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Atmospheric Wet Deposition of PCDD/Fs in the Ambient Air

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ABSTRACT

Wet deposition is an important mechanism for removing air pollutants from the atmosphere. The total PCDD/Fs-WHO_{2005}-TEQ wet deposition from 2018–2020 was investigated for Beijing and Tianjin City in this study. In addition, the gas-particle partitioning of wet deposition, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, and the $PM_{2.5}$ concentration were also studied for Beijing and Tianjin City, respectively. Between 2018 to 2020, as a whole, the average seasonal variations in PCDD/F wet deposition fluxes in spring, summer, fall, and winter were 50.50, 41.47, 23.03 and 16.76 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively, while in Tianjin, they were 35.30, 42.40, 13.37, and 14.77 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. Because the two cities have more rainfall in spring and summer than they do in fall and winter, rainfall has a significant influence on the wet deposition flux. In regard to PCDD/Fs-WHO_{2005}-TEQ in the rain, in Beijing, the average total PCDD/Fs-WHO_{2005}-TEQ concentration in the rain in spring, summer, fall, and winter were 1.70, 0.39, 1.42, and 1.52 pg WHO_{2005}-TEQ L^{-1}, respectively, while those in Tianjin, were 1.73, 0.42, 1.35, and 1.88 pg WHO_{2005}-TEQ L^{-1}, respectively. The above results show that the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain are significantly lower in summer, which is mainly due to the fact that in summer, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the air are lower, and the proportion of the gas phase is increased. When the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the air are washed away by a heavy rainfall, the PCDD/F concentrations are diluted.

Keywords: $PM_{2.5}$, Wet deposition, PCDD/Fs, Beijing, Tianjin

1 INTRODUCTION

Persistent organic pollutants (POPs) have characteristics that include persistence, toxicity, and biological accumulation and have a great impact on air quality (Hao *et al*, 2021; Sari *et al*, 2021). As a sub-group of persistent organic pollutants, PCDD/Fs (polydibenzo-*p*-dioxins and polydibenzofurans) are high similar in terms of structure and properties (Schecter *et al*., 2006). The toxicity of PCDD/Fs is estimated using the toxicity equivalent (TEQ) of 17 biotoxic homologues. They have high chemical stability and are difficult to be degraded in the atmosphere, soil, and other environmental media (Lee *et al*., 2016). PCDDs are derivatives of polydibenzo-*p*-dioxins; there are 75 compounds in total, seven of which have strong biotoxicity, while PCDFs, derivatives of polydibenzofuran, have 135 compounds, ten of which are toxic. Studies have shown that the long-term, stable existence of PCDD/Fs in the human body through the food chain will not only cause irreversible mutations, carcinogenesis, and teratogenicity, but will also affect fetal development (Vilavert *et al*., 2015). The sources of PCDD/Fs mainly include both natural and artificial sources. Industrial activities account for the largest proportion of PCDD/F human sources (Hagenmaier *et al*., 1994; Alcock *et al*., 1998). Waste incineration, the pharmaceutical industry,

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and pesticide production processes containing aromatic hydrocarbons and organochlorine all lead to the formation of PCDD/Fs (Ma *et al*., 2019; Zhan *et al*., 2019; Qiu *et al*., 2020). The exhaust gas from automobiles and motorcycles also contains PCDD/Fs (Chen *et al*., 2020). Studies show that the retarding fuel injection timing will lead to increased emissions, and the emissions of PCDD/Fs of heavy-duty diesel engines are different at different exhaust gas recirculation rates (Chen *et al*., 2019; Zhao *et al*., 2019).

The atmospheric environment is the main PCDD/F migration and transformation route. In addition, atmospheric deposition, which involves both dry and wet deposition, is one of the important ways to remove POPs (Redfern *et al*., 2017). Among them, wet deposition has a significant effect on removing PCDD/Fs in the particle and gas phases in the atmosphere (Moon *et al.*, 2005; Melymuk *et al*., 2011). In the case of atmospheric wet deposition, precipitation in the atmosphere such as rain, snow, or other forms of water vapor condensates can remove PCDD/Fs (Lohmann and Jones, 1998). Wet deposition flux is related to the rainfall intensity, temperature, and concentration of environmental pollutants (Kaupp and McLachlan, 1998). The concentrations of total PCDD/Fs-WHO_{2005}-TEQ in rain can be calculated from rainfall intensity and wet deposition flux.

PCDD/Fs produced by combustion exists in the atmosphere in the form of gas and particle phases (Li *et al*., 2008). The results show that the distribution behavior of the gas and particle phases is the main factor that determines the environmental trend of PCDD/Fs. Similarly, some studies have shown that PCDD/Fs in the gas phase exist in small amounts due to the degradation reaction in the atmospheric environment, while PCDD/Fs in the particulate phase mainly enter the ecosystem through the atmospheric environment. The gas-particle partition of PCDD/Fs in the ambient air is related to the ambient temperature, humidity, air pressure, and the compounds themselves (Wang *et al.,* 2010; Cheruiyot *et al*., 2015). Generally speaking, the gas phase ratio of PCDD/Fs in summer is much higher than that in winter, and the gas phase ratio of low-molecular homologues is higher than that of high molecular homologues. This is mainly determined by the cold vapor pressure of the compound. The undercooled vapor pressure of low-chlorinated dioxins is higher than that of high-chlorinated dioxins. Therefore, low-chlorinated dioxins are more likely to accumulate in the gas phase. When the temperature drops, part of the gas phase PCDD/Fs is exchanged and transferred to the particulate phase (Oh *et al*., 2001).

Particulate matter (PM) is an aerosol, which refers to a mixture of solid particles and liquid droplets (Ghosh *et al*., 2014). In many cities, it has become the primary pollutant affecting air quality and which has attracted extensive attention from people and governments around the world (Querol *et al*., 2004; Zavala *et al*., 2013; Wang *et al*., 2017). The biological effects of PM are mainly related to particle size, which can be divided into coarse particulate matter (PM_{10}), fine particulate matter ($PM_{2.5}$), and ultrafine particulate matter (UFPM) according to the aerodynamic diameter (Chow *et al*., 2015; Lu *et al*., 2016). The main sources include motor vehicle exhaust, dust, outdoor pollution from industrial and agricultural production processes, and indoor pollution from the burning of fuels as firewood and coal (Bilos *et al*., 2001; Kong *et al*., 2014; Alghamdi *et al*., 2015). $PM_{2.5}$ has small diameter, significant activity, strong penetrating power, can easily carry toxic and harmful substances, and is more harmful to human health than PM_{10} (Dai *et al*., 2015). In addition, $PM_{2.5}$ is an important factor causing haze, and regional air pollution seriously affects environmental air quality (Li *et al*., 2015). Studied have shown a strong link between air pollution from coal burning and lung cancer mortality (He *et al*., 1991). A positive correlation between $PM_{2.5}$ and first hospital diagnoses for Alzhemimer's disease and Parkinson's disease dementia has also been found (Kioumourtzoglou *et al*., 2016).

In this work, the atmospheric PCDD/F wet deposition, the gas-particle PCDD/F partition in wet deposition, the concentration of total PCDD/Fs-WHO_{2005}-TEQ in the rain, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content and $PM_{2.5}$ concentration in Beijing (39°56'N, 116°20'E) and Tianjin (39°13'N, 117°2'E) in northern China were investigated for the period 2018–2020.

2 METHODS

The monthly mean concentrations of both $PM_{2.5}$ and PM_{10}, and the monthly temperature and precipitation in both cities were obtained from local air quality monitoring stations, the local weather bureau and the Statistics Yearbook of China.

The total PCDD/F concentrations were simulated using a regression analysis of the PM_{10} concentration. Tang *et al*. (2017) reported that there is a high correlation between PM_{10} values and total PCDD/F mass concentrations. Their research included the following two regression equations:

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

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

where Y_1, Y_2 represent the total PCDD/F concentration (pg m^{-3}), and x represents the PM_{10} concentration in the ambient air (μg m^{-3}).

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

2.1 Atmospheric Wet Deposition of PCDD/Fs

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 through wet deposition (Huang *et al*., 2011). The wet deposition flux of PCDD/Fs is a combination of both vapor dissolution into rain and removal of suspended particulates through precipitation (Koester and Hites, 1992)

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

$$F_{w,T} = F_{w,dis} + F_{w,p} \quad (3)$$

$$F_{w,dis} = C_{rain,dis} \times \text{Rainfall} \quad (4)$$

$$F_{w,p} = C_{rain,particle} \times \text{Rainfall}, \quad (5)$$

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

2.2 Gas-Particle Partitioning

The PCDD/F concentrations in the gas and particle phases, respectively, were calculated using a gas-particle partitioning model as shown in Eq. (6) (Yamasaki *et al*., 1982; Pankow, 1987; Pankow and Bidleman, 1992):

$$K_p = \frac{F/TSP}{A}, \quad (6)$$

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

Plotting log K_p against the logarithm of the subcooled liquid vapor pressure, P_L^0, gives

$$\log K_p = m_r \times \log P_L^0 + b_r, \quad (7)$$

where P_L^0 is the subcooled liquid vapor pressure (Pa); m_r is the cited slope, and b_r is the cited y-intercept.

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

A previous study correlated the P_L^0 of PCDD/Fs with gas chromatographic retention indexes (GC-RI) on a nonpolar (DB-5) GC-column using p,p'-DDT as a reference standard. The correlation

has been re-developed as follows (Hung *et al*., 2002):

$$\log P_L^0 = \frac{-1.34(RI)}{T} + 1.67\times10^{-3}(RI) - \frac{1320}{T} + 8.087 \quad (8)$$

where *RI* represents the gas chromatographic retention indexes (Donnelly *et al*., 1987), *T* is the ambient temperature (K).

3 RESULTS AND DISCUSSION

3.1 Wet Deposition Flux of PCDD/Fs

From 2018–2020, the monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Beijing and Tianjin in the ambient air are shown in Figs. 1 (a) and 1(b). And wet deposition of total PCDD/Fs-WHO_{2005}-TEQ in some countries and regions in the world is shown in Table 1. The monthly rainfall in Beijing and Tianjin from 2018–2020 is shown in Table 2.

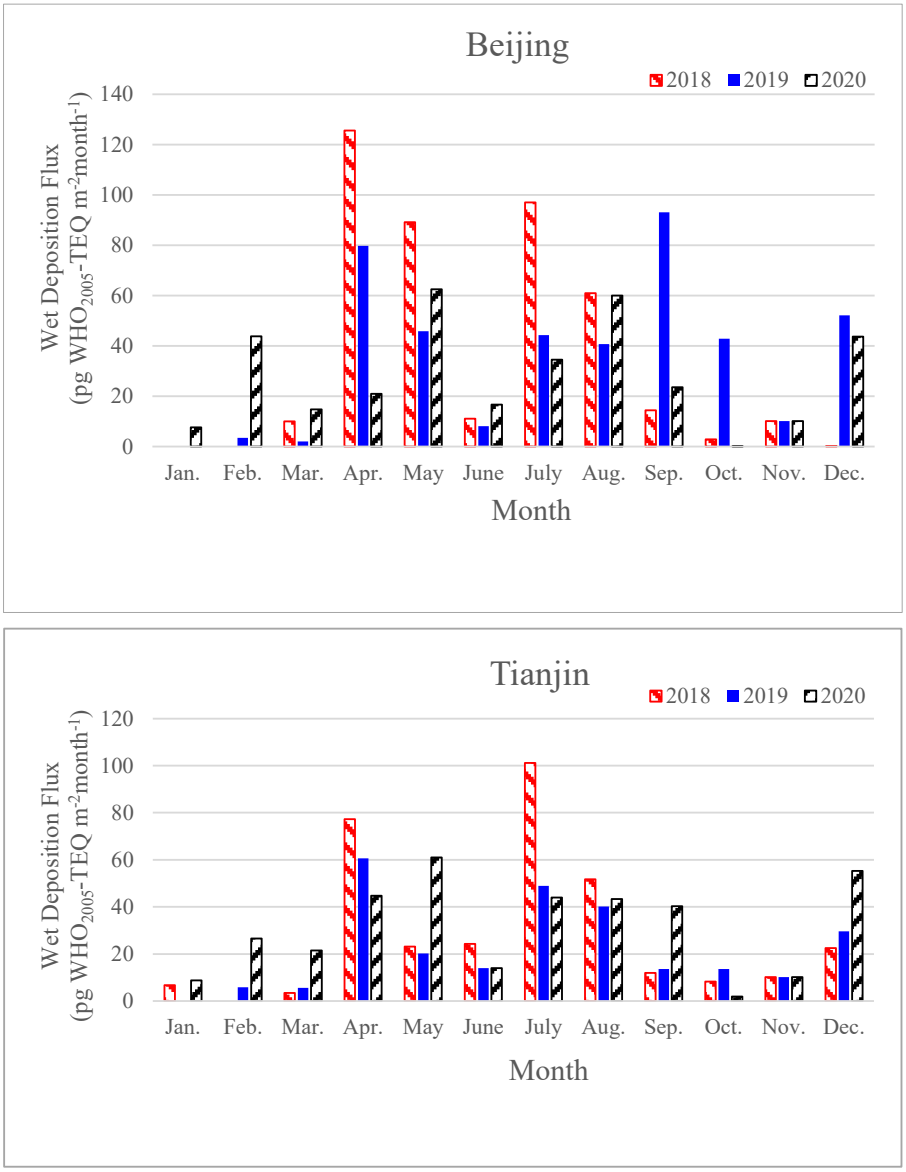

Fig. 1. Monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Beijing and Tianjin in 2018, 2019, and 2020, respectively.

Table 1. Wet deposition of total PCDD/Fs-WHO_{2005}-TEQ in some countries and regions in the world.

Country	City	Wet deposition (pg WHO_{2005}-TEQ m^{-2} $month^{-1}$)	Reference
USA	Houston	360.0	Correa *et al*., 2006
Canada	Burlington	18.5	Backus *et al*., 2004
China	Harbin	30.8	Zhu *et al*, 2017b
China	Shijiazhuang	51.8	Zhu *et al*, 2017b
China	Guangzhou	47.5	Zhu *et al*, 2017a
China	Nanjing	103.3	Zhu *et al*, 2017a
China	Wuhan	128.2	Wang *et al*, 2018
China	Bengbu	127.1	Wang *et al*, 2018
China	Beijing	32.8	This study, 2021
China	Tianjin	27.1	This study, 2021

Table 2. Monthly Rainfall (mm) of Beijing and Tianjin from 2018 to 2020.

Month	Beijing			Tianjin		
	2018	2019	2020	2018	2019	2020
Jan.	0.0	0.0	5.1	3.5	0.0	3.7
Feb.	0.0	2.1	34.6	0.0	2.5	19.7
Mar.	3.8	1	11.6	1.4	2.8	13.5
Apr.	53.1	39.4	15.2	37.1	29.3	27.2
May	7.9	35.7	65.4	20.3	15.3	62.3
June	59.8	15.9	29.1	44.8	26.3	21.2
July	298.8	128.1	98.5	304	160.4	113.5
Aug.	185	151.2	200	153.8	125.6	135.7
Sept.	26	139.2	63.5	18.6	17	84.9
Oct.	2.6	33.9	0.1	6.5	9.1	1.3
Nov.	2.4	28.6	30.9	10.1	14.5	32.8
Dec.	0.1	4.7	0.7	2.1	7.1	0.1

In 2018, the monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Beijing ranged from 0.0 to 125.6 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 421.3 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. In 2019, those values ranged from 0.0 to 93.09 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 422.2 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. In 2020, those values ranged from 0.13 to 62.50 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 338.3 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. Rainfall has a great influence on the wet deposition flux of PCDD/Fs. Generally, an increase in rainfall will lead to an increase in the wet deposition flux. In 2018, the highest wet deposition flux in Beijing occurred in April (125.6 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), with 51.33 mm of rainfall. The lowest value occurred in January and February (almost zero), when there both cities had no rainfall. The month with the largest rainfall in that year was July (298.9 mm). In 2019, the highest wet deposition flux of PCDD/Fs occurred in September (93.09 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), when the monthly rainfall was 139.2 mm. The lowest value occurred in January, at nearly zero. The month with the highest rainfall in the year was August (151.2mm). In 2020, the highest wet deposition flux of PCDD/Fs occurred in May (62.50 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), when the monthly rainfall was 65.4 mm. The lowest value occurred in October (0.133 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), with 0.1 mm of rainfall. The month with the highest rainfall in the year is August (200 mm). The above results indicate that the rainfall intensity, the PM size, and the PCDD/F concentration were key factors affecting the wet deposition flux of PCDD/Fs. In terms of the seasonal variations, the four seasons were defined as spring (March, April, May), summer (June, July, August), fall (September, October, November), and winter (January, February and December). In 2018, the average wet deposition fluxes of PCDD/Fs in spring, summer, fall, and winter were 74.91, 56.33, 9.13, 0.06 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. In 2019, the average wet deposition fluxes were 42.51, 31.03, 48.68,

18.52 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively, and in 2020, the average wet deposition fluxes were 32.73, 37.06, 11.28, 31.69 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively.

As for Tianjin, the monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in 2018 ranged from 0.0 to 101.20 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 340.8 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. In 2019, the values ranged from 0 to 60.60 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 262.6 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. In 2020, the values ranged from 1.96 to 61.03 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 371.8 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. In terms of the influence of rainfall on wet deposition, in 2018, the highest wet deposition flux in Tianjin occurred in July (101.2 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), with 304 mm of rainfall. The lowest value occurred in February (almost zero), with zero rainfall. The rainfall in that month was the lowest of the year. In 2019, the highest wet deposition flux occurred in April (60.60 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), when the monthly rainfall was 29.3 mm. The lowest value occurred in January (almost zero), with zero rainfall. The month with the highest rainfall in the year was July (160.4 mm). In 2020, the highest wet deposition flux occurred in May (61.04 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which was 62.3 mm of monthly rainfall. The lowest value occurred in October (1.96 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which was 1.3 mm of rainfall. The month with the highest rainfall in the year was August (135.7 mm). The above results demonstrated that rainfall has a great influence on wet deposition flux of PCDD/Fs, but it is not the only factor. With regard to seasonal variations, in 2018, the average wet deposition fluxes in spring, summer, fall, and winter were 34.62, 59.07, 10.16, 9.75 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. In 2019, they were 28.83, 34.36, 12.48, 11.87 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. In 2020, they were 42.44, 33.78, 17.46, 22.69 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively.

By analyzing the data from the two cities, it can be seen that, in Beijing from 2018 to 2020, the average wet deposition fluxes of PCDD/Fs in spring, summer, fall, and winter were 50.50, 41.47, 23.03 and 16.76 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The highest value was found in spring, followed by summer and fall, and the lowest value was found in winter. However, in Tianjin, from 2018 to 2020, the seasonal average wet deposition fluxes of PCDD/Fs were 35.30, 42.40, 13.37 and 14.77 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The highest value was found in summer, followed by spring and winter, and the lowest value was found in fall. This is because there are many factors affecting the wet deposition flux of PCDD/Fs, such as rainfall intensity, PCDD/F concentration, $PM_{2.5}$ concentration, temperature, and wind speed, all of which will affect the results of PCDD/F wet deposition flux (Suryani *et al*., 2015).

Table 1 showed the wet deposition of total PCDD/Fs-WHO_{2005}-TEQ in some countries and regions in the world. The area with more precipitation did have more amount of PCDD/F wet deposition.

3.2 Rainfall Intensity

The rainfall intensity of Beijing and Tianjin in 2018, 2019 and 2020 are shown in Table 2. As for Beijing, in 2018, the rainfall ranged between 0.0 and 298.8 mm and averaged 53.29 mm; in 2019, it ranged from 0.0 to 151.2 mm and averaged 48.32 mm, and in 2020, it ranged between 0.1 and 200.0 mm, with an average of 46.23 mm. In 2018, the highest rainfall occurred in July (298.8 mm) and the lowest values occurred in January (almost zero) and February (almost zero). In both 2019 and 2020, the highest rainfall occurred in August, where the values were 151.2 and 200.0 mm, respectively. In 2019 and 2020, the lowest rainfall occurred in January and October, respectively, when the rainfall was almost zero.

For Tianjin, in 2018, the rainfall ranged between 0.0 and 153.8 mm and averaged 50.18 mm; in 2019, it ranged from 0.0 to 160.4 mm and averaged 34.16 mm; while in 2020, it ranged between 0.1 and 135.7 mm and averaged 42.99 mm. In 2018, the highest rainfall occurred in August (153.8 mm), and the lowest value occurred in February (almost zero). In 2019, the highest rainfall occurred in July (160.4 mm), and the lowest value occurred in January (almost zero). In 2020, the highest rainfall occurred in August (135.7 mm) and the lowest value occurred in December (0.1 mm).

With regard to seasonal variations, for Beijing, in 2018, the rainfall in spring, summer, fall and winter were 21.6, 181.2, 47.00, and 40.67 mm respectively; in 2019, in that order, it was 25.37, 98.40, 67.23, 2.27 mm, respectively, and in 2020, it was were 30.73, 109.2, 31.50, 13.47 mm.,

respectively. The results shown that in Tianjin, during 2018, the rainfall in spring, summer, fall, and winter was 19.60, 167.5, 11.87, 1.87 mm, respectively. In 2019 it was 15.8, 104.1, 13.53, 3.20 mm, respectively, and in 2020 it was 34.33, 90.13, 39.67, 7.83 mm, respectively. On the whole, in Beijing, the average rainfall in spring, summer, fall, and winter from 2018 to 2020 was 25.90, 129.6, 48.58, and 18.74 mm, respectively, while in Tianjin, it was 23.24, 93.13, 21.69, and 4.3 mm respectively. In general, the rainfall in both cities showed obvious seasonal changes, which were all the highest in summer and lowest in winter. According to the previous discussion on wet deposition flux, the lowest value usually occurs in winter, which indicates that the influence of precipitation and number of rainy days on PCDD/F wet deposition flux varies significantly (Lee *et al.*, 2016).

Beijing's climate is a typical semi-humid continental monsoon climate in the northern temperate zone, with summers characterized by high temperatures and rain and winters that are cold and dry. The seasonal distribution of precipitation is very uneven. In 2020, more than 59.0% of the precipitation was concentrated in June, July, and August. Specifically, 17.8% of the annual rainfall was in July, and 36.1% was in August. Tianjin is located in the north temperate zone on the east coast of the Eurasian continent in the middle latitudes. It is mainly dominated by monsoon circulation, and it is a region where the East Asian monsoon is prevalent. It is in the warm temperate zone characterized by a semi-humid monsoon climate. Because it is close to Bohai Bay, the influence of the marine climate on Tianjin is obvious. The main climate features are four distinct seasons: a windy spring with drought and little rain, a hot summer, during which the rain is concentrated, a cool, moderate fall and a cold dry winter with little snow.

3.3 Gas-Particle Partitioning of Wet Deposition

Wet deposition refers to the process in which precipitation such as rain, snow and other forms of water vapor in the atmosphere play a role in removing air pollutants. The gas-particle partitioning of PCDD/Fs is an important factor affecting atmospheric dry and wet deposition. Table 3 shows the monthly gas-particle partition contribution on wet deposition in the ambient air in Beijing and Tianjin for the period 2018–2020.

In 2018, 2019 and 2020, in Beijing, the monthly contribution in the particle phase ranged between 73.5% and ~100%, 73.6% and ~100% and 75.5% and 99.7%, respectively and averaged 91.4%. In Tianjin, the monthly contributions ranged between 71.4% and 100%, 73.5% and ~100% and 74.5% and 99.7%, respectively and averaged 90.8%. In terms of seasonal variations, during 2018–2020, in Beijing, the seasonal contribution fractions on PCDD/F wet deposition flux by the gas phase were 2.8%, 22.4%, 5.4% and 0.2% in spring, summer, fall, and winter, respectively, and those for Tianjin were 3.4%, 2.5%, 5.5% and 0.2%, respectively. The above results revealed that the higher temperature in summer (21.3°C–27.8°C) led to a higher seasonal percentage of PCDD/F wet deposition flux in the gas phase, while the lower temperature (between –7.0°C and 0.7°C) in winter resulted in a lower percentage contribution by the gas phase.

3.4 Total PCDD/Fs-WHO_{2005}-TEQ Concentration in the Rain

The monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain can be calculated from the rainfall and wet deposition fluxes. The monthly average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain from 2018–2020 in Beijing and Tianjin are shown in Figs. 2(a) and 2(b), respectively. It can be seen that the monthly average total PCDD/Fs-WHO_{2005}-TEQ concentration in the rain in Beijing in 2018 ranged between 0.32 and 4.23 pg WHO_{2005}-TEQ L^{-1} and averaged 1.52 pg WHO_{2005}-TEQ L^{-1}, where July had the lowest (0.32 pg WHO_{2005}-TEQ L^{-1}), and November had the highest (4.23pg WHO_{2005}-TEQ L^{-1}). As to the seasonal variations, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were 2.13, 0.40, 1.97, and 1.60 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. During 2019, the monthly average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain ranged between 0.26 and 2.02 pg WHO_{2005}-TEQ L^{-1} and averaged 1.25 pg WHO_{2005}-TEQ L^{-1}, where August had the lowest (0.26 pg WHO_{2005}-TEQ L^{-1}), and April had the highest (2.02 pg WHO_{2005}-TEQ L^{-1}). As to the seasonal variations, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were 1.76, 0.37, 1.25, and 1.61 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. During 2020, the monthly average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain ranged between 0.30 and 1.48 pg WHO_{2005}-TEQ L^{-1} and averaged 1.00 pg WHO_{2005}-TEQ L^{-1}, where August had the lowest (0.30 pg WHO_{2005}-TEQ L^{-1}), and

Table 3. Monthly gas-particle partition of wet deposition fluxes in the ambient air in Beijing and Tianjin in 2018, 2019, and 2020, respectively.

	City	Beijing		Tianjin	
Year	Month	Gas(%)	Particle (%)	Gas(%)	Particle (%)
2018	Jan.	0.0	~100	0.2	99.8
	Feb.	0.0	~100	0.0	~100
	Mar.	1.1	98.9	1.2	98.8
	Apr.	3.1	96.9	3.6	96.4
	May	8.3	91.7	10.1	89.9
	June	20.3	79.7	22.0	78.0
	July	26.5	73.5	28.7	71.3
	Aug.	25.2	74.8	26.7	73.3
	Sep.	13.2	86.8	13.9	86.1
	Oct.	4.2	95.8	4.9	95.2
	Nov.	3.6	96.4	1.3	98.7
	Dec.	0.3	99.7	0.6	99.4
2019	Jan.	0.0	~100	0.0	100
	Feb.	0.4	99.6	0.4	99.7
	Mar.	1.5	98.5	1.9	98.1
	Apr.	2.9	97.1	3.2	96.8
	May	8.4	91.6	9.7	90.3
	June	19.6	80.4	20.6	79.4
	July	26.4	73.6	26.5	73.5
	Aug.	24.7	75.3	24.7	75.3
	Sep.	14.8	85.2	15.2	84.8
	Oct.	3.9	96.1	4.6	95.4
	Nov.	0.9	99.1	1.3	98.7
	Dec.	0.4	99.6	0.6	99.4
2020	Jan.	0.3	99.7	0.3	99.7
	Feb.	0.6	99.4	0.8	99.2
	Mar.	1.9	98.1	1.9	98.1
	Apr.	4.2	95.8	4.0	96.0
	May	9.2	90.8	9.5	90.5
	June	10.9	89.1	18.8	81.2
	July	23.7	76.3	24.4	75.6
	Aug.	24.5	75.5	25.5	74.5
	Sep.	17.2	82.8	16.6	83.4
	Oct.	4.0	96.0	4.1	95.9
	Nov.	1.3	98.7	1.6	98.4
	Dec.	0.4	99.6	0.4	99.6

January had the highest (1.48 pg WHO_{2005}-TEQ L^{-1}). As to the seasonal variations, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were 1.20, 0.41, 1.04, and 1.34 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively.

As for Tianjin, during 2018, the monthly average concentration of total PCDD/Fs-WHO_{2005}-TEQ in the rain ranged between 0.33 and 2.49 pg WHO_{2005}-TEQ L^{-1} and averaged 1.36 pg WHO_{2005}-TEQ L^{-1}, which indicated that the July had the lowest (0.33 pg WHO_{2005}-TEQ L^{-1}), and March had the highest (2.49 pg WHO_{2005}-TEQ L^{-1}). As to the seasonal variations, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were 1.91, 0.40, 1.39, and 1.73 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. During 2019, the monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain ranged between 0.30 and 2.55 pg WHO_{2005}-TEQ L^{-1} and averaged 1.43 pg WHO_{2005}-TEQ L^{-1}, which indicated that July had the lowest (0.30 pg WHO_{2005}-TEQ L^{-1}), and January had the highest (2.55 pg WHO_{2005}-TEQ L^{-1}). As to the seasonal variations, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were 1.80, 0.39, 1.45, and 2.09 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. During 2020, the monthly

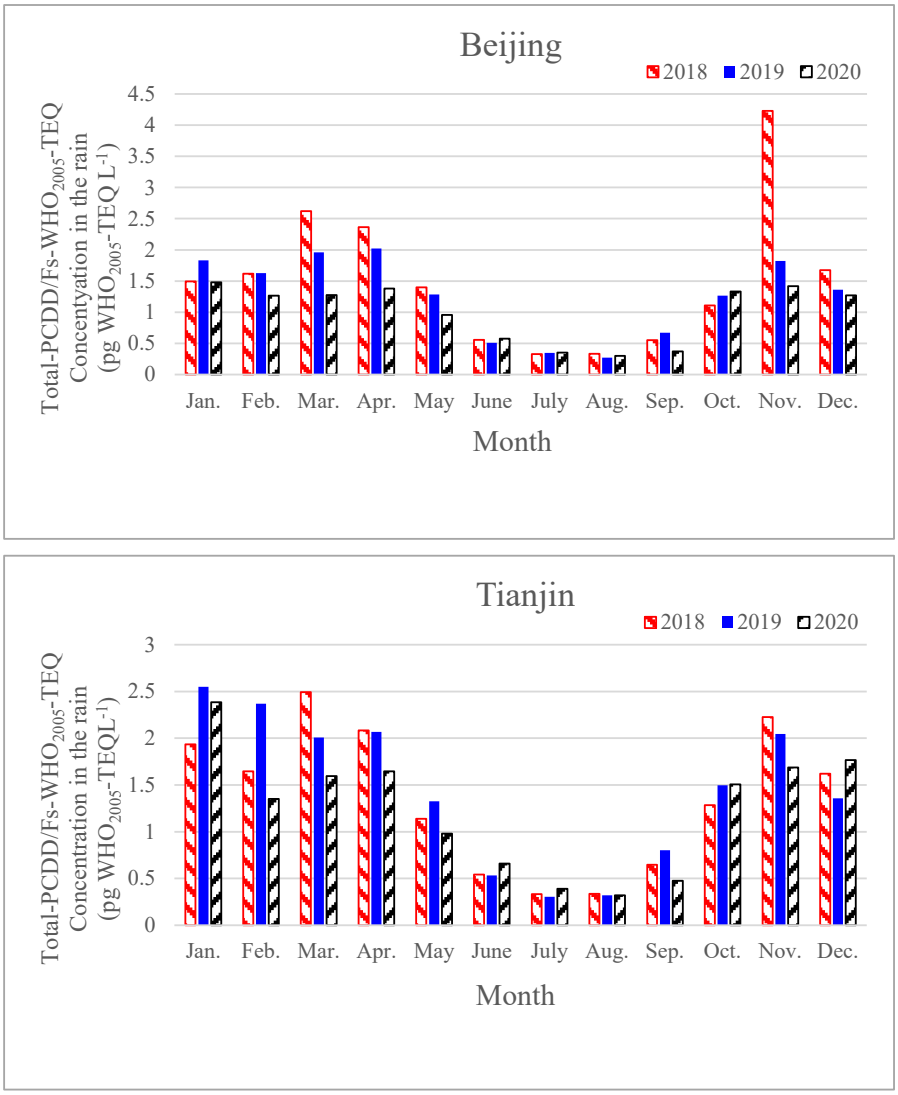

Fig. 2. Monthly average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain in Beijing and Tianjin in 2018, 2019, and 2020.

average concentrations of total PCDD/Fs-WHO_{2005}-TEQ in the rain ranged between 0.32 and 2.39 pg WHO_{2005}-TEQ L^{-1} and averaged 1.23 pg WHO_{2005}-TEQ L^{-1}, which indicated that August had the lowest (0.32 pg WHO_{2005}-TEQ L^{-1}), and January had the highest (2.39 pg WHO_{2005}-TEQ L^{-1}). As to the seasonal variations, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were 1.48, 0.46, 1.22, and 1.83 pg WHO_{2005}-TEQ L^{-1} in spring, summer, fall, and winter, respectively. The above results showed that the total PCDD/Fs-WHO_{2005}-TEQ concentration in the rain was significantly lower in summer (averaged 0.03 pg-WHO_{2005}-TEQ m^{-3}), which was mainly because in summer the total PCDD/Fs-WHO_{2005}-TEQ concentration in the air is lower (averaged 0.41 pg WHO_{2005}-TEQ L^{-1}), and the proportion of the gas phase is increased. When the total PCDD/Fs-WHO_{2005}-TEQ concentration in rain is diluted by a heavy rainfall, the PCDD/F concentration is diluted by this greater amount of rain (Wang *et al*, 2018). However, in winter, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in rain were higher, which was due to the low temperature in winter, which resulted in a greater more fraction of PCDD/Fs being adsorbed into the granular phase and easily scavenged by the precipitation (Zhu *et al.*, 2017b).

3.5 $PM_{2.5}$-bound Total PCDD/Fs-WHO_{2005}-TEQ Content

Relevant studies have shown that in summer, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in counties in Asia accounts for 36.4% to 71.5% of the total concentration in the particle phase.

The $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in Beijing and Tianjin in 2018, 2019, and 2020 are shown in Figs. 3(a) and 3(b).

As for Beijing, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content ranged between 0.10 and 0.84 ng-WHO_{2005}- TEQ g^{-1}, with an average of 0.51 ng-WHO_{2005}-TEQ g^{-1} in 2018. It ranged between 0.13 and 0.78 ng-WHO_{2005}-TEQ g^{-1}, with an average of 0.51 ng-WHO_{2005}-TEQ g^{-1} in 2019. In 2020, the level of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ ranged between 0.12 and 0.86 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.48 ng-WHO_{2005}-TEQ g^{-1}. In the three years under observation, the highest $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in 2018 and 2020 was in December (0.84 ng-WHO_{2005}-TEQ g^{-1}, 0.86 ng-WHO_{2005}-TEQ g^{-1}), and the highest $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents in 2019 was in November (0.78 ng-WHO_{2005}-TEQ g^{-1}). July was consistently the month with the lowest $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content (0.10 ng-WHO_{2005}-TEQ g^{-1}, 0.13 ng-WHO_{2005}-TEQ g^{-1}, 0.12 ng-WHO_{2005}-TEQ g^{-1}).

With regard to Tianjin, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in 2018 ranged between 0.12 and 0.87 ng-WHO_{2005}-TEQ g^{-1}, with an average of 0.47 ng-WHO_{2005}-TEQ g^{-1}. In 2019, it ranged from 0.10 to 0.78 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.48 ng-WHO_{2005}-TEQ g^{-1}. During 2020, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content ranged between 0.13 and 0.75 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.45 ng-WHO_{2005}-TEQ g^{-1}. In the three years under observation,

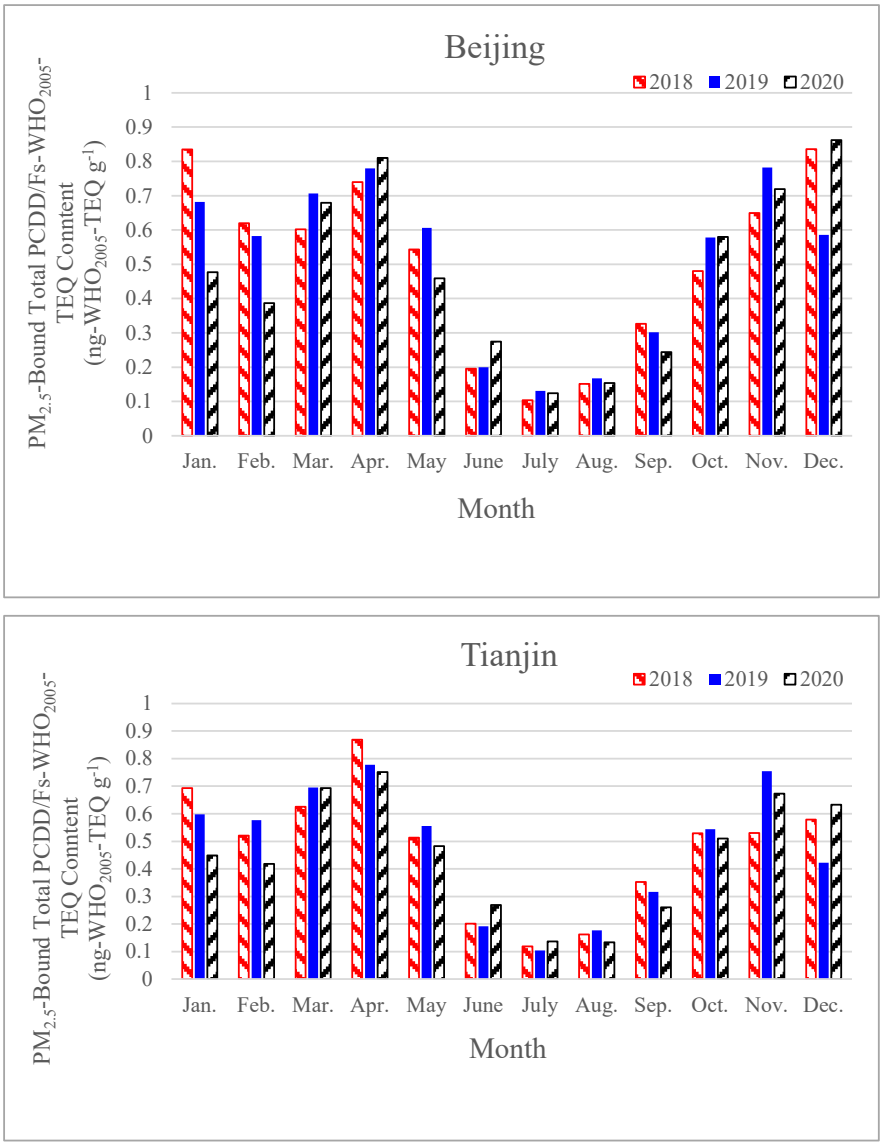

Fig. 3. $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in Beijing and Tianjin in 2018, 2019, and 2020, respectively.

the highest $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents were all occurred in April (0.87 ng-WHO_{2005}-TEQ g^{-1}, 0.78 ng-WHO_{2005}-TEQ g^{-1} and 0.75 ng-WHO_{2005}-TEQ g^{-1}). The lowest $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in 2018 and 2019 was in July (0.10 ng-WHO_{2005}-TEQ g^{-1}, 0.13 ng-WHO_{2005}-TEQ g^{-1}), and the lowest $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in 2020 was in August (0.13 ng-WHO_{2005}-TEQ g^{-1}).

When regard to seasonal variations, for Beijing, in 2018, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in spring, summer, fall and winter was 0.63, 0.15, 0.49, and 0.76 ng-WHO_{2005}-TEQ g^{-1}, respectively, and in 2019, it was 0.7, 0.17, 0.55 and 0.62 ng-WHO_{2005}-TEQ g^{-1}, respectively. In 2020, it was 0.22, 0.06, 0.19, and 0.25 ng-WHO_{2005}-TEQ g^{-1}, respectively. It was found that, in Beijing from 2018 to 2020, the average $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in summer was approximately 76.8%, 69.7%, and 77.2% lower than that in the other three seasons (spring, fall and winter). The three-year average summer ambient temperature in Beijing was 26.2°C, which was much higher than the 14.5°C in spring, 13.0°C in fall and –3.2°C in winter. For Tianjin, in 2018, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in spring, summer, fall, and winter was 0.67, 0.16, 0.47, and 0.60 ng- WHO_{2005}-TEQ g^{-1}, respectively. In 2019, it was 0.68, 0.16, 0.54, and 0.53 ng-WHO_{2005}-TEQ g^{-1}, respectively, and in 2020, it was 0.03, 0.01, 0.02, and 0.04 ng-WHO_{2005}-TEQ g^{-1}, respectively. The average $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in summer in Tianjin was approximately 76.4%, 68.5%, and 72.2% lower than that in the other three seasons (spring, fall and winter). In Tianjin, the situation was similar. The three-year average summer ambient temperature in Tianjin was 27.1°C, which was much higher than the 15.5°C in spring,16.9°C in fall and 0°C in winter. This was because with the increase in ambient temperature, more of the total PCDD/Fs-WHO_{2005}-TEQ concentration evaporates from the particle phase to the gas phase, resulting in a decrease in the average $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content.

3.6 $PM_{2.5}$ Concentration

The $PM_{2.5}$ concentration not only affects human health, but also can reflect the PCDD/F concentration in a given area. This is because the total PCDD/Fs-WHO_{2005}-TEQ concentration in ambient air mainly comes from the particulate phase. The monthly average $PM_{2.5}$ concentrations in the ambient air of Beijing and Tianjin are shown in Table 4 for the period between 2018 and 2020. As shown in the Table 4, during 2018–2020 in Beijing, the $PM_{2.5}$ concentration ranged between 28.0 and 82.0, between 23.0 and 53.0, and between 24.0 and 62.0 μg m^{-3}, and averaged 47.3, 42.0 and 37.9 μg m^{-3}, respectively. Over the three years examined, the highest $PM_{2.5}$ concentration occurred in 2018, followed by 2019, and the lowest was in 2020. The annual average $PM_{2.5}$ concentration was compared between 2018 and 2020, where it was concluded that it was reduced by approximately 19.9%. The decrease in $PM_{2.5}$ concentration is partly due to the stricter emission regulations for pollutants in Beijing, the capital of China, and partly due to people's increased awareness of environmental protection and efforts. As a whole, the $PM_{2.5}$ three-year average concentration in Beijing ranged between 23.0 and 82.0 μg m^{-3}, with an average of 42.4 μg m^{-3}. The highest monthly concentration of $PM_{2.5}$ occurred in March (82.0 μg m^{-3}), while the lowest one occurred in August (23.0 μg m^{-3}).

As for Tianjin, over the course of the three years, the highest $PM_{2.5}$ concentration occurred in 2019, at 25-81 μg m^{-3}, with an average of 51.3 μg m^{-3}; followed by 2018, which range from 27–79 μg m^{-3}, with an average of 48.6 μg m^{-3}. In 2020, ranged from 29–101 μg m^{-3}, with an average of 48.0 μg m^{-3}. These results indicate that from 2018 to 2020 the annual average $PM_{2.5}$ concentration fell from 51.3 to 48.0 μg m^{-3}, falling by approximately 6.4%. However, the highest values increased by 27.8% from 2020 (101 μg m^{-3}) to 2018 (79 μg m^{-3}). As a whole, the $PM_{2.5}$ concentrations for these three years in Tianjin ranged between 25 and 101 μg m^{-3}, with an average of 49.3 μg m^{-3}. This indicates that the $PM_{2.5}$ level in Tianjin was higher than that in Beijing. This may due to the fact that Tianjin's coal consumption is much higher than Beijing's, and Beijing has better air pollution control measures. Although Tianjin's air quality has improved annually, these $PM_{2.5}$ concentrations were still well above the WHO air quality regulated standard (10 μg m^{-3}). Therefore, effectively control of $PM_{2.5}$ emissions must be accomplished to achieve sustainable economic and environmental development in Tianjin.

As for Beijing, the average $PM_{2.5}$ concentrations in spring, summer, fall, and winter were 62.0, 39.3, 47.0, and 40.7 μg m^{-3} in 2018, respectively, and those during 2019 were 45.7, 33.0, 40.0,

Table 4. Monthly atmospheric $PM_{2.5}$ concentration in Beijing and Tianjin in 2018, 2019, and 2020, respectively.

	City	Beijing			Tianjin		
Year	Month	Range	Average	RSD (%)	Range	Average	RSD (%)
2018	Jan.	6.0–133	34	46	9.0–143	53	37
	Feb.	7.0–168	50	47	11–142	60	40
	Mar.	17–233	82	37	14–272	75	37
	Apr.	10–206	59	47	14–163	44	40
	May	12–128	45	32	14–80	38	23
	June	8,0–110	43	30	10–77	40	18
	July	9.0–74	44	23	8.0–61	38	20
	Aug.	8.0–74	31	35	8.0–62	29	25
	Sep.	5.0–100	28	49	8.0–73	30	35
	Oct.	4.0–153	42	59	7.0–116	44	43
	Nov.	13–220	71	43	20–225	79	35
	Dec.	6.0–182	38	48	10–243	53	41
2019	Jan.	9.0–217	51	48	13–248	81	43
	Feb.	8.0–136	53	39	10–237	78	42
	Mar.	4.0–183	52	57	10–142	54	42
	Apr.	9.0–106	48	32	15–124	49	34
	May	6.0–91	37	37	8.0–102	41	31
	June	5.0–74	39	26	10–92	42	24
	July	7.0–83	37	33	22–86	41	21
	Aug.	3.0–76	23	40	6.0–58	26	27
	Sep.	6.0–77	36	33	16–78	41	27
	Oct.	6.0–124	40	36	12–145	50	36
	Nov.	7.0–138	44	37	14–157	51	37
	Dec.	6.0–183	44	50	10–175	61	45
2020	Jan.	10–107	59	42	15–239	101	43
	Feb.	3.0–207	62	48	6.0–169	61	47
	Mar.	5.0–117	35	44	8.0–112	43	31
	Apr.	6.0–90	31	40	9.0–84	40	31
	May	5.0–108	36	38	17–66	35	24
	June	7.0–66	32	28	10–80	38	22
	July	14–85	41	25	25–75	41	16
	Aug.	9.0–61	28	29	14–69	34	26
	Sep.	5.0–82	24	46	8.0–65	29	34
	Oct.	5.0–138	42	48	8.0–133	54	42
	Nov.	6.0–107	37	50	12–122	47	40
	Dec.	6.0–72	28	38	8.0–149	53	44

and 49.3 μg m^{-3}, respectively. Those in 2020 were 34.0, 36.7, 34.3, and 49.7 μg m^{-3}, respectively. For Tianjin, during 2018, the average $PM_{2.5}$ concentrations in spring, summer, fall, and winter were 52.3, 35.7, 51.0, and 55.3 μg m^{-3}, respectively, and those in 2019 were 48.0, 36.3, 47.3, and 73.3 μg m^{-3}, respectively. Those in 2020 were 39.3, 37.7, 43.3, and 71.7 μg m^{-3}, respectively. As can be seen from the above results, the $PM_{2.5}$ concentration undergoes significant seasonal variations. The highest values always occurred in winter and the lowest always occurred in summer, while the values in spring and fall were similar and in the middle levels. As a whole, Beijing's $PM_{2.5}$ concentration in summer (35.3 μg m^{-3}) was 32.0% lower than that in winter (46.6 μg m^{-3}) based on a three-year average, and the values of Tianjin in summer (36.6 μg m^{-3}) were 82.5% in magnitude lower than those in winter (66.8 μg m^{-3}).

The $PM_{2.5}$ concentration is highest in winter for the following reasons. Cold temperatures in winter enhance the stability of the atmosphere and hinder the vertical convection of the air, thus resulting in the accumulation of $PM_{2.5}$ in the ambient air. In addition, heating in northern cities in winter significantly increases the amount of both coal and biomass that are burned, and lower

temperatures increase vehicle exhaust emissions as well. The lowest $PM_{2.5}$ concentrations in summer were due to the hot, rainy summer when the instability of the atmosphere is conducive to vertical dispersion and increases in atmospheric humidity, which reduce the $PM_{2.5}$ concentration in the ambient air.

4 CONCLUSION

1. In Beijing from 2018 to 2020, the average wet deposition fluxes of PCDD/Fs in spring, summer, fall, and winter were 50.50, 41.47, 23.03, and 16.76 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The highest value was found in spring, and the lowest value was found in winter. In Tianjin, from 2018 to 2020, the seasonal average wet deposition fluxes of PCDD/Fs were 35.30, 42.40, 13.37, and 14.77 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The highest value was found in summer, and the lowest value was found in fall. This is because the two cities have more rainfall in spring and summer, but less in fall and winter, and rainfall has a great influence on the wet deposition flux.
2. As to the seasonal variations, from 2018–2020, in Beijing, the percentages of seasonal contributions to PCDD/F wet deposition fluxes by the gas phase were 2.8%, 22.4%, 5.4%, and 0.2% in spring, summer, fall, and winter, respectively, and those of in Tianjin were 3.4%, 2.5%, 5.5%, and 0.2%, respectively. The above results revealed that a higher temperature in summer (between 21.3°C and 27.8°C) led to a higher seasonal percentage of PCDD/F wet deposition fluxes by the gas phase, while those in winter with lower temperature (between –7.0°C and 0.7°C) resulted in a lower contribution by the gas phase.
3. As to seasonal variations, in Beijing, the average concentration of total PCDD/Fs-WHO_{2005}-TEQ in the rain in spring, summer, fall and winter were 1.70, 0.39, 1.42, and 1.52 pg WHO_{2005}-TEQ L^{-1} from 2018–2020, respectively. In Tianjin, the values in spring, summer, fall and winter were 1.73, 0.42, 1.35, and 1.88 pg WHO_{2005}-TEQ L^{-1} from 2018–2020, respectively. The above results showed that the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain were significantly lower in summer, which is mainly because in summer, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the air are lower, and the proportion of the gas phase is increased. When the total PCDD/Fs-WHO_{2005}-TEQ concentration in rain is diluted by a heavy rainfall, the PCDD/F concentration is also diluted by this greater amount of rain. However, in winter, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in rain were higher, which was due to the low temperature in winter, which caused more PCDD/Fs to be adsorbed into the granular phase and to be more easily scavenged by the precipitation.
4. With regard to seasonal variations, for Beijing, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in spring, summer, fall, and winter were 0.51, 0.13, 0.41, and 0.55 ng-WHO_{2005}-TEQ g^{-1} from 2018–2020, respectively. During the three years, the average $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents in summer were was 76.2%,69.7%, and 77.2% lower than that in the other three seasons (spring, fall and winter). For Tianjin, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents in spring, summer, fall and winter was 0.46, 0.11, 0.34 and 0.39 ng-WHO_{2005}-TEQ g^{-1} from 2018–2020, respectively. The average $PM_{2.5}$-bound total PCDD/FsWHO$_{2005}$-TEQ content in summer in Tianjin was approximately 76.4%,68.5% and 72.2% lower than that in the other three seasons (spring, fall and winter). This was because with an increase in the ambient temperature, more total PCDD/F mass evaporates from the granular phase to the gas phase, resulting in a decrease in the average $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents.
5. As a whole, from 2018–2020, three-year averages of the $PM_{2.5}$ concentration in Beijing in spring, summer, fall, and winter were 47.2, 36.3, 37.3, and 139.7 μg m^{-3}, respectively. Beijing's $PM_{2.5}$ concentration in summer (35.3 μg m^{-3}) was 32.0% lower than that in winter (46.6 μg m^{-3}) based on a three-year average. In Tianjin, the values in spring, summer, fall and winter were 46.5, 36.6, 47.2, and 66.8 μg m^{-3}, respectively. The values of Tianjin in summer (36.6 μg m^{-3}) were 82.5% lower than those in winter (66.8 μg m^{-3}) during the three-year period. This means that the $PM_{2.5}$ concentration exhibits significant seasonal variations. The highest level always occurred in winter and was the lowest in summer, which was related to the atmospheric temperature inversion in the winter time.

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