89	33	67	79	21
1,2,3,4,7,8-HxCDF	74	26	36	64	66	34	95	5	71	29	32	68	63	37	94	6
1,2,3,6,7,8-HxCDF	75	25	37	63	67	33	95	5	72	28	33	67	64	36	95	5
1,2,3,7,8,9-HxCDF	83	17	49	51	76	24	97	3	80	20	44	56	73	27	97	3
2,3,4,6,7,8-HxCDF	80	20	44	56	72	28	96	4	77	23	40	60	69	31	96	4
1,2,3,4,6,7,8-HpCDF	93	7	73	27	89	11	99	1	92	8	69	31	87	13	99	1
1,2,3,4,7,8,9-HpCDF	97	3	85	15	94	6	100	0	96	4	82	18	93	7	99	1
OCDF	99	1	95	5	99	1	100	0	99	1	94	6	98	2	100	0

Table 13. Seasonal Gas-Particle Partitioning of Individual PCDD/F Mass Concentrations in Various Cities during 2014.

PCDD/Fs	Guangzhou								Nanning							
	Spring		Summer		Fall		Winter		Spring		Summer		Fall		Winter	
	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)
2,3,7,8-TeCDD	8	92	2	98	6	94	29	71	9	91	3	97	8	92	37	63
1,2,3,7,8-PeCDD	32	68	11	89	26	74	71	29	32	68	12	88	32	68	77	23
1,2,3,4,7,8-HxCDD	68	32	38	62	62	38	93	7	66	34	39	61	67	33	95	5
1,2,3,6,7,8-HxCDD	69	31	39	61	63	37	94	6	67	33	41	59	69	31	95	5
1,2,3,7,8,9-HxCDD	71	29	42	58	66	34	94	6	62	38	44	56	71	29	96	4
1,2,3,4,6,7,8-HpCDD	92	8	78	22	89	11	99	1	91	9	79	21	92	8	99	1
OCDD	98	2	95	5	98	2	100	0	98	2	95	5	98	2	100	0
2,3,7,8-TeCDF	6	94	2	98	4	96	21	79	6	94	2	98	6	94	28	72
1,2,3,7,8-PeCDF	20	80	6	94	16	84	55	45	20	80	6	94	20	80	63	37
2,3,4,7,8-PeCDF	25	75	8	92	20	80	63	37	25	75	8	92	25	75	70	30
1,2,3,4,7,8-HxCDF	54	46	25	75	48	52	88	12	53	47	26	74	55	45	91	9
1,2,3,6,7,8-HxCDF	55	45	26	74	49	51	88	12	54	46	27	73	56	44	91	9
1,2,3,7,8,9-HxCDF	66	34	36	64	60	40	93	7	65	35	38	62	66	34	94	6
2,3,4,6,7,8-HxCDF	62	38	31	69	56	44	91	9	60	40	33	67	62	38	93	7
1,2,3,4,6,7,8-HpCDF	84	16	61	39	80	20	98	2	83	17	63	37	84	16	98	2
1,2,3,4,7,8,9-HpCDF	91	9	77	23	89	11	99	1	91	9	78	22	91	9	99	1
OCDF	98	2	92	8	97	3	100	0	97	3	93	7	97	3	100	0

Table 14. Seasonal Gas-Particle Partitioning of Individual PCDD/F Mass Concentrations in Various Cities during 2014.

PCDD/Fs	Fuzhou								Haikou							
	Spring		Summer		Fall		Winter		Spring		Summer		Fall		Winter	
	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)	P^a (%)	G^b (%)
2,3,7,8-TeCDD	13	87	2	98	6	94	27	73	2	98	2	95	2	98	11	89
1,2,3,7,8-PeCDD	44	56	11	89	25	75	69	31	12	88	11	89	13	87	41	59
1,2,3,4,7,8-HxCDD	79	21	38	62	59	41	93	7	39	61	35	65	42	58	78	22
1,2,3,6,7,8-HxCDD	80	20	39	61	60	40	93	7	40	60	36	64	43	57	79	21
1,2,3,7,8,9-HxCDD	81	19	42	58	63	37	94	6	43	57	38	62	46	54	81	19
1,2,3,4,6,7,8-HpCDD	96	4	77	23	88	12	99	1	77	23	71	29	80	20	96	4
OCDD	99	1	95	5	97	3	100	0	94	6	92	8	95	5	99	1
2,3,7,8-TeCDF	9	91	2	98	4	96	20	80	2	98	2	98	2	98	8	92
1,2,3,7,8-PeCDF	29	71	6	94	15	85	53	47	7	93	6	94	7	93	26	74
2,3,4,7,8-PeCDF	36	64	8	92	19	81	61	39	9	91	8	92	9	91	33	67
1,2,3,4,7,8-HxCDF	67	33	25	75	46	54	87	13	26	74	24	76	28	72	66	34
1,2,3,6,7,8-HxCDF	68	32	26	74	47	53	88	12	27	73	25	75	30	70	67	33
1,2,3,7,8,9-HxCDF	77	23	36	64	57	43	92	8	37	63	34	66	40	60	77	23
2,3,4,6,7,8-HxCDF	74	26	32	68	53	47	90	10	33	67	30	70	35	65	73	27
1,2,3,4,6,7,8-HpCDF	91	9	61	39	78	22	97	3	61	39	55	45	65	35	91	9
1,2,3,4,7,8,9-HpCDF	95	5	76	24	87	13	99	1	76	24	70	30	79	21	95	5
OCDF	99	1	92	8	96	4	100	0	92	8	89	11	93	7	99	1

(27.9°C), Nanchang (27.7°C) and Changsha (27.6°C), with the average temperature being 28.2°C, and this all had lower particle-bound PCDD/Fs fractions, and particularly for the low molecular weight PCDD/F congeners. For example, the particle phase fractions of both 2,3,7,8-TeCDD and 2,3,7,8-TeCDF averaged 3% and 2%, respectively; the middle molecular weight PCDD/Fs, those of both 1,2,3,7,8-PeCDD and 1,2,3,7,8-PeCDF, were 13% and 7%, respectively; however, the high molecular weight PCDD/Fs were still had more fractions in the particle phase, and those of both OCDD and OCDF averaged 95% and 93%, respectively, in the particle phase.

During fall 2014 (September, October and November), the top six average temperatures were found to be in Haikou (26.1°C), Fuzhou (23.7°C), Guangzhou (23.9°C), Nanning (23.4°C), Nanchang (21.1°C) and Changsha (20.4°C), which an average of 23.1°C. The fractions of particle-bound PCDD/Fs, for the low molecular weight PCDD/F congeners, both 2,3,7,8-TeCDD and 2,3,7,8-TeCDF, averaged 8% and 6%, respectively; for the middle molecular weight PCDD/Fs, both 1,2,3,7,8-PeCDD and 1,2,3,7,8-PeCDF, were 30% and 19%, respectively; however, for the high molecular weight PCDD/Fs, both OCDD and OCDF were 98 % and 97%, respectively.

However, during winter 2014 (January, February, and December), the six cities with the lowest average temperatures were Hefei (4.5°C), Nanjing (5.0°C), Wuhan (5.1°C), Guiyang (5.3°C), Shanghai (6.2°C) and Chengdu (6.2°C), with the average being 5.4°C, and all of PCDD/F congeners were mainly with more particle-bound PCDD/Fs fractions, especially the low molecular weight PCDD/Fs, for which the particle phase fractions of both 2,3,7,8-TeCDD and 2,3,7,8-TeCDF averaged 67% and 57%, respectively; those of the middle molecular weight PCDD/Fs, both 1,2,3,7,8-PeCDD and 1,2,3,7,8-PeCDF, were 93% and 86%, respectively; while those of the high molecular weight PCDD/Fs, both OCDD and OCDF in the cold winter season were both almost 100% in the particle phase.

In addition, during spring 2014 (March, April, and May), the six cities with the highest temperatures were Hefei (17.2°C), Nanjing (16.9°C), Wuhan (17.4°C), Guiyang (14.9°C), Shanghai (16.3°C) and Chengdu (16.7°C), with an average of 16.5°C. The particle-bound PCDD/Fs fractions, for the low molecular weight PCDD/F congeners, both 2,3,7,8-TeCDD and 2,3,7,8-TeCDF averaged 24% and 18%, respectively; for the middle molecular weight PCDD/Fs, both 1,2,3,7,8-PeCDD and 1,2,3,7,8-PeCDF, were 61% and 45%, respectively; however, for the high molecular weight PCDD/Fs, both OCDD and OCDF, were 100% and 99%, respectively.

During winter 2014, the six cities (Hefei, Nanjing, Wuhan, Guiyang, Shanghai and Chengdu) with the lowest temperatures in winter (an average of 5.4°C) had average particle phase fractions of total-PCDD/Fs-WHO$_{2005}$-TEQ that were approximately 76%, 53%, 71% and 93% in the spring, summer, fall and winter season, respectively. In contrast, for the six cities (Haikou, Fuzhou, Guangzhou, Nanning, Nanchang and Changsha) with the highest temperatures in summer (an average of 16.5°C), their average particle phase fractions of total-PCDD/Fs-WHO$_{2005}$-TEQ were approximately 61%, 42%, 57% and 81% in the spring, summer, fall and winter, respectively.

These results prove that the low molecular weight PCDD/Fs were primarily in the gas phase, while the high molecular weight PCDD/F congeners were associated with the particulates (Wu *et al.*, 2009a; Lin *et al.*, 2010; Huang *et al.*, 2011a, b; Mi *et al.* 2012; Suryani R. *et al.*, 2015). In the summer, the temperatures were higher, and thus more of the PCDD/F mass fractions were in the gas phase, while in the winter the temperatures were quite low and more PCDD/F mass fractions were in the particle phase.

The results of this study provided information showing the trends of both atmospheric PM$_{2.5}$ and PCDD/Fs in 23 cities in southern China, and this can be useful for the establishment of better control strategies.

CONCLUSIONS

The results of this study illustrate that by using PM$_{10}$ concentration data, we can simulate PCDD/F concentrations in different areas, determine the spatial and temporal trends of PCDD/Fs, and further evaluate the effects of atmospheric PCDD/F deposition in the future. The results of this study can be summarized as follows:

1. Among the 23 cities in southern China examined in this work, those with higher latitudes had higher PM$_{2.5}$ concentrations than those with lower latitudes. During 2014–2016, the lowest annual average concentrations of PM$_{2.5}$ were in Sanya and Haikou and were 18.5 and 22.3 µg m^{-3} in 2014, 17.1 and 21.5 µg m^{-3} in 2015 and 13.6 and 21.2 µg m^{-3} in 2016, respectively. In contrast, the highest concentrations of PM$_{2.5}$ occurred in Wuhan and Luzhou, and were 80.5 and 64.5 µg m^{-3} in 2014, 68.8 and 60.7 µg m^{-3} in 2015 and 57.0 and 64.0 µg m^{-3} in 2016, respectively.
2. With regard to the annual average concentrations of PM$_{2.5}$ in the 23 cities, those in 2016 were the lowest (ranging between 13.6 and 64.0 µg m^{-3} and an average of 41.3 µg m^{-3}), followed by 2015 (ranging between 17.1 and 68.8 µg m^{-3} and an average of 42.9 µg m^{-3}), and the highest in 2014 (ranging between 18.5 and 80.5 µg m^{-3} and an average of 49.8 µg m^{-3}). Overall, the result showed that the atmospheric PM$_{2.5}$ levels in southern China have fallen, although slowly.
3. When comparing the PM$_{2.5}$ concentration in 2015 with those in 2016, it was found that the levels in most of the cities decreased, but there were still five cities in which they increased: Chengdu increased from 59.9 to 62.7 µg m^{-3}, Luzhou from 60.7 to 64.0 µg m^{-3}, Nanchang from 41.6 to 42.8 µg m^{-3}, Qujing from 30.0 to 31.0 µg m^{-3}, and Quanzhou from 26.9 to 28.2 µg m^{-3}. This means that during 2015 and 2016, these still had poor air pollution control.
4. With regard to the monthly averages, during December the PM$_{2.5}$ concentrations of most cities decreased. However, the concentrations of PM$_{2.5}$ in Nanjing increased from 63.3 to 94.2 µg m^{-3}, Hangzhou from 71.8 to 90.5 µg m^{-3}, Hefei from 68.2 to 112.9 µg m^{-3},

Wuhan from 83.7 to 108.6 μg m^{-3}, and Chengdu from 83.8 to 90.2 μg m^{-3}; while in the other months they fell. This was probably because the temperature in December 2015 was lower than that in December 2014 and so the vertical atmospheric convection was hindered and slower.

5. The R_M values showed that the top six R_M for each year were between 5.4 and 8.1 and averaged 6.5; this means that the average $PM_{2.5}$ concentrations were up to 6.5 times the WHO air quality standard. However, during 2014–2016, the six lowest R_M were in the range between 1.36 and 3.5, and averaged 2.78. In addition, when we compared the values among 2014–2016, the average R_M values were 5.20, 4.49 and 4.13 in 2014, 2015 and 2016, respectively. These results indicate that during last three years the $PM_{2.5}$ concentrations in these cities of southern China slowly fell, but remained high and still well above WHO air quality standard.
6. In general, there was a higher $PM_{2.5}$ concentration with a higher $PM_{2.5}/PM_{10}$ ratio. During 2014–2016, the six cities with the highest $PM_{2.5}$ concentrations were Wuhan, Hefei, Chengdu, Luzhou, Changsha and Nanjing, and the $PM_{2.5}$ concentration averaged 64.4 μg m^{-3} and the $PM_{2.5}/PM_{10}$ ratio averaged 0.66, However, the six cities with lowest $PM_{2.5}$ concentrations were Sanya, Haikou, Quanzhou, Fuzhou, Kunming and Shenzhen, which had an average $PM_{2.5}$ concentration of 26.0 μg m^{-3} and average $PM_{2.5}/PM_{10}$ ratio of 0.53.
7. During 2014–2016, the simulation results showed that the six highest three-year averages of total-PCDD/Fs-WHO_{2005}-TEQ concentrations were 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. However, the six lowest three-year averages of total-PCDD/Fs-TEQ concentrations were 0.0201, 0.0245, 0.0313, 0.0335, 0.0342 and 0.0349 pg-WHO_{2005}-TEQ m^{-3} in Sanya, Haikou, Shenzhen, Qujing, Quanzhou and Fuzhou, respectively.
8. Generally, during winter, particle-bound PCDD/Fs increased in fractions, but fell in the summer due to the influence of temperature. Additionally, higher chlorinated PCDD/Fs were associated with the particulate phase, while lower chlorinated congeners were predominantly in the gas phase.
9. As for the phase distributions of total-PCDD/Fs-WHO_{2005}-TEQ, during winter 2014 (January, February, and December), the six cities (Hefei, Nanjing, Wuhan, Guiyang, Shanghai and Chengdu) with the lowest temperatures in winter had average particle phase fractions of total-PCDD/Fs-WHO_{2005}-TEQ of approximately 76%, 53%, 71% and 93% in the spring, summer, fall and winter season, respectively. In contrast, the six cities (Haikou, Fuzhou, Guangzhou, Nanning, Nanchang and Changsha) with the highest temperatures in summer had average particle phase fractions of total-PCDD/Fs-WHO_{2005}-TEQ of approximately 61%, 42%, 57% and 81% in the spring, summer, fall and winter, respectively.
10. The results of this study provide the information of great importance showing the trends of both atmospheric $PM_{2.5}$ and PCDD/Fs in the 23 cities of southern China. They will thus be useful for the establishment of control strategies in the future.

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