

# Atmospheric Concentration, Particle-bound Content, and Dry Deposition of PCDD/Fs

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## ABSTRACT

In this study, the atmospheric total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations, gas-particle partitioning, PM<sub>2.5</sub> concentration, PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content and dry deposition flux in Shanghai and Nanjing were investigated from 2018-2020. In Shanghai, the total PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration dropped from 0.0291 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup> from 2018–2019 to 0.0250 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup> in 2020, while in Nanjing, it dropped from 0.0423 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup> to 0.0338 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>. The average concentrations of PCDD/Fs-WHO<sub>2005</sub>-TEQ in spring and winter in Shanghai and Nanjing were 47.6% and 53.8% higher than those in summer, respectively. From 2018-2019, the average particle phase fractions of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai and Nanjing were 50.3% and 57.5%, respectively, while in 2020, they were 47.8% and 55.1%, respectively. From 2018-2019, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content was 0.342 and 0.493 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> in Shanghai and Nanjing, respectively, while in 2020, it was 0.312 and 0.489 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively. In Shanghai and Nanjing, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content in spring and winter was 77.5% and 73.2% higher than that in summer, respectively. From 2018–2019, the dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ was 316.3 and 460.5 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup> in Shanghai and Nanjing, respectively, while in 2020, it was 272.5 and 368.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. The average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in spring and winter was 47.6% and 53.8% higher than that summer in Shanghai and Nanjing, respectively. The above results indicate that COVID-19 in 2020 had a positive effect on air quality improvement in PCDD/Fs. On average, more than 98.88% of the total PCDD/Fs-WHO<sub>2005</sub>-TEQ dry deposition flux was primarily contributed by the particle phase. This was attributed to the fact that dry deposition of particle phase PCDD/Fs was mainly due to gravitational settling accompanied by higher dry deposition velocities, while the gas phase PCDD/Fs were deposited mostly by diffusion at a lower dry deposition velocity.

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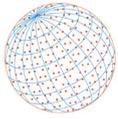
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**Keywords:** PCDD/Fs, PM<sub>2.5</sub>, Particle-bound, Phase distribution, Dry deposition

## 1 INTRODUCTION

PCDD/Fs are common persistent organic pollutants (POPs) in the environment. They are the general name for polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) (Alcock *et al.*, 1996). In addition, PCDD/Fs have strong thermal stability, high decomposition temperatures above 700°C, are extremely insoluble in water, and are soluble in most organic solvents (Wielgosiński *et al.*, 2011). There are 210 homologous PCDD/F isomers, of which 17 (2, 3, 7, and 8 are all replaced by chlorine atoms) are considered to be harmful to human health (Li *et al.*, 2016). The sources of PCDD/Fs in nature are mainly volcanic eruptions and forest fires, and dioxins can also be produced by human activities, such as burning of waste, chemical manufacturing, metal



smelting and automobile exhaust (Qiu *et al.*, 2020). PCDD/Fs in the environment can enter the biological system through respiration, absorption and ingestion.

The concept of Toxic Equivalent Quantity (TEQ) is often used in the international toxicity evaluation of PCDD/Fs, which is expressed by the amount of 2,3,7, and 8-TCDD (Hsieh *et al.*, 2018). PCDD/Fs are highly toxic and can exist in environmental media for a long time (Wang *et al.*, 2020). Moreover, PM<sub>2.5</sub> can be used as the transmission medium for long-distance transport through the atmosphere and ocean currents and seriously affects human health. The elimination of PCDD/Fs occurs mainly through migration, diffusion and deposition, which is closely related to the distribution of the gas-solid phase and temperature in the atmospheric environment. The gas phase can be removed from the environment by photodegradation and OH radical reactions, while the particle phase mainly entered the soil and aquatic environment through sedimentation. (Koester *et al.*, 1992; Lohmann *et al.*, 2007).

Particulate matter (PM) is one of the most important components of air pollution in China. Particulate matter is a type of aerosol. It refers to solid or liquid particles in the atmosphere and can be further classified according to its size (Ghosh *et al.*, 2014). PM<sub>2.5</sub> is a type of fine particulate matter with a diameter of less than 2.5 μm. PM<sub>2.5</sub> is also known as a fine particulate that can directly enter the lungs. It is used as the main pollutant index that characterizes the ambient air quality (Chow *et al.*, 2015). Increases in PM<sub>2.5</sub> concentrations have significantly reduced urban visibility, worsened climate conditions, and seriously damaged the environment (Menon *et al.*, 2002). High concentrations of PM<sub>2.5</sub> are also harmful to human health. Studies have shown that the risk of acute lower respiratory tract infection increases by 1.12% for every 10 μg m<sup>-3</sup> increase in the annual PM<sub>2.5</sub> concentration (Mehta *et al.*, 2013; Xing *et al.*, 2016). At present, aerosols in cities are mainly emitted directly from human activities such as combustion and automobile exhaust (Kong *et al.*, 2014).

In this study, the concentration, gas-particle partitioning and PM<sub>2.5</sub>-bound content of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ, PM<sub>2.5</sub> concentrations, and dry deposition of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in two cities (Shanghai and Nanjing) in the south of China from 2018 to 2020 were investigated, compared and discussed.

## 2. METHODS

The air quality from 2018 to 2020 in Shanghai and Nanjing was investigated, including total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations, PCDD/F gas-particle partitioning, PM<sub>2.5</sub> concentration, PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content and dry deposition of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ.

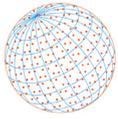
Shanghai City is located in the Yangtze River Delta region, at a 120°52′–122°12′ east longitude and a 30°40′–31°53′ north latitude. It has a subtropical monsoon climate. The annual temperature in Shanghai ranges between 2.0 and 34 and averages 17.6°C; the annual average sunshine totals approximately 1,663 hours, and the annual average precipitation is 1,173.4 mm. More than 60% of the annual rainfall is concentrated into the flood season, which runs from May to September.

Nanjing is located in the southwest of Jiangsu Province, at a 118°22′–119°14′ east longitude and a 31°14′–32°37′ north latitude. It has a humid climate and is located in the northern subtropics. The annual average temperature in Nanjing ranges between –2.0 and 33 and averages 15.4°C. The annual average sunshine is approximately 1,944 hours, and the annual average precipitation is 1,106.5 mm. More than 60% of the annual rainfall is concentrated in the rainy season, which runs from May to September.

### 2.1 PCDD/F Concentration

In the absence of measured data, the concentration of PCDD/Fs can be simulated using a regression analysis. For the purposes of this study, two regression analysis equations were selected, for which the results were averaged. The two equations are as follows (Wang *et al.*, 2010; Huang *et al.*, 2011):

$$Y_1 = 0.0138X \pm 0.0472 \quad (1)$$



$$Y_2 = 0.0117X - 0.021, \quad (2)$$

where  $Y_1$  and  $Y_2$  represent the concentration of total PCDD/Fs, and  $X$  represents the concentration of  $PM_{10}$  in the urban atmosphere.

The goodness-of-fit of regression equation is  $R^2=0.9855$  (Suryani *et al.*, 2015; Lee *et al.*, 2016). The results indicated good reliability in terms of prediction and goodness of fit. In this study, the regression was used to obtain the PCDD/F concentration. The concentration of total PCDD/Fs was obtained from the mean value of  $Y_1$  and  $Y_2$ , and the PCDD/Fs were analyzed and discussed by combining the meteorological data for the local cities.

## 2.2 Gas-Particle Partitioning

The gas and particle partitioning of PCDD/Fs were evaluated by multiplying the gas-particle distribution by the total concentration of PCDD/Fs. The gas-particle partitioning constant ( $K_p$ ) is calculated as follows (Yamasaki *et al.*, 1982; Pankow *et al.*, 1992):

$$K_p = \frac{F / TSP}{A} \quad (3)$$

where TSP represents the concentration of total suspended particulate matter ( $\mu\text{g m}^{-3}$ );  $F$  represents the concentration of the compounds of interest bound to particles ( $\text{pg m}^{-3}$ ), and  $A$  represents the gaseous concentration of the compound of interest ( $\text{pg m}^{-3}$ ).

Plotting  $\log K_p$  against the logarithm of the subcooled liquid vapor pressure,  $P_L^0$ , gives (Hung *et al.*, 2002):

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

where  $P_L^0$  represents the subcooled liquid vapor pressure (Pa);  $m_r$  represents the cited slope,  $-1.29$ , and  $b_r$  represents the cited y-intercept,  $-7.2$  (Chao *et al.*, 2004).

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

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

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

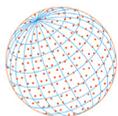
## 2.3 Dry Deposition Flux of PCDD/Fs

The dry sedimentation flux is a combination of the diffusion of gaseous matter and the sedimentation of granular matter.

$$F_T = F_g + F_p \quad (6)$$

$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \quad (7)$$

where  $F_T$  represents the total dry deposition flux ( $\text{pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$ );  $F_g$ : represents the diffusion of gaseous matter producing dry deposition flux ( $\text{pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$ );  $F_p$  represents the gravitational settling of particulate matter producing dry deposition flux ( $\text{pg WHO}_{2005}\text{-TEQ m}^{-2} \text{ month}^{-1}$ );  $C_T$  represents the total concentration of PCDD/Fs in the atmosphere ( $\text{pg m}^{-3}$ );  $V_{d,T}$  represents the dry deposition rate of PCDD/Fs,  $0.42 \text{ cm s}^{-1}$  (Shih *et al.*, 2006);  $C_g$  represents the calculated concentration of PCDD/Fs in the gas phase ( $\text{pg m}^{-3}$ );  $V_{d,g}$  represents the dry deposition rate of PCDD/Fs in gas phase,  $0.01 \text{ cm s}^{-1}$  (Sheu *et al.*, 1996);  $C_p$  represents the calculated concentration of PCDD/Fs in the particle phase ( $\text{pg m}^{-3}$ ), and  $V_{d,p}$  represents the dry deposition rate of PCDD/Fs in the particle phase ( $\text{cm s}^{-1}$ ).

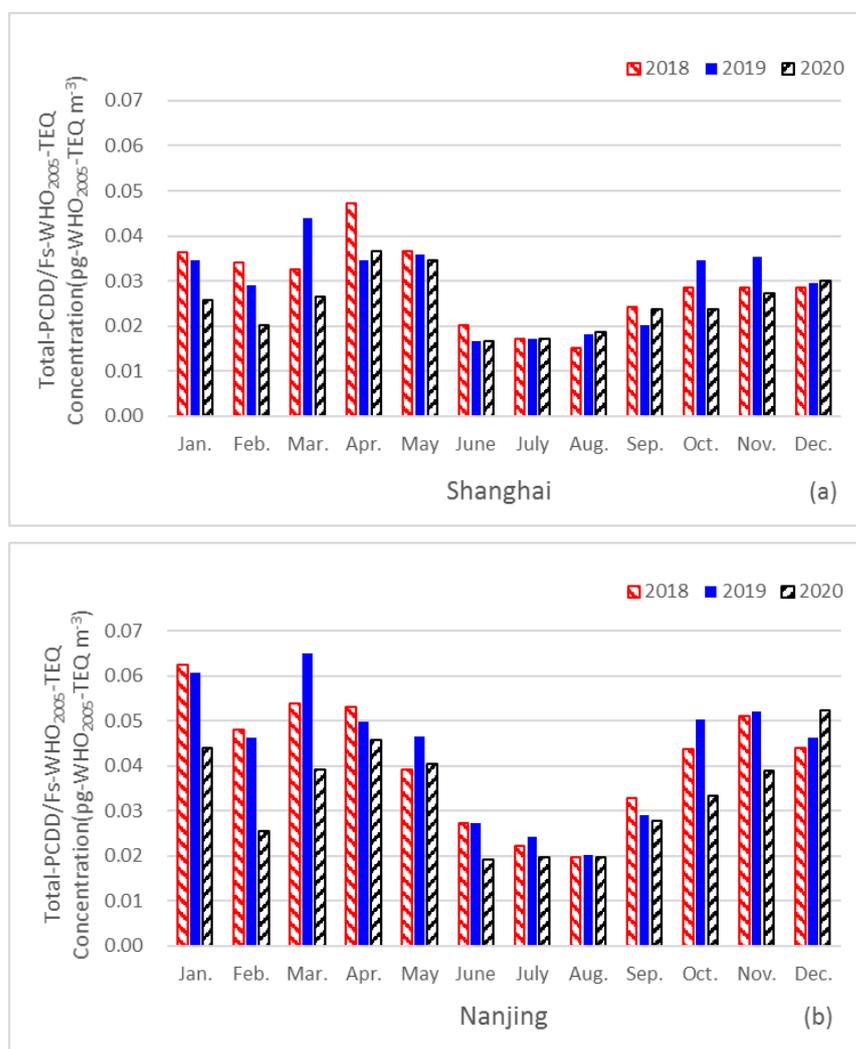


### 3. RESULTS AND DISCUSSION

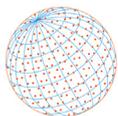
#### 3.1 Total-PCDD/Fs-WHO<sub>2005</sub>-TEQ Concentration

The total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations were calculated based on the combination of the PCDD/Fs mass concentration and the toxicity equivalence factor (TEF) following the World Health Organization (WHO) guidelines. The average monthly total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in Shanghai and Nanjing during 2018-2020 are shown in Fig. 1.

As shown in Fig. 1(a), the total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in Shanghai in the four seasons (spring, summer, autumn, and winter) of 2018 ranged between 0.0326 and 0.0472, between 0.0150 and 0.0202, between 0.0243 and 0.0285, and between 0.0284 and 0.0362 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>, and averaged 0.0388, 0.0174, 0.0271, and 0.0329 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>, respectively. In 2018 in Shanghai, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration (0.0358 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>) in spring and winter was 51.4% higher than that in summer (0.0174 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>), indicating that the lowest value usually occurs in summer. As can be seen, in summer, due to an increase in the ambient temperature in Shanghai, the concentration of PCDD/Fs in the gas phase also increases. In winter, with the decrease in the temperature, the atmospheric density increases, and part of the PCDD/Fs in the gas phase is transferred to the particle phase. This may also be related to coal combustion and atmospheric inversion in winter, where the temperature inversion indicates that the air temperature rises with an increase in altitude, which promotes the accumulation of particulate



**Fig. 1.** Monthly total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in Shanghai and Nanjing in 2018, 2019, and 2020, respectively.



matter on the ground and causes significant air pollution. In addition, according to the monthly  $PM_{2.5}$  concentration and total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration comparison, it was found that a higher  $PM_{2.5}$  concentration was highly correlated with a higher total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration.

In Shanghai, in the four seasons (spring, summer, autumn, and winter) of 2019, total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations ranged from 0.0345 to 0.0438, from 0.0165 to 0.0181, from 0.0200 to 0.0352, and from 0.0290 to 0.0346  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , and averaged 0.0381, 0.0172, 0.0299, and 0.0310  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2019, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration (0.0346  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ) in spring and winter was 50.1% higher than that in summer (0.0172  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ).

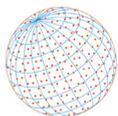
The concentrations in Shanghai in 2020 (spring, summer, autumn, and winter) ranged from 0.0266–0.0365, 0.0165–0.0186, 0.0237–0.0273, and 0.0201–0.0301  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , with an average of 0.0326, 0.0174, 0.0249, and 0.0253  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2020, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations (0.0289  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ) in spring and winter were 39.8% higher than that of summer (0.0174  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ). This was down 15.3%, 12.8%, and 20.9% from the same period in the spring, autumn, and winter of 2018–2019, respectively, while it rose by 0.5% in the summer. In addition, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in 2018 and 2019 was 0.0290 and 0.0291  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2020, the average concentration was 0.0250  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , which was significantly lower than that in 2018–2019. Based on the data from Shanghai in the last three years, the average concentration for 2020 was 13.9% lower than the average for 2018–2019. Shanghai began to implement strict epidemic prevention measures in February 2020, and the concentration of PCDD/Fs in February 2020 was 36.3% lower than that in 2018–2019. Under the control measures, factories were closed, and employees were on leave, so industrial waste gas and traffic emissions were significantly reduced, and air quality was significantly improved.

As shown in Fig. 1(b), the total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in Nanjing in the four seasons (spring, summer, autumn, and winter) in 2018 ranged between 0.0392 and 0.0538, between 0.0196 and 0.0274, between 0.0328 and 0.0510, and between 0.0440 and 0.0624  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , and averaged 0.0487, 0.0231, 0.0425, and 0.0515  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2018, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations (0.0501  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ) in spring and winter were 53.9% higher than those in summer (0.0231  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ), indicating that high temperatures can vaporize PCDD/Fs from the particle phase to the gas phase. In the four seasons (spring, summer, autumn, and winter) of 2019, total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations ranged from 0.0465 to 0.0651, from 0.0202 to 0.0274, from 0.0291 to 0.0522, and from 0.0463 to 0.0607  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , and averaged 0.0538, 0.0239, 0.0439, and 0.0511  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2019, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations (0.0524  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ) in spring and winter were 54.3% higher than those in summer (0.0239  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ). Those during 2020 were ranged from 0.0392–0.0458, 0.0191–0.0196, 0.0279–0.0388, and 0.0256–0.0524  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , and averaged 0.0418, 0.0195, 0.0334, and 0.0407  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2020, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations (0.0413  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ) in spring and winter were 52.8% higher than those in summer (0.0195  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ ) and were 18.4% (spring), 17.2% (summer), 22.7% (autumn), and 20.6% (winter) lower than those from 2018–2019. In addition, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in 2018 and 2019 were 0.0414 and 0.0432  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , respectively. In 2020, the average concentration was 0.0338  $pg\text{-}WHO_{2005}\text{-}TEQ\ m^{-3}$ , which was 18.3% and 21.6% lower than those in 2018 and 2019, respectively. Based on data from Nanjing in the last three years, the average concentration for 2020 was 20.0% lower than that the average for the period 2018–2019. Nanjing began to implement strict epidemic prevention measures in February 2020, and the concentration of PCDD/Fs in February 2020 was 45.5% lower than the average for the period 2018–2019. Because Nanjing's air quality was worse than that in Shanghai, the city needs a big improvement.

Table 1 shows total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentrations in atmospheric environments in some countries and cities globally. The results of this study were in a similar range compared with those found in other countries.

### 3.2 Gas-Particle Partitioning of PCDD/Fs

The gas-particulate distribution of PCDD/Fs plays an important role in the wet and dry deposition

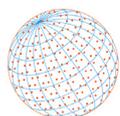
**Table 1.** Total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration range in some regions in the world.

Country	City	PCDD/Fs-WHO <sub>2005</sub> -TEQ Con. (pg-WHO <sub>2005</sub> -TEQ m <sup>-3</sup> )	Reference
Germany	Berlin	0.02–0.40	Christman <i>et al.</i> , 1989
Sweden	Stockholm	0.003–0.024	Broman <i>et al.</i> , 1991
Italy	Rome	0.05–0.28	Turrio-Baldassarri <i>et al.</i> , 1994
England	Manchester	0.03–0.22	Coleman <i>et al.</i> , 1997
England	London	0.07–0.20	Coleman <i>et al.</i> , 1997
Portugal	Oporto	0.02–0.55	Coutinho <i>et al.</i> , 2001
Korea	-	0.17–0.88	Park and Kim, 2002
Greece	Thessaloniki	0.12–0.40	Mandalakis <i>et al.</i> , 2002
China	Hong Kong	0.02–0.43	Sin <i>et al.</i> , 2002
USA	Houston	0.009–0.032	Correa <i>et al.</i> , 2004
Brazil	Sao Paulo	0.05–0.75	de Assunção <i>et al.</i> , 2005
Spain	Catalonia	0.005–1.20	Abad <i>et al.</i> , 2007
Belgium	Flanders	0.02–0.38	Zhou <i>et al.</i> , 2014
USA	Phoenix	0.09–0.45	Correa <i>et al.</i> , 2004
China	Wuhu	0.021–0.085	Wang <i>et al.</i> , 2018
	Chongqing	0.030–0.059	Xing <i>et al.</i> , 2017
	Shanghai	0.015–0.047	This study, 2021
	Nanjing	0.019–0.065	This study, 2021

removal rate. The gas-particle distribution of semi-volatile organic compounds is affected by the temperature, the nature of the particulate matter, and its interaction with environmental organic matter (Pankow *et al.*, 1987). Monthly minimum, maximum, and average temperatures in Shanghai and Nanjing during the period 2018–2020 are shown in Table 2. The average monthly maximum temperatures in Shanghai in 2018, 2019, and 2020 were 33.0°C, 32.0°C, and 34.0°C, respectively. The average monthly minimum temperatures were 2.0°C, 4.0°C, and 4.0°C, respectively, and the annual average temperatures were 18.0°C, 17.5°C, and 18.0°C, respectively. The three-year average temperatures in spring, summer, fall, and winter were 17.0, 27.5, 19.7 and 6.9°C, respectively. In Nanjing, The average monthly maximum temperatures in 2018, 2019, and 2020 were 33.0°C, 32.0°C, and 33.0°C; the average monthly minimum temperatures were –2.0°C, 1.0°C, and 1.0°C; the annual average temperatures were 16.6°C, 16.6°C, and 16.5°C, and the three-year average temperatures in spring, summer, fall, and winter were 16.7, 27.1, 18.0, and 4.7°C, respectively. Because of its geographical location, the average temperature in Nanjing was lower than that in Shanghai.

As shown in Figs. 2(a), 2(b), and 2(c), the average particle phase fractions for the phase distribution of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai in 2018 were approximately 57.1%, 16.4%, 40.9%, and 84.2% in the spring, summer, autumn, and winter, respectively. Similarly, those in 2019 were 60.0%, 19.0%, 43.1%, and 81.8%, respectively, and those in 2020 were 56.1%, 17.2%, 40.9%, and 76.9%, respectively. From 2018 to 2020, the fractions of particle-bound PCDD/Fs for the low molecular weight PCDD/F homologues were calculated. For example, in 2018, 2019, and 2020, for 2,3,7,8-TCDD, they were 24.3%, 23.2% and 17.9%; for 2,3,7,8-TCDF, they were 14.5%, 13.9% and 10.2%, respectively. For the middle molecular weight PCDD/F homologues, they were as follows: For 1,2,3,7,8-PeCDD, they were 46.5%, 48.3%, and 41.2%, and for 1,2,3,7,8-PeCDF, they were 36.2%, 36.6% and 29.8%, respectively. However, for the high molecular weight PCDD/F homologues they were as follows: For 1,2,3,4,6,7,8,9-OCDD, they were 97.3%, 97.8%, and 97.1% and for 1,2,3,4,6,7,8,9-OCDF, they were 97.8%, 98.2% and 97.7%, respectively (Fig. 3). The above results indicate that low molecular weight PCDD/F homologues mainly exist in the gas phase, while high molecular weight PCDD/F homologues are typically associated with particulates. This is due to the fact that a low molecular weight PCDD/F homologue has a higher vapor pressure.

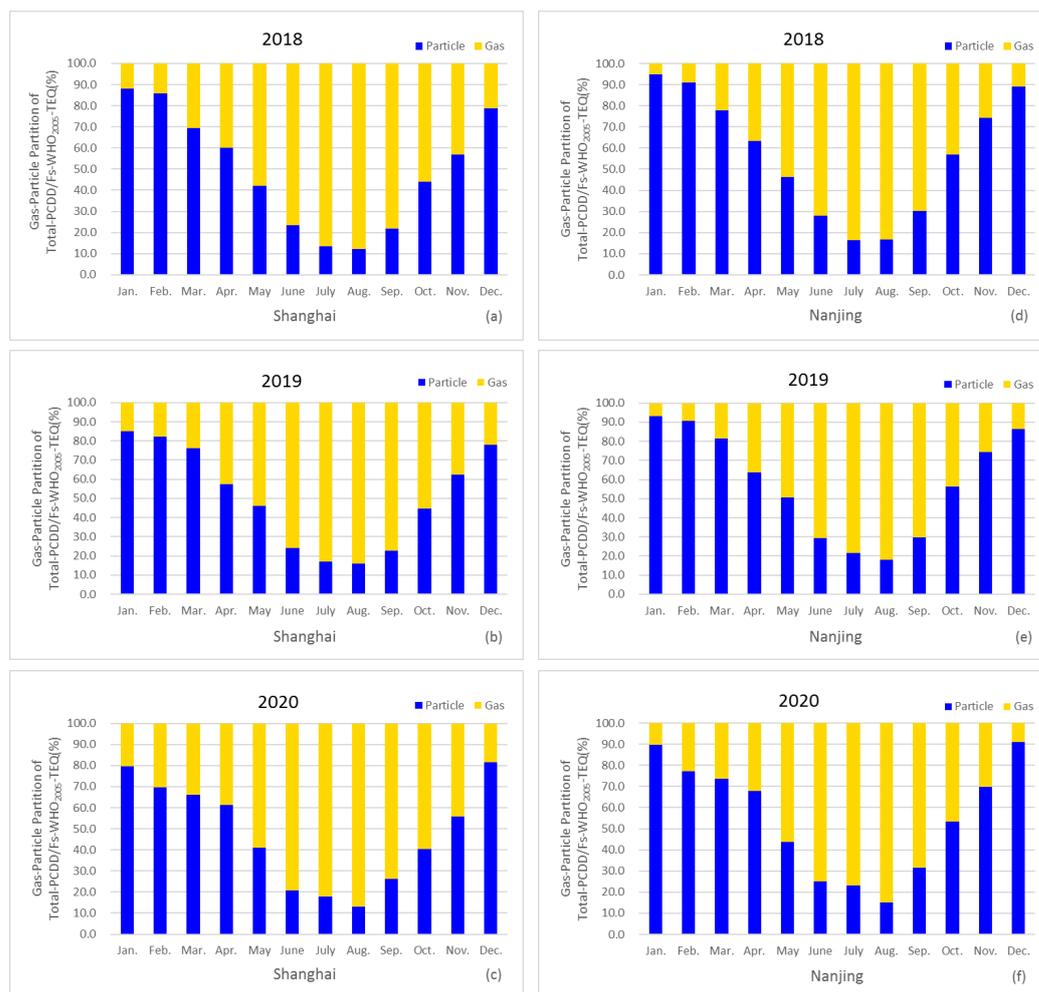
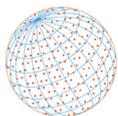
As shown in Figs. 2(d), 2(e), and 2(f), the average particle phase fractions of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Nanjing in 2018 were approximately 62.5%, 20.4%, 53.8%, and 91.6% in the spring, summer, autumn, and winter season, respectively. In the four seasons (spring, summer, autumn, and winter) of 2019, the average particle phase fractions were 65.3%, 23.0%, 53.5%, and 90.0%,



**Table 2.** Monthly minimum, maximum and average temperatures in Shanghai and Nanjing from 2018–2020 (Unit: °C).

City		Shanghai			Nanjing		
Year	Month	Minimum	Maximum	Average	Minimum	Maximum	Average
2018	Jan.	2.0	7.0	4.5	−2.0	5.0	1.5
	Feb.	2.0	9.0	5.5	0.0	8.0	4.0
	Mar.	9.0	16.0	12.5	7.0	17.0	12.0
	Apr.	14.0	22.0	18.0	13.0	22.0	17.5
	May	19.0	26.0	22.5	18.0	25.0	21.5
	June	22.0	29.0	25.5	21.0	30.0	25.5
	July	26.0	33.0	29.5	26.0	33.0	29.5
	Aug.	27.0	32.0	29.5	25.0	32.0	28.5
	Sep.	23.0	29.0	26.0	21.0	28.0	24.5
	Oct.	16.0	22.0	19.0	13.0	22.0	17.5
	Nov.	12.0	18.0	15.0	9.0	16.0	12.5
	Dec.	6.0	10.0	8.0	2.0	8.0	5.0
2019	Jan.	4.0	8.0	6.0	1.0	6.0	3.5
	Feb.	5.0	8.0	6.5	2.0	6.0	4.0
	Mar.	8.0	15.0	11.5	7.0	16.0	11.5
	Apr.	14.0	20.0	17.0	13.0	21.0	17.0
	May	17.0	25.0	21.0	16.0	26.0	21.0
	June	21.0	27.0	24.0	21.0	29.0	25.0
	July	25.0	30.0	27.5	24.0	31.0	27.5
	Aug.	25.0	32.0	28.5	24.0	32.0	28.0
	Sep.	22.0	27.0	24.5	20.0	28.0	24.0
	Oct.	17.0	23.0	20.0	14.0	23.0	18.5
	Nov.	12.0	17.0	14.5	8.0	17.0	12.5
	Dec.	6.0	11.0	8.5	3.0	11.0	7.0
2020	Jan.	5.0	9.0	7.0	2.0	7.0	4.5
	Feb.	7.0	12.0	9.5	4.0	12.0	8.0
	Mar.	9.0	16.0	12.5	7.0	17.0	12.0
	Apr.	12.0	20.0	16.0	10.0	20.0	15.0
	May	19.0	26.0	22.5	17.0	28.0	22.5
	June	23.0	28.0	25.5	21.0	28.0	24.5
	July	24.0	30.0	27.0	23.0	28.0	25.5
	Aug.	27.0	34.0	30.5	26.0	33.0	29.5
	Sep.	21.0	27.0	24.0	19.0	27.0	23.0
	Oct.	16.0	22.0	19.0	13.0	21.0	17.0
	Nov.	13.0	17.0	15.0	9.0	16.0	12.5
	Dec.	4.0	10.0	7.0	1.0	8.0	4.5

respectively. Those during 2020 were, on average, 61.8%, 21.0%, 51.6%, and 86.0%, respectively. From 2018-2020, the fractions of particle-bound PCDD/Fs for the low molecular weight PCDD/F homologues were as follows: For 2,3,7,8-TCDD, they were 38.0%, 36.2%, and 31.2% on average, and for 2,3,7,8-TCDF, they were 25.0%, 23.6% and 19.7%, on average, respectively. The middle molecular weight PCDD/F homologues were as follows: For 1,2,3,7,8-PCDD, they were 58.5%, 59.1%, and 54.4%, on average, and for 1,2,3,7,8-PeCDF, they were 49.0%, 48.8% and 43.8%, on average, respectively. However, for the high molecular weight PCDD/F homologues, they were as follows: for 1,2,3,4,6,7,8,9-OCDD, they were 98.2%, 98.6%, and 98.2%, on average, and for 1,2,3,4,6,7,8,9-OCDF, they were 98.7%, 98.8% and 98.6%, on average, respectively. The average particle phase fractions of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2018 and 2019 were 57.1% and 58.0%. In 2020, the average particle phase fractions was 55.1%, which was lower than in previous years. Based on data from Nanjing in the last three years, the average particle phase fractions of gas-particle partitioning of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ for 2020 was significantly lower than it

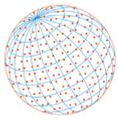


**Fig. 2.** Monthly gas-particle partition of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai and Nanjing in 2018, 2019, and 2020, respectively.

was in 2018 and 2019. The results show that the particle phase fractions of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ were obviously higher than those in the gas phase in spring and winter, but were significantly lower than those in the gas phase in summer. However, in autumn, the difference between the gas and particle phase fractions was less. In general, lower molecular weight PCDD/Fs congeners occur mainly in the gas phase, while the particle phase is usually combined with the higher molecular weight PCDD/Fs congeners. The distribution proportion of compounds in the particle phase in 2020 was significantly lower than that in 2018-2019. When the control measures were implemented in 2020, the particle phase fractions were significantly reduced, indicating that the number of PCDD/Fs congeners in high polymers was obviously reduced, which is conducive to environmentally friendly development.

### 3.3 PM<sub>2.5</sub> Concentration

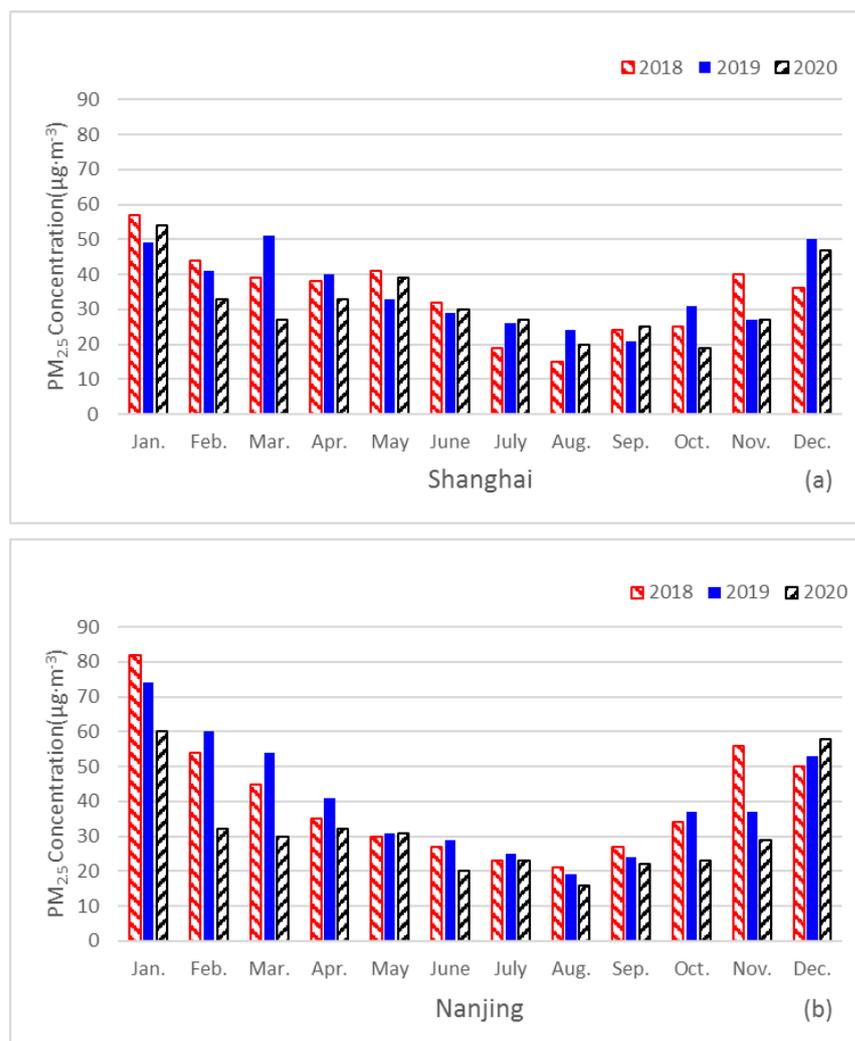
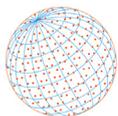
High concentrations of PM<sub>2.5</sub> are harmful to human health. The PM<sub>2.5</sub> levels in Shanghai and Nanjing in the period from 2018-2020 are shown in Fig 4. As shown in Fig. 4(a), the PM<sub>2.5</sub> concentrations in Shanghai in the four seasons (spring, summer, autumn, and winter) of 2018 ranged between 38 and 41, between 15 and 32, between 24 and 40, and between 36 and 57  $\mu\text{g m}^{-3}$ , and averaged 39.3, 22.0, 29.7, and 45.7  $\mu\text{g m}^{-3}$ , respectively. In 2018 in Shanghai, the average PM<sub>2.5</sub> concentrations (42.5  $\mu\text{g m}^{-3}$ ) in spring and winter were 48.2% higher than in summer (22.0  $\mu\text{g m}^{-3}$ ). In the four seasons of 2019, PM<sub>2.5</sub> concentrations ranged from 33 to 51, from 24 to 29, from 21 to 31, and from 41 to 50  $\mu\text{g m}^{-3}$ , and averaged 41.3, 26.3, 26.3, and 46.7  $\mu\text{g m}^{-3}$ , respectively. In 2019, the average PM<sub>2.5</sub> concentration (44.0  $\mu\text{g m}^{-3}$ ) of



**Fig. 3.** Seasonal gas-particle partitioning of PCDD/Fs in Shanghai and Nanjing from 2018–2020.

those spring and winter was 40.2% higher than that in summer ( $26.3 \mu\text{g m}^{-3}$ ). While during 2020, the concentrations ranged from 27–39, 20–30, 19–27, and 33–54  $\mu\text{g m}^{-3}$ , and averaged 33.0, 25.7, 23.7, and 44.7  $\mu\text{g m}^{-3}$ , respectively. In 2020, the average  $\text{PM}_{2.5}$  concentrations ( $38.8 \mu\text{g m}^{-3}$ ) in spring and winter were 40.2% higher than those in summer ( $25.7 \mu\text{g m}^{-3}$ ). In addition, the average  $\text{PM}_{2.5}$  concentrations in 2018 and 2019 were 34.2 and 35.2  $\mu\text{g m}^{-3}$ , respectively, which indicated no significant difference. In 2020, the average  $\text{PM}_{2.5}$  concentration was 31.8  $\mu\text{g m}^{-3}$ , which was 8.4% lower than the average in 2018 and 2019.

As shown in Fig. 4(b), the  $\text{PM}_{2.5}$  concentrations in Nanjing in the four seasons (spring, summer, autumn, and winter) of 2018 ranged between 30 and 45, between 21 and 27, between 27 and 56, and between 50 and 82  $\mu\text{g m}^{-3}$ , and averaged 36.7, 23.7, 39.0, and 62.0  $\mu\text{g m}^{-3}$ , respectively. In 2018 in Nanjing, the average  $\text{PM}_{2.5}$  concentrations ( $49.3 \mu\text{g m}^{-3}$ ) in spring and winter were 52.0% higher than those in summer ( $23.7 \mu\text{g m}^{-3}$ ). In the four seasons of 2019, the  $\text{PM}_{2.5}$  concentrations ranged from 31 to 54, from 19 to 29, from 24 to 37, and from 53 to 74  $\mu\text{g m}^{-3}$ , and averaged 42.0, 24.3, 32.7, and 62.3  $\mu\text{g m}^{-3}$ , respectively. In 2019, the average  $\text{PM}_{2.5}$  concentrations ( $52.2 \mu\text{g m}^{-3}$ ) in



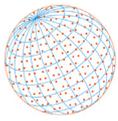
**Fig. 4.** Monthly PM<sub>2.5</sub> concentration in Shanghai and Nanjing in 2018, 2019, and 2020, respectively.

spring and winter were 53.4% higher than those in summer ( $24.3 \mu\text{g m}^{-3}$ ). Those during 2020 ranged from 30–32, 16–23, 22–29, and 32–60  $\mu\text{g m}^{-3}$ , and averaged 31.0, 19.7, 24.7, and 50.0  $\mu\text{g m}^{-3}$ , respectively. In 2020, the average PM<sub>2.5</sub> concentrations ( $40.5 \mu\text{g m}^{-3}$ ) in spring and winter were 51.4% higher than those in summer ( $19.7 \mu\text{g m}^{-3}$ ). In addition, the average PM<sub>2.5</sub> concentrations in 2018 and 2019 were both  $40.3 \mu\text{g m}^{-3}$ , and the average PM<sub>2.5</sub> concentration in 2020 was  $31.3 \mu\text{g m}^{-3}$ , which was 22.3% lower than that in the period from 2018–2019.

Due to the COVID-19 outbreak in February 2020, PM<sub>2.5</sub> concentrations in Shanghai and Nanjing were 22.4% and 43.9% lower than those in February 2018–19, respectively. This is because in February of 2020, enterprises and factories were shut down, and the government called on people to quarantine at home. The control measures resulted in a significant reduction in vehicle exhaust emissions, and the PM<sub>2.5</sub> concentration dropped significantly. Based on the above results, the control measures under the COVID-19 epidemic had a significant positive influence on air quality.

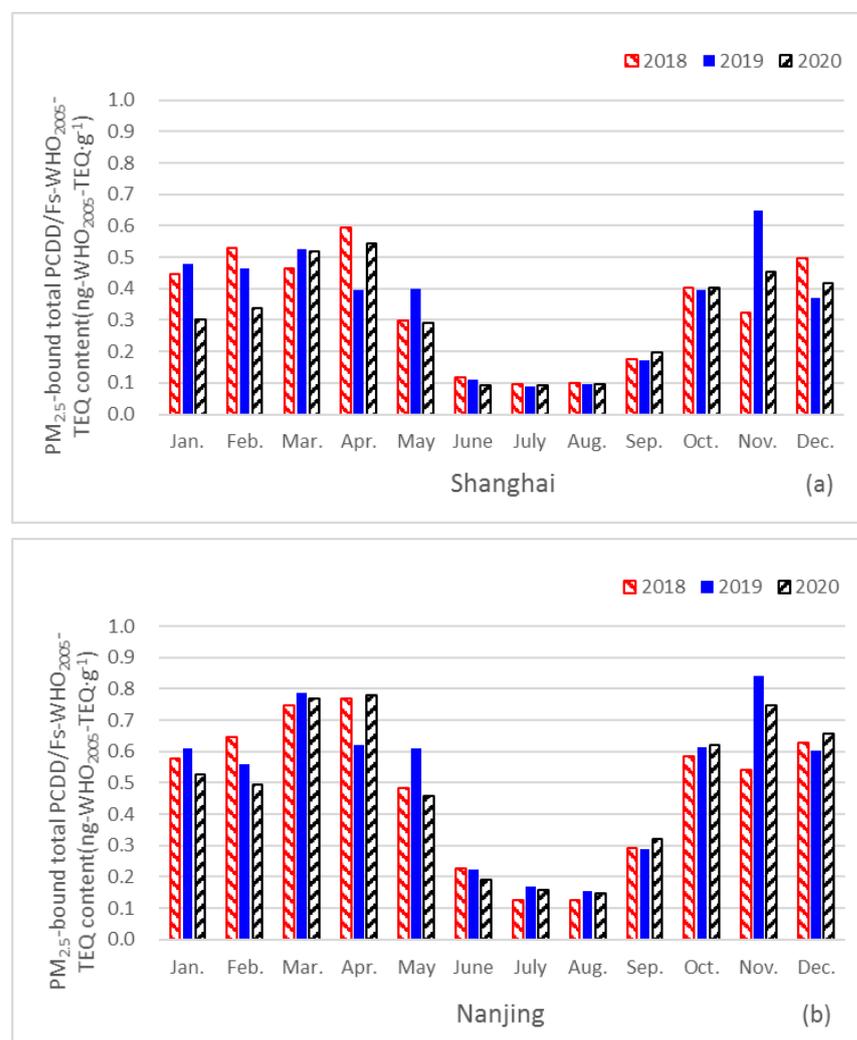
### 3.4 PM<sub>2.5</sub>-bound Total PCDD/Fs-WHO<sub>2005</sub>-TEQ Content

The content of PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai and Nanjing during the period from 2018–2020 is shown in Fig. 5. In 2018, in Shanghai, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content ranged between 0.097 and 0.595 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> and averages 0.337 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>. In 2018 in Shanghai, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content ( $0.472 \text{ ng-WHO}_{2005}\text{-TEQ g}^{-1}$ ) in the spring and winter was 77.9% higher than that in summer ( $0.105 \text{ ng-WHO}_{2005}\text{-TEQ g}^{-1}$ ). In 2019, the content of PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ

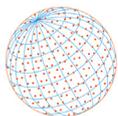


ranged between 0.089 and 0.649 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> and averaged 0.346 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>. In 2019, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content (0.439 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) in spring and winter was 77.7% higher than that in summer (0.098 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>). In 2020, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content ranged between 0.091 and 0.543 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> and averaged 0.312 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>. In 2020, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content (0.402 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) in spring and winter was 77.0% higher than that in summer (0.093 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>). As for seasonal variations, for Shanghai, in 2018, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content in spring, summer, fall, and winter was 0.453, 0.105, 0.301 and 0.492 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively, and in 2019 it was 0.441, 0.098, 0.406 and 0.438 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively. In 2020, it was 0.451, 0.093, 0.352 and 0.353 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively. Compared with the same period in the summer and winter of 2018–2019, the content decreased by 8.7% and 24.1%, while the decrease was slower or even increased in the spring and autumn. Overall, the annual PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content in 2020 (0.312 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) was lower than the average from 2018–2019 (0.342 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>).

As shown in Fig. 5(b), in Nanjing, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content ranged between 0.126 and 0.771 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> and averaged 0.479 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> in 2018. In 2018 in Nanjing, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ contents (0.672 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) in spring and winter was 75.0% higher than that in summer (0.160 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>). In 2019, the content of PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged between 0.153



**Fig. 5.** Monthly PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content in Shanghai and Nanjing in the period from 2018–2020, respectively.



and 0.841 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> and averaged 0.506 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>. In 2019, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content (0.631 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) in spring and winter was 71.3% higher than that in summer (0.181 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>). In 2020, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content ranged between 0.148 and 0.779 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup> and averaged 0.489 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>. In 2020, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content (0.614 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) in spring and winter was 73.1% higher than that in summer (0.165 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>). As for the seasonal variations, for Nanjing, in 2018, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content in spring, summer, fall, and winter was 0.666, 0.160, 0.473 and 0.617 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively. In 2019 it was 0.672, 0.181, 0.581 and 0.591 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively, and in 2020, it was 0.669, 0.165, 0.563 and 0.560 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>, respectively. Compared with the same period in the summer and winter in the period from 2018-2019, the content decreased by 3.0% and 7.3%, but it increased in the autumn. Overall, the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2020 (0.489 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) were slightly lower than during 2018-2019 (0.493 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>).

On the whole, the content of the PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ was the lowest in summer and the highest in winter, which was due to the fact that high temperatures in summer caused the evaporation of a large amount of PCDD/Fs from the particle to the gas phase, so the content of PCDD/Fs particle-bound in PM<sub>2.5</sub> was reduced.

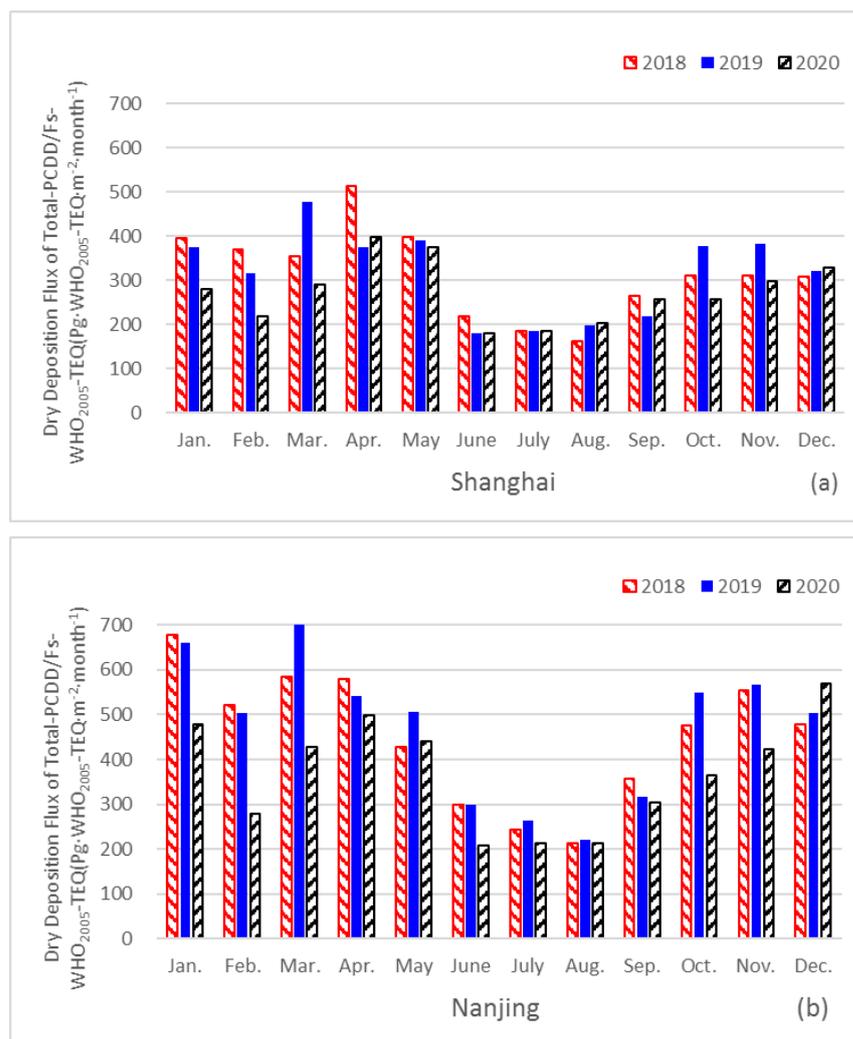
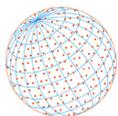
### 3.5 Dry Deposition of Total-PCDD/Fs-WHO<sub>2005</sub>-TEQ

The dry deposition of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in the gas phase occurs mainly through diffusion, while in the particle phase, it occurs mainly through gravitational settling. Table 3 shows the dry deposition of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in atmospheric environments in some countries and cities globally. The monthly dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai and Nanjing in 2018, 2019, and 2020 are presented in Figs. 6(a) and 6(b).

As shown in Fig. 6(a), the dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai in the four seasons (spring, summer, autumn, and winter) of 2018 ranged between 354.4 and 513.4, between 163.2 and 219.4, between 264.3 and 310.5, and between 309.5 and 394.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, and averaged 421.9, 189.4, 295.1, and 358.0 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. In 2018 in Shanghai, the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (389.9 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter was 51.4% higher than that in summer (189.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). In the four seasons of 2019, the dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged from 376.1 to 477.3, from 180.1 to 196.9, from 218.1 to 383.2,

**Table 3.** Dry deposition of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in some countries and regions in the world.

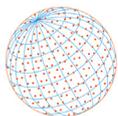
Country	City	Dry deposition (ng m <sup>-2</sup> year <sup>-1</sup> )	Reference
Belguim	Flander	1.48–12.96	Van <i>et al.</i> , 2001
Italy	Venice	0–3.31	Guerzoni <i>et al.</i> , 2004
Korea	Daeyeon	2.1–8.2	Moon <i>et al.</i> , 2005
USA	Houston	0.08–0.25	Correa <i>et al.</i> , 2006
Japan	Kanazawa	0.03–15.48	Oka <i>et al.</i> , 2006
Thailand	Chiangmai	1.0–1.6	Trinh <i>et al.</i> , 2018
China	Beijing	2.4–84.1	Trinh <i>et al.</i> , 2018
Vietnam	Danang	3.1–19.1	Trinh <i>et al.</i> , 2018
	Sonla	1.5–7.8	Trinh <i>et al.</i> , 2018
Iran		0.58–1.19	Momeniha <i>et al.</i> , 2011
France	Marseille	2–80.8	Castro-Jimenez <i>et al.</i> , 2017
Tunisia	Bizerte	2–69.2	Castro-Jimenez <i>et al.</i> , 2017
Taiwan Area	-----	0.67–7.06	Lee <i>et al.</i> , 2018
Turkey	Istanbul	16.07	Gunes <i>et al.</i> , 2019
Czech Republic		0.42–0.57	Degrendele <i>et al.</i> , 2020
China	Shanghai	2.0–5.7	This study, 2021
	Nanjing	2.5–8.5	This study, 2021



**Fig. 6.** Monthly dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai and Nanjing in 2018, 2019 and 2020, respectively.

and from 315.5 to 376.2 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, and averaged 414.6, 187.6, 325.9, and 337.8 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. In 2019, the average dry deposition fluxes of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (376.2 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter were 50.1% higher than those in summer (187.6 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). Those during 2020 ranged from 289.3–397.8, 180.1–202.6, 257.7–297.3, and 218.5–327.7 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, and averaged 354.4, 189.4, 270.9, and 275.1 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. In 2020, the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (314.7 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter was 39.8% higher than that in summer (189.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). Shanghai began to implement strict epidemic prevention measures in February 2020, and the dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in February 2020 was 32.0% lower than that in 2018–2019. The average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2018 and 2019 was 316.1 and 316.5 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. In 2020, the average dry deposition flux was 272.5 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, which was 13.9% lower than in 2018–2019.

As shown in Fig. 6(b), the dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Nanjing in the four seasons (spring, summer, autumn, and winter) of 2018 ranged between 426.7 and 585.7, between 213.8 and 298.1, between 356.8 and 554.9, and between 479.3 and 679.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, and averaged 530.3, 251.3, 462.4, and 560.1 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. In 2018 in Nanjing, the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (545.2 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter was 53.9% higher than that in summer (251.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). In the four seasons of 2019, the dry deposition flux of total-



PCDD/Fs-WHO<sub>2005</sub>-TEQ ranged from 506.2 to 708.6, from 219.4 to 298.1, from 317.1 to 568.1, and from 503.5 to 661.2 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, and averaged 585.7, 260.7, 477.8, and 556.1 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. In 2019, the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (570.9 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter was 54.3% higher than that in summer (260.7 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). In 2020, it ranged from 426.7–499.0, 208.2–213.8, 303.9–422.8, and 279.1–570.9 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, and averaged 455.6, 211.9, 363.4, and 442.9 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, respectively. In 2020, the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (449.2 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter was 52.8% higher than that in summer (211.9 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). Due to the COVID-19 outbreak, the dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in February 2020 in Nanjing was 17.8% lower than the average in 2018–2019. The average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2018 and 2019 was 451.0 and 470.1 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>. In 2020, the average dry deposition flux was 368.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>, which was 20.0% lower than that in 2018–2019.

The dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ contributed by gas and particle phase PCDD/Fs, respectively, in Shanghai and Nanjing is shown in Table 4. It can be seen that the dry deposition flux was mainly contributed by the particle phase (averaged 98.88%), and the contribution of the particle phase fraction on the PCDD/F dry deposition flux decreased with increases in the temperature. This could have been due to the fact that more of the particle-bound PCDD/Fs were shifted to the gas phase during the high temperature season. The proportion of particle phase deposition flux in 2020 (average 98.84%) of 4.65% and 7.26% was smaller than that in 2018–2019 (average 98.89% and 98.92%, respectively). Effective control measures can thus significantly reduce particulate pollutants in the atmosphere and improve the air quality.

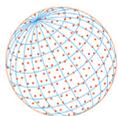
On average, more than 98.88% of the total PCDD/Fs-WHO<sub>2005</sub>-TEQ dry deposition flux was primarily contributed by the particle phase. This was due to the fact that dry deposition of particle phase PCDD/Fs are mainly caused by gravitational settling due to higher dry deposition velocities, while that of gas phase PCDD/Fs are deposited mostly by diffusion, which is due to a lower dry deposition velocity.

It can be seen that seasonal variation of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ dry deposition flux was the highest in winter, followed by spring and summer, in that order. In addition, the annual variations in concentration and the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2020 for both cities (320.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) were lower than those in 2018 (383.6 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) and 2019 (393.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>). It can be seen that the control measures under the epidemic had a significant positive impact on air quality.

## 4. CONCLUSIONS

The results of this study can be summarized as follows:

1. The total PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration in Shanghai in 2018–2019 (0.0291 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>) was 13.9% in magnitude higher than that in 2020 (0.0250 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>), and that for Nanjing in 2018–2019 (0.0423 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>) was 20.0% in magnitude higher than that in 2020 (0.0338 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>).
2. In Shanghai, the average total-PCDD/Fs-WHO<sub>2005</sub>-TEQ concentration (0.0331 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>) in spring and winter were 47.6% of magnitude higher than those in summer (0.0173 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>). Those in Nanjing in spring and winter (0.479 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>) were 53.8% in magnitude higher than in summer (0.222 pg-WHO<sub>2005</sub>-TEQ m<sup>-3</sup>), indicating that the lowest value usually occurs in summer.
3. In Shanghai, the average for the particle phase fractions of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2018–2019 was 50.3%, while the average in 2020 was 47.8%. In Nanjing, the average particle phase fractions of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in 2018–2019 was 57.5%, and in 2020, the average for the particle phase fractions was 53.2%.
4. In Shanghai, the PM<sub>2.5</sub> concentration dropped from 34.7 μg m<sup>-3</sup> in the period 2018–2019 to 31.8 μg m<sup>-3</sup> in 2020, while in Nanjing, it dropped from 40.3 μg m<sup>-3</sup> to 31.3 μg m<sup>-3</sup>. In 2020, the PM<sub>2.5</sub> concentrations in Shanghai and Nanjing were 13.9% and 20.0% lower than the average in 2018–19, respectively. Shanghai and Nanjing had better air quality in 2020 than in

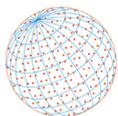


**Table 4.** Monthly gas-bound and particle-bound ratio of the dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai and Nanjing in 2018, 2019, and 2020 (Unit: %).

City	Shanghai		Nanjing		
	Year	Month	Gas-bound Ratio	Particle-bound Ratio	Gas-bound Ratio
2018	Jan.	0.28	99.72	0.12	99.88
	Feb.	0.34	99.66	0.21	99.79
	Mar.	0.73	99.27	0.53	99.47
	Apr.	0.95	99.05	0.87	99.13
	May	1.38	98.62	1.28	98.72
	June	1.82	98.18	1.71	98.29
	July	2.06	97.94	1.99	98.01
	Aug.	2.09	97.91	1.98	98.02
	Sep.	1.86	98.14	1.66	98.34
	Oct.	1.33	98.67	1.03	98.97
	Nov.	1.03	98.97	0.61	99.39
	Dec.	0.51	99.49	0.26	99.74
2019	Jan.	0.35	99.65	0.17	99.83
	Feb.	0.42	99.58	0.22	99.78
	Mar.	0.56	99.44	0.44	99.56
	Apr.	1.01	98.99	0.86	99.14
	May	1.28	98.72	1.17	98.83
	June	1.81	98.19	1.68	98.32
	July	1.98	98.02	1.87	98.13
	Aug.	2.00	98.00	1.95	98.05
	Sep.	1.84	98.16	1.68	98.32
	Oct.	1.32	98.68	1.04	98.96
	Nov.	0.90	99.10	0.61	99.39
	Dec.	0.52	99.48	0.33	99.67
2020	Jan.	0.49	99.51	0.25	99.75
	Feb.	0.72	99.28	0.54	99.46
	Mar.	0.81	99.19	0.63	99.37
	Apr.	0.92	99.08	0.76	99.24
	May	1.40	98.60	1.34	98.66
	June	1.89	98.11	1.79	98.21
	July	1.95	98.05	2.38	97.62
	Aug.	2.07	97.93	2.02	97.98
	Sep.	1.76	98.24	1.63	98.37
	Oct.	1.42	98.58	1.11	98.89
	Nov.	1.05	98.95	0.72	99.28
	Dec.	0.44	99.56	0.21	99.79

2018–19, especially in February. This was due to the positive effects of COVID-19 restrictions on improvement in air quality.

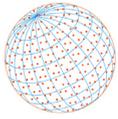
5. The average PM<sub>2.5</sub> concentrations (41.8 μg m<sup>-3</sup>) in spring and winter were 41.0% higher than those in summer (24.7 μg m<sup>-3</sup>) in Shanghai, and for Nanjing in spring and winter (47.3 μg m<sup>-3</sup>), they were 52.3% higher than that in summer (22.6 μg m<sup>-3</sup>).
6. The annual PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ content in Shanghai in 2020 (0.312 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) was 8.6% lower than the average in 2018-2019 (0.342 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>). In Nanjing in 2020 (0.489 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>), it was slightly lower than that in 2018-2019 (0.493 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>).
7. In Shanghai, the average PM<sub>2.5</sub>-bound total PCDD/Fs-WHO<sub>2005</sub>-TEQ contents (0.438 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) in spring and winter was 77.5% higher than that in summer (0.098 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>), and that for Nanjing in spring and winter (0.629 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>) was 73.2% higher than that in summer (0.169 ng-WHO<sub>2005</sub>-TEQ g<sup>-1</sup>).



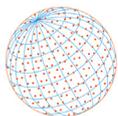
8. The average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ in Shanghai in 2018–2019 (316.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) was higher than that in 2020 (272.5 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>), and that in Nanjing in 2018-2019 (460.5 WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) was higher than that in 2020 (368.4 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>).
9. In Shanghai, the average dry deposition flux of total-PCDD/Fs-WHO<sub>2005</sub>-TEQ (360.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) in spring and winter was 47.6% higher than that in summer (188.8 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>), and that in Nanjing in spring and winter (521.8 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>) was 53.8% higher than that in summer (241.3 pg WHO<sub>2005</sub>-TEQ m<sup>-2</sup> month<sup>-1</sup>).

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