



Sensitivity Analysis of PM_{2.5}-Bound Total PCDD/Fs-TEQ Content: In the Case of Wuhu City, China

Weiwei Wang¹, Kangping Cui^{1*}, Rong Zhao¹, Jinning Zhu^{1**}, Qianli Huang¹, Wen-Jhy Lee^{1,2***}

¹ School of Resources and Environmental Engineering, Hefei University of Technology, Hefei 246011, China

² Department of Environmental Engineering, National Cheng Kung University, Tainan 70101, Taiwan

ABSTRACT

During 2015–2017, the atmospheric PM_{2.5}, PM_{2.5}/PM₁₀, PCDD/Fs, PCDD/F phase distribution and PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in Wuhu and Bengbu were investigated in this study. In addition, the sensitivity analysis for PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ of Wuhu was also studied. During 2015–2017, the three-year average PM_{2.5} concentration in Wuhu was 53.0 μg m⁻³, and in Bengbu was 61.4 μg m⁻³; the results also showed the annual average PM_{2.5} concentrations of these two cities had declined, but the levels were still far above the WHO annual PM_{2.5} standard (10 μg m⁻³). In addition, in Wuhu, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in summer (0.166 ng-WHO₂₀₀₅-TEQ g⁻¹) were approximately only 68.8% in magnitude lower than the average value of other three seasons (0.532 ng-WHO₂₀₀₅-TEQ g⁻¹), and that of Bengbu in summer (0.187 ng-WHO₂₀₀₅-TEQ g⁻¹) was approximately 66.7% in magnitude lower than the average value of other three seasons (0.561 ng-WHO₂₀₀₅-TEQ g⁻¹). Sensitivity analysis showed that the PCDD/F concentration was the most positively correlated sensitive factor for PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ, and when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +106%. The second positively correlated sensitive factor was PM₁₀ concentration, and when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +72%. This was followed by atmospheric temperature, and its effect was negatively correlated, when ΔP/P was changed from -50% to +50%, ΔS/S responded from +73% to -112%. The last sensitive parameter was PM_{2.5} concentration, with the impact divided into two stages: when ΔP/P was changed from 0% to +70%, ΔS/S responded from 0% to +33%, but when ΔP/P was changed from +70% to +100%, ΔS/S responded from +33% to +25%. The results of this study provide useful information that can be used to achieve more insights into both atmospheric PM_{2.5} and PCDD/Fs.

Keywords: PM_{2.5}; PM₁₀; PCDD/Fs; Phase distribution; Sensitivity analysis.

INTRODUCTION

The particulate matter (PM) and polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) in the ambient air have received great public concern due to their significant correlation with the incidences of pulmonary and cardiac diseases (Schwartz *et al.*, 1996; Ito *et al.*, 2006).

Particulate matter (PM) is a suspension of solid or liquid particles in the atmosphere, which is a kind of aerosol (Ghosh

et al., 2014). According to the aerodynamic diameters of PM, it can be divided into TSP (range from ~0 to 100 μm), PM₁₀ (range from ~0 to 10 μm) and PM_{2.5} (range from ~0 to 2.5 μm) (Chow *et al.*, 2015; Lu *et al.*, 2016). The sources of PM can be natural or anthropogenic, with forest burning, dust storms and volcanic eruptions are the main natural sources, while industrial activities, vehicle exhausts and the construction industry are the major anthropogenic sources (Bilos *et al.*, 2001; Kong *et al.*, 2014; Alghamdi *et al.*, 2015). The PM in ambient air is a conglomerate of organic and inorganic carbon, mineral element, nitrates, ammonium, sulfate and so on (Zhu *et al.*, 2017). While the PM emitted into atmosphere by primary sources, the secondary aerosol forms with a specific ratio and environmental conditions (Lee *et al.*, 2016). Previous studies of the chemical characteristics of PM in the atmosphere indicated that it not only had adverse effects on air quality and even global climate, but also impacts on human health due to its particle toxicity (Chen *et al.*, 2014; Huang *et al.*, 2014; Wang *et al.*, 2014; Liu *et al.*, 2016).

PCDD/Fs are well-known persistent organic pollutants

* Corresponding author.

Tel.: +86-186-5595-3355

E-mail address: cuikangping@163.com

** Corresponding author.

Tel.: +86-159-7217-0421

E-mail address: zhujinning@whu.edu.cn

*** Corresponding author.

Tel.: +86-151-5596-2356; +886-913-027-189

E-mail address: wjlee@mail.ncku.edu.tw

(POPs) and semi-volatile organic compounds (SOCs), which can transport over long distances and interact for long periods in the environment (Wu *et al.*, 2009b; Chen *et al.*, 2014; Lee *et al.*, 2016; Redfern, *et al.*, 2017), as well as bio-accumulate in the fatty tissues and bio-magnify within the food chain (Shih *et al.*, 2009). PCDD/Fs are extremely hazardous chemicals, and ingestion, inhalation and dermal contact are the primary pathways for them to get into human bodies (Shih *et al.*, 2009; Chen *et al.*, 2010), thus posing risks to the immune system, interfering with hormones and even leading to cancer (Lin *et al.*, 2010; Chi *et al.*, 2011). PCDD/Fs were detected in the emissions of municipal solid waste incinerators (MSWIs) for the first time (Olie *et al.*, 1977), and since then have become one of the most controversial environmental pollutants. Combustion processes and some industrial activities are the most dominant sources of PCDD/Fs released to the environment, such as power and heating facilities, metal smelting processes and waste incineration (Schuhmacher *et al.*, 2000; Wang *et al.*, 2003; Lin *et al.*, 2007; Hsieh *et al.*, 2009; Chuang *et al.*, 2010, 2011). PCDD/Fs are complex mixture of different congeners, there are 210 possible congeners and 17 of these have been shown to be more toxic, with the 2,3,7,8 positions attached by chlorine atoms, and the toxicities are estimated by the toxic equivalent quantity (TEQ) (Cheruiyot, *et al.*, 2016).

In the atmosphere, after being emitted from combustion facilities, the PCDD/Fs are distributed into both gas and particle phases (Li *et al.*, 2008b; Chen *et al.*, 2011a; Kou *et al.*, 2015). Many studies show that the gas-particle partitioning of PCDD/Fs is highly dependent on their vapor pressures, ambient temperatures and other parameters (Wu *et al.*, 2009a; Wang *et al.*, 2010; Cheruiyot *et al.*, 2015). More fractions of PCDD/Fs are volatilized into gas with an increase in temperature (Fiedler, 1996; Oh *et al.*, 2001). The degradation of PCDD/Fs depends on chemical and photochemical reactions, and their removal mainly relies on the atmospheric deposition (Giorgi, 1988; Chi *et al.*, 2009; Wu *et al.*, 2009a; Huang *et al.*, 2011a; Mi *et al.*, 2012).

This study investigated the PM_{2.5} concentrations, PM_{2.5}/PM₁₀ ratios, PCDD/F concentrations, gas-particle partitioning, and PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content, while the sensitivity analysis of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents was also studied and discussed.

METHODS

Two cities, namely Wuhu (31°33'N, 118°38'E) and Bengbu (32°93'N, 117°34'E) in Anhui province, China, were evaluated in this study. The monthly mean concentrations of both PM_{2.5} and PM₁₀, monthly temperature and precipitation in both cities were obtained from local air quality monitoring stations and the Statistics Yearbook of China.

The total PCDD/F concentration was simulated by regression analysis of the PM₁₀ concentration. Tang *et al.* (2017) reported that there is a high correlation between PM₁₀ values and total PCDD/F mass concentrations. Tang *et al.* (2017) included the following two regression equations:

$$Y_1 = 0.0138x + 0.0472 \quad (1)$$

$$Y_2 = 0.0117x - 0.021 \quad (2)$$

Y_1, Y_2 : total PCDD/F concentration (pg m⁻³);
 x : PM₁₀ concentration in ambient air (μg m⁻³).

The final total PCDD/F concentration was the average of Y_1 and Y_2 .

Gas-Particle Partitioning

The PCDD/F concentrations in the gas and particle phases, respectively, were calculated by using gas-particle partitioning model as Eq. (3) (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 (m³ μg⁻¹);
 TSP: concentration of total suspended particulate matter, which was multiplied by PM₁₀ 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^0 , gives

$$\log K_p = m_r \times \log P_L^0 + b_r \quad (4)$$

P_L^0 : 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 have 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^0 of PCDD/Fs with gas chromatographic retention indexes (GC-RI) on a nonpolar (DB-5) GC-column using p,p'-DDT as a reference standard. The correlation has been re-developed as follows (Hung *et al.*, 2002).

$$\log P_L^0 = \frac{-1.34(RI)}{T} + 1.67 \times 10^{-3}(RI) - \frac{1320}{T} + 8.087 \quad (5)$$

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 PM_{2.5} concentration not only has a significant correlation with air visibility and human health, but can also reflect the PCDD/F concentration of a region. For the period 2015–2017, the monthly average PM_{2.5} concentrations in the ambient air of Wuhu and Bengbu are shown in

Figs. 1(a), 1(b) and 1(c). As for Wuhu, over the three years examined, the lowest PM_{2.5} concentration occurred in 2017, at 27.0–99.0 $\mu\text{g m}^{-3}$, and with an average of 48.9 $\mu\text{g m}^{-3}$; followed by 2016, which was in the range of 25.0–90.0 $\mu\text{g m}^{-3}$ and with an average of 53.0 $\mu\text{g m}^{-3}$; and in 2015, in the range of 32.0–108.0 $\mu\text{g m}^{-3}$, with an average of 57.3 $\mu\text{g m}^{-3}$. Comparing the annual average PM_{2.5} concentrations, we can see that the highest values occurred in 2015, followed by 2016, and the lowest occurred in 2017. The PM_{2.5} annual average concentration, compared to that for 2015 to 2017, was reduced by approximately 14.7%. The tendency of the PM_{2.5} level to decline slowly may be because of social efforts to improve the quality of the environment. As a whole, the PM_{2.5} concentration of the three-year average in Wuhu ranged between 25.0 and 108.0 $\mu\text{g m}^{-3}$, with an average of 53.0 $\mu\text{g m}^{-3}$. It also can be seen that even though the air quality of Wuhu was improved significantly, the PM_{2.5} concentrations were still far above the WHO air quality regulated standard (10 $\mu\text{g m}^{-3}$), so more efforts are needed issue.

With regard to Bengbu (Figs. 1(a), 1(b) and 1(c)), the monthly average PM_{2.5} concentrations in 2017, which was between 34.0 and 98.0 $\mu\text{g m}^{-3}$, with an average of 61.1 $\mu\text{g m}^{-3}$; and those in 2016 ranged from 33.0 to 101.0 $\mu\text{g m}^{-3}$ and averaged 59.8 $\mu\text{g m}^{-3}$; during 2015, the PM_{2.5} concentration ranged between 47.0 and 88.0 $\mu\text{g m}^{-3}$ and averaged 63.2 $\mu\text{g m}^{-3}$. These results reveal that from 2015 to 2016 the annual average PM_{2.5} concentration fell from 63.2 to 59.8 $\mu\text{g m}^{-3}$, falling by approximately 5.4%. However, the highest values increased by 14.8% from 2015 (88.0 $\mu\text{g m}^{-3}$) to 2016 (101.0 $\mu\text{g m}^{-3}$). As a whole, the PM_{2.5} concentration for these three years in Bengbu ranged between 33.0 and 101.0 $\mu\text{g m}^{-3}$, with an average of 61.4 $\mu\text{g m}^{-3}$. This indicates that the PM_{2.5} level in Wuhu was slightly lower than that in Bengbu. This is probably due to the development of industry with poor air pollution control, as well as more pollutants being emitted from mobile and stationary sources.

When regard to the seasonal variations, the four seasons were defined as spring (March, April, May), summer (June, July, August), fall (September, October, November) and winter (January, February and December). For Wuhu, during 2015, the average PM_{2.5} concentrations in spring, summer, fall and winter were 49.7, 34.7, 60.3 and 84.3 $\mu\text{g m}^{-3}$, respectively; and those in 2016 were 52.3, 32.0, 51.0 and 76.7 $\mu\text{g m}^{-3}$, respectively; while those in 2017 were 47.7, 30.3, 46.7 and 71.0 $\mu\text{g m}^{-3}$, respectively. For Bengbu, during 2015, the average PM_{2.5} concentrations in spring, summer, fall and winter were 57.3, 48.0, 69.0 and 78.3 $\mu\text{g m}^{-3}$, respectively; and those in 2016 were 60.0, 37.7, 53.3 and 90.0 $\mu\text{g m}^{-3}$, respectively; while those in 2017 were 60.0, 40.0, 52.3 and 92.0 $\mu\text{g m}^{-3}$, respectively. This indicates that the PM_{2.5} concentrations varied from season to season, the highest values always occurred in winter and the lowest in summer, while the values for spring were very similar with fall and both in the middle levels. As a whole, the three-year average of PM_{2.5} concentration of Wuhu in summer (32.3 $\mu\text{g m}^{-3}$) was 58.2% in magnitude lower than that in winter (77.3 $\mu\text{g m}^{-3}$); and the values of Bengbu in summer (41.9 $\mu\text{g m}^{-3}$) was 51.7% in magnitude lower than that in winter (86.8 $\mu\text{g m}^{-3}$).

During the cold seasons, cities at a higher latitude have a very low ground temperature, so it is easier for high stability vertical atmospheric convection, to occur, which hinders the dispersion of air pollutants. As a result, an accumulation of PM_{2.5} concentrations occurs in regions with human activity. In addition, the parts of north China require more coal and other fossil fuel combustion for heating during winter, and thus have more air pollutant emissions. Therefore, the air current blowing from the northern cities, which have quite high PM_{2.5} concentration levels, may increase the PM_{2.5} concentrations in both Wuhu and Bengbu, cities located in central China. And in warm season the air temperature is higher and the vertical transport of air current is more violent, which can accelerate the dispersion of air PM_{2.5}. Moreover, in summer both rainfall and wind speed are stronger, enhancing the effects of rainfall scavenging and wind blowing in removing the PM_{2.5} from the atmosphere.

PM_{2.5}/PM₁₀ Ratio

The PM_{2.5}/PM₁₀ ratio can reflect the contribution of fine particulate matter to the ambient air pollutants, and thus mirror the status of air pollution. The monthly average of PM_{2.5}/PM₁₀ ratio in the ambient air in Wuhu and Bengbu are presented in Figs. 2(a), 2(b) and 2(c).

The monthly PM_{2.5}/PM₁₀ ratios of Wuhu were in the range of 0.53–0.73, with an average of 0.67 in 2015, of 0.61–0.74 and with an average of 0.66 in 2016; and of 0.42–0.89 and with an average of 0.54 in 2017. As for Bengbu, the annual PM_{2.5}/PM₁₀ ratios ranged between 0.61 and 0.79 and averaged 0.70 in 2015, ranged between 0.57 and 0.79 and averaged 0.65 in 2016, and between 0.43 and 0.72 and averaged 0.59 in 2017, respectively. The PM_{2.5}/PM₁₀ ratio of the three-year range in Wuhu was 0.42–0.89, with an average of 0.63, was 0.43–0.79, and with an average of 0.65 in Bengbu. These results show that the annual average of PM_{2.5}/PM₁₀ ratios in both Wuhu and Bengbu were all lower than those reported in the Beijing-Tianjin-Hebei region (0.83), the Yangtze River Delta region (0.76) and the Pearl River Delta region (0.74) (Chen *et al.*, 2017). This result was also consistent with the conclusion of Zhou *et al.* (2015), which reported there was a strong positive correlation between PM_{2.5} and PM₁₀ mass concentration in the atmosphere. In particular, the trend of a decreasing PM_{2.5}/PM₁₀ ratio was similar to that seen for the PM_{2.5} concentration.

For Wuhu, during 2015, the three highest monthly averages of PM_{2.5}/PM₁₀ ratios were 0.73 in November, 0.72 in December and 0.70 in January; while in 2016, the three highest monthly averages of PM_{2.5}/PM₁₀ ratios were 0.74 in December, 0.72 in July and 0.72 in January; in 2017 the three highest monthly averages of the PM_{2.5}/PM₁₀ ratios were 0.89 in December, 0.67 in November and 0.65 in February. However, in 2015, the three lowest monthly averages of the PM_{2.5}/PM₁₀ concentration ratios were 0.53 in June, 0.64 in July and 0.66 in September; while in 2016, the three lowest monthly averages of the PM_{2.5}/PM₁₀ concentration ratios were 0.61 in April, 0.62 in September and 0.64 in August; in 2017, the three highest monthly averages of the PM_{2.5}/PM₁₀ ratios were 0.42 in April, 0.44 in May and 0.51 in July.

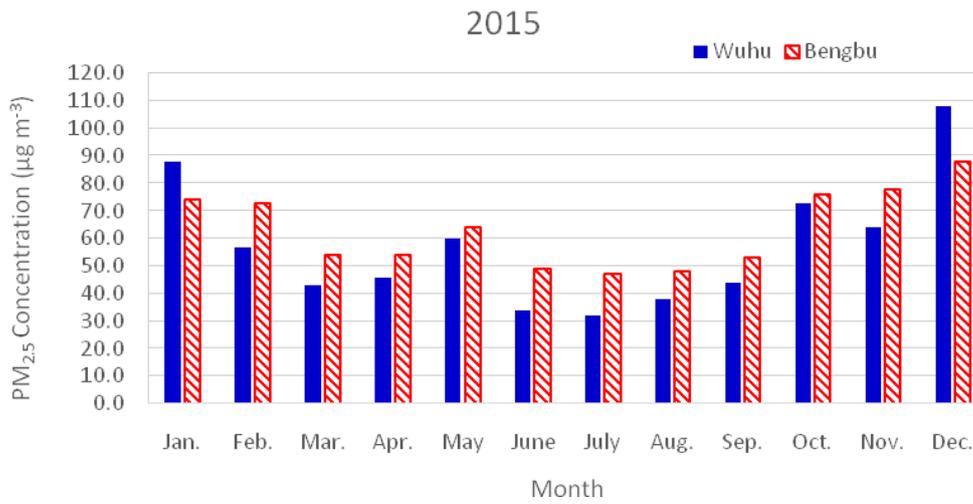


Fig. 1(a). Monthly average atmospheric PM_{2.5} concentration in Wuhu and Bengbu during 2015.

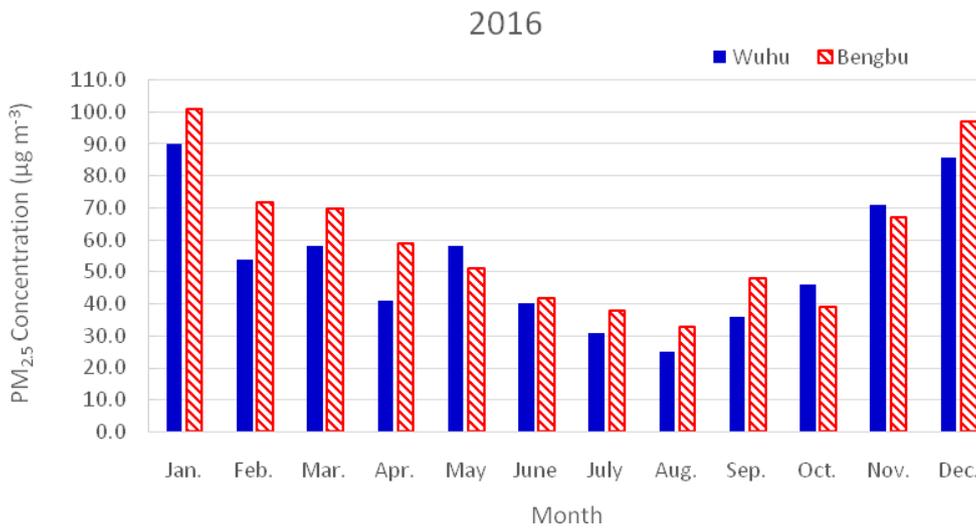


Fig. 1(b). Monthly average atmospheric PM_{2.5} concentration in Wuhu and Bengbu during 2016.

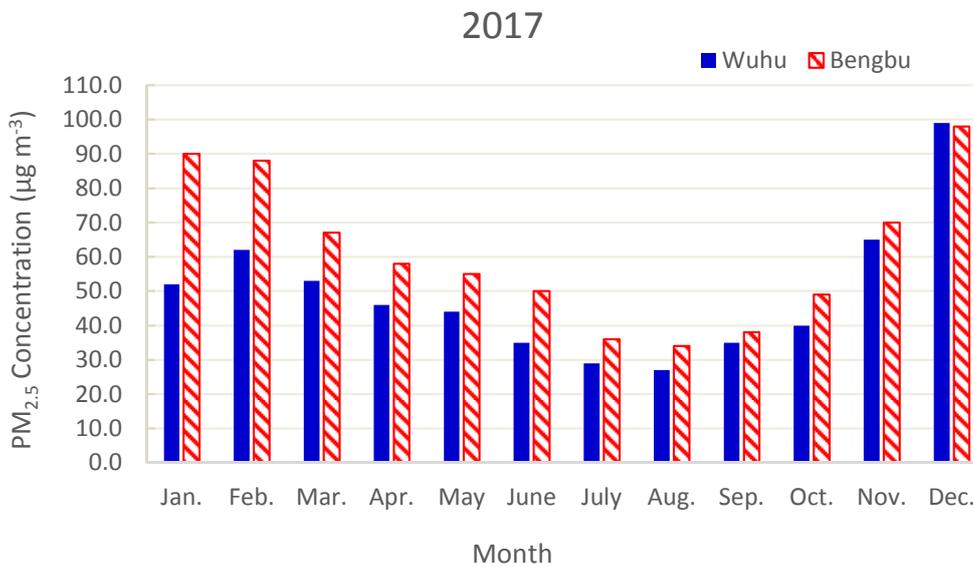


Fig. 1(c). Monthly average atmospheric PM_{2.5} concentration in Wuhu and Bengbu during 2017.

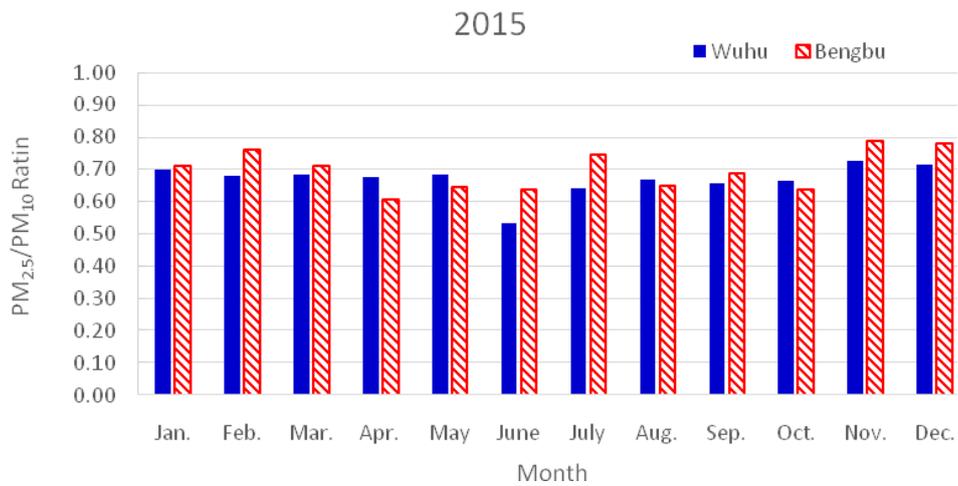


Fig. 2(a). Monthly Average $PM_{2.5}/PM_{10}$ Ratio in Wuhu and Bengbu during 2015.

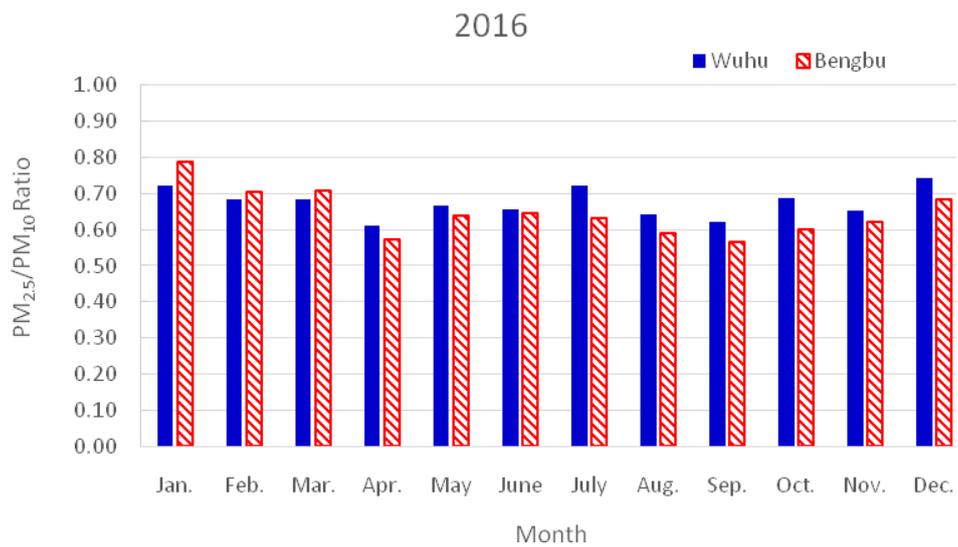


Fig. 2(b). Monthly Average $PM_{2.5}/PM_{10}$ Ratio in Wuhu and Bengbu during 2016.

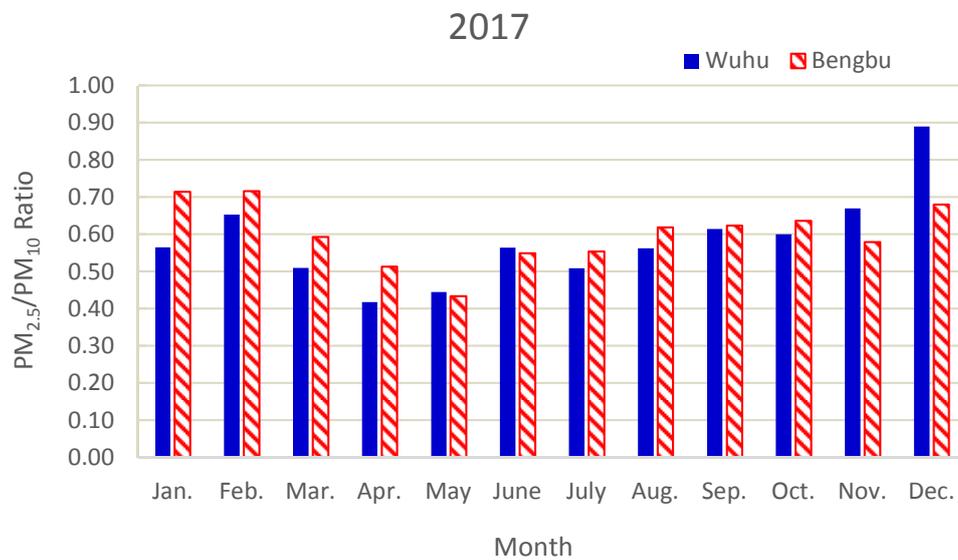


Fig. 2(c). Monthly Average $PM_{2.5}/PM_{10}$ Ratio in Wuhu and Bengbu during 2017.

For Bengbu, during 2015, the three highest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.79 in November, 0.78 in December and 0.76 in February; while in 2016, the three highest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.57 in September, 0.57 in April and 0.59 in August; in 2017 the three highest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.72 in February, 0.71 in January and 0.68 in December. However, in 2015, the three lowest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.61 in April, 0.64 in June and 0.64 in October; while in 2016, the three lowest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.57 in September, 0.57 in April and 0.59 in August; in 2017 the three lowest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.43 in May, 0.51 in April and 0.55 in June, respectively.

In general, it was found that a higher $PM_{2.5}/PM_{10}$ ratio always accompanied a higher $PM_{2.5}$ concentration. This demonstrated that $PM_{2.5}$ is the major portion of atmospheric particles. A previous study (Tang *et al.*, 2017) reported the atmospheric particles were mostly due to the gas-particle transformation, going through the condensation and flocculation processes, and then the accumulation mode of $PM_{2.5}$. Furthermore, a certain fraction of the $PM_{2.5}$ concentration in the ambient air is due to the resuspension of road dust or entrainment of naked lands. A higher $PM_{2.5}/PM_{10}$ ratio signifies more harmful air pollution to human health, and thus should be a case for more concern.

PCDD/F Concentration

Previous studies have shown the strong correlation between PM_{10} and PCDD/F mass concentrations, and Lee *et al.* (2016), Suryani *et al.* (2015) and Huang *et al.* (2011a) reported the correlation coefficients were as high as 0.98, 0.99 and 0.94, respectively. In Tang *et al.* (2017), the monthly concentrations of PCDD/Fs in the ambient air were simulated by PM_{10} using the regression analyses (Wang *et al.*, 2010). Based on the PM_{10} concentrations, the total PCDD/F mass concentrations were calculated using Eq. (1) and Eq. (2).

The results shown that in Wuhu, the total PCDD/F mass concentrations were in the range of 0.65–1.94 $pg\ m^{-3}$ and with an average of 1.09 $pg\ m^{-3}$ in 2015, of 0.51–1.61 $pg\ m^{-3}$ and with an average of 1.01 $pg\ m^{-3}$ in 2016, and of 0.63–1.43 $pg\ m^{-3}$ and with an average of 1.07 $pg\ m^{-3}$ in 2017; while in terms of concentrations of toxicity equivalent quantity (TEQ), those ranged between 0.026 and 0.085 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ in 2015, ranged between 0.021 and 0.070 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ in 2016, and ranged between 0.025 and 0.074 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ in 2017, and the corresponding average values were 0.050, 0.047 and 0.050 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively. The time variation results show that both the average of the total PCDD/F mass and total-PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentrations decreased slowly, and this may be due to a better control of domestic emissions. Comparing with previous studies, the values of the total-PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentrations were all at the same levels of those seen in the Kaohsiung area, ranging between 0.021 and 0.077 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ and with an average of 0.048 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ for 2014, and between 0.021 and 0.072 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ and with an average of 0.044 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ for 2015 in southern

Taiwan (Lee *et al.*, 2016).

In Bengbu, the total PCDD/F mass concentrations were in the range of 0.82–1.53 $pg\ m^{-3}$ with an average of 1.17 $pg\ m^{-3}$, of 0.73–1.82 $pg\ m^{-3}$ and with an average of 1.17 $pg\ m^{-3}$, of 0.71–1.85 $pg\ m^{-3}$ and with an average of 1.31 $pg\ m^{-3}$ in 2015, 2016 and 2017, respectively; while in terms of concentrations of toxicity equivalent quantity ranged between 0.033 and 0.073 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ in 2015, between 0.029 and 0.080 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ in 2016, ranged between 0.029 and 0.085 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ in 2017, and the corresponding average values were 0.054, 0.054 and 0.061 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively. On the whole, both the average of the total PCDD/F mass and total-PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentrations were all at high levels.

With regard to the seasonal variations, for Wuhu, during 2015, the average total-PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentrations in spring, summer, fall and winter were 0.049, 0.030, 0.054 and 0.068 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively; and those in 2016 were 0.054, 0.025, 0.048 and 0.060 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively; while those in 2017 were 0.054, 0.029, 0.045 and 0.056 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively. For Bengbu, during 2015, the total-PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentrations in spring, summer, fall and winter were 0.059, 0.037, 0.060 and 0.059 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively; and those in 2016 were 0.063, 0.032, 0.053 and 0.070 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively; while those in 2017 were 0.079, 0.037, 0.053 and 0.074 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$, respectively. The total PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentration of Wuhu in summer (0.028 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$) was 48.1% in magnitude lower than the average value of other three seasons (0.054 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$); and the values of Bengbu in summer (0.035 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$) was 44.4% of magnitude lower than the average value of other three seasons (0.063 $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$).

These results show that the lowest total PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentration of the two cities were all occurred in summer, which means that the levels of particulate matter had a significant effect on the total PCDD/F concentrations. Therefore, a higher total PCDD/Fs- $WHO_{2005}\text{-}TEQ$ concentration always accompanied with a higher level of particulate matter. As such, controlling the PM emissions from sources will subsequently result in reductions in ambient dioxin levels.

Gas-Particle Partitioning of PCDD/Fs

The gas-particle partitioning of PCDD/F plays an important role in the efficiency of the atmospheric wet and dry deposition (Bidleman and Harner, 2000). Several factors are important here, such as the ambient temperature, PCDD/F concentrations, vapor pressure and the atmospheric particulate concentration (Hoff *et al.*, 1996). The gas-particle partition was calculated by meteorological data using Eq. (3), Eq. (4) and Eq. (5), and the seasonal gas-particle partitioning of total PCDD/Fs- $WHO_{2005}\text{-}TEQ$ in the ambient air of Wuhu and Bengbu during 2015–2017 are shown in Figs. 3(a), 3(b) and 3(c).

In Wuhu, during 2015, the seasonal average temperatures were 17.3, 26.7, 18.6 and 6.5°C in spring, summer, fall and

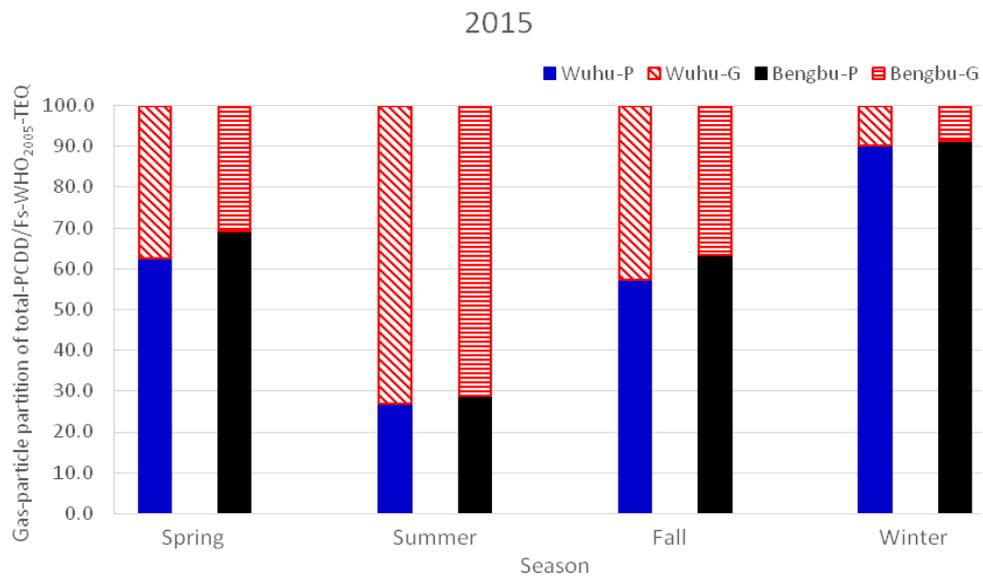


Fig. 3(a). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Wuhu and Bengbu during 2015.

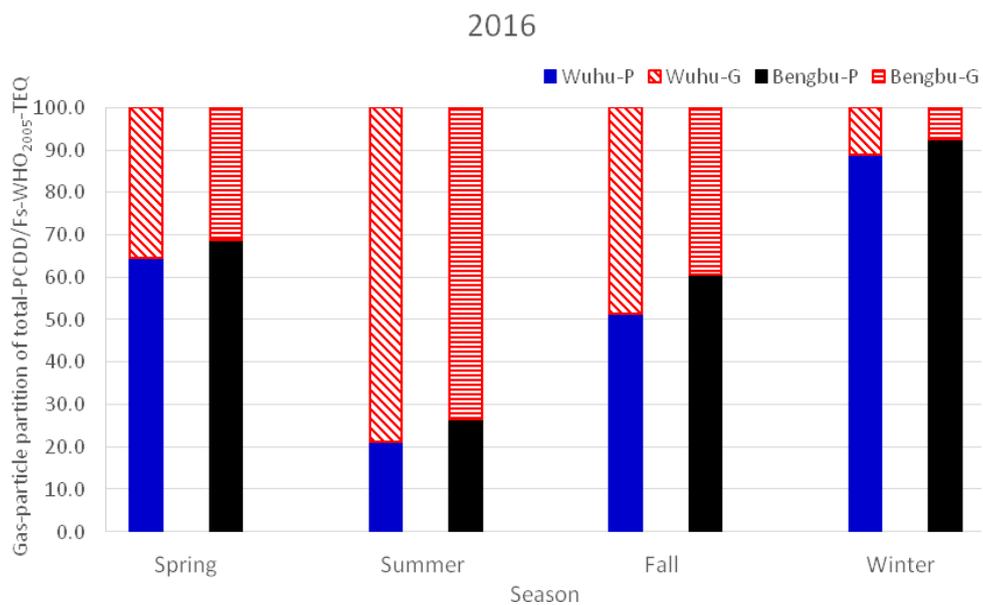


Fig. 3(b). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Wuhu and Bengbu during 2016.

winter, respectively; in 2016, the seasonal average temperatures of spring, summer, fall and winter were 17.1, 28.3, 19.4 and 6.4°C, respectively; while in 2017, the average temperatures of spring, summer, fall and winter were 17.5, 28.6, 17.6 and 6.6°C, respectively. As for the seasonal variations of gas-particle partitioning, the fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in spring, summer, fall and winter were 37.4%, 73.0%, 42.7% and 9.9% in 2015; 35.5%, 78.7%, 48.5% and 11.0% in 2016; and 32.8%, 77.4%, 43.1% and 11.3% in 2017.

In Bengbu, the seasonal average temperatures during 2015 were 15.9, 27.5, 16.9 and 5.0°C in spring, summer, fall and winter, respectively; in 2016, the seasonal average

temperatures in spring, summer, fall and winter were 16.3, 27.3, 16.9 and 4.5°C, respectively; while in 2017, the average temperatures of spring, summer, fall and winter were 16.5, 28.0, 16.2 and 4.9°C, respectively. The fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in spring, summer, fall and winter were 30.7%, 71.5%, 36.5% and 8.7% in 2015; 31.2%, 73.5%, 39.9% and 7.4% in 2016; and 28.6%, 72.7%, 37.3% and 7.2% in 2017. The above results show that the fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ of the both cities were all lower than that in the particle phase in spring, fall and winter, but were significantly higher than that in particle phase in summer (Figs. 3(a), 3(b) and 3(c)).

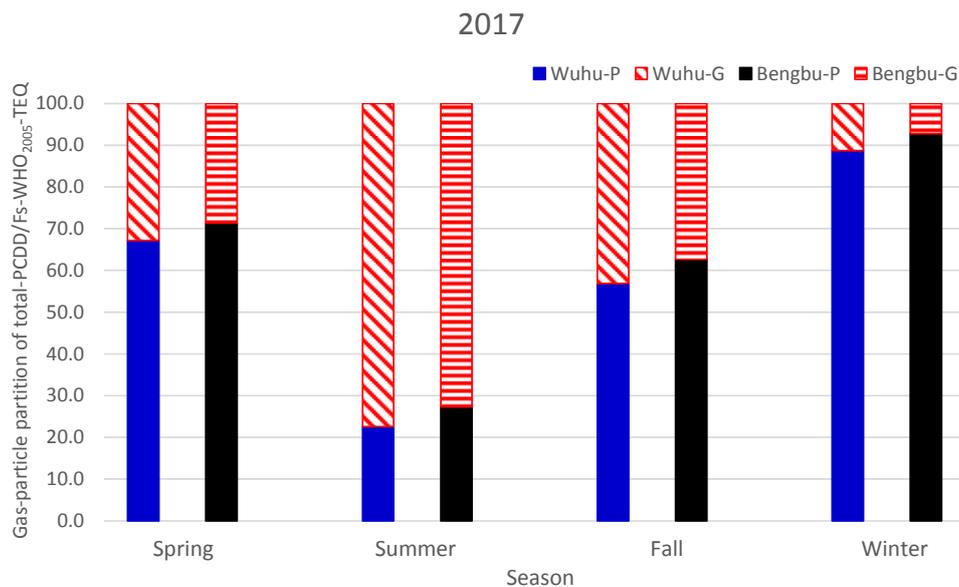


Fig. 3(c). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Wuhu and Bengbu, respectively, during 2017.

As for Wuhu, the three-year average fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ in spring, summer, fall and winter were 35.2%, 44.8%, 55.2% and 88.3%, respectively; the three-year average temperatures in spring, summer, fall and winter were 17.3, 27.8, 18.5 and 6.5°C, respectively. While in Bengbu, the three-year average fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ in spring, summer, fall and winter were 30.2%, 72.6%, 37.9% and 7.8%, respectively; and the three-year average temperatures in spring, summer, fall and winter were 16.2, 27.6, 16.7 and 4.8°C, respectively. The above results indicate that the fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ in summer were highest and those in winter were lowest, while the values of spring and fall were both in the middle levels, but the former were slightly lower than the latter. The fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ had a significant, positive correlation with air temperature. As the temperature increased, a certain fraction of particle phase PCDD/F evaporated into the gas phase, and thus the gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ increased with an increasing temperature. In addition, the average temperature of Wuhu in spring, summer, fall and winter were all slightly higher than those in Bengbu, while the corresponding fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ were slightly higher than those in Bengbu (Figs. 3(a), 3(b) and 3(c)).

With regard to the gas-particle partitioning of PCDD/F-WHO₂₀₀₅-TEQ of each congener, more gas phase PCDD/Fs-WHO₂₀₀₅-TEQ always occurred in summer, particularly for the low molecular weight PCDD/Fs. For instance, the three-year averages for the fractions of 2,3,7,8-TeCDD and 2,3,7,8-TeCDF in the gas phase were 97.2% and 98.1% in summer, but were 39.2% and 49.9% in winter, respectively; those fractions of the middle molecular weight PCDD/Fs for both 1, 2,3,6,7,8-HxCDD and 1, 2,3,6,7,8-HxCDF were 57.6% and 70.9% in summer, but were 1.6% and 3.1% in winter, in the gas phase; while for the high molecular weight

PCDD/Fs of OCDD and OCDF, they were 5.1% and 7.4% in summer and 0.0% and 0.1% in winter, respectively.

In Bengbu the situation was very similar to Wuhu, during 2015, 2016 and 2017, for the average fractions of gas phase PCDD/Fs, the low molecular weight PCDD/Fs of 2,3,7,8-TeCDD and 2,3,7,8-TeCDF were 96.4% and 97.6% in summer, but were 30.1% and 40.2% in winter, respectively; for the middle molecular weight PCDD/Fs, such as 1, 2,3,6,7,8-HxCDD and 1, 2,3,6,7,8-HxCDF, were 51.2% and 65.3% in summer, but were 1.0% and 2.0% in winter, respectively; however, the high molecular weight PCDD/Fs of OCDD and OCDF still had lower fractions in the gas phase, and were 3.8% and 5.6% in summer and both 0.0% in winter, respectively.

The results indicate that lower molecular weight PCDD/F congeners were primarily in the gas phase, while the particle phase was usually associated with higher molecular weight PCDD/F congeners (Wu *et al.*, 2009a; Lin *et al.*, 2010; Huang *et al.*, 2011a; Mi *et al.*, 2012; Suryani R. *et al.*, 2015). The gas phase PCDD/Fs had higher fractions in summer than in winter, similar to in previous studies (Xu *et al.*, 2009; Wang *et al.*, 2010; Huang *et al.*, 2011a; Lee *et al.*, 2016). This may due to lower molecular weight PCDD/Fs usually having higher vapor pressure (Wang *et al.*, 2010; Huang *et al.*, 2011a). As the ambient temperature increased, the fraction of gas phase PCDD/Fs also rose, while when the temperature decreased some of the gas PCDD/Fs were exchanged and transferred into the particle phase. As a result, lower molecular weight PCDD/Fs primarily existed in the gas phase, and the gas phase PCDD/Fs fractions increased with increasing temperature.

PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content

The contents of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ of Wuhu and Bengbu in 2015, 2016 and 2017 are shown in Figs. 4(a), 4(b), and 4(c).

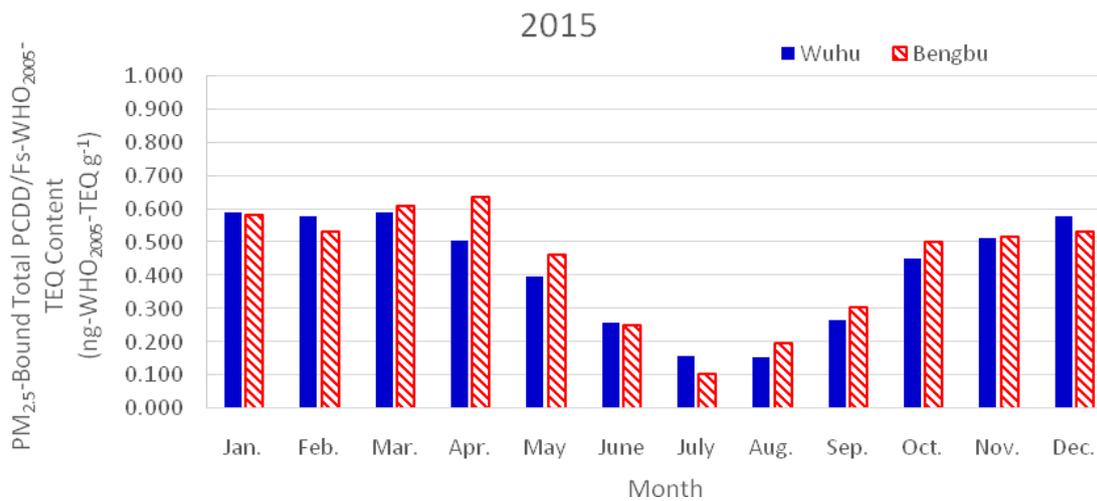


Fig. 4(a). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content of Wuhu and Bengbu during 2015.

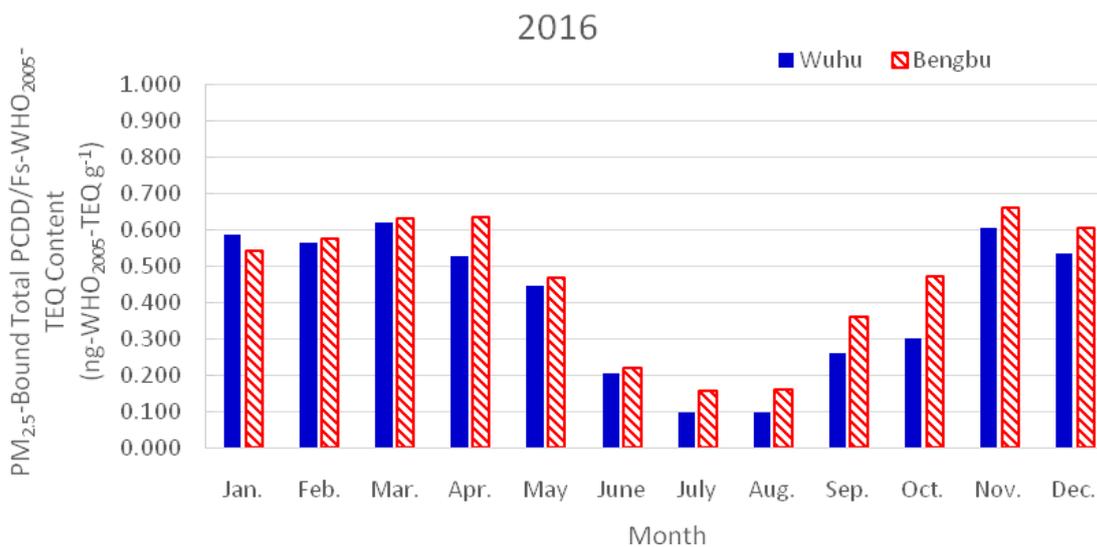


Fig. 4(b). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content of Wuhu and Bengbu during 2016.

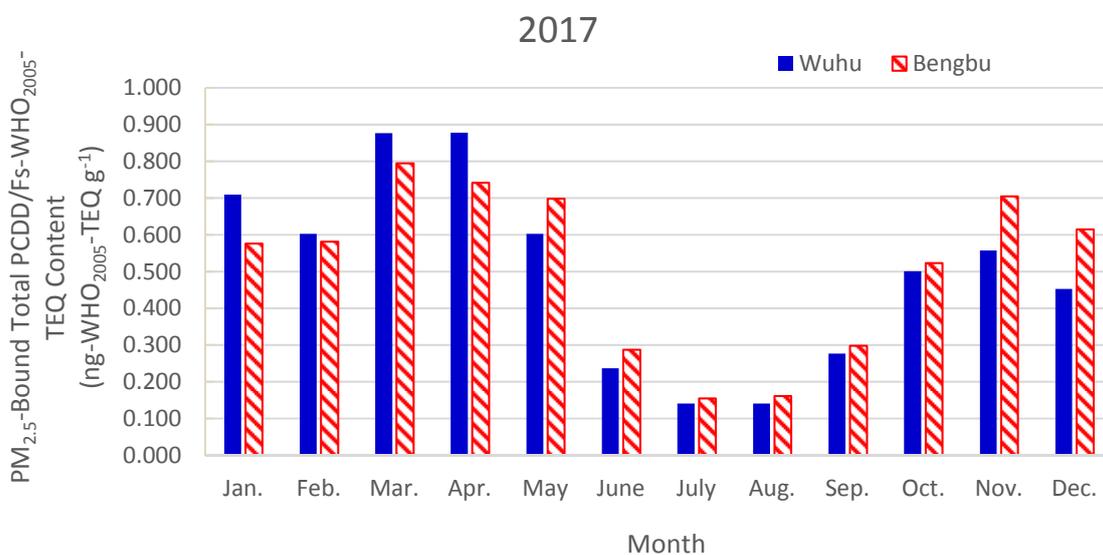


Fig. 4(c). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content of Wuhu and Bengbu during 2017.

During 2015, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were between 0.154 and 0.588 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.419 ng-WHO₂₀₀₅-TEQ g⁻¹ in Wuhu, were between 0.104 and 0.633 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.435 ng-WHO₂₀₀₅-TEQ g⁻¹ in Bengbu. While in 2016, the level of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ in Wuhu ranged between 0.097 and 0.619 ng-WHO₂₀₀₅-TEQ g⁻¹ and averaged 0.404 ng-WHO₂₀₀₅-TEQ g⁻¹, and the level of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ in Bengbu ranged between 0.158 and 0.659 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.457 ng-WHO₂₀₀₅-TEQ g⁻¹. And in 2017, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were between 0.141 and 0.878 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.498 ng-WHO₂₀₀₅-TEQ g⁻¹ in Wuhu, were between 0.155 and 0.795 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.511 ng-WHO₂₀₀₅-TEQ g⁻¹ in Bengbu.

During 2015, the lowest three monthly PM_{2.5} concentrations were for July (32.0 μg m⁻³), June (34.0 μg m⁻³) and August (38.0 μg m⁻³) in Wuhu, and July (47.0 μg m⁻³), August (48.0 μg m⁻³) and June (49.0 μg m⁻³) in Bengbu; however, the lowest three monthly PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents occurred in July, August and June in both Wuhu and Bengbu, and the levels were 0.158, 0.154, 0.259 ng-WHO₂₀₀₅-TEQ g⁻¹ and 0.104, 0.197, 0.248 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. But in 2016, the three months with the lowest PM_{2.5} concentrations were August (25.0 μg m⁻³), July (31.0 μg m⁻³) and September (36.0 μg m⁻³) in Wuhu, accompanied by the lowest three monthly PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, which were 0.097, 0.099 and 0.206 ng-WHO₂₀₀₅-TEQ g⁻¹ in July, August and June, respectively; and the three months with the lowest PM_{2.5} concentrations were August (33.0 μg m⁻³), July (38.0 μg m⁻³) and October (39.0 μg m⁻³) in Bengbu, accompanied by the lowest three monthly PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, which were 0.158, 0.159 and 0.218 ng-WHO₂₀₀₅-TEQ g⁻¹ in July, August and June, respectively. And in 2017, the lowest three monthly PM_{2.5} concentrations were for August (27.0 μg m⁻³), July (29.0 μg m⁻³) and September (35.0 μg m⁻³) and in Wuhu, and August (34.0 μg m⁻³), July (36.0 μg m⁻³) and September (38.0 μg m⁻³) in Bengbu; however, the lowest three monthly PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents occurred in July, August and June in both Wuhu and Bengbu, and the levels were 0.141, 0.141, 0.238 ng-WHO₂₀₀₅-TEQ g⁻¹ and 0.155, 0.161, 0.287 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. It can be seen that in the ambient air the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content was not closely correlated with the PM_{2.5} concentration, and similar results were reported in previous studies (Xing et al., 2017).

As for the seasonal variation, for Wuhu, in 2015, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in spring, summer, fall and winter were 0.496, 0.190, 0.410 and 0.580 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; and in 2016 they were 0.530, 0.134, 0.389 and 0.561 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; while in 2017 they were 0.786, 0.173, 0.445 and 0.588 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. It was found that, in Wuhu, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in summer were roughly only 68.8% in magnitude

lower than the average value of the other three seasons (spring, fall and winter). For Bengbu, in 2015, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in spring, summer, fall and winter were 0.569, 0.183, 0.441 and 0.549 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; while in 2016, they were 0.578, 0.178, 0.496 and 0.575 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; and in 2017, they were 0.745, 0.201, 0.509 and 0.591 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. The results indicated that in Bengbu the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in summer was approximately 66.7% in magnitude lower than the average values of other three seasons (spring, fall and winter). This was because that the ambient temperature in summer was much higher than the average values of other three seasons (spring, fall and winter) and more of the PCDD/Fs bound to the particles were evaporated to the gas phase, and so the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents decreased.

Sensitivity Analysis

Sensitivity analysis can provide a better basis for confirming which environmental parameters are important to the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents. Several parameters, such as the ambient temperature, PM_{2.5} concentration, PM₁₀ concentration and total PCDD/F mass concentration, could affect the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content. In this study, sensitivity analyses were carried out depending on the initial values of ambient air temperature = 22.2°C, PM_{2.5} = 58.0 μg m⁻³, PM₁₀ = 87.0 μg m⁻³ and total-PCDD/F mass concentration = 1.12 pg m⁻³. The parametric sensitivity for the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents of Wuhu is shown in Fig. 5.

where P: initial value of parameters;

ΔP: increase or reduction in parameters;

S: predicted value in each of the parameters at the initial value;

ΔS: response in each of the parameters.

The sensitivity analysis indicated that the total PCDD/F mass concentration was the most sensitive parameter for atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents. When ΔP/P was changed from 0% to +20%, +50%, ΔS/S responded from 0% to +42%, +106%. This may be because PCDD/Fs are the root cause of total PCDD/Fs-WHO₂₀₀₅-TEQ, so the change in PCDD/Fs mass concentration has a significant effect on PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents.

From Fig. 5, it also can be seen that the PM₁₀ concentration is also an important sensitive parameter for the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents. When ΔP/P was changed from 0% to +20%, +50%, ΔS/S responded from 0% to +29%, +72%. There was a strong correlation between PM₁₀ and total PCDD/F mass concentrations, and the change in PM₁₀ concentration has a great impact on the PCDD/F mass concentration, and thus it can affect the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in a significant manner.

The sensitivity analysis also demonstrated that an increase in air temperature has a significant, negative effect on atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ

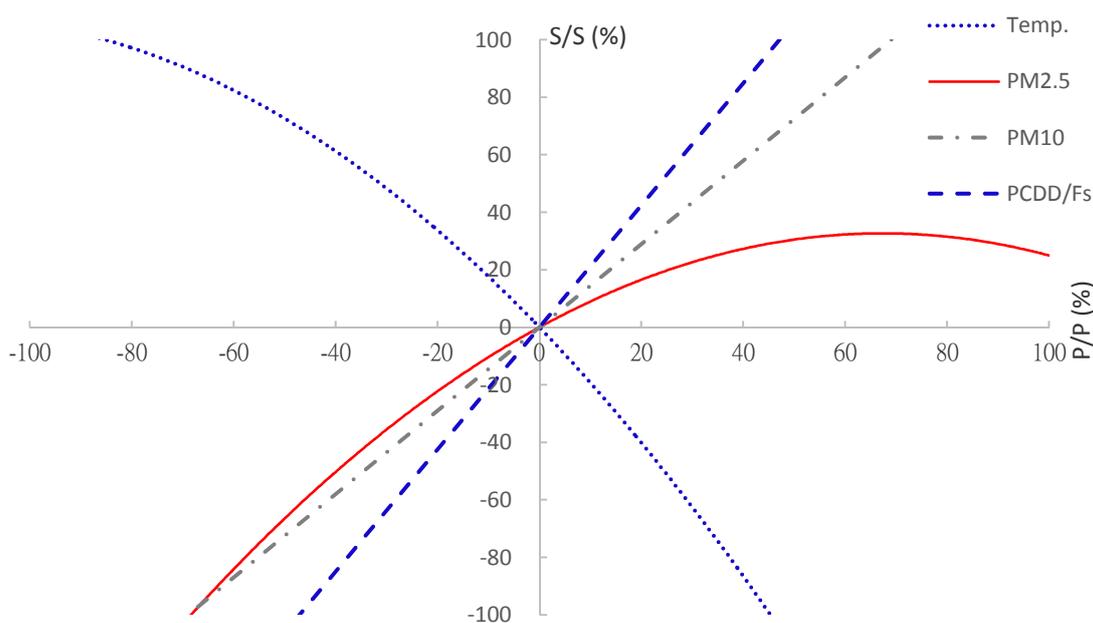


Fig. 5. Sensitivity analysis for the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ content of Wuhu.

contents, when $\Delta P/P$ was changed from 0% to 50%, $\Delta S/S$ responded from 0% to -112%. But a decrease in air temperature has less effect on the atmospheric $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, when $\Delta P/P$ was changed from 0% to -50%, $\Delta S/S$ responded from 0% to +73%. The temperature affects the atmospheric $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents through changing the gas-particle partitioning of PCDD/Fs, the high molecular weight PCDD/Fs have a large contribution to the total PCDD/F mass concentration and primarily existed in the particle phase. When the air temperature was increasing, the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents decreased obviously as more of the particle-bound PCDD/Fs were evaporated to the gas phase; when the temperature was not high, the PCDD/Fs were mostly existed in the particle phase, when the temperature decreased, the rest of the gas phase PCDD/Fs changed into the particle phase, and thus the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents increased. This is consistent with the conclusion that the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in the summer season was only approximately 34.1% in magnitude lower than the average of the other three seasons (spring, fall and winter).

A decrease in the $PM_{2.5}$ concentration had a negative or positive correlation with the atmospheric $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, when $\Delta P/P$ was changed from 0% to -50%, $\Delta S/S$ responded from 0% to -66%. The effect of increasing $PM_{2.5}$ can also be divided into two stages: when $\Delta P/P$ was changed from 0% to +30% and +70%, $\Delta S/S$ responded from 0% to +23% and +33%, respectively, but when $\Delta P/P$ was changed from +70% to +100%, $\Delta S/S$ responded from +33% to +25%. The $PM_{2.5}$ concentration affects the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents mainly in terms of the particle-bound PCDD/Fs. Lower $PM_{2.5}$ concentrations were always accompanied by a better atmospheric stability, which is beneficial to the

dispersion of air pollutants, and thus PCDD/Fs also decreased obviously, and the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ content was the value of the total-PCDD/Fs-WHO₂₀₀₅-TEQ/ $PM_{2.5}$ ratio, and the atmospheric $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents also decreased slowly. However, when the $PM_{2.5}$ concentrations were higher than $86 \mu\text{g m}^{-3}$, because the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were calculated by the ratio of the total PCDD/Fs-TEQ concentration divided by that of $PM_{2.5}$, a higher $PM_{2.5}$ concentration was always accompanied by a low air temperature, and thus the $PM_{2.5}/PM_{10}$ ratio was high, which means the rate of increase of the $PM_{2.5}$ concentrations was greater than that of PM_{10} , and also higher than that of the total PCDD/F mass concentration, and the rate of increase of the $PM_{2.5}$ concentration was greater than that of total PCDD/Fs-WHO₂₀₀₅-TEQ concentration, and therefore the $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents decreased when the $PM_{2.5}$ concentration continuous to rise.

The result of the sensitivity analysis suggested that atmospheric $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were most sensitive to total PCDD/F mass concentration, followed by PM_{10} concentration, and then the air temperature and $PM_{2.5}$ concentration.

CONCLUSION

The results of the current investigation of both $PM_{2.5}$ and PCDD/Fs in Wuhu and Bengbu are summarized as follows:

1. The $PM_{2.5}$ concentration of the focal three years in Wuhu ranged between 25.0 and $108.0 \mu\text{g m}^{-3}$, with an average of $53.0 \mu\text{g m}^{-3}$; for Bengbu, the figures were 33.0 – $101.0 \mu\text{g m}^{-3}$ and with an average of $61.4 \mu\text{g m}^{-3}$. The $PM_{2.5}$ level in Wuhu was lower than that in Bengbu. In general, the time variation of the $PM_{2.5}$ concentration in both Wuhu and Bengbu fell slowly, but was still above the WHO air quality regulated standard ($10 \mu\text{g m}^{-3}$).

2. As for the seasonal variations, the three-year average of PM_{2.5} concentration for Wuhu in summer (32.3 μg m⁻³) was 58.2% in magnitude lower than that in winter (77.3 μg m⁻³); and the value for Bengbu in summer (41.9 μg m⁻³) was 51.7% in magnitude lower than that in winter (86.8 μg m⁻³). The high temperature in summer contributed to more violent vertical air current transport, which can accelerate PM_{2.5} dispersion, while stronger rainfall and wind speed also enhanced the removal of PM_{2.5}. In winter, the low temperature hindered PM_{2.5} dispersion, and polluted air from northern cities caused the PM_{2.5} concentration to increase.
3. The PM_{2.5}/PM₁₀ ratios of the three-year range in Wuhu were in the range of 0.42–0.89, with an average of 0.63, and were 0.43–0.79 with an average of 0.65 in Bengbu. A higher PM_{2.5}/PM₁₀ ratio was always associated with a higher PM_{2.5} concentration. This means that PM_{2.5} is the major portion of atmospheric particles.
4. The total PCDD/Fs-WHO₂₀₀₅-TEQ concentration of Wuhu in summer (0.028 pg-WHO₂₀₀₅-TEQ m⁻³) was 48.1% in magnitude lower than that in other three seasons (0.054 pg-WHO₂₀₀₅-TEQ m⁻³); and the value for Bengbu in summer (0.035 pg-WHO₂₀₀₅-TEQ m⁻³) was 44.4% in magnitude lower than that in other three seasons (0.063 pg-WHO₂₀₀₅-TEQ m⁻³). The values for the total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations of these two cities were all higher than those in previous studies of Taiwan (0.046 pg-WHO₂₀₀₅-TEQ m⁻³).
5. In Wuhu, the three-year average fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in summer (76.4%) was 2.53 times than that in the other seasons (30.2%); while in Bengbu, the fraction in summer (72.6%) was 2.87 times than that in the other seasons (25.3%). As such, the fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ had a significant, positive correlation with air temperature.
6. In Wuhu, the three-year average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in summer (0.166 ng-WHO₂₀₀₅-TEQ g⁻¹) was approximately only 68.8% in magnitude lower than the average value of the other three seasons (0.532 ng-WHO₂₀₀₅-TEQ g⁻¹); while in Bengbu, that of in summer (0.187 ng-WHO₂₀₀₅-TEQ g⁻¹) was approximately 66.7% in magnitude lower than the average value of the other seasons (0.561 ng-WHO₂₀₀₅-TEQ g⁻¹). Due to the higher temperature in summer, more of the particle-bound PCDD/Fs evaporated to the gas phase, and the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents thus decreased.
7. The sensitivity analysis of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ in Wuhu showed that the PCDD/F concentration was the most positively correlated sensitive factor, when ΔP/P was changed from 0% to +20%, +50%, ΔS/S responded from 0% to +42%, +106%. The second positively correlated sensitive factor was the PM₁₀ concentration, when ΔP/P was changed from 0% to +20%, +50%, ΔS/S responded from 0% to +29%, +72%. This was followed by the atmospheric temperature, and its effect was negatively correlated, when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to

–112%; when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to +73%. The last sensitive parameter was the PM_{2.5} concentration, and the impact was divided into two stages: when ΔP/P was changed from 0% to +30%, +70%, ΔS/S responded from 0% to +23%, +33%; but when ΔP/P was changed from +70% to +100%, ΔS/S responded from +33% to +25%.

8. The results of this study can provide useful information in the search for more insights into both atmospheric PM_{2.5} and PCDD/Fs.

REFERENCES

- Alghamdi, M.A., Almazroui, M., Shamy, M., Redal, M.A., Alkhalaf, A.K., Hussein, M.A. and Khoder, M.I. (2015). Characterization and elemental composition of atmospheric aerosol loads during springtime dust storm in western Saudi Arabia. *Aerosol Air Qual. Res.* 15: 440–453.
- Bidleman, T.F. and Harner, T. (2000). Sorption to aerosols. In *Handbook of property estimation methods for chemicals: Environmental and health sciences*, Boethling, R.S. and Mackay, D. (Eds.), Lewis Publishers. New York, pp. 233–260.
- Bilos, C., Colombo, J.C., Skorupka, C.N. and Rodriguez Presa, C.N. (2001). Sources, distribution and variability of airborne trace metals in Plata city area, Argentina. *Environ. Pollut.* 111: 149–158.
- Chao, M.R., Hu, C.W., Chen, Y.L., Chang-Chien, G.P., Lee, W.J., Chang, L.W., Lee, W.S. and Wu, K.Y. (2004). Approaching gas-particle partitioning equilibrium of atmospheric PCDD/Fs with increasing distance from an incinerator: Measurements and observations on modeling. *Atmos. Environ.* 38: 1501–1510.
- Chen, J., Qiu, S., Shang, J., Wilfrid, O.M., Liu, X., Tian, H. and Boman, J. (2014). Impact of relative humidity and water soluble constituents of PM_{2.5} on visibility impairment in Beijing, China. *Aerosol Air Qual. Res.* 14: 260–268.
- Chen, Y.C., Tsai, P.J., Wang, L.C., Shih, M. and Lee, W.J. (2010). An integrated approach for identification of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) pollutant sources based on human blood contents. *Environ. Sci. Pollut. Res. Int.* 17: 759–769.
- Cheruiyot, N.K., Lee, W.J., Mwangi, J.K., Wang, L.C., Lin, N.H., Lin, Y.C., Cao, J., Zhang, R. and Chang-Chien, G.P. (2015). An overview: Polycyclic aromatic hydrocarbon emissions from the stationary and mobile sources and in the ambient air. *Aerosol Air Qual. Res.* 15: 2730–2762.
- Cheruiyot, N.K., Lee, W.J., Yan, P., Mwangi, J.K., Wang, L.C., Gao, X., Lin, N.H., Chang-Chien G.P., (2016). An overview of PCDD/F inventories and emission factors from stationary and mobile sources: What we know and what is missing. *Aerosol Air Qual. Res.* 16: 2965–2988.
- Chi, K.H., Hsu, S.C., Lin, C.Y., Kao, S.J. and Lee, T.Y. (2011). Deposition fluxes of PCDD/Fs in a reservoir system in northern Taiwan. *Chemosphere* 83: 745–752.
- Chi, K.H., Liu, K.T., Chang, S.H. and Chang, M.B. (2009). Atmospheric deposition of PCDD/Fs measured via automated and traditional samplers in Northern Taiwan.

- Chemosphere* 77: 1184–1190.
- Chow, J.C., Yang, X., Wang, X., Kohl, S.D., Hurbain, P.R., Chen, L.A. and Watson, J.G. (2015). Characterization of ambient PM₁₀ bioaerosols in a California agricultural town. *Aerosol Air Qual. Res.* 15: 1433–1447.
- Chuang, S.C., Chen, S.J., Huang, K.L., Wu, E.M.Y., Chang-Chien, G.P. and Wang, L.C. (2010). Gas/particle partitioning of dioxins in exhaust gases from automobiles. *Aerosol Air Qual. Res.* 10: 489–496.
- Ghosh, S., Gupta, T., Rastogi, N., Gaur, A., Misra, A., Tripathi, S.N., Paul, D., Tare, V., Prakash, O. and Bhattu, D. (2014). Chemical characterization of summertime dust events at Kanpur: Insight into the sources and level of mixing with anthropogenic emissions. *Aerosol Air Qual. Res.* 14: 879–891.
- Giorgi, F. (1988). Dry deposition velocities of atmospheric aerosols as inferred by applying a particle dry deposition parameterization to a general circulation model. *Tellus Ser. B* 40: 23–41.
- Hoff, R., Strachan, W., Sweet, C., Chan, C., Shackleton, M., Bidleman, T., Brice, K., Burniston, D., Cussion, S. and Gatz, D. (1996). Atmospheric deposition of toxic chemicals to the Great Lakes: A review of data through 1994. *Atmos. Environ.* 30: 3505–3527.
- Hsieh, L.T., Wang, Y.F., Kuo, G.H., Wang, L.C. and Chang-Chien, G.P. (2009). Cluster analysis for polychlorinated dibenzo-*p*-dioxins and dibenzofurans concentrations in southern Taiwan. *J. Air Waste Manage. Assoc.* 59: 1474–1480.
- Huang, C.J., Chen, K.S., Lai, Y.C., Wang, L.C. and Chang-Chien, G.P. (2011). Characterization of atmospheric dry deposition of polychlorinated dibenzo-*p*-dioxins/dibenzofuran in a rural area of Taiwan. *Aerosol Air Qual. Res.* 11: 448–459.
- Huang, R.J., Zhang, Y., Bozzetti, C., Ho, K.F., Cao, J.J., Han, Y., Daellenbach, K.R., Slowik, J.G., Platt, S.M. and Canonaco, F. (2014). High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* 514: 218–222.
- Hung, H., Blanchard, P., Poole, G., Thibert, B. and Chiu, C.H. (2002). Measurement of particle-bound polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in Arctic air at Alert, Nunavut, Canada. *Atmos. Environ.* 36: 1041–1050.
- Ito, K., Christensen, W.F., Eatough, D.J., Henry, R.C., Kim, E., Landen, F., Lall, R., Larson, T.V., Neas, L., Hopke, P.H. and Thurston, G.D. (2006). PM source apportionment and health effects: 2. An investigation of intermethod variability in associations between source-apportioned fine particle mass and daily mortality in Washington, DC. *J. Exposure Sci. Environ. Epidemiol.* 16: 300–310.
- Kong, S., Ji, Y., Lu, B., Zhao, X., Han, B. and Bai, Z. (2014). Similarities and differences in PM_{2.5}, PM₁₀ and TSP chemical profiles of fugitive dust sources in a coastal oilfield city in China. *Aerosol Air Qual. Res.* 14: 2017–2028.
- Lee, K.L., Lee, W.J., Mwangi, J.K., Wang, L.C., Gao, X. and Chang-Chien, G.P. (2016). Atmospheric PM_{2.5} and depositions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in Kaohsiung area, Southern Taiwan. *Aerosol Air Qual. Res.* 16: 1775–1791.
- Lin, L.F., Lee, W.J., Li, H.W., Wang, M.S. and Chang-Chien, G.P. (2007). Characterization and inventory of PCDD/F emissions from coal-fired power plants and other sources in Taiwan. *Chemosphere* 68: 1642–1649.
- Lin, L.F., Shih, S.I., Su, J.W., Shih, M., Lin, K.C., Wang, L.C. and Chang-Chien, G.P. (2010). Dry and wet deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans on the drinking water treatment plant. *Aerosol Air Qual. Res.* 10: 231–244.
- Liu, Q., Baumgartner, J., Zhang, Y. and Schauer, J.J. (2016). Source apportionment of Beijing air pollution during a severe winter haze event and associated proinflammatory responses in lung epithelial cells. *Atmos. Environ.* 126: 28–35.
- Lu, H.Y., Lin, S.L., Mwangi, J.K., Wang, L.C. and Lin, H.Y. (2016). Characteristics and source apportionment of atmospheric PM_{2.5} at a coastal city in Southern Taiwan. *Aerosol Air Qual. Res.* 16: 1022–1034.
- Mi, H.H., Wu, Z.S., Lin, L.F., Lai, Y.C., Lee, Y.Y., Wang, L.C. and Chang-Chien, G.P. (2012). Atmospheric dry deposition of polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in southern Taiwan. *Aerosol Air Qual. Res.* 12: 1016–1029.
- Oh, J.E., Choi, J.S. and Chang, Y.S. (2001). Gas/particle partitioning of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in atmosphere; evaluation of predicting models. *Atmos. Environ.* 35: 4125–4134.
- Olie, K., Vermeulen, P.L. and Hutzinger, O. (1977). Chlorodibenzo-*p*-dioxins and chlorodibenzofurans are trace components of fly ash and flue gas of some municipal incinerators in the Netherlands. *Chemosphere* 6: 455–459.
- Pankow, J.F. (1987). Review and comparative analysis of the theories on partitioning between the gas and aerosol particulate phases in the atmosphere. *Atmos. Environ.* 21: 2275–2283.
- Pankow, J.F. and Bidleman, T.F. (1991). Effects of temperature, TSP and percent non-exchangeable material in determining the gas-particle partitioning of organic compounds. *Atmos. Environ.* 25: 2241–2249.
- Pankow, J.F. and Bidleman, T.F. (1992). Interdependence of the slopes and intercepts from log-log correlations of measured gas-particle partitioning and vapor pressure—I. theory and analysis of available data. *Atmos. Environ.* 26: 1071–1080.
- Redfern, F.M., Lee, W.J., Yan, P., Mwangi, J.K., Wang, L.C. and Shih, C.H. (2017). Overview and perspectives on emissions of polybrominated diphenyl ethers on a global basis: Evaporative and fugitive releases from commercial PBDE mixtures and emissions from combustion sources. *Aerosol Air Qual. Res.* 17: 1117–1131.
- Schuhmacher, M., Granero, S., Rivera, J., Müller, L., Llobet, J. and Domingo, J. (2000). Atmospheric deposition of PCDD/Fs near an old municipal solid waste incinerator: Levels in soil and vegetation. *Chemosphere* 40: 593–600.
- Schwartz, J., Dochery, D.W. and Neas, L.M. (1996). Is

- daily mortality associated specifically with fine particles? *J. Air Waste Manage. Assoc.* 46: 927–939.
- Shih, S.I., Wang, I.C., Wu, K.Y., Li, H.W., Wang, L.C. and Chang-Chien, G.P. (2009). Uptake of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in laying ducks. *J. Environ. Sci. Health Part A* 44: 799–807.
- Suryani R., C., Lee, W.J., Endah Mutiara M.P., Mwangi, J.K., Wang, L.C., Lin, N.H. and Chang-Chien, G.P. (2015). Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans at coastal and high mountain areas in Taiwan. *Aerosol Air Qual. Res.* 15: 1390–1411.
- Tang, H.Y., Cui, K.P., Xing, J., Zhu, J.N., Lee, W.J., John, K.M. and Lee, Y.C. (2017). Part I: PM_{2.5} and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the ambient air of southern China. *Aerosol Air Qual. Res.* 17: 1550–1569.
- Wang, L.C., Lee, W.J., Lee, W.S., Chang-Chien, G.P. and Tsai, P.J. (2003). Characterizing the emissions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans from crematories and their impacts to the surrounding environment. *Environ. Sci. Technol.* 37: 62–67.
- Wang, Q., Jin, Y., Li, X., Chen, J., Lu, S., Chen, T., Yan, J., Zhou, M. and Wang, H. (2014). PCDD/F emissions from hazardous waste incinerators in China. *Aerosol Air Qual. Res.* 14: 1152–1159.
- Wang, Y.F., Hou, H.C., Li, H.W., Lin, L.F., Wang, L.C. Chang-Chien, G.P. and You, Y.S. (2010). Dry and wet depositions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the atmosphere in Taiwan. *Aerosol Air Qual. Res.* 10: 378–390.
- Wu, Y.L., Lin, L.F., Hsieh, L.T., Wang, L.C. and Chang-Chien, G.P. (2009a). Atmospheric dry deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the vicinity of municipal solid waste incinerators. *J. Hazard. Mater.* 162: 521–529.
- Wu, Y.L., Lin, L.F., Shih, S.I., Yu, K.M., Hsieh, L.T., Wang, L.C. and Chang-Chien, G.P. (2009). Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans on the soils in the vicinity of municipal solid waste incinerators. *J. Environ. Sci. Health Part A* 44: 1327–1334.
- Xing, J., Cui, K.P., Tang, H.Y., Lee, W.J., Wang, L.C., Zhu, J.N. and Huang, Q.L. (2017). Part II: PM_{2.5} and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the ambient air of northern China. *Aerosol Air Qual. Res.* 17: 2010–2016.
- Xu, M.X., Yan, J.H., Lu, S.Y., Li, X.D., Chen, T., Ni, M.J., Dai, H.F., Wang, F. and Cen, K.F. (2009). Gas/particle partitioning of atmospheric PCDD/Fs in a satellite town in Eastern China. *Chemosphere* 76: 1540–1549.
- Yamasaki, H., Kuwata, K. and Miyamoto, H. (1982). Effects of ambient temperature on aspects of airborne polycyclic aromatic hydrocarbons. *Environ. Sci. Technol.* 16: 189–194.
- Zhu, J.N., Tang, H.Y., Xing, J., Lee, W.J., Yan, P. and Cui, K.P. (2017). Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in two cities of southern China. *Aerosol Air Qual. Res.* 17: 1798–1810.

Received for review, November 16, 2017

Revised, January 3, 2018

Accepted, January 8, 2018