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Sensitivity Analyses for the Atmospheric Dry Deposition of Total PCDD/Fs-TEQ for Handan and Kaifeng Cities, China

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ABSTRACT

During the period 2016–2017, the atmospheric wet, dry, and total deposition fluxes and scavenging ratios of the total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng were investigated. In addition, a sensitivity analysis for the dry deposition fluxes of the total PCDD/Fs-WHO_{2005}-TEQ was conducted. The annual wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan ranged between 51.1 and 83.5 and averaged 72.3 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was approximately 1.04 times of magnitude higher than that in Kaifeng (69.3 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). From 2016–2017, the contribution fraction of dry deposition to the total PCDD/Fs-WHO_{2005}-TEQ deposition flux ranged between 60.8% and 100% and averaged 80.4%. Dry deposition fluxes were more dominant than wet deposition fluxes. In terms of the seasonal variations in total PCDD/Fs-WHO_{2005}-TEQ dry deposition fluxes (the mean values for 2016 and 2017) in Handan, those in spring, summer, fall, and winter were 1084, 563, 964, and 1325 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively, while in Kaifeng, they were 963, 428, 715, and 1016 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The total PCDD/Fs-WHO_{2005}-TEQ deposition fluxes in winter was approximately 2.0 times of magnitude higher than that in summer. The sensitivity analysis of total PCDD/FsWHO_{2005}-TEQ dry deposition fluxes in Handan and Kaifeng showed that the PM_{10} concentration was the most positively correlated sensitive factor. When ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +46.1% and +46.3%, respectively. The second positively correlated sensitive factor was the $PM_{2.5}$ concentration, where when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +47.8% and +40.8%, respectively. For PCDD/Fs mass concentration, when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +32.2% and +28.1%, respectively. This was followed by the atmospheric temperature, and its effect was negatively correlated. When ΔP/P was changed from –50% to +50%, ΔS/S responded from +46.4% to –26.9% and +57.0% to –30.5%, respectively. The results of this study provide useful information that can be used to achieve more insights into both atmospheric deposition of total PCDD/Fs-WHO_{2005}-TEQ and the sensitive factors for dry deposition fluxes.

Keywords: Dry deposition; Wet deposition; Scavenging ratio; Sensitivity analysis.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) are persistent organic pollutants (POPs) because they are toxic, persistent, and bio-accumulative (Micheletti *et al.*, 2007; White and Birnbaum, 2009). They are extremely hazardous chemicals that form both naturally and anthropogenically (Hashimoto *et al.*, 1990; Brzuzy and Hites, 1996; Kim *et al.*, 2003). Combustion processes in nature, such as forest fires and volcanoes as well as anthropogenic activities are the main sources of the PCDD/Fs released into the environment (Chi *et al.*, 2011). Of these two sources, anthropogenic activities are the most dominant sources contributing to the presence of PCDD/Fs in the environment, which include many manufacturing processes related to products for humans. The main sources of PCDD/Fs have been found to mostly come from the emissions of waste combustion, chemical plants, thermal sources, metal smelting process, and vehicles (Schuhmacher *et al.*, 2000; Wang *et al.*, 2003; Lin *et al.*, 2007; Hsieh *et al.*, 2009; Chuang *et al.*, 2010, 2011). PCDD/Fs can enter the human body via ingestion, inhalation, and dermal contact (Shih *et al.*, 2009;

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Chen *et al.*, 2010). In the human body, PCDD/Fs pose health risks to the human immune system, interfere with developmental, reproductive, and regulatory hormones, and even create the risk of cancer (Lin *et al.*, 2010; Chi *et al.*, 2011). They are chemically stable, have low solubility in the water, and have been shown to accumulate in the food chain (Shih *et al.*, 2009). PCDD/Fs are emitted into the atmosphere, where they are transformed, degraded, and transported from the source to receptor sites (Chi *et al.*, 2009; Xu *et al.*, 2009; Fang *et al.*, 2011). In the atmosphere, PCDD/Fs are partitioned between gas and particle phases through a process that is dependent on their vapor pressures, ambient temperatures, and other parameters (Wu *et al.*, 2009; Wang *et al.*, 2010; Cheruiyot *et al.*, 2015). PCDD/Fs can be degraded by chemical reactions controlled by OH radicals as well as by photochemical reactions (Chi *et al.*, 2009). Removal of PCDD/Fs from the atmosphere occurs via both dry and wet deposition (Giorgi, 1988; Chi *et al.*, 2009; Wu *et al.*, 2009; Huang *et al.*, 2011; Mi *et al.*, 2012). Since they were first found in the fly ash of municipal solid waste incinerators (MSWIs), researchers have paid more attention to various emissions sources (Wang *et al.*, 2003; Wang *et al.*, 2010; Chi *et al.*, 2015; Cheruiyot *et al.*, 2015, 2016; Wei *et al.*, 2016; Li *et al.*, 2017).

Wet deposition is the process by which atmospheric pollutants are removed via rainfall, cloud droplets, or snow (Lohmann and Jones, 1998) and is responsible for much of the higher chlorinated homologues in environmental sinks (Shih *et al.*, 2006; Wang *et al.*, 2010). The wet deposition flux of SVOCs is a combination of both vapor dissolution into rain and removal of suspended particulates by precipitation (Bidleman, 1988; Koester and Hltes, 1992). The dry deposition of PCDD/Fs is a combination of both gas- and particle-phase fluxes. The dry deposition fluxes of PCDD/Fs are usually higher than the wet deposition fluxes, demonstrating that dry deposition is the major PCDD/Fs removal mechanism in the atmosphere (Wang *et al.*, 2010; Tseng *et al.*, 2014; Lee *et al.*, 2016; Zhu *et al.*, 2017a, b). The ambient temperature, rainfall, vapor pressure, and particle size will also affect the atmospheric deposition process (Wu *et al.*, 2009; Wang *et al.*, 2010; Chang *et al.*, 2004). This is usually evaluated using the total scavenging ratio (Stot), the ratio of the concentration in the precipitation to the concentration in the atmosphere.

Accumulation due to heavy industry in northern China, the winter cold in the north, and the use of coal for energy have led to significant coal combustion emissions and serious environmental pollution. In addition, the north temperate monsoon climate, characterized by a cold winter and dry, poor air flow, leads to accumulation of atmospheric pollution that is not easily disseminated, which exacerbates air pollution. Zhu *et al.* (2017) studied the PCDD/Fs, wet/dry deposition of total PCDD/Fs-WHO_{2005}-TEQ, the total deposition of total PCDD/Fs-WHO_{2005}-TEQ, and the scavenging ratio in the air of northern China's cities (Shijiazhuang and Harbin) over a period of time in 2014, and there have been few studies in recent years. Handan is an industrial city with pillar industries including steel, coal, and cement. All of these industries are characterized by high energy consumption and high air pollutant emission. Handan is also the hardest hit areas in terms of particulate matter (Ren *et al.*, 2004; Wang *et al.*, 2012; Zhao *et al.*, 2012). Kaifeng City is a key city designated by the state environmental protection administration for the prevention and control of national air pollution. With the acceleration of the industrialization process and the rapid development of tertiary industries, in recent years, there have been 222 newly built, reconstructed and expanded coal-fired boilers in urban areas, together with 386 original ones, totaling 608. These boilers burn 2 million tons of coal each year, generating 430,000 tons of slag and releasing 18,000 tons of soot and 15,000 tons of sulfur dioxide into the atmosphere. Therefore, two northern cities, Handan and Kaifeng, were selected for this study to investigate their wet/dry deposition of total PCDD/Fs-WHO_{2005}-TEQ, the total deposition of total PCDD/Fs-WHO_{2005}-TEQ, and the scavenging ratios in both 2016 and 2017, thus providing valuable current data for the air pollution in northern cities.

METHODS

Two cities, Handan and Kaifeng in Hebei and Henan province, China, respectively, were selected and evaluated in this study. The monthly average of $PM_{2.5}$ and PM_{10} concentrations, and related meteorological information including monthly average temperature and precipitation from 2016 and 2017 in Handan and Kaifeng were obtained from local air quality monitoring stations and the Weather Underground website.

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 adding both gas- and particle-phase deposition fluxes,
where
$F_{d,g}$: 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}) was as proposed by Shih *et al.* (2006). Dry deposition of gas-phase PCDD/Fs occurs mainly by 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) and used by Lee *et al.* (1996), was used in the current work to calculate the PCDD/F dry deposition flux contributed by its gas phase. Dry deposition of particle-phase PCDD/Fs is mainly achieved by gravitational settling, and the dry deposition velocity of particle-phase PCDD/Fs, $V_{d,g}$, can be calculated using Eq. (1).

Scavenging Ratios

In the case of slightly soluble trace organic compounds, 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; Ligocki *et al.*, 1985b; Cheruiyot *et al.*, 2015; Cheruiyot *et al.*, 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:

$$S_g = \frac{RT}{H}, \tag{2}$$

where
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:

$$S_g = \frac{C_{rain,dis}}{C_g}, \tag{3}$$

where
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 by:

$$S_p = \frac{C_{rain,particle}}{C_p}, \tag{4}$$

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

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

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

where
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 through precipitation (rainfall and cloud droplets). Precipitation scavenging accounts for the majority of removal of PCDD/Fs 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 the removal of suspended particulates through precipitation (Bidleman *et al.*, 1988; Koester and Hites *et al.*, 1992).

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

$$F_{w,T} = F_{w,dis} + F_{w,p} \tag{6}$$

$$F_{w,dis} = F_{rain,dis} + Rainfall \tag{7}$$

$$F_{w,p} = C_{rain,particle} + Rainfall, \tag{8}$$

where
$F_{w,T}$: 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

Wet Deposition

Wet deposition of PCDD/Fs is defined as the removal of both particle-phase and vapor-phase PCDD/Fs from the atmosphere through rainfall or other precipitation (Lee *et al.*, 2016). The monthly average of wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Handan and Kaifeng cities from 2016 to 2017 are presented in Figs. 1(a) and 1(b). Monthly rainfall in Handan and Kaifeng from 2016 to 2017 are shown in Table 1. In the case of Handan, the monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ is shown to decrease annually. In 2016, the monthly average wet deposition was 82.4 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, ranging from almost zero to 333.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$; the annual wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ was

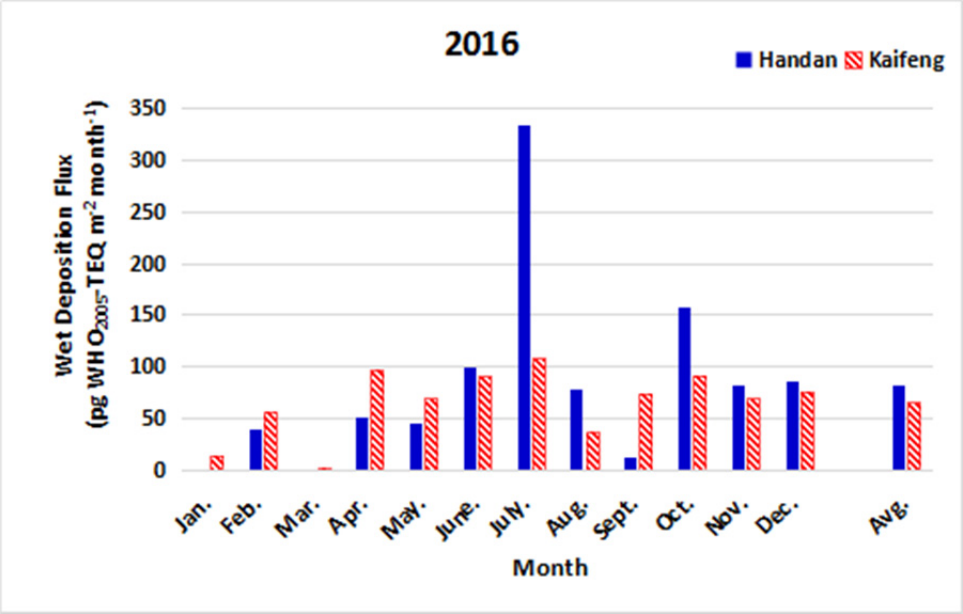

Fig. 1(a). Monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2016.

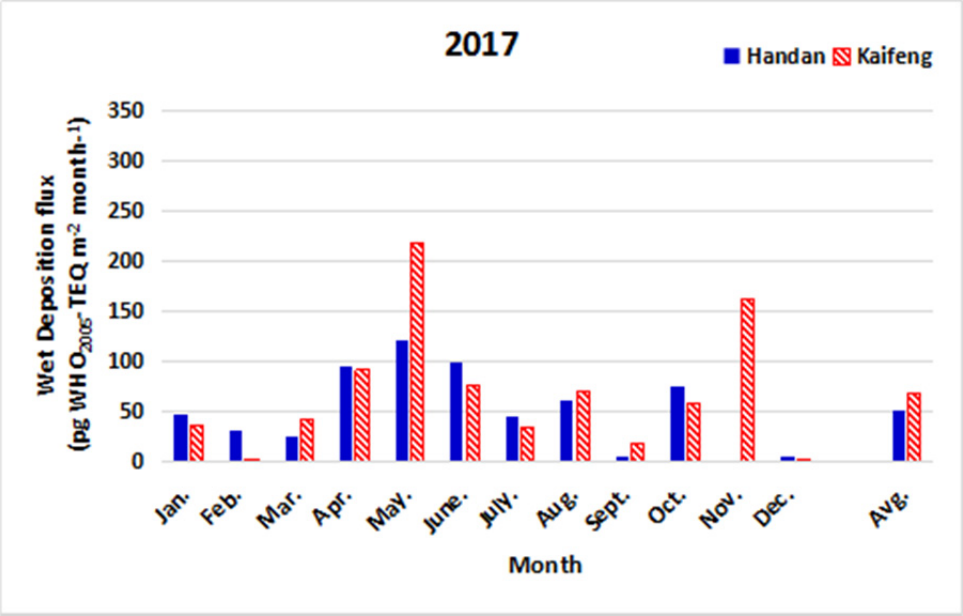

Fig. 1(b). Monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2017.

988.7 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was 1.49 times of magnitude higher than Lunbei City (397 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in the coastal area of central Taiwan according to Chen *et al.* (2017), who found in the same year (2017) the monthly average was 51.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with concentrations ranging between 1.1 and 122.1 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, where the annual wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 613.7 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was very similar to the value of 622 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in Shijiazhuang in 2014 (Zhu *et al.*, 2017). The impact of rainfall on wet deposition is significant. Under normal conditions, an increase in rainfall may cause a rise in wet deposition (Chen *et al.*, 2017). In 2016, the maximum wet deposition in Handan occurred in July (333.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), while the minimum occurred in March (almost zero), with an annual wet deposition flux of 988.7 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, and the rainfall was 414.6,0 and 786.4 mm, respectively. In 2017, the maximum wet deposition occurred in May (122.1 pg WHO_{2005}–TEQ m^{-2} $month^{-1}$) was approximately 111 orders of magnitude higher than the minimum occurring in November (1.1 WHO_{2005}-TEQ m^{-2} $month^{-1}$), with an annual wet deposition flux of 613.7 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ and rainfall of 37.6, 0.3, and 318.4 mm, respectively. From Table 1 we can clearly see that the maximum rainfall occurring in June (71 mm)

Table 1. Monthly Rainfall (mm) of Handan and Kaifeng from 2016 to 2017.

Month	Handan		Kaifeng	
	2016	2017	2016	2017
Jan.	0.4	7.1	3.1	12.2
Feb.	14.9	6.8	17.8	6.4
Mar.	0	8.3	0.3	13.0
Apr.	12.8	29.2	30.2	13.5
May	27.5	37.6	42.4	36.8
June	110.5	71	122.1	26.2
July	414.6	66.4	212.3	72.9
Aug.	107.3	56.5	79.5	140.0
Sept.	7.5	4.1	53.9	59.7
Oct.	62.8	29.9	72.1	42.9
Nov.	16.3	0.3	20.1	0.5
Dec.	11.8	1.2	18.8	0.8

did not occur in May (37.6 mm) in 2017, indicating that rainfall was not the only factor affecting wet deposition fluxes and that the concentration and PM size also affected the amount of the wet deposition (Chandra Suryani *et al.*, 2015). In 2016, the average wet deposition fluxes were 31.9, 170.8, 84.2, and 42.7 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively, which were associated with rainfall of 13.4, 210.8, 28.9 and 9.0 mm, respectively. In 2017, the average wet deposition fluxes were 80.5, 68.8, 27.4, and 28.0 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively, which were associated with rainfall of 25.0, 64.6, 11.4, and 5.0 mm, respectively.

In the case of Kaifeng, in 2016, the monthly average wet deposition flux was 65.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, ranging from 1.0 to 107.0 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$. The annual wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 783.4 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was a 43.0% order of magnitude higher than that of Taisi City (548 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in the coastal area of central Taiwan according to Chen *et al.*'s investigating in the same year. In 2017, the monthly average was 67.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with wet deposition fluxes between 2.3 and 216.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$. The annual wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 814.8 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was a 1.2 times of magnitude increased by that of Harbin (369 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in 2014 (Zhu *et al.*, 2017). As for the impact of rainfall on wet deposition, in 2016, the maximum wet deposition occurred in July (107.0 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) with monthly rainfall of 212.4 mm, which was far above the minimum occurring in March (1.0 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), with monthly rainfall of 0.3 mm and an annual wet deposition flux of 783.4 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ and annual rainfall of 672.8 mm. In 2017, the maximum wet deposition occurred in May (216.9 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) (with monthly rainfall of 36.8 mm), which was approximately 94.3 orders of magnitude higher than the minimum occurring in December (2.3 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) (0.8 mm). The annual wet deposition flux was 814.8 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, with annual rainfall of 424.9 mm. Similarly, from Table 1, we can see that the maximum rainfall in 2017 occurred in August (140.0 mm) and the minimum occurred in November (0.5 mm), which were different from May, in which the maximum value of wet deposition fluxes occurred (36.8 mm) and the minimum occurring in December (0.8 mm). This further proves that rainfall is an important factor affecting wet deposition fluxes but that it is not the only factor. In 2016, the average wet deposition fluxes were 55.9, 78.4, 78.3 and 48.6 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively, which were associated with rainfall of 24.3, 138.0, 48.7 and 13.2 mm, respectively. In 2017, the average wet deposition fluxes were 117.2, 60.7, 79.9 and 13.7 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively, which were associated with rainfall of 21.1, 79.7, 34.4 and 6.5 mm, respectively, since the summer season has more rainfall compared to the winter season, which has fewer rainy days (Lee *et al.*, 2016). In general, the summer wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ are greater than those for winter. The highest wet deposition fluxes occurred in the spring, but the maximum of rainfall occurred in the summer, which further proved that wet deposition is not only affected by rainfall, but also by factors such as particulate matter concentration, temperature, and wind speed (Wang *et al.*, 2010; Huang *et al.*, 2011).

Based on the above monthly rainfall as obtained from local air quality monitoring stations and monthly wet deposition fluxes, the concentrations of total PCDD/Fs-WHO_{2005}-TEQ in rain can be calculated by the wet deposition fluxes divided by rainfall intensity. The monthly average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain in Handan and Kaifeng from 2016 to 2017 are presented in Figs. 1(c) and Fig. 1(d). As the results show, in Handan, the monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain were ranged between 0 (March) and 7.38 (December) pg WHO_{2005}-TEQ L^{-1}, and averaged 2.87 pg WHO_{2005}-TEQ L^{-1} in 2016. This value was 8.3 times of magnitude higher than Kaohsiung in Taiwan in 2015, which averaged 0.307 pg WHO_{2005}-TEQ L^{-1} (Lee *et al.*, 2016). In 2017, the monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain were ranged between 0.67 (July) and 6.70 (January) pg WHO_{2005}-TEQ L^{-1}, and averaged 2.98 pg WHO_{2005}-TEQ L^{-1}, which was approximately 1.3 orders of magnitude higher than that of Harbin in 2014, which averaged 2.28 pg WHO_{2005}-TEQ L^{-1} (Zhu *et al.*, 2017). As for seasonal variations, the average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain were 2.38, 0.81, 2.91, and 4.74 pg WHO_{2005}-TEQ L^{-1}, in spring, summer, fall, and winter, respectively, in 2016. In 2017, the average concentrations were 3.22, 1.07, 2.40, and 5.60 pg WHO_{2005}-TEQ L^{-1}, in spring, summer, fall, and winter, respectively. It can be seen that the highest value occurred in the summer and the lowest occurred in the winter. This is related to the fact that summer is the rainy season.

In Kaifeng, the monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain were ranged between 0.47 (August) and 4.49 (January) pg WHO_{2005}-TEQ L^{-1}, with an average of 2.30 pg WHO_{2005}-TEQ L^{-1} in 2016.

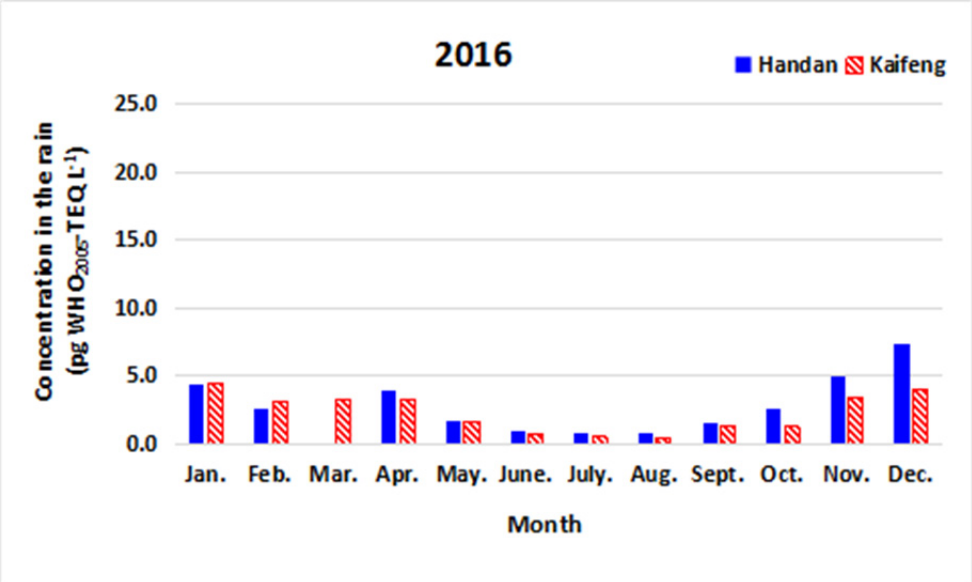

Fig. 1(c). Monthly average concentration of total PCDD/Fs-WHO_{2005}-TEQ in the Rain of Handan and Kaifeng in 2016.

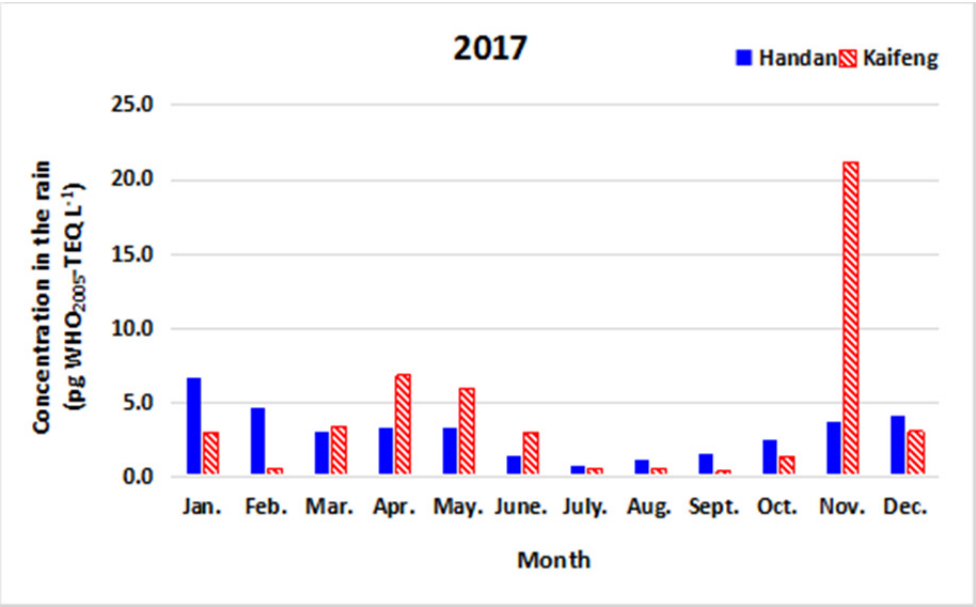

Fig. 1(d). Monthly average concentration of total PCDD/Fs-WHO_{2005}-TEQ in the Rain of Handan and Kaifeng in 2017.

This value was 13.7 times of magnitude higher than that in Meinong, Taiwan in 2015 (averaged 0.167 pg WHO_{2005}-TEQ L^{-1}) (Lee *et al.*, 2016). In 2017, the monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain were ranged between 0.49 (July) and 21.2 (November) pg WHO_{2005}-TEQ L^{-1}, with an average 4.15 pg WHO_{2005}-TEQ L^{-1}, which was 1.1 times higher than that of Shijiazhuang in 2014 (averaged 3.95 pg WHO_{2005}-TEQ L^{-1}) (Zhu *et al.*, 2017). As for seasonal variations, in 2016, the average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain were 2.30, 0.57, 1.61, and 3.68 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively, and those in 2017, were 5.37, 1.38, 7.67, and 2.18 pg WHO_{2005}-TEQ L^{-1}, respectively. In summer, more rain and lower PCDD/F concentrations in the atmosphere resulted in a lower PCDD/F concentration in the rain.

The total scavenging ratios (Stot) in Handan City and Kaifeng City from 2016 to 2017 are shown in Figs. 1(e) and 1(f). It can be clearly seen that the scavenging ratio (Stot) trend in the two cities in 2016 and 2017 was similar. In Handan, the scavenging ratio (Stot) of total PCDD/Fs-WHO_{2005}-TEQ ranges from 22970 (July) to 41350 (January) and from 21040 (July) to 41400 (January), with an average of 33040 and 32980 in 2016 and 2017, respectively. In Kaifeng, the scavenging ratio (Stot) of total PCDD/Fs-WHO_{2005}-TEQ ranged between 20120 (July) and 41110 (January) and ranged between 18980 (July) and 40730 (January), averaging 31750 and 31890 in 2016 and 2017, respectively. The

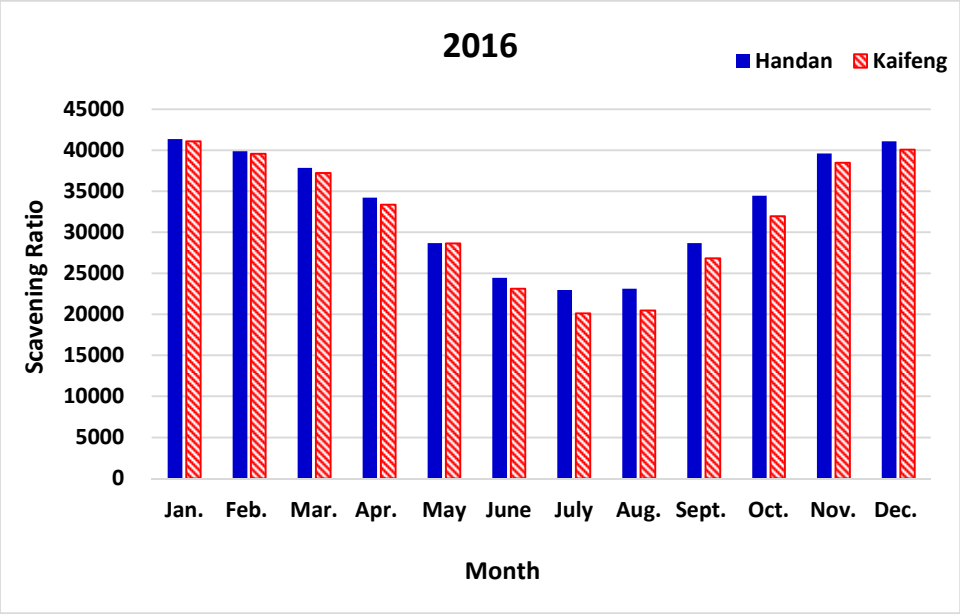

Fig. 1(e). Monthly average scavenging ratio of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2016.

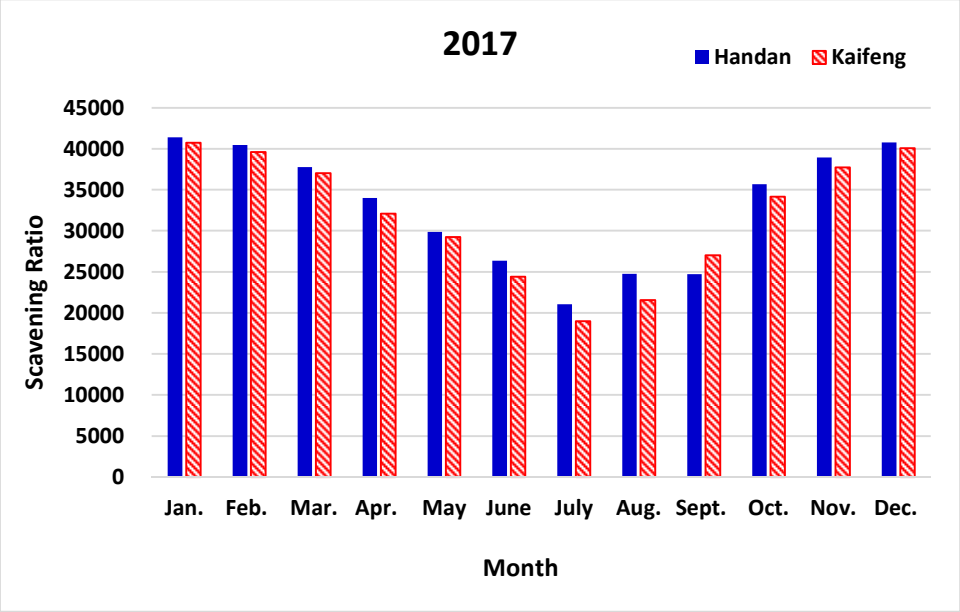

Fig. 1(f). Monthly average scavenging ratio of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2017.

maximum and minimum values for both cities occurred in January (winter) and July (summer), respectively. The above results were very similar to those of previous studies, where the total scavenging ratio (Stot) increased with decreased in temperature (Chen *et al.*, 2017).

Dry Deposition

The dry deposition fluxes of PCDD/Fs are contributed by both the gas and particle phases. The monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in the ambient air of Handan and Kaifeng from 2016 to 2017 are shown in Figs. 2(a) and 2(b), respectively. For Handan, in 2016, the monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged between 467 and 1959 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual dry deposition fluxes of 11590 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was approximately 2.9 times higher than those in Lunbei, Taiwan (3970 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in the same year (Chen *et al.*, 2017). The maximum monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ (1959 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) occurred in December at a level that was approximately 2.8 times higher than the minimum level (467 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which occurred in August. In 2017, the monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged between 489 and 1765 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual

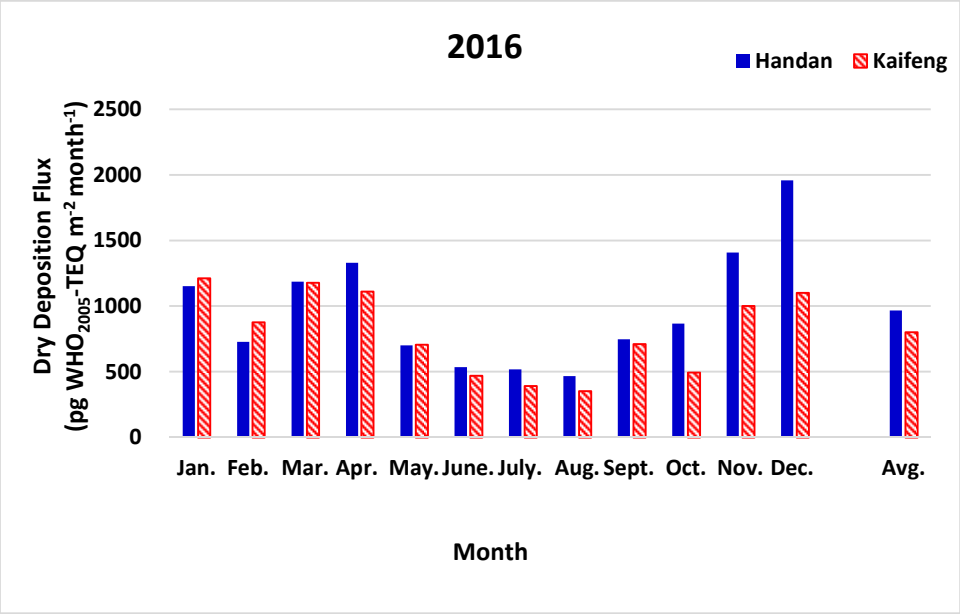

Fig. 2(a). Monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2016.

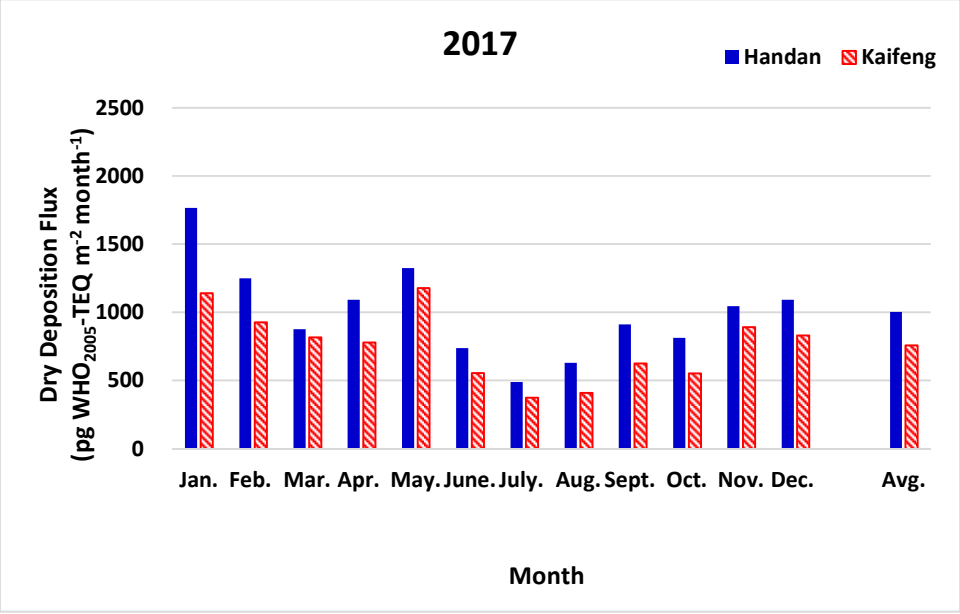

Fig. 2(b) Monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2017.

dry deposition flux of 12020 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was a 22.0% of magnitude decreased by that of Shijiazhuang (15400 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in 2014 (Zhu *et al.*, 2017). The maximum monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ (1765 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) occurred in January at a level that was approximately 3.6 times higher than the minimum level (489 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which occurred in July. In terms of seasonal variations, the average values of the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ were in the following order: winter (1324 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) > spring (1084 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) > fall (964 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) > summer (562 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). The value in summer (562 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) was 57.6% lower than the value in winter (1324 WHO_{2005}-TEQ m^{-2} $month^{-1}$). The maximum for both occurred in winter, and the minimum for both occurred in the summer. This is due to the fact that a higher temperature will cause a greater fraction of PCDD/Fs in the gas phase in summer and the gas phase scavenging ratio will be less than that of the particle phase (Chen *et al.*, 2017).

For Kaifeng, in 2016, the monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged from 354 to 1213 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual dry deposition flux of 9630 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$,

which was approximately 2.7 times higher than that in Taisi, Taiwan (3580 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in the same year (Chen *et al.*, 2017). The maximum monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ (1213 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) occurred in January, with a level that was approximately 3.4 times higher than the minimum level (354 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which occurred in August. In 2017, the monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged between 377 and 1140 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual dry deposition flux of 9094 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was a 1.1 times of magnitude higher than Harbin (8240 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in 2014 (Zhu *et al.*, 2017). The maximum monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ (1140 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) occurred in January, at a level that was approximately 3.0 times higher than the minimum level (377 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which occurred in July. Similar to the seasonal variations in Handan City, the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ were in the following order: (1016 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) > spring (963 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) > fall (714 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) > summer (427 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). The value in summer (427 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) was a 60.0% of magnitude lower than the value in winter (1016 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). It was also verified that as the temperature increased, the dry deposition fluxes were reduced. Similar to previous studies, in Taiwan, the seasonal variations in dry deposition fluxes were high in winter and low in summer (Wu *et al.*, 2009; Wang *et al.*, 2010; Mi *et al.*, 2012; Zhu *et al.*, 2017).

Total (Dry +Wet) Deposition

The total (dry + wet) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ were calculated by adding the dry and wet deposition fluxes, and shown in Figs. 3(a) and 3(b), respectively. Figs. 3(c) and 3(d) show the contribution fractions for the wet deposition and dry deposition fluxes, respectively, to the total deposition fluxes in Handan and Kaifeng for 2016-2017. For Handan, the monthly total (dry + wet) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged from 545 (August) to 2046 (December) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ as well as the annual 12580 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2016, which were approximately 2.9 times higher than those in Lunbei, Taiwan (4360 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in the same year (Chen *et al.*, 2017). The contribution fractions of dry deposition to the total deposition fluxes ranged from 60.8% to 100%, where the maximum occurred in March (100%), and the minimum occurred in July (60.8%). This is because the maximum and minimum rainfall for 2016 occurred in July (414.6 mm) and March (0 mm) respectively, resulting in a variation in the dry deposition contribution fraction to the total (dry + wet) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ. In 2017, the monthly total (dry + wet) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged from 534 (July) to 1813 (January) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual value of 12630 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was a 21.5% of magnitude decreased by that in Shijiazhuang (16100 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in 2014 (Zhu *et al.*, 2017). The contribution fraction of dry deposition to total deposition ranged from 88.0% to 99.9%; the maximum occurred in November (99.9%). and the minimum occurred in June (88.0%). In these two months, the rainfall was 71 mm and 0.3 mm, respectively. As to the seasonal variations in Handan, the averages for these two years were 1141, 682, 1020, and 1360 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and that of winter was approximately 2.0 times higher than that of summer. Previous studies have mentioned that the total dry deposition flux increases as the ambient air temperature decreases (Shih *et al.*, 2006; Huang *et al.*, 2011). When ambient air temperature decreases in the winter season, the PM_{10}, $PM_{2.5}$ and total PCDD/F concentration will increase and thus results in higher total deposition fluxes in January than that occur in August (Lee *et al.*, 2016).

For Kaifeng, the monthly total (dry + wet) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged from 391 (August) to 1227 (January) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual value of 10410 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2016, which was approximately 2.5 times higher than that in Taisi, Taiwan (4130 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in the same year (Chen *et al.*, 2017). The contribution fraction of dry deposition to the total deposition ranged from 78.5% to 99.9%, and averaged 90.9%; the maximum occurred in March (99.9%), and the minimum occurred in July (78.5%), and the maximum and minimum rainfall for 2016 occurred in July (212.4 mm) and March (0.25 mm) respectively. In 2017, the monthly total (dry + wet) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ ranged from 412 (July) to 1395 (May) pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, with an annual value of 9909 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was a 15% order magnitude higher than that in Harbin (8610 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) in 2014 (Zhu *et al.*, 2017). The contribution fraction of dry deposition to the total deposition ranged from 84.5% to 99.7% and averaged 91.8%; the maximum occurred in December (99.7%), and the minimum occurred in May (84.5%), and the rainfall for these two months was 36.8 mm and 0.8 mm, respectively. As to the seasonal variations, the average values for 2016 and 2017 of total PCDD/Fs-WHO_{2005}-TEQ deposition fluxes in Handan, were 1049, 497, 794, and 1047 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in spring, summer, fall, and winter, respectively, for which the values for spring and winter were approximately 2.1 orders of magnitude higher than that for summer.

Sensitivity Analysis

The parameters that can affect gas-particle partitioning will be the parameters influencing the dry deposition fluxes. The sensitivity analysis of this study is focused on the ambient temperature, the $PM_{2.5}$ concentration, the PM_{10} concentration, and the total PCDD/F concentration, which are sensitive variables for altering dry deposition fluxes. The sensitivity analyses conducted in this study in Handan and Kaifeng were dependent on the initial values of ambient air temperature = 13.5°C, $PM_{2.5}$ = 68.0 μg m^{-3}, PM_{10} =130.0 μg m^{-3}, total PCDD/F mass concentration = 1.76 pg m^{-3}, ambient air temperature = 13.7°C, $PM_{2.5}$ = 54.0 μg m^{-3}, PM_{10} = 107.0 μg m^{-3}, and total PCDD/F mass

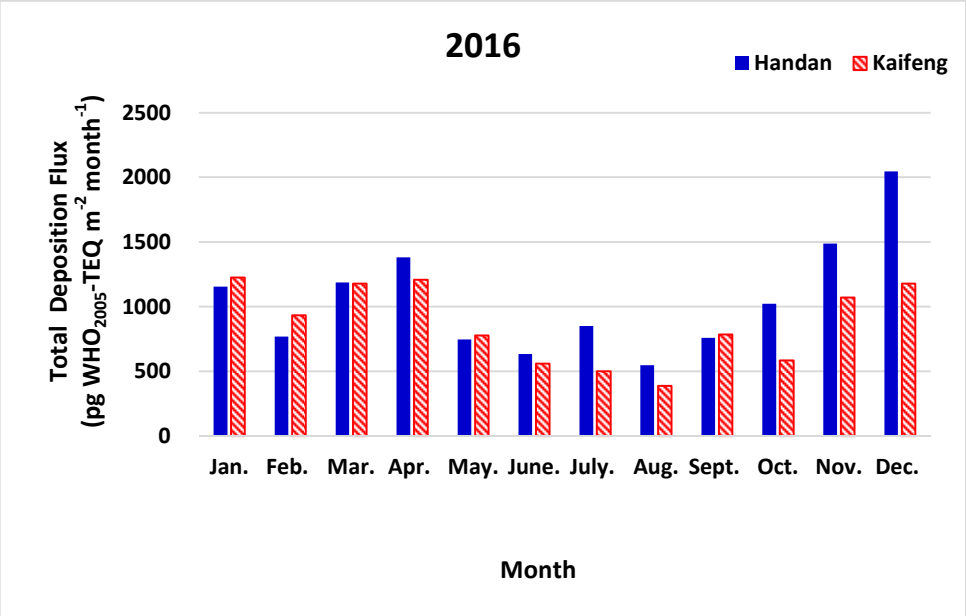

Fig. 3(a). Monthly average total (wet + dry) deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2016.

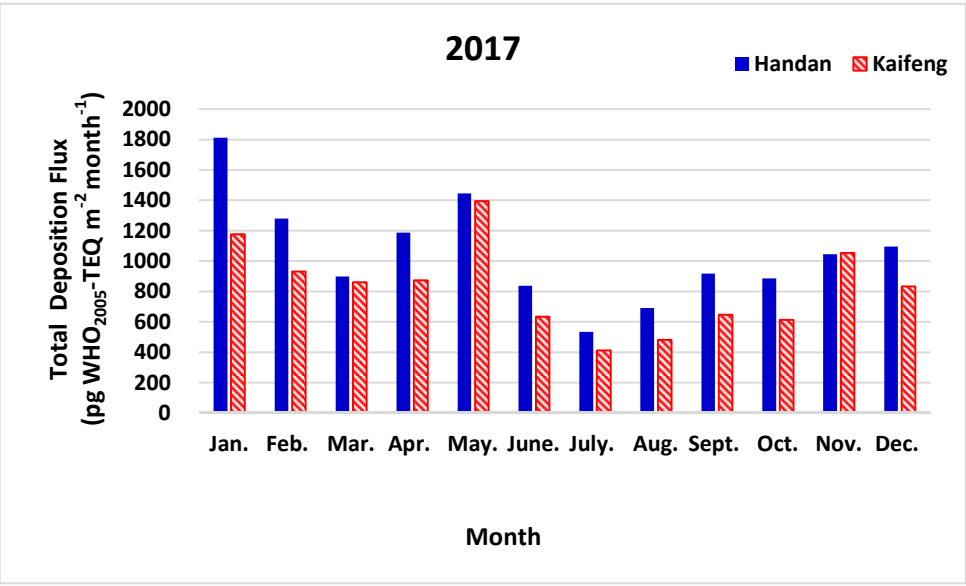

Fig. 3(b). Monthly average total (wet+dry) fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng in 2017.

concentration =1.75 pg m^{-3}, respectively. The parametric sensitivity for the dry deposition fluxes of Handan and Kaifeng are shown in Figs. 4(a) and 4(b), respectively,
where
P: initial value of parameters;
ΔP: increase or decrease in parameters;
S: predicted value in each of the parameters at the initial value;
ΔS: response in each of the parameters.

The $PM_{2.5}$ and PM_{10} concentrations have equivalently important sensibility to the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ. The effect of both PM_{10} concentration and PCDD/Fs can be divided into two parts: For PM_{10} concentration, whenΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –46.1%; when ΔP/P was changed from 0% to +50% and +80%, ΔS/S responded from 0% to +46.1% and from 0% to +73.7%, respectively. In terms of $PM_{2.5}$ concentration, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –53.8%; when ΔP/P was changed from 0% to +50% and +80%, ΔS/S responded from 0% to +47.8% and from 0% to +73.6%, respectively in Handan. In the case of Kaifeng, for PM_{10}, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –46.3%; when ΔP/P was changed from 0% to +50% and +80%, ΔS/S responded from 0% to +46.3% and from 0% to +74.1%, respectively. In terms of

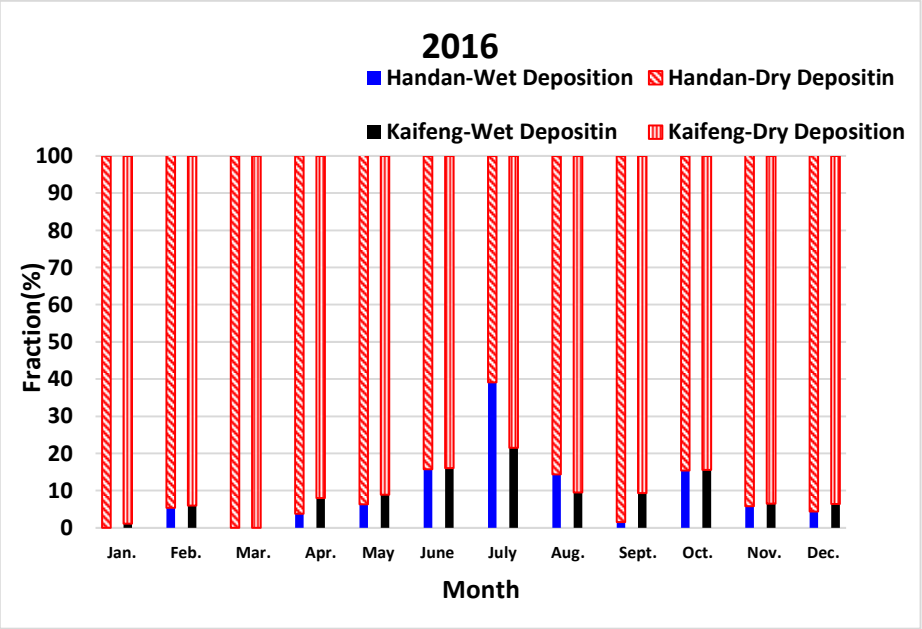

Fig. 3(c). The fraction of total deposition flux in total PCDD/Fs-WHO$_{2005}$-TEQ contributed by the dry and wet deposition, in Handan and Kaifeng in 2016.

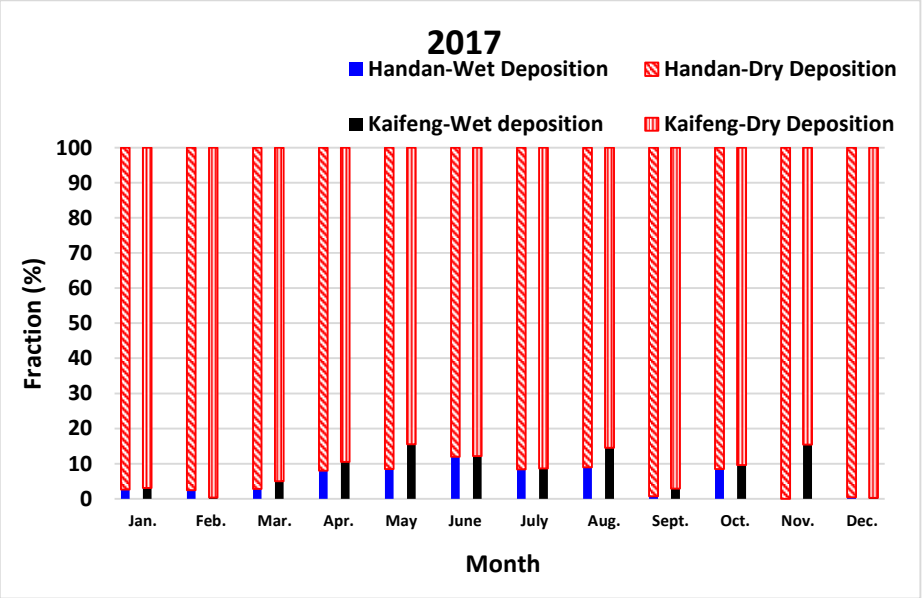

Fig. 3(d). The fraction of total deposition flux in total PCDD/Fs-WHO$_{2005}$-TEQ contributed by the dry and wet deposition, in Handan and Kaifeng in 2017.

$PM_{2.5}$ concentration, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –65.3%; when ΔP/P was changed from 0% to +50% and +80%, ΔS/S responded from 0% to +40.8% and from 0% to +53.6%, respectively The PM_{10} can reflect the status of particle phase PCDD/Fs, where at low concentrations, the effect of PM_{10} concentration on dry deposition is greater than that of $PM_{2.5}$, but as the concentration of PM increases, the effect of $PM_{2.5}$ concentration on dry deposition gradually increases. In addition, after the $PM_{2.5}$ concentration reaches a certain level, the effect of increasing concentration on the dry deposition flux decreases. This because when the $PM_{2.5}$ concentrations is further increased, its sensitivity is weaker than that incurred when the PM_{10} concentration is decreased.

The sensitivity analysis indicated that PCDD/Fs mass concentration has a significant, positive effect on the dry deposition fluxes of total PCDD/Fs-WHO$_{2005}$-TEQ. In the case of Handan, the effect of PCDD/Fs mass concentration can be divided into two parts: When ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –69.7%; when ΔP/P was changed from 0% to +50% and +100%, ΔS/S responded from 0% to +32.2% and +27.0%, respectively.

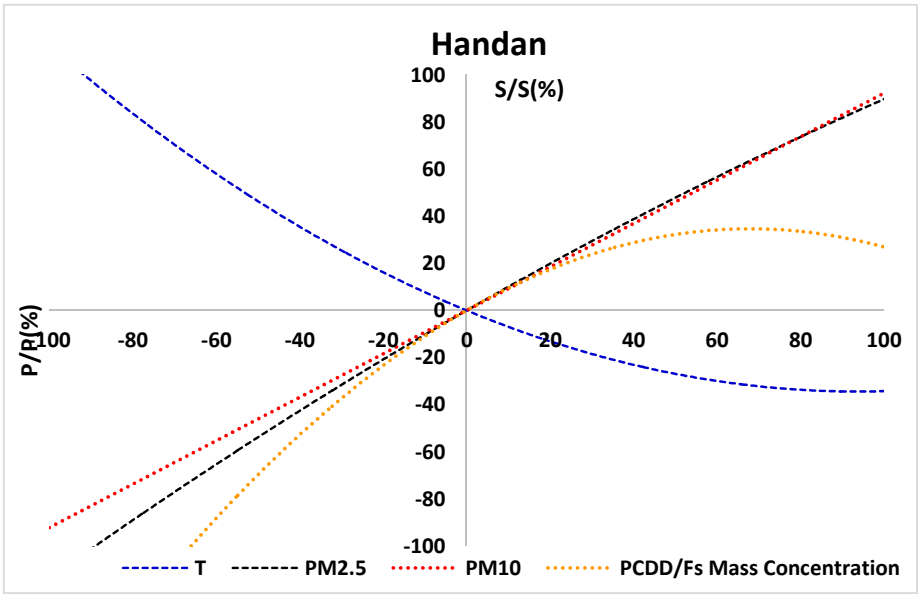

Fig. 4(a.) Sensitivity analysis for the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan from 2016 to 2017.

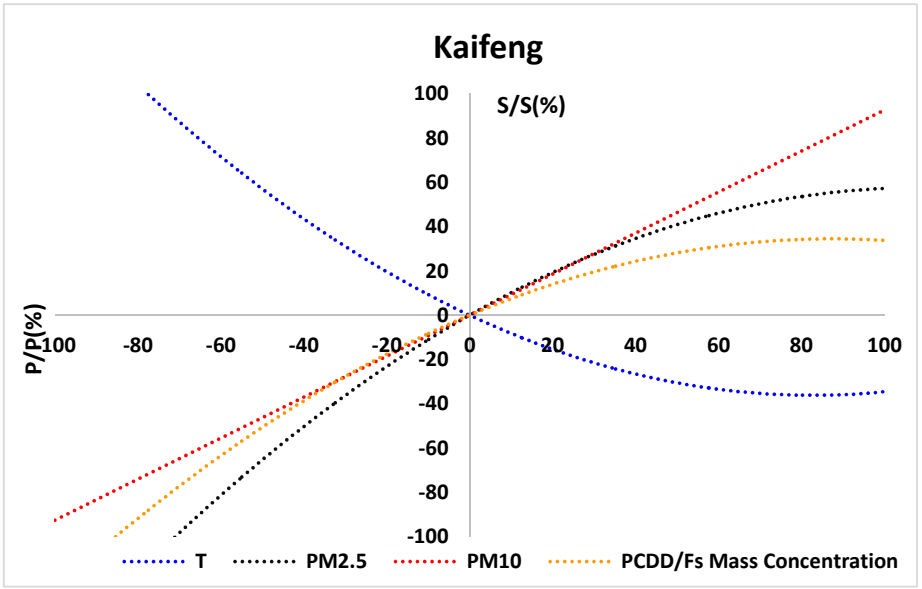

Fig. 4(b) Sensitivity analysis for the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Kaifeng from 2016 to 2017.

In Kaifeng, when ΔP/P was changed from 0% to –50%, ΔS/S responded from 0% to –50.6%; when ΔP/P was changed from 0% to +50% and +100%, ΔS/S responded from 0% to +28.1% and +33.8%, respectively. It can be seen that at low concentrations, the mass concentration of PCDD/Fs has a significant effect on the dry deposition fluxes, but the effect is weakened at high concentrations, indicating that the mass concentration of PCDD/Fs has a saturation value in the case of the dry deposition fluxes, so this factor is weaker than other factors.

Figs. 4(a) and 4(b) both show that air temperature has a negative correlation with the dry deposition of total PCDD/Fs-WHO_{2005}-TEQ. In the case of Handan, when ΔP/P was changed from –50% to +50%, ΔS/S responded from +46.4% to –26.9%. In Kaifeng, when ΔP/P was changed from –50% to +50%, ΔS/S responded from +57.0% to –30.5%. The temperature will affect the vapor pressure of PCDD/Fs and influence gas-particle partitioning because a higher temperature will cause a greater fraction of PCDD/Fs in the gas phase in the summer, and the gas phase scavenging ratio will be less than that of the particle phase (Chen *et al.*, 2017). As the air temperature increases, this will result in a greater amount of particle-bound PCDD/Fs evaporating to the gas phase, and the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ will decrease with increases in air temperature.

The above results suggest that the most sensitive parameters for the dry deposition of total PCDD/Fs-WHO_{2005}-TEQ are atmospheric $PM_{2.5}$ and PM_{10} concentrations, followed by the PCDD/Fs mass concentration and air temperature,

which had a negative correlation with the dry deposition of total PCDD/Fs-WHO_{2005}-TEQ.

CONCLUSION

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

1. In 2016, the annual wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan was 988.7 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was 1.3 times higher than that in Kaifeng (783.4 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). In 2017, the annual wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Handan was 613.7 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was approximately 24.7% lower than that in Kaifeng (814.8 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$).
2. In Handan, the monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain ranged between null and 7.38 and between 0.67 and 6.70 pg WHO_{2005}-TEQ L^{-1} in 2016 and 2017, respectively. In Kaifeng, the monthly average concentrations ranged between 0.47 and 4.49 and between 0.49 and 21.2 pg WHO_{2005}-TEQ L^{-1}, respectively.
3. The trend of the scavenging ratios (Stot) for the two cities in 2016 and 2017 was similar. The average total scavenging ratios of total PCDD/Fs-WHO_{2005}-TEQ in Handan were 33040 and 32980, respectively. The maximum and minimum values for both cities occurred in January in winter and July in summer, respectively.
4. In 2016, the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Handan was 11590 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was 1.2 times higher than that in Kaifeng (9630 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). In 2017, the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Handan was 12020 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was approximately 1.3 times higher than that in Kaifeng (9094 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). For the seasonal distribution, the average values of the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ were in the following order: winter > spring > fall > summer in both Handan and Kaifeng from 2016 to 2017. This is due to the fact that a higher temperature will cause a greater fraction of PCDD/Fs in the gas phase in summer, and the gas phase scavenging ratio will be less than that of the particle phase.
5. In 2016, the annual total (wet + dry) deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Handan was 12580 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was 1.2 times higher than that in Kaifeng (10410 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). In 2017, the annual total (wet + dry) deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Handan was 12630 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was approximately 1.3 times higher than that in Kaifeng (9900 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$). In addition, the average contribution fraction of dry deposition to the total deposition was 92.7% and 91.4% for Handan and Kaifeng in both 2016 and 2017, respectively.
6. The sensitivity analysis of the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Handan and Kaifeng showed that the $PM_{2.5}$ and PM_{10} concentrations were the most positively correlated sensitive factor. When ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +46.1% and +46.3%, respectively. The second positively correlated sensitive factor was the $PM_{2.5}$ concentration. When ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +47.8% and +40.8%. For PCDD/Fs mass concentration, when ΔP/P was changed from 0% to +50%, ΔS/S responded from 0% to +32.2% and +28.1%, respectively. This was followed by the atmospheric temperature, for which the effect was negatively correlated. When ΔP/P was changed from –50% to +50%, ΔS/S responded from +46.4% to –26.9% and +57.0% to –30.5%.

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