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Sensitivity Analyses for Atmospheric Scavenging Ratio of Total PCDD/Fs-TEQ Wet Deposition: Case of Wuhu City, China

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ABSTRACT

This study investigated the monthly average dry, wet and total deposition fluxes in total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu and Bengbu, respectively. In addition, sensitivity analyses for both scavenging ratio and wet deposition of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu were also conducted. In 2015, the annual dry deposition fluxes, wet deposition fluxes, and total deposition fluxes were 6407, 1538, and 7945 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in Wuhu, and were 7101, 1525, and 8626 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in Bengbu. The annual scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ were 30144 and 31267 in Wuhu and Bengbu, respectively. As to the sensitivity analysis for the scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ, the most positive sensitivity parameters were PCDD/Fs concentration and PM_{10} concentration; the second positively correlated sensitivity parameter was $PM_{2.5}$ concentration, then, followed by atmospheric temperature, which was negatively correlated; the last negative correlated sensitivity factor was the rainfall. In terms of the sensitivity analysis for wet deposition of total-PCDD/Fs-WHO_{2005}-TEQ, the most positive sensitivity parameter was air temperature; the second positive or negative correlated sensitivity factor was atmospheric PCDD/Fs mass concentration, followed by $PM_{2.5}$ and PM_{10} concentration, which were positively correlated; the last positive correlated sensitivity factor was the rainfall. This study led to greater insight into the parameters affecting the atmospheric wet deposition of PCDD/Fs, which will benefit development of appropriate control strategies for PCDD/Fs and provided useful information for the scientific community.

Keywords: Dry deposition; Wet deposition; Scavenging ratio; Sensitivity analyses; PCDD/Fs; Wuhu; Bengbu.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and other POPs are persistent, toxicological and bioaccumulative contaminants (Oh *et al*., 2001; Huang *et al*., 2014; Cheruiyot *et al.*, 2015, 2016; Redfern *et al*., 2017). Since PCDD/Fs are mostly emitted from anthropogenic activities into the environment (Moon *et al*., 2005; Kulkarni *et al*., 2008; Wang *et al*., 2010; Huang *et al*., 2011), they can be distributed between the gas and particulate phases and transport over long distances in the atmosphere, and then can enter terrestrial and aquatic environments through both dry and wet deposition (Jurado *et al*., 2004; Lohmann *et al*., 2007; Oh *et al*., 2011), which are the fundamental pathways by which to remove PCDD/Fs and other POPs from air (Lohmann and Jones, 1998; Zhu *et al*., 2017). Therefore, the role of PCDD/Fs as environmental pollutants has received much public attention and has become one of the most controversial issues over the last decade (Huang *et al*., 2011; Suryani *et al*., 2015). This study mainly concentrates on deposition processes, including dry deposition and wet deposition, which are effective and important mechanisms for removal of atmospheric POPs (Cheruiyot *et al.*, 2015, 2016; Redfern *et al*., 2017).

Dry deposition is a combination used for the removal of the gas-phase and fine particle-phase, as well as a large fraction of the coarse particle-phase from the atmosphere during non-precipitation days (Giorgi *et al.*, 1988; Moon *et al.*, 2005). The mechanisms of dry deposition include turbulent diffusion, sedimentation, Brownain motion, interception, inertial forces, thermophoresis, electrical migration and diffusiophoresis. According to dry deposition process, while gas phase deposition absorption at the air-liquid or air-solid interfaces, thus dry deposition depends significantly on the gas-particle partition and gravitational settling (Lohmann *et*

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al., 1999; Chang *et al.*, 2004; Oh *et al.*, 2005). The factors including air temperature, wind, the concentration of PCDD/Fs as well as the characteristics particle phase (Hoff *et al.*, 1996; Oh *et al.*, 2002; Moon *et al*., 2005; Giorgi *et al.*, 2008; Wang *et al.*, 2010) will govern the dry deposition process of PCDD/Fs (Lee *et al.*, 2016).

Wet deposition is also an efficient mechanism of removing both particle and gaseous phase PCDD/Fs from atmosphere, especially the fine particle phase (Moon *et al.*, 2005; Melymuk *et al.*, 2011). The wet deposition process, vaporous PCDD/Fs dissolve into rain and suspended particulates removed in atmosphere via precipitation, which in form of rainfall, snow or cloud droplets (Lohmann and Jones, 1998). Furthermore, higher chlorinated homologues of POPs in the environmental sinks can be majorly attributed to wet deposition (Shih *et al.*, 2006; Lin *et al.*, 2010a; Wang *et al.*, 2010), and it is more relevant in the period of heavy rainy days and seasons with higher precipitation rates.

To evaluate the wet deposition of PCDD/Fs, the total-PCDD/Fs-WHO_{2005}-TEQ concentration in the rain and its scavenging ratio should be addressed. Scavenging ratio is the combined concentration of gas and particulates dissolving into raindrops divided by those in the atmosphere during precipitation events (Huang *et al.*, 2011b; Tseng *et al.*, 2014). As scavenging occurs, parts of the vaporous phase dissolve into raindrops, where the particulate phase can be obtained through precipitation, and the total-PCDD/Fs-WHO_{2005}-TEQ concentration in the rain varies with rainfall intensity and the level of PCDD/Fs, especially in the particle phase. On the other hand, the total wet deposition flux of PCDD/Fs largely depends on the intensity and frequency of rainfall (Shih *et al.*, 2006; Wang *et al.*, 2010), the PCDD/F concentration, and the gas-particle partition. Furthermore, previous studies have indicated that precipitation scavenging can account for the vast majority of wet deposition of hydrophobic organic chemicals, and it is more significant than dry deposition during days with precipitation (Tseng *et al.*, 2014).

China has a large territory with clear monsoon and continental climates. About 98% of the land area stretches between a latitude of 20°N and 50°N, from the subtropical zones in the south to the temperate zones (including warm and cool temperatures) in the north (Zhao *et al.*, 2015). Some factors affecting the climate of China include location, topography, seasonal winds, and human activity. The air temperature is lower in the winter and in the cold north due to the fact that the strength of the sun light is different at all latitude zones. On the other hand, the rainfall decreases from the southeast coast to the northwest inlands. Therefore, the main features of China's climate include being cold and dry in the winter and warm and humid in the summer. The above climate characteristics of China are a result of variations in atmospheric stability in the north and south parts of the country.

The objectives of this study include an investigation of the monthly, seasonal, and annual variations in dry, wet, and total deposition fluxes of atmospheric PCDD/Fs in two typical cities, Wuhu and Bengbu, located in the mid-latitude region of China. In addition, the monthly average total-PCDD/Fs-WHO_{2005}-TEQ concentration in the rain and the wet deposition scavenging ratio in these two cities are also discussed. Furthermore, the effects of various parameters on the scavenging ratio and wet deposition of total PCDD/Fs-WHO_{2005}-TEQ are evaluated by using a sensitivity analysis.

METHODS

Two cities, Wuhu (31°33′N, 118°38′E) and Bengbu (32°93′N, 117°34′E) in Anhui province, China, were selected and evaluated in this study. The monthly average of $PM_{2.5}$ and PM_{10} concentrations, and related meteorological information including monthly temperature and precipitation from January 2015 to December 2015 in Wuhu and Bengbu were obtained from local air quality monitoring stations and the Statistic Yearbook of China.

Atmospheric Dry Deposition of PCDD/Fs

The atmospheric dry deposition flux of PCDD/Fs is a combination of both gas- and particle-phase fluxes, which are given by:

$$F_{d,T} = F_{d,g} + F_{d,p}$$
$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \quad (1)$$

$F_{d,T}$: the total PCDD/F deposition flux contributed by the summation of both gas- and particle-phase fluxes;
$F_{d,g}$: the PCDD/F deposition flux contributed by the gas phase;
$F_{d,p}$: the PCDD/F deposition flux contributed by the particle phase;
C_T: the measured concentration of total PCDD/Fs in the ambient air;
$V_{d,T}$: the dry deposition velocity of total PCDD/Fs;
C_g: the calculated concentration of PCDD/Fs in the gas phase;
$V_{d,g}$: the dry deposition velocity of gas-phase PCDD/Fs;
C_p: the calculated concentration of PCDD/Fs in the particle phase;
$V_{d,p}$: the dry deposition velocity of particle -phase PCDD/Fs.

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

Scavenging Ratios

For a slightly soluble trace organic compound, such as PCDD/Fs and other semi-volatile organic compounds, it is commonly believed that equilibrium partitioning occurs

between the compound in the gas phase and that in a falling rain drop (Ligocki *et al.*, 1985a, b; Cheruiyot *et al.*, 2015, 2016; Redfern *et al*., 2017). The scavenging ratio is defined as the concentration of the pollutant in the raindrop divided by the concentration of the same pollutant in the surrounding air during precipitation. The gas scavenging ratio, S_g, can be estimated by:

$$S_g = \frac{RT}{H} \quad (2)$$

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

On the other hand, particle scavenging largely depends on meteorological factors and particle characteristics. The gas scavenging ratio is the ratio of the dissolved phase concentration in the raindrop divided by the gas phase concentration in the air, S_g, and can be calculated by:

$$S_g = \frac{C_{rain,dis}}{C_g} \quad (3)$$

S_g: the gas scavenging ratio of PCDD/Fs (dimensionless);
$C_{rain,dis}$: the dissolved-phase concentration of PCDD/Fs in the raindrop;
C_g: the concentration of PCDD/Fs in the gas phase.

The particle scavenging ratio is the ratio of the particle phase concentration in a raindrop divided by the particle phase concentration in the air, S_p, which can be calculated as:

$$S_p = \frac{C_{rain,particle}}{Cp} \quad (4)$$

S_p: the particle scavenging ratio of PCDD/Fs (dimensionless);
$C_{rain,particle}$: the particle-phase concentration of PCDD/Fs in the raindrop;
C_p: the concentration of PCDD/Fs in the particle phase.

The total scavenging of precipitation is the sum of gas and particle scavenging, S_{tot}, which can be calculated as:

$$S_{tot} = S_g\,(1-\phi) + S_p \times \phi \quad (5)$$

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

Because of a lack of measured data for the particle scavenging ratios of PCDD/Fs, the S_p (S_p is 42,000) and the values of OCDD and OCDF as measured by Eitzer and Hites (1989) were averaged and used here.

Wet Deposition

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

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

$$F_{w,T} = F_{w,dis} + F_{w,p} \quad (6)$$

$$F_{w,dis} = C_{rain,dis} + Rainfall \quad (7)$$

$$F_{w,p} = C_{rain,particle} + Rainfall \quad (8)$$

$F_{w,T}$: the wet deposition flux of PCDD/Fs from both vapor dissolution into rain and removal of suspended particulates by precipitation;
$F_{w,dis}$: the wet deposition flux contributed by vapor dissolution into rain;
$F_{w,p}$: the wet deposition flux contributed by removal of suspended particulates by precipitation; *Rainfall*: monthly rainfall (m).

RESULTS AND DISCUSSION

Dry Deposition

During non-precipitation days, the deposition of PCDD/Fs in the atmosphere depend on dry deposition, which is a combination of small fractions bound to fine particles and the gaseous phase. It is also primarily the removal of coarse particle-bound PCDD/Fs (Giorgi *et al.*, 1988; Moon *et al.*, 2005). Based on the deposition Eq. (1), the monthly mean dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu and Bengbu are presented in Fig. 1.

During 2015, the monthly average dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu ranged from 287 to 922 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ was 6407 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was similar to the result found in Nanjing (6480 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), and was approximately only 21.5% and 58.0% order of magnitude lower than those in Harbin (8240 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Shijiazhuang (15400 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhu *et al.*, 2017b), respectively; but were 1.6 and 2.6 times higher than those in Yunlin (4568 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Guangzhou (2470 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), respectively (Chen *et al.*, 2017; Zhu *et al.*, 2017a). These levels were much higher than those in the coastal and high mountain areas in Taiwan, which was 9.4 times higher than those in Hengchun (685 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and 47.6 times higher than those in Lulin (135 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Suryani *et al.*, 2015).

As for Bengbu, the monthly average dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 360 and 793 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ was 7101 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was approximately only 13.8% and 53.9% of an order of magnitude lower than those in Harbin (8240 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and

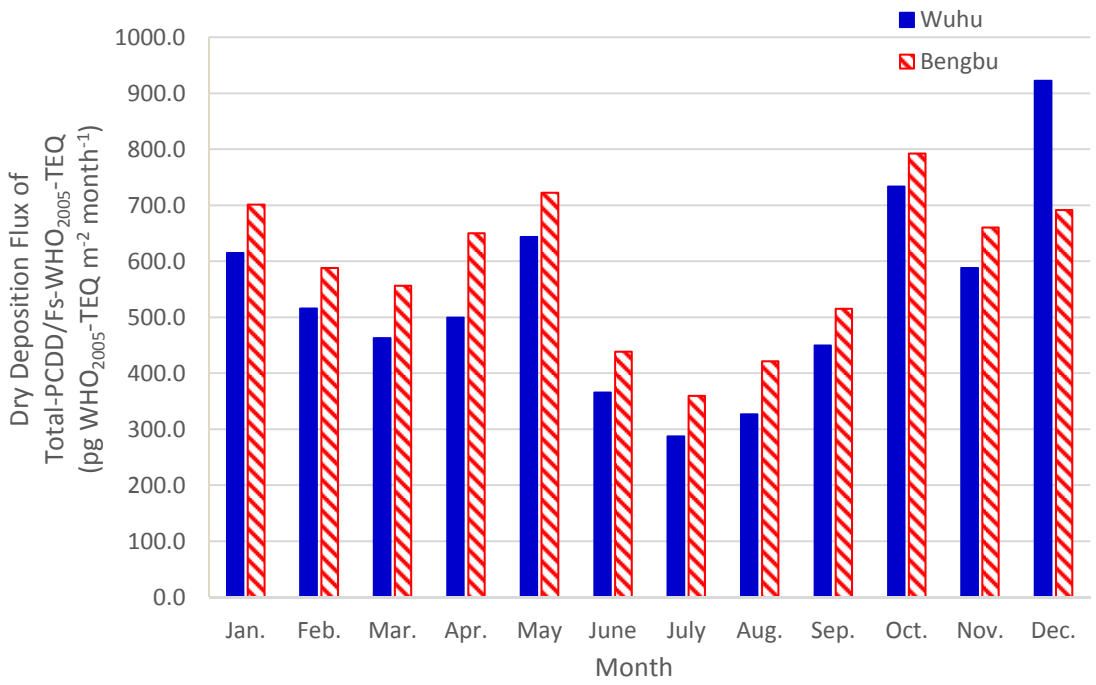

Fig. 1. Monthly average dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ during 2015.

Shijiazhuang (15400 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhu *et al.*, 2017b), respectively; but was 1.1, 1.4 and 2.9 times higher than those in Nanjing (6480 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Yunlin (4568 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Guangzhou (2470 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), respectively (Chen *et al.*, 2017; Zhu *et al.*, 2017a). These levels were much higher than that in coastal and high mountain areas in Taiwan, which was 10.4 times higher than those in Hengchun (685 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and 52.7 times higher than those in Lulin (135 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Suryani *et al.*, 2015).

With regard to the maximum and minimum monthly average dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu, the highest values occurred in December (922 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), and the lowest values occurred in July (287 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$); while in Bengbu, the highest dry deposition fluxes were in October (793 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), and the lowest values were in July (360 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). For the purpose of a comparison, Suryani *et al.* (2015) reported that the lowest dry deposition fluxes in Hengchun and Lulin both occurred in August (33.6 and 4.2 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively); these similar results were comparable with those of Zhu *et al.* (2017b), where the lowest dry deposition fluxes of Harbin and Shijiazhuang also occurred in August (308 and 699 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively). This indicated that the lowest dry deposition occurred in the warm season, as similar with the previous study of Zhu *et al.* (2017a), the estimated monthly fluctuations of PCDD/Fs dry deposition fluxes reached the highest level in cold season and the lowest in the warm season (Sugita *et al.*, 1994; Shih *et al.*, 2006; Huang *et al.*, 2011a).

For the seasonal variations, the average dry deposition fluxes for Wuhu were 535, 326, 590 and 684 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively, and in Bengbu, these values were 644, 407, 656 and 660 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively. These values were similar to those for Nanjing (625, 208, 477 and 848 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively), due to the fact that these three cities were located in the mid-latitude region. Thus, the air conditions were significantly similar, including atmospheric temperature, PCDD/Fs concentration, and particulate matter concentration in the air (Tang *et al.*, 2017; Xing *et al.*, 2017; Zhu *et al.*, 2017b; Wang *et al.*, 2018). However, the dry deposition fluxes in the high latitude cities of Harbin (552, 335, 890, and 950 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively) and Shijiazhuang (1400, 800, 1190, and 1750 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively) were higher in all seasons, reaching the highest levels in winter. This was probably because a significant amount of coal heating and biomass burning produced more pollutant, and an atmospheric temperature inversion occurred more easily with a low ground temperature, which would hinder the diffusion and dispersion of pollutants. Hence, the accumulated total-PCDD/Fs-WHO_{2005}-TEQ was mainly removed by dry deposition during light rainfall intensity seasons. In Guangzhou, which is a low latitude city with higher air temperature, showed lower dry deposition fluxes (186, 64.3, 177 and 396 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively). These results were similar to those found in previous studies of dry deposition fluxes of PCDD/Fs in rural areas in Taiwan (Shih *et al.*, 2006; Wu *et al.*, 2009; Mi *et al.*, 2012; Li *et al.*, 2016; Zhu *et al.*, 2017).

In general, the monthly average dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ in Bengbu (7101 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) was slightly higher than that in Wuhu (6407 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), this may due to a lower average temperature and a higher atmospheric $PM_{2.5}$ concentration (Zhu *et al.*, 2017). The above results also suggest that the levels for the both cities were similar to those of the mid-latitude city of Nanjing, were lower than those in the higher latitude cities of Harbin and Shijiazhuang, but were higher than those in lower the latitude cities of Guangzhou and Taiwan. Furthermore, the lowest dry deposition fluxes of these cities occurred in summer and the highest levels occurred in winter, which was consistent with previous studies (Sugita *et al.*, 1994; Shih *et al.*, 2006; Huang *et al.*, 2011a; Zhu *et al.*, 2017). The regional and seasonal differences may be the result of a combined effect of pollutant concentration and atmospheric temperature. The higher latitude region with higher total-PCDD/Fs concentrations in the air, occurred where inertial collisions are more likely to occur. Inertial collisions are a mechanism of dry deposition, thus the dry deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ were higher than those of middle and lower latitude regions. On the other hand, previous studies have indicated that most dry deposition fluxes in PCDD/Fs are bound to the particle phase, and there has been a negative relationship found between temperature and particle phase (Lee *et al.*, 2008; Wang *et al.*, 2010). This means that there are greater amounts of particle-bound PCDD/Fs under conditions of low air temperature. Owing to gravity, more particles are removed by dry deposition, so it the total dry deposition flux of PCDD/Fs decrease as the temperature rises (Shih *et al.*, 2006; Huang *et al.*, 2011a; Suryani *et al.*, 2015).

Wet Deposition

Wet deposition is also an efficient way to remove PCDD/Fs during days with precipitation. The gaseous phase PCDD/Fs dissolve into raindrops, and suspended particles are removed via precipitation in the form of rainfall, cloud droplets, and snow (Lohmann and Jones *et al.*, 1998; Moon *et al.*, 2005; Melymuk *et al.*, 2011). The monthly mean wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu and Bengbu were estimated by using Eq. (7), as shown in Fig. 2.

During 2015, the monthly average wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu ranged from 46.2 to 373.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ was 1538 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. As for Bengbu, the average wet deposition fluxes were between 13.6 and 278.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual wet deposition flux of 1525 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. Compared with the other cities in China, the annual wet deposition flues in Wuhu and Bengbu were all approximately 4.1, 4.1, 2.5, 2.7 and 1.4 times higher than those in Harbin (369 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Yunlin (373 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Shijiazhuang (622 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Guangzhou (570 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Nanjing (1240 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Chen *et al.*, 2017; Zhu *et al.*, 2017a, b) and were also much higher than those in coastal and high mountain areas in Taiwan, which were 10.9 times higher than those in Hengchun (140.1 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and 15.7 times higher than those in Lulin (97.4 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Suryani *et al.*, 2015). It was found that these levels in both Wuhu and Bengbu were similar to those in Nanjing, but were higher than those in Harbin and Shijiazhuang, northern cities in China, and Guangzhou, southern cities in China. This was due to the fact that there are several key factors affecting wet deposition of total-PCDD/Fs-WHO_{2005}-TEQ in ambient air like PM_{10}, $PM_{2.5}$, snow, temperature, and wind speed, in addition to rainfall (Wang *et al.*, 2010; Huang *et al.*, 2011b; Suryani *et al.*, 2015; Zhu *et al.*, 2017). Based on monthly $PM_{2.5}$ concentration and rainfall data from the local air quality

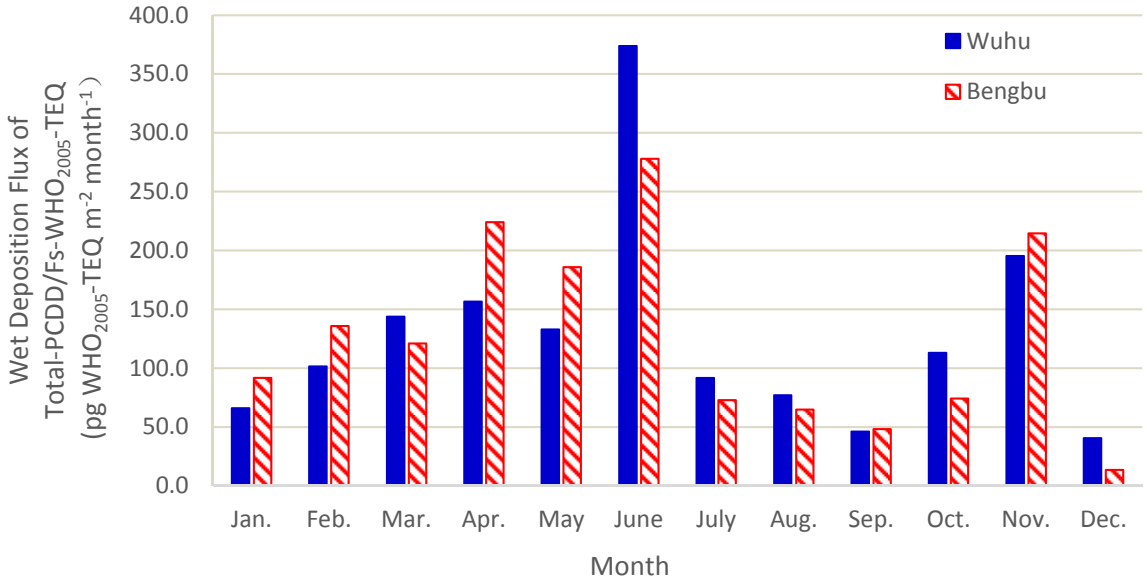

Fig. 2. Monthly average wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ during 2015.

monitoring stations, the values in Wuhu ($PM_{2.5}$ concentration = 57.3 μg m^{-3} and rainfall = 976.2 mm) and Bengbu (63.2 μg m^{-3} and 1222.3 mm) were similar with those of Nanjing (56.6 μg m^{-3} and 1091.1 mm). Thus, the wet deposition levels of these three cities were very similar, and the higher $PM_{2.5}$ concentrations with less rainfall occurred in both Harbin (69.7 μg m^{-3} and 415.8 mm) and Shijiazhuang (88.0 μg m^{-3} and 534.5 mm) and could have resulted in a lower wet deposition flux. As for Guangzhou, even though there was higher rainfall (2234 mm), the lower $PM_{2.5}$ concentration (38.9 μg m^{-3}) also led to a lower wet deposition flux.

In Wuhu, the highest monthly average wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ occurred in the highest rainfall (191.4 mm) period in June (373.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), with a lower $PM_{2.5}$ concentration (34 μg m^{-3}); and the lowest values occurred in December (40.5 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which had the highest $PM_{2.5}$ concentration (108 μg m^{-3}), but the rainfall (38.8 mm) was very low. The lowest rainfall (27.4 mm) occurred in January, while the $PM_{2.5}$ concentration (88 μg m^{-3}) was very high. Thus, its wet deposition was not the lowest, and the lowest $PM_{2.5}$ concentration (32 μg m^{-3}) occurred in July, when the rainfall (139.2 mm) was very high, and the wet deposition fluxes also not the lowest. Both wet deposition flux and rainfall fluctuated from month to month, and the variations for both were very similar.

In Bengbu, the highest wet deposition fluxes also occurred in the month with the highest rainfall (368.7 mm) in June (278.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which also had a higher $PM_{2.5}$ concentration (49 μg m^{-3}), although the $PM_{2.5}$ concentration (88 μg m^{-3}) was high in December due to having the lowest rainfall (5.5 mm), resulting in the lowest wet deposition (13.6 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). The above results indicate that rainfall and $PM_{2.5}$ concentration may affect the wet deposition fluxes significantly, but the maximum and minimum wet deposition mainly occurred in the highest or lowest rainfall period, which means that the rainfall had the strongest influence on wet deposition (Suryani *et al.*, 2015).

The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ varied from season to season. In Wuhu, the seasonal average wet deposition fluxes were 144.4, 180.8, 118.2 and 69.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively. The highest wet deposition flux obtained in summer and the lowest value occurred in winter were strongly influenced by the seasonal distribution of rainfall, which were 89.5, 139.5, 56.2 and 40.2 mm in spring, summer, fall and winter, respectively. And in Bengbu, the seasonal average wet deposition fluxes were 177.0, 138.5, 112.4 and 80.4 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively. The rainfall in Bengbu is mainly concentrated in spring and summer, and fall and winter always are characterized by less rainfall, which was 99.6, 222.1, 61.9 and 35.9 mm in spring, summer, fall, and winter, respectively. For seasonal values seen in previous studies, the wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ of Guangzhou in spring, summer, fall and winter were 127, 32.7, 10.8 and 31.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively (Zhu *et al.*, 2017a), were 60.4, 60.1, 71.2 and 15.8 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Shijiazhuang, and were 32.0, 48.8, 27.2 and 15.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Harbin, respectively (Zhu *et al.*, 2017b). The lowest wet deposition always occurred in winter, where previous studies demonstrated that the amount of precipitation and the number of rainy days could affect seasonal variations in wet deposition fluxes more than ambient temperature (Suryani *et al.*, 2015; Lee *et al.*, 2016).

The monthly average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain are shown in Fig. 3. It can be seen that the monthly average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain in Wuhu ranged between 0.36 and 3.29 pg WHO_{2005}-TEQ L^{-1} and averaged 1.41 pg WHO_{2005}-TEQ L^{-1}, where July had the lowest, while December had the highest. As to the seasonal variations, the average total-PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain in Wuhu were 1.347, 0.443, 1.443 and 2.415 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. The average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain in Bengbu ranged between 0.38 and 2.53 pg WHO_{2005}-TEQ L^{-1} and averaged 1.61 pg WHO_{2005}-TEQ L^{-1}, which indicated that the July had the lowest, while January had the highest. The seasonal average of total-PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain in Bengbu were 1.774, 0.584, 1.737, and 2.354 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. The above results were similar to those for Nanjing (averaged 1.70 pg WHO_{2005}-TEQ L^{-1}), but were higher than those in southern cities in China (averaged 0.49 pg WHO_{2005}-TEQ L^{-1} in Guangzhou) (Zhu *et al.*, 2017a) and were lower than those in northern cities in China (averaged 3.95 and 2.28 pg WHO_{2005}-TEQ L^{-1} in Shijiazhuang and Harbin, respectively) (Zhu *et al.*, 2017b). These values were much greater than those found in Taiwan (averaged 0.064 pg WHO_{2005}-TEQ L^{-1} in Hengchun and 0.027 pg WHO_{2005}-TEQ L^{-1} in Lulin, respectively) (Suryani *et al.*, 2015). The above results showed that the total-PCDD/Fs-WHO_{2005}-TEQ concentration in the rain of winter were highest, this is probably attributed to less precipitation and higher concentration of total-PCDD/Fs-WHO_{2005}-TEQ lead to greater fraction of PCDD/Fs absorbed to particle at cooler temperature (Koester and Hites, 1992; Zhu *et al.*, 2017b), and those were lowest in summer; while the values for spring and fall were both in the middle levels.

The scavenging ratio is the pollutant concentration in a raindrop divided by the concentration in the ambient air. The scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ are presented in Fig. 4. As the results indicate, the monthly average scavenging ratio ranged between 19580 and 39370 and averaged 30140 (annual temperature was 17.3°C) in Wuhu, with 17130–39400 and an average of 31270 (annual temperature was 16.3°C) in Bengbu. These values were similar to those of Shijiazhuang (30700) and Nanjing (30950), but were lower than those in Harbin (31900) and Yunlin (41700), higher than those in Guangzhou (24080), as well as much greater than those found in Hengchun (8015) and Lulin (13450) in Taiwan (Suryani *et al.*, 2015; Chen *et al.*, 2017; Zhu *et al.*, 2017a, b). This may be attributed to differences in air temperature, which were 14.9°C for

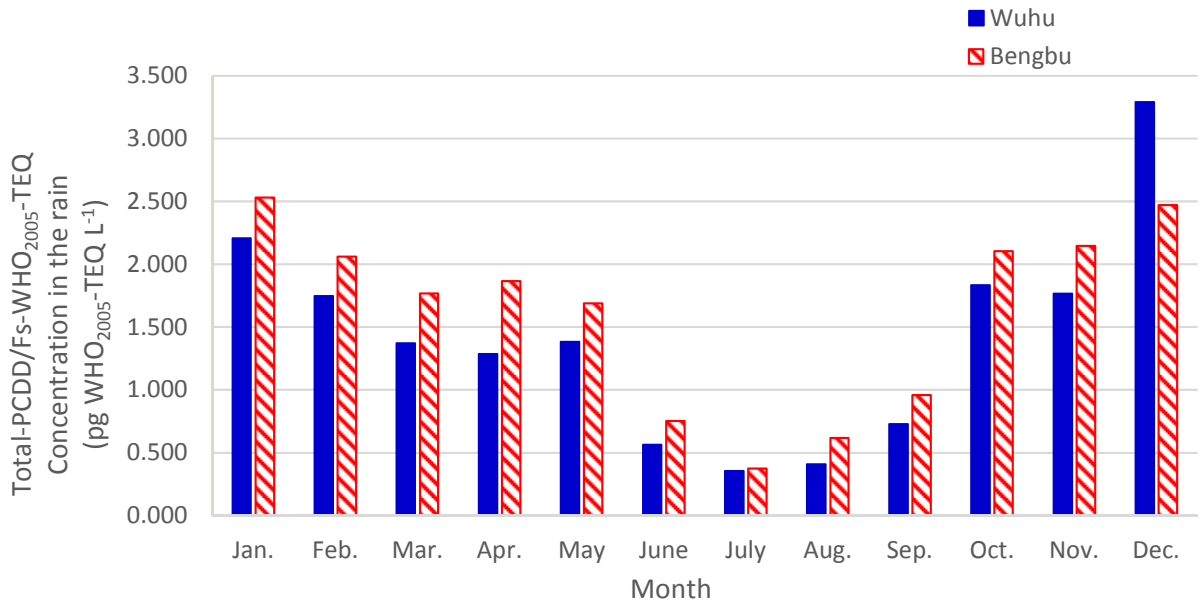

Fig. 3. Monthly average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain during 2015.

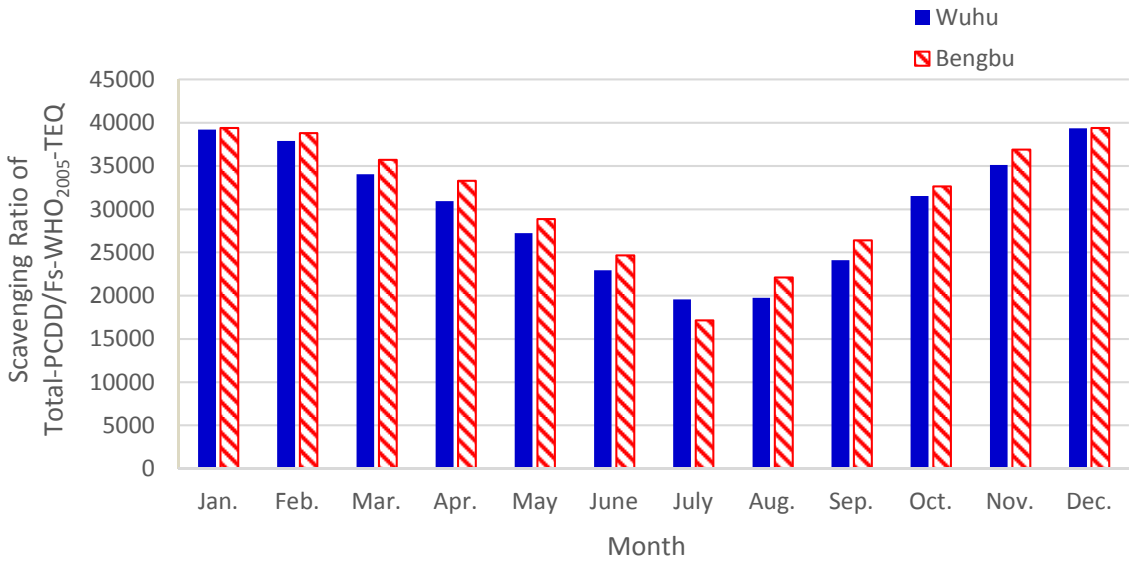

Fig. 4. Monthly average scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ during 2015.

Shijiazhuang, 16.4°C for Nanjing, 5.1°C for Harbin and 21.7°C for Guangzhou. The total scavenging ratios had a negative correlation with the ambient temperature due to the fact that much of the particle phase scavenging occurred between ground level and clouds, where the gas-particle partitioning of PCDD/Fs could be affected by air temperature. Furthermore, the seasonal average air temperature of Wuhu were 17.3, 26.7, 18.6 and 6.5°C in spring, summer, fall and winter, respectively; and these of Bengbu were 15.9, 27.5, 16.9 and 5.0°C in spring, summer, fall and winter, respectively. The air temperature is higher in summer and the vertical transport of air current is more violent, which can accelerate the dispersion of air contaminants, thus the concentration of total-PCDD/Fs-WHO_{2005}-TEQ are in lower levels (Wang *et al.*, 2018). Due to more amount of particle-bound PCDD/Fs can evaporate into the gas phase with a higher air temperature, the levels of particle-bound PCDD/Fs are also lower, thus the rainfall can scavenge less both particle phase and total PCDD/Fs and the scavenging ratio of atmospheric PCDD/Fs in the summer was much lower. Similar results can be found in previous studies (Koester and Hites, 1992; Van Ry *et al.*, 2002; Wang *et al.*, 2010; Huang *et al.*, 2011; Tseng *et al.*, 2014).

Total (Dry +Wet) Deposition

The total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were the combination of dry and wet depositions. The monthly average total (dry + wet) deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in both Wuhu and Bengbu were presented in Fig. 5.

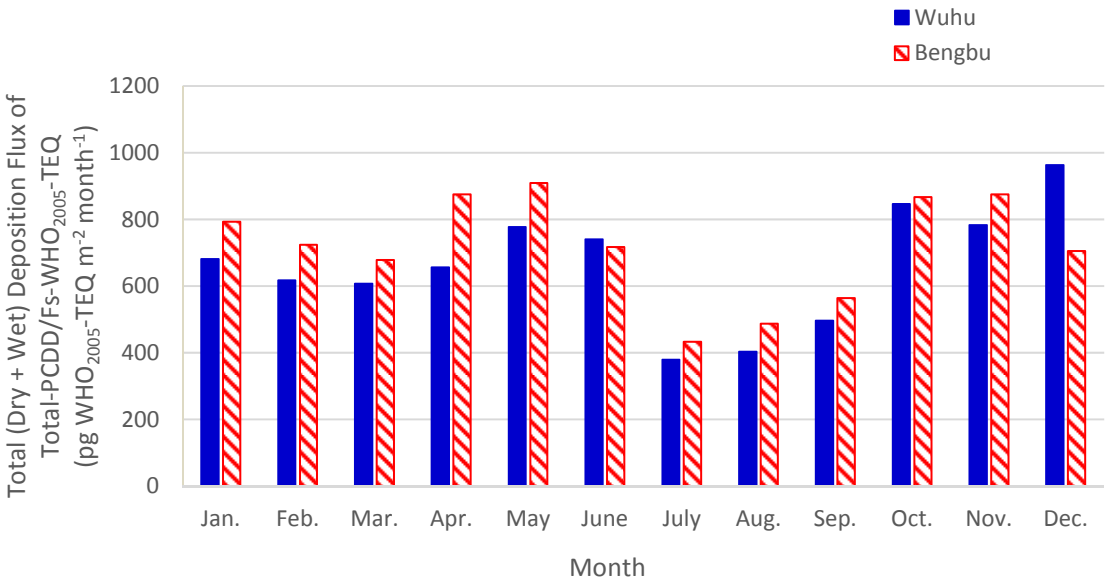

Fig. 5. Monthly average total (dry + wet) deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ during 2015.

In Wuhu, the total deposition fluxes ranged from 379 to 962 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total deposition flux was 7945 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$; while in Bengbu, the total deposition fluxes ranged between 433 and 909 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total deposition flux was 8626 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. These values were similar to the result found in Nanjing (7716 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhu *et al.*, 2017a) and Harbin (8610 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), and were approximately only a 48.9% order of magnitude lower than those for Shijiazhuang (16100 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhu *et al.*, 2017b), but were approximately 1.7, 2.7, 10.1, and 35.7 times higher than those in Guangzhou (3043 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Yunlin (4941 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Hengchun (825 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Lulin (232 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Suryani *et al.*, 2015; Chen *et al.*, 2017; Zhu *et al.*, 2017a), respectively. The distribution of total deposition flux was similar for the corresponding dry deposition flux as discussed above.

In terms of seasonal variation, the total deposition fluxes of Wuhu were 679, 507, 708 and 753 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively, and in Bengbu, these values were 821, 545, 769 and 741 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. Comparing these values with Nanjing (744, 277, 582 and 970 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall and winter, respectively), Harbin (584, 404, 917 and 965 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively), Shijiazhuang (1460, 860, 1270 and 1770 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively), and Guangzhou (314, 97, 188 and 415 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively) (Zhu *et al.*, 2017a, b), the lowest total deposition fluxes in these cities occurred in summer, and the highest levels occurred in winter, which was consistent with the dry deposition results. The above results indicated that dry deposition was the main contributor to the total deposition. Zhu *et al.* (2017b) reported that the dry deposition fluxes are the major removal mechanism of the total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air.

The fraction of total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ contributed by the dry and wet deposition, respectively, in Wuhu and Bengbu are shown in Fig. 6. In Wuhu, the fraction of wet deposition contributing to total deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ were 4.2% in December and 50.6% in June, with an annual average of 19.7%; in Bengbu, this was 1.9% in December and 38.8% in June, with an annual average of 17.2%. Compared to the other cities, the contribution fractions of wet deposition fluxes to that of total deposition in Wuhu and Bengbu were similar to those in Nanjing, which were 1.4% in December and 33.5% in July, with an annual average of 17.6%. These percentages were slightly lower than that in Guangzhou, which were 0.2% in December and 49.4% in July, with an annual average of 21.4% (Zhu *et al.*, 2017a). Compared with the northern cities in China, the contribution fractions of wet deposition fluxes to the total deposition in Wuhu and Bengbu were higher than that in Harbin, which were ~0% in January and 49.4% in May, and in Shijiazhuang, which were 0.2% in December and 49.4% in September, respectively (Zhu *et al.*, 2017b). In general, the fraction of total deposition contributed by wet deposition in the lower latitude region was higher than that in the higher latitude region, and it was also found that the highest fraction of wet deposition occurred in the hot season and the lowest occurred in the cold season because of climate variations and more precipitation in the summer. In China, from north to south, the climate includes cold temperate zones, temperate zones, warm temperate zones, subtropical zones, and tropical zones, where the rainfall intensity gradually becomes stronger in that order. Moreover, the monsoon climate in China significantly contributes to the increased rainfall in summer and there being less in winter. Thus, the particles were also scavenged more by rainfall via wet deposition in seasons with greater precipitation than was the case for dry deposition.

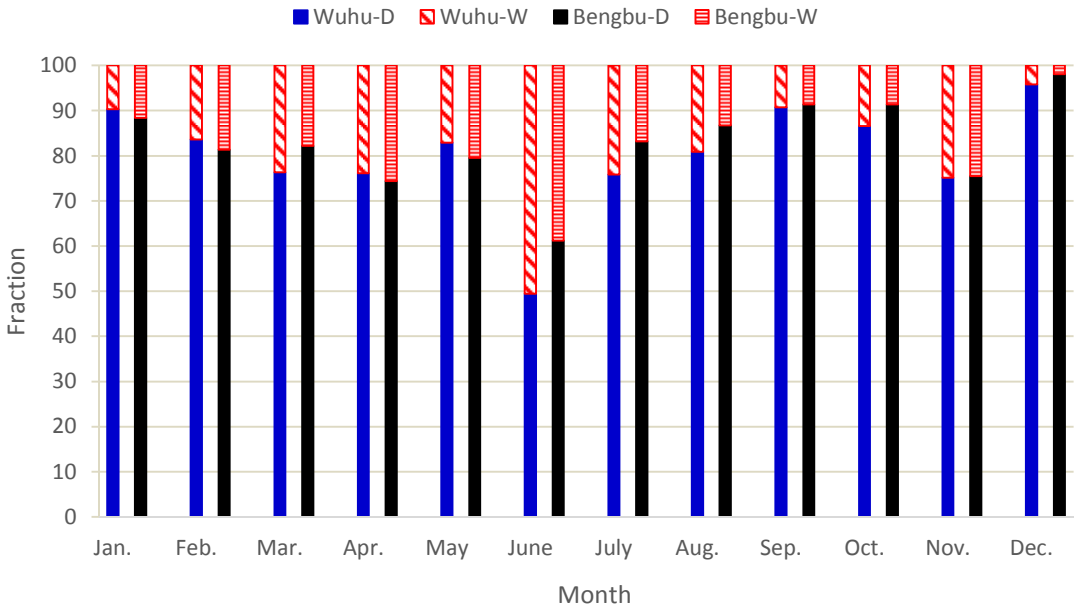

Fig. 6. The fraction of total deposition flux in total-PCDD/Fs-WHO_{2005}-TEQ contributed by the dry and wet deposition, in Wuhu and Bengbu, respectively.

Sensitivity Analysis

The parameters most likely to affect gas-particle partitioning will be the most influential parameters on the scavenging ratio and wet deposition flux. The sensitivity analysis conducted in this study was focused on the ambient temperature, the $PM_{2.5}$ concentration, the PM_{10} concentration, total PCDD/F concentrations, and rainfall, which are sensitivity variables that contribute to alterations in both the scavenging ratio and wet deposition flux. In the sensitivity analyses conducted in this study, in Wuhu, depending on the initial values of ambient air temperature = 20.0°C, $PM_{2.5}$ = 46.0 μg m^{-3}, PM_{10} = 67.0 μg m^{-3}, total-PCDD/F mass concentration = 1.12 pg m^{-3} and rainfall = 72.5 mm. The parametric sensitivity for the scavenging ratio for Wuhu are shown in Fig. 7(a), and the that for the wet deposition flux are shown in Fig. 7(b).

where P: initial value of parameters;

ΔP: increase or decrease in parameters;

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

ΔS: response in each of the parameters.

From Fig. 7(a), the PM_{10} concentration and PCDD/Fs mass concentration have equivalently important sensitivity to the total-PCDD/Fs-WHO_{2005}-TEQ scavenging ratio. The effect of both PM_{10} concentration and PCDD/Fs can be divided into two parts: For PM_{10} concentration, where ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –65.9%; when ΔP/P was changed from 0% to +50% and +70%, ΔS/S responded from 0% to +25.4% and from 0% to +27.6%, respectively. In terms of the mass concentration of PCDD/Fs, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –67.0%; when ΔP/P was changed from 0% to +50% and +70%, ΔS/S responded from 0% to +25.5% and from 0% to +27.5%, respectively. The PM_{10} can reflect the status of particle phase PCDD/Fs, where a lower PM_{10} concentration means the air condition are beneficial to the dispersion of air pollutants, hence, the PM_{10} and total PCDD/F concentration decreased, and fewer pollutants could be removed by rainfall, as a result, the scavenging ratio decreased significantly. While the scavenging ratio rose with increases in the PM_{10} and PCDD/Fs, which usually occurred in winter with less rainfall. This kind of positive effect more significantly elevated the scavenging ratio.

The sensitivity analysis indicated that $PM_{2.5}$ concentration has a significant, positive effect on the scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ. The effect of $PM_{2.5}$ can be divided into two parts, where when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –57.0%; when ΔP/P was changed from 0% to +50% and +100%, ΔS/S responded from 0% to +23.5% and +28.6%, respectively. For comparison, the change in $PM_{2.5}$ concentration had a similar positive effect on the scavenging ratio with the PM_{10} concentration, but when the $PM_{2.5}$ concentration was further reduced, its sensitivity was weaker than the decreasing PM_{10} concentration, which was due to the fact that $PM_{2.5}$ is not as representative as PM_{10} in terms of reflecting the status of particle phase PCDD/Fs.

Fig. 7(a) shows that air temperature has a negative correlation with the scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ, where when ΔP/P was changed from –50% to +50%, ΔS/S responded from +28.6% to –28.6%. Because the temperature will affect the vapor pressure of PCDD/Fs and influence gas-particle partitioning. The mean fractions of particle scavenging of PCDD/Fs in spring, summer, fall, and winter were 94.5%, 78.4%, 92.3% and 99.3%, respectively, indicating particle scavenging was dominant in total PCDD/Fs. This was due to the fact that a large fraction of particle-bound PCDD/Fs formed at lower temperatures (Koester and Hites, 1992). Huang *et al.* (2011) also observed

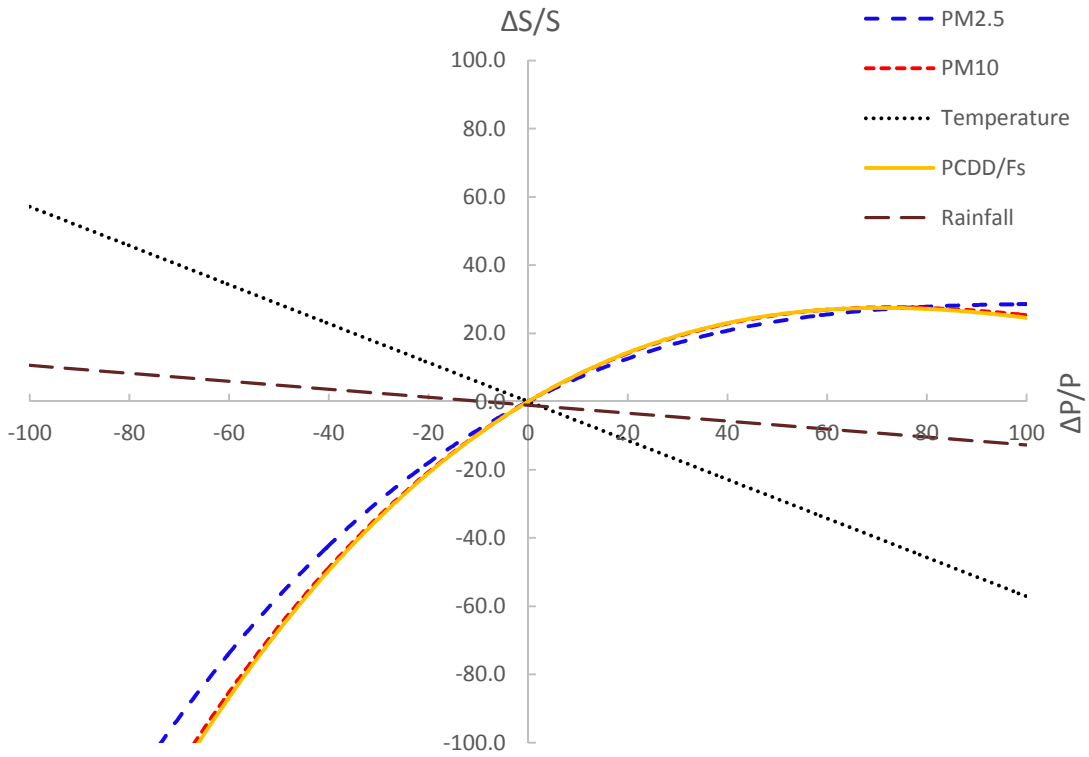

Fig. 7(a). Sensitivity analysis for the scavenging ratio of total PCDD/Fs-WHO_{2005}-TEQ in Wuhu.

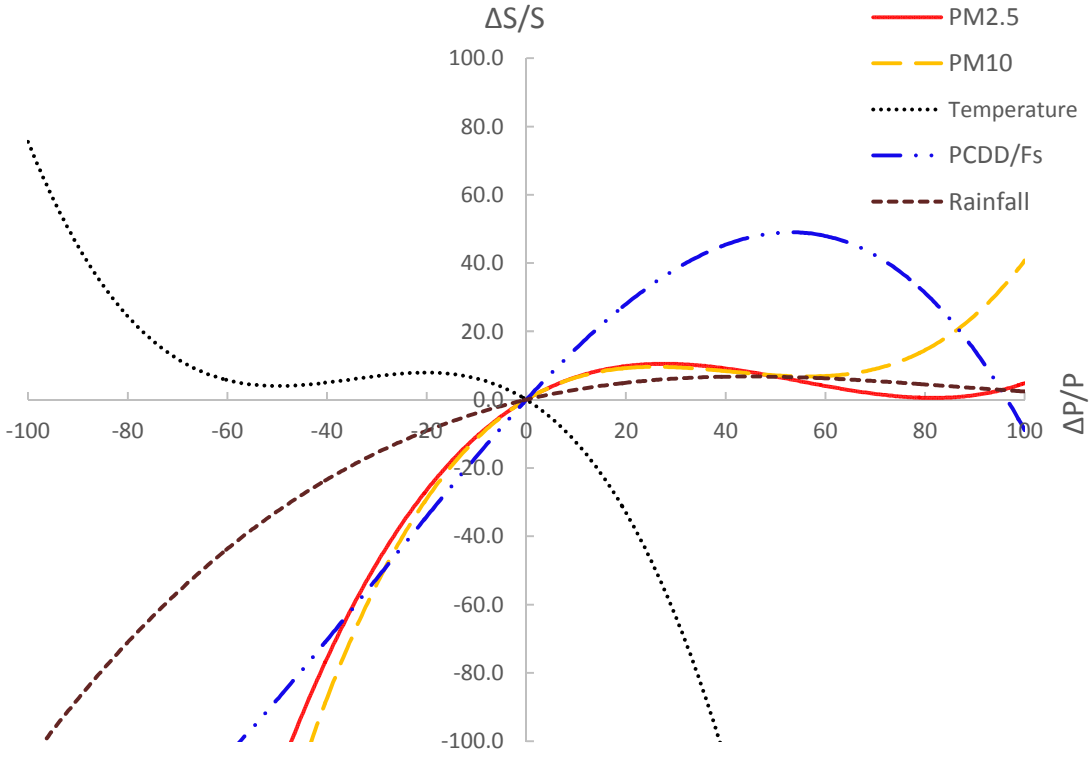

Fig. 7(b). Sensitivity analysis for wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Wuhu.

that the highest chlorinated congener is associated with the highest value of particle scavenging ratio. Therefore, as the air temperature increases, this will result in a greater amount of particle-bound PCDD/Fs evaporating to the gas phase. The total scavenging ratio of PCDD/Fs was decreased when few particle-phase PCDD/Fs were removed by raindrops. Hence, the total scavenging ratio was decreased with increases in air temperature.

Fig. 7(a) also demonstrates that rainfall has a negative effect on the scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ, where when ΔP/P was changed from –50% to +50%, ΔS/S responded from +5.1% to –6.6%. This may be attributed to the fact that PCDD/F concentration plays a more important role in the scavenging ratio than rainfall. In spite of tremendous rainfall intensity always occurring in the summer, the total and particle-bound PCDD/F concentration was at a low level in these periods. In addition, during the summer time, when temperatures were higher, less vapor and suspended particulates were removed by raindrops, which led to a decrease in the concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain and a slight reduction in the total scavenging ratio. When the fraction of total and particle-phase PCDD/F concentration increased, less rainfall was accompanied with higher total-PCDD/Fs-WHO_{2005}-TEQ in the rain, thus the total scavenging ratio of PCDD/Fs displayed an increasing tendency in cold winters.

The above results suggest that the most sensitive parameters for the scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ is atmospheric PCDD/F concentration and PM_{10} concentration, followed by the $PM_{2.5}$ concentration, and air temperature in that order. The last sensitivity parameter was rainfall.

With regard to the sensitivity analyses for the wet deposition flux of PCDD/Fs, Fig. 7(b) show that the air temperature was the most sensitive parameter for the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ, which can be divided into three stages: when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to –161.5%; when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to +4.0%; and when ΔP/P was changed from –50% to –100%, ΔS/S responded from +4.0% to +75.5%. Due to the fact that increasing air temperatures will increase the amount of particle-phase PCDD/Fs and will cause PCDD/Fs with higher molecular weight to be evaporated into the gas phase, in the first stage, this results in a significantly negative effect on wet deposition flux. As for the second stage, the PCDD/F mass mainly existed in the particle phase, when the temperature was not very high (between 10°C and 20°C), and less gas phase PCDD/Fs were able to be shifted into the particle phase. As a result, when the temperature rose, and the atmospheric total PCDD/Fs increased only slightly. Therefore, increasing temperatures had less effect on the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ. However, when the temperature decreased continuously, the wet deposition increased slowly and was affected by other parameters such as snow and fog.

From Fig. 7(b), it can be seen that the atmospheric PCDD/F mass concentration has a positive or negative correlation with the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ, where when ΔP/P was changed from –50% to +50%, ΔS/S responded from –87.7% to +48.8%; but when ΔP/P was changed from +50% to +100%, the ΔS/S responded from +48.8% to –8.9%. An increase in PCDD/Fs means that there are more gas phase and particle phase PCDD/Fs removed by wet deposition, but when the atmospheric PCDD/F mass concentration is higher than 1.47 pg m^{-3}, this period is usually accompanied with significantly less rainfall, and therefore, the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ decreases as the PCDD/F mass concentration continues to rise.

The sensitivity analysis also indicated that both $PM_{2.5}$ and PM_{10} concentrations have almost equivalent sensitivity to the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ. The effect of both $PM_{2.5}$ and PM_{10} concentrations is as follows: In the case of the $PM_{2.5}$ concentration, when ΔP/P was changed from 0% to –50%, the ΔS/S responded from 0% to –110.7%; when ΔP/P was changed from 0% to +30% and +50%, ΔS/S responded from 0% to +10.4% and from 0% to +6.8%, respectively. In terms of PM_{10} concentration, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –130.6%; when ΔP/P was changed from 0% to +30% and +50%, ΔS/S responded from 0% to +9.5% and from 0% to +7.1%, respectively. Lower levels of $PM_{2.5}$ concentrations usually occurred in the warm season, during which the rainfall intensity was higher, and more particle-bound PCDD/Fs were removed by wet deposition. However, a higher $PM_{2.5}$ concentration usually occurred in the cold season, where, because of the limitation of other parameters such as rainfall, snow, and fog in this period, the effect of increasing $PM_{2.5}$ concentrations on the wet deposition of the flux of total-PCDD/Fs-WHO_{2005}-TEQ was minor.

A decrease in rainfall has a slightly positive correlation with the wet deposition flux, but increasing rainfall is not very sensitive to the wet deposition flux. These two stages can be described as follows: When ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –32.7%; but when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +6.7%. In general, a higher PCDD/F mass concentration was accompanied by less rainfall in the cold season, where when the rainfall decreased, less PCDD/Fs were removed by raindrops, and consequently, the wet deposition flux was reduced. However, the wet deposition flux was also affected by the change in PCDD/F concentration, so the positive correlation of rainfall in wet deposition was less obvious than that for the total PCDD/F concentration. However, the atmospheric PCDD/Fs concentration was at a low level in the rainy warm season, where increasing rainfall intensity caused a minor effect on the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ, which was limited by the low level of PCDD/Fs concentration in the atmosphere. In terms of seasonal variation, the lowest values of the PCDD/F wet deposition (64.6, 61.9 and 38.7 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in 2015, 2016 and 2017, respectively) and the highest rainfall intensity (139.5, 159.6 and 101.5 mm in 2015, 2016 and 2017, respectively) are occurred in summer. From the view of monthly average levels, in summer, the rainfall intensity in June (191.4, 243.6 and 101.3 mm in 2015, 2016 and 2017, respectively) and July (139.2, 201.6 and 133.8 mm in 2015, 2016 and 2017, respectively) are very strong, while the rainfall in August (87.9, 33.5 and 69.5 mm in 2015, 2016 and 2017, respectively) is very low. But the concentration of PCDD/Fs achieve the lowest levels in July (0.65, 0.56 and 0.74 pg m^{-3} in 2015, 2016 and 2017, respectively) and August (0.74, 0.51 and 0.63 pg m^{-3} in 2015, 2016 and 2017, respectively), this means that the PCDD/Fs concentration can significantly affect wet deposition

of PCDD/Fs. This result is similar to the report of coastal and high mountain areas in Taiwan, particle matter concentration also plays an important role in the wet deposition, higher particle matter concentration means that more particulates can be scavenged by wet deposition, in addition, other parameters effect like snow or fog also should consider (Suryani *et al*., 2015). So the rainfall intensity is not the most sensitive parameter for the PCDD/F wet deposition.

The above discussion suggests that the most sensitive parameter for the wet deposition of total-PCDD/Fs-WHO_{2005}-TEQ was ambient air temperature, followed by the atmospheric PCDD/F mass concentration, the $PM_{2.5}$ concentration, and the PM_{10} concentration in that order. The rainfall intensity was the last sensitivity parameter.

CONCLUSION

The results of this study on the atmospheric deposition in Wuhu and Bengbu can be summarized as follows:

1. During 2015, the monthly dry deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 287 and 922 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Wuhu, and between 360 and 793 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Bengbu. The annual total dry deposition flux in Bengbu (7101 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) was approximately 1.11 times higher than that in Wuhu (6407pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). In general, the levels in both Wuhu and Bengbu were similar to those in the mid-latitude city of Nanjing, were lower than those in the higher latitude cities of Harbin and Shijiazhuang, and were higher than those in the lower latitude cities of Guangzhou and Taiwan.
2. The wet deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 46.2 and 373.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Wuhu, and between 13.6 and 278.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Bengbu. The annual total dry deposition fluxes were 1538 and 1525 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in Wuhu and Bengbu, respectively. Furthermore, these levels in both cities were similar to those in Nanjing (1240 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), but were higher than those in Harbin (369 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Yunlin (373 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Shijiazhuang (622 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Guangzhou (570 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Hengchun (140.1 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Lulin (97.4 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$).
3. The monthly average concentration of total-PCDD/Fs-WHO_{2005}-TEQ in the rain were 0.36 (July) –3.29 (December) pg WHO_{2005}-TEQ L^{-1} in Wuhu, and were 0.38 (July) –2.53 (January) pg WHO_{2005}-TEQ L^{-1} in Bengbu, and the average were 1.41 and 1.61 pg WHO_{2005}-TEQ L^{-1} in Wuhu and Bengbu, respectively. The results showed that the total-PCDD/Fs-WHO_{2005}-TEQ concentration in the rain was lowest in summer and highest in winter, while the values for spring and fall were both in the middle levels.
4. The total scavenging ratios of total-PCDD/Fs-WHO_{2005}-TEQ had a negative correlation with the ambient temperature. The monthly average scavenging ratio ranged between 19576–39370, with an average of 30144 (17.3°C) in Wuhu, and between 17131–39401, with an average of 31267 (16.3°C) in Bengbu. These values were similar to those of Shijiazhuang (30700, 14.9°C) and Nanjing (30950, 16.4°C), but were lower than that in Harbin (31900, 5.1°C) and higher than that in Guangzhou (24080, 21.7°C).
5. The total (dry and wet) deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ were 379–962 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Wuhu, and were 433–909 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in Bengbu, for which the annual values were 7945 and 8626 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in Wuhu and Bengbu, respectively. These values were similar to the results found in Nanjing (7716 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) and Harbin (8610 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), and were approximately only 48.9% of an order of magnitude lower than that in Shijiazhuang (16100 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), but were approximately 1.7, 2.7, 10.1, and 35.7 times higher than those in Guangzhou (3043 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Yunlin (4941 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Hengchun (825 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), and Lulin (232 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$).
6. The total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ in Wuhu were 679, 507, 708 and 753 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively, where in Bengbu, these values were 821, 545, 769 and 741 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively.
7. The fraction of wet deposition contributed to annual total deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ were 19.7% and 17.2% in Wuhu and Bengbu, respectively. Because of being dominated by the variations in climate, the fraction contributed by wet deposition in the lower latitude region was higher than that in the higher latitude region, and the highest fraction of wet deposition occurred in warm summers and the lowest occurred in cold winters.
8. The most sensitive parameters for the scavenging ratio of total-PCDD/Fs-WHO_{2005}-TEQ were atmospheric PCDD/F mass concentration and PM_{10} concentration. When $\Delta P/P$ was changed from –50% to +50%, $\Delta S/S$ responded from –67.0% to +25.5% for PCDD/Fs; and when $\Delta P/P$ was changed from –50% to +50%, $\Delta S/S$ responded from –65.9% to +25.4% for PM_{10}. The second positive correlated sensitivity factor was $PM_{2.5}$ concentration. When $\Delta P/P$ was changed from –50% to +50%, $\Delta S/S$ responded from –57.0% to +23.5%. This was followed by air temperature, where when $\Delta P/P$ was changed from –50% to +50%, $\Delta S/S$ responded from +28.6% to –28.6%. The last sensitive parameter was rainfall, where when $\Delta P/P$ was changed from –50% to +50%, $\Delta S/S$ responded from +5.1% to –6.6%.
9. The most sensitivity parameter for the wet deposition flux of total-PCDD/Fs-WHO_{2005}-TEQ was air temperature, which can be divided into three stages: when $\Delta P/P$ was changed from 0% to +50%, $\Delta S/S$ responded from 0% to –161.5%; but when $\Delta P/P$ was changed from 0% to –50%, $\Delta S/S$ responded from 0% to +4.0%. When $\Delta P/P$ was changed from –50% to –100%, $\Delta S/S$ responded from +4.0% to +75.5%. The second positive or negative correlated sensitivity factor was atmospheric PCDD/F

mass concentration, when ΔP/P was changed from –50% to +50%, ΔS/S responded from –87.7% to +48.8%. this was followed by the $PM_{2.5}$ and PM_{10} concentration, in the case of the $PM_{2.5}$ concentration, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –110.7%; when ΔP/P was changed from 0% to +30% and +50%, ΔS/S responded from 0% to +10.4% and +6.8%, respectively. In terms of the PM_{10} concentration, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –130.6%; when ΔP/P was changed from 0% to +30% and +50%, ΔS/S responded from 0% to +9.5% and +7.1%, respectively. The last sensitivity parameter was rainfall, where when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –32.7%, and when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +6.7%.

10. The results of this study provided useful information for the establishment of PCDD/F control strategies and as a reference for scientific communities.

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