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Atmospheric $PM_{2.5}$, Total PCDD/Fs-WHO_{2005}-TEQ Level and Wet Deposition: Cases of Jinan and Weihai Cities, China

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ABSTRACT

The atmospheric $PM_{2.5}$, total PCDD/Fs-WHO_{2005}-TEQ concentrations, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, gas-particle partitioning and wet deposition of PCDD/Fs in Jinan and Weihai, from 2015 to 2017, were investigated, respectively. Furthermore, a regression analysis of the air quality index (AQI) and total PCDD/Fs-WHO_{2005}-TEQ concentrations was studied as well. From 2015–2017, the three-year average $PM_{2.5}$ concentrations in Jinan and Weihai were 76.1 and 32.9 µg m^{-3}, respectively. The three-year average total PCDD/Fs-WHO_{2005}-TEQ concentrations in Jinan and Weihai were 0.0866 and 0.0396 pg-WHO_{2005}-TEQ m^{-3}, respectively, indicating that maritime climate regulation contributes to the spread of pollutants. The three-year average fraction of gas phase total PCDD/Fs-WHO_{2005}-TEQ concentrations in spring, summer, autumn, and winter in Jinan were 26.1%, 63.8%, 30.5% and 4.62%, respectively; while, those in Weihai were 30.7%, 74.5%, 44.4%, and 8.41%, respectively. In 2016, the annual wet deposition fluxes in Jinan and Weihai were 1288 and 483 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, respectively. The annual scavenging ratios of total PCDD/Fs-WHO_{2005}-TEQ were 33080 and 30870 in Jinan and Weihai, respectively. The total concentration of PCDD/Fs-WHO_{2005}-TEQ increased with an increase of AQI and showed a positive correlation; the R^2 values of three years (2015, 2016, and 2017) were 0.873, 0.847, and 0.744, respectively. Therefore, knowing the AQI value, the total concentration of PCDD/Fs-WHO_{2005}-TEQ in the ambient air can be roughly estimated using a regression equation.

Keywords: $PM_{2.5}$; PM_{10}; PCDD/Fs; Wet deposition; Scavenging ratio; AQI; Regression analysis; TEQ.

INTRODUCTION

Atmospheric PM_{10}, $PM_{2.5}$, and $PM_{1.0}$ are known as particulate matter (PM), which is a general term for various solid and liquid particulate materials present in the atmosphere. All kinds of particulate matter are evenly dispersed in the air, forming a relatively stable and huge suspension system, known as an aerosol system. Therefore, atmospheric particulate matter is also known as atmospheric aerosol (Ghosh *et al.*, 2014). According to aerodynamic diameter, atmospheric particulate matter can be divided into total suspended particulate matter (TSP), PM_{10}, and $PM_{2.5}$. The World Health Organization (WHO) calls PM_{10} as thoracic particle, and the term respirable particles as defined by Pooley and Gibbs (1996) refers to particles that can enter the human alveoli, meaning $PM_{2.5}$. That of $PM_{2.5}$ can be inhaled to cause pneumonia and can dissolve in the bloodstream and cause heart and reproductive system diseases (Yang *et al.*, 2017). The particle size of $PM_{2.5}$ is small, so it is easier for it to enrich toxic substances, and it stays in the ambient air for a long time. In addition to significantly reducing the visibility of the atmosphere, which has a significant impact on the air environment. The $PM_{2.5}$ by human inhalation will also cause significant health issues. The annual average guideline value of $PM_{2.5}$ regulated by the World Health Organization (WHO) Air Quality Guidelines is 10 µg m^{-3}.

The full name of dioxin is polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). A benzene ring in which two chlorine atoms are bonded by two oxygen atoms is polychlorinated dibenzodioxins (PCDDs), and a benzene ring in which two

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chlorine atoms are bonded by one oxygen atom is polychlorinated dibenzofurans (PCDFs). PCDD/Fs is one kind of the persistent organic pollutants (POPs) (Schecter *et al.*, 2006; Redfern *et al.*, 2017). Natural microbes and hydrolysis have little effect on the molecular structure of dioxins. Therefore, it is difficult for natural dioxin to be eliminated by natural degradation. Moreover, the PCDD/Fs diffuses rapidly in the atmosphere and often exists in the ambient air, soil and water with fine particles and gas phase. Therefore, it is very important to carry out regional monitoring of the atmospheric environment for PCDD/F pollution (Fiedler *et al.*, 1992; Hutzinger *et al.*, 1992). The PCDD/Fs includes 210 congeners. The toxicity of 2,3,7,8 TCDD is approximately 900 times that of arsenic (Van den Berg *et al.*, 1998; Li *et al.*, 2016). It is known as the most toxic poison of the century. One ten thousandth or even one hundredth of a gram of dioxin can cause serious harm to health (Li *et al.*, 2008). It is classified as a human carcinogen by the International Cancer Center (IARC, 1997; US Department of Health and Human Services, 2000; Paustenbach, 2002; Cole *et al.*, 2003). In addition to carcinogenic toxicity, PCDD/Fs also have reproductive toxicity and genetic toxicity, which directly endanger the health and life of future generations (Zhou *et al.*, 2002). The toxicity equivalent factor (TEF) is given here, and the other 16 homologs have TEF values set relative to this. In addition, these TEFs are multiplied by the corresponding individual homolog concentrations in order to generate toxicity equivalents (TEQs) (Lohmann and Jones, 1998; Van den Berg *et al.*, 1998). The studies found that the PCDD/F concentration can be reduced in ambient air by dry and wet deposition (Huang *et al.*, 2011; Mi *et al.*, 2012; Redfern *et al.*, 2017).

Wet deposition refers to the process of removing particulate matter from the atmosphere by rain, snow, and other form of water droplets. It is an effective method for removing atmospheric particulate matter and trace gaseous pollutants (Lohmann and Jones, 1998; Huang *et al.*, 2011b). In addition, the main cause of higher chlorinated homologous of persistent organic pollutants in the environment is due to wet deposition (Shih *et al.*, 2006; Lin *et al.*, 2010a; Wang *et al.*, 2010). The ambient temperature, rainfall, vapor pressure, and particle size may also be the factors affecting the wet deposition process (Wu *et al.*, 2009; Wang *et al.*, 2010; Chang *et al.*, 2004). The wet deposition of PCDD/Fs is related to the intensity and amount of precipitation. Wet deposition is typically evaluated using the total scavenging ratio (Stot), which is the ratio of the concentration of a given compound in the precipitation to the concentration in the atmosphere.

The AQI (Air Quality Index) is a parameter that reports the daily air quality and describes the extent to which air is clean or contaminated, as well as the impact of the current air quality on human health. The focus of the AQI is to assess the health effects of breathing air for hours or days since people may be breathing polluted air for hours or days. The EPA calculates the air quality index through five major pollution criteria, including ground-level ozone, particulate matter, carbon monoxide, sulfur dioxide, and nitrogen dioxide. The Environmental Protection Agency established national environmental air quality standards to protect public health for five pollutants. Ground ozone and particles in the air constitute the greatest threat to human health in China. AQI is divided into six levels, from first, second, third grade light pollution, and fourth grade moderate pollution, where grades 5 and 6 are considered heavily polluted. The air pollution index is divided into 0–50, 51–100, 101–150, 151–200, 201–300, and more than 300.

This study investigated two cities in China, Jinan and Weihai. From 2015 to 2017, $PM_{2.5}$ data from two cities were obtained from the environmental monitoring website. The total concentrations of PCDD/Fs-WHO_{2005}-TEQ in the environment were estimated using the equations given in a previous study (Tang *et al.*, 2017), which demonstrated the relationship between the PM_{10} value and the total PCDD/F concentration. Then, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content was calculated using the gas-particle phase distribution formula. This study also investigated the monthly and seasonal variation in atmospheric PCDD/F wet deposition flux. The monthly average total PCDD/Fs-WHO_{2005}-TEQ concentration in the rain and the wet deposition scavenging ratio in these two cities was also discussed. In addition, a linear regression analysis was performed on total PCDD/Fs-WHO_{2005}-TEQ and AQI in ambient air.

METHODS

Two cities, Jinan and Weihai in Shandong province, China, were evaluated in this study. The monthly average concentrations of $PM_{2.5}$ and PM_{10}, monthly average temperature, and monthly average precipitation in the two cities were obtained from the environmental monitoring website and the China Statistical Yearbook.

Through a regression analysis of PM_{10} concentrations and total PCDD/F concentrations, it was concluded that there is a high correlation between them (Tang *et al.*, 2017). The following two regression equations are included:

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

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

Y_1, Y_2: total PCDD/Fs concentration (pg m^{-3});
x: PM_{10} concentration in ambient air (μg m^{-3}).

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

Gas-Particle Partitioning

The concentration of PCDD/Fs in the gas and particle phases were calculated by using Eq. (3) (Yamasaki *et al.*, 1982; Pankow, 1987; Pankow and Bidleman, 1991, 1992):

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

K_p: temperature-dependent partitioning constant (m^3 μg^{-1});
TSP: concentration of total suspended particulate matter,

multiplied by PM_{10} concentration with 1.24 (μg m^{-3});
F: concentration of the compounds of interest bound to particles (pg m^{-3});
A: gaseous concentration of the compound of interest (pg m^{-3}).

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

$$\log K_p = m_r \times \log P_L^0 + b_r \tag{4}$$

P_L^0: subcooled liquid vapor pressure (Pa);
m_r: cited slope;
b_r: cited y-intercept.

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

A previous study correlated the P_L^0 of PCDD/Fs with gas chromatographic retention indexes (GC-RI) on a nonpolar (DB-5) GC-column using p,p′-DDT as a reference standard. The correlation has been redeveloped as the following (Hung *et al.*, 2002):

$$\log P_L^0 = \frac{-1.34(RI)}{T} + 1.67\times10^{-3}(RI) - \frac{1320}{T} + 8.087 \tag{5}$$

RI: gas chromatographic retention indexes developed by Donnelly *et al.* (1987) and Hale *et al.* (1985);
T: ambient temperature (K).

Scavenging Ratios

Studies have shown that for micro-soluble trace organic compounds, such as PCDD/Fs and other semi-volatile organic compounds, that equilibrium partitioning occurs between the compound in the gas phase and that in a falling raindrop (Ligocki *et al.*, 1985a, b; Cheruiyot *et al.*, 2015, 2016; Redfern *et al.*, 2017). The scavenging ratio is defined as the concentration of the pollutant in the raindrop divided by the concentration of the same pollutant in the surrounding air during precipitation. The gas scavenging ratio, S_g, can be estimated by:

$$S_g = \frac{RTP}{H} \tag{6}$$

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

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

$$S_g = \frac{C_{rain,dis}}{C_g} \tag{7}$$

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

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

$$S_p = \frac{C_{rain,partical}}{C_p} \tag{8}$$

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

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

$$S_{tot} = S_g(1 - \phi) + S_p \times \phi \tag{9}$$

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

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

Wet Deposition

Wet deposition refers to the process of the removal of particulate matter from the atmosphere by rain, snow, and the like. It is an effective method for removing atmospheric particulate matter and trace gaseous pollutants (Lohmann and Jones, 1998; Huang *et al.*, 2011b). The wet deposition flux of PCDD/Fs is a combination of both vapor dissolution into rain and removal of suspended particulates through precipitation (Bidleman *et al.*, 1988; Koester and Hites, 1992).

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

$$F_{w,T} = F_{w,dis} + F_{w,p} \tag{10}$$

$$F_{w,dis} = F_{rain,dis} + Rainfall \tag{11}$$

$$F_{w,p} = F_{rain,partical} + Rainfall \tag{12}$$

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

RESULTS AND DISCUSSION

$PM_{2.5}$ Concentration

From 2015 to 2017, the corresponding monthly average $PM_{2.5}$ concentrations in Jinan and Weihai are shown in Figs. 1(a), 1(b) and 1(c).

For Jinan, the $PM_{2.5}$ concentration was the highest in 2015, which was in the range of 57.0–159 µg m^{-3}, and with an average of 89.4 µg m^{-3}; then in 2016, ranged from 41.0 to 128 µg m^{-3} and averaged 74.9 µg m^{-3}; and during 2017, ranged between 37.0 and 131 µg m^{-3} and averaged 64.0 µg m^{-3}. By comparing the annual average $PM_{2.5}$ concentrations, we found that the annual average $PM_{2.5}$ concentration in 2016 decreased by about 16.2% than 2015 and the annual average $PM_{2.5}$ concentration in 2017 decreased by about 14.6% than 2016. The slow decline of $PM_{2.5}$ level may be due to the reduction of coal consumption and the use of new energy vehicles. Although the ambient air quality in Jinan has improved significantly, the concentration of $PM_{2.5}$ in Jinan is still far higher than the WHO air quality regulated standard (10.0 µg^{-3}) (Wang *et al.*, 2018).

In regard to Weihai, the monthly average $PM_{2.5}$ concentrations ranged from 21.0 to 64.0 µg m^{-3} and averaged 37.4 µg m^{-3} in 2015; and those was in the range of 20.0–46.0 µg m^{-3} and averaged 33.3 µg m^{-3} in 2016; during 2017, the $PM_{2.5}$ concentration ranged between 14.0 and 42.0 µg m^{-3} and averaged 28.1 µg m^{-3}. Between 2015 and 2017, we can see that the annual average $PM_{2.5}$ concentration is highest in 2015, followed by 2016 and the lowest in 2017. The annual average $PM_{2.5}$ concentration in 2016 decreased by 11.0% compared with 2015, and the annual average $PM_{2.5}$ concentration in 2017 decreased by 15.6% compared with 2016. In general, the average concentration of $PM_{2.5}$ in Weihai was 32.9 µg m^{-3} in these three years, and the $PM_{2.5}$ level was much lower than that in Jinan. It was also much lower than Bengbu (61.4 µg m^{-3}) (Wang *et al.*, 2018). This may be due to Weihai is located in the eastern part of Shandong Province, surrounded by the sea

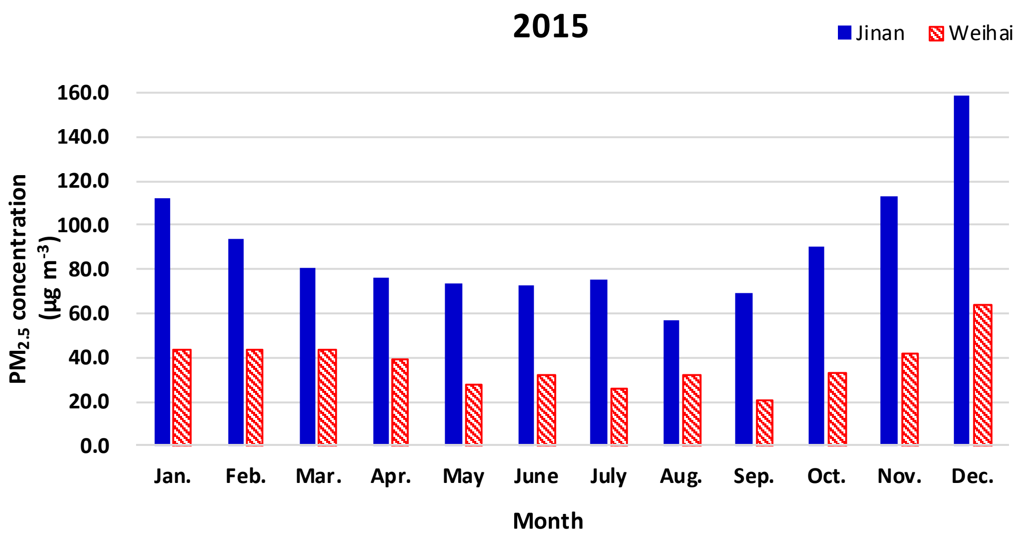

Fig. 1(a). Monthly average atmospheric PM $_{2.5}$ concentration in Jinan and Weihai during 2015.

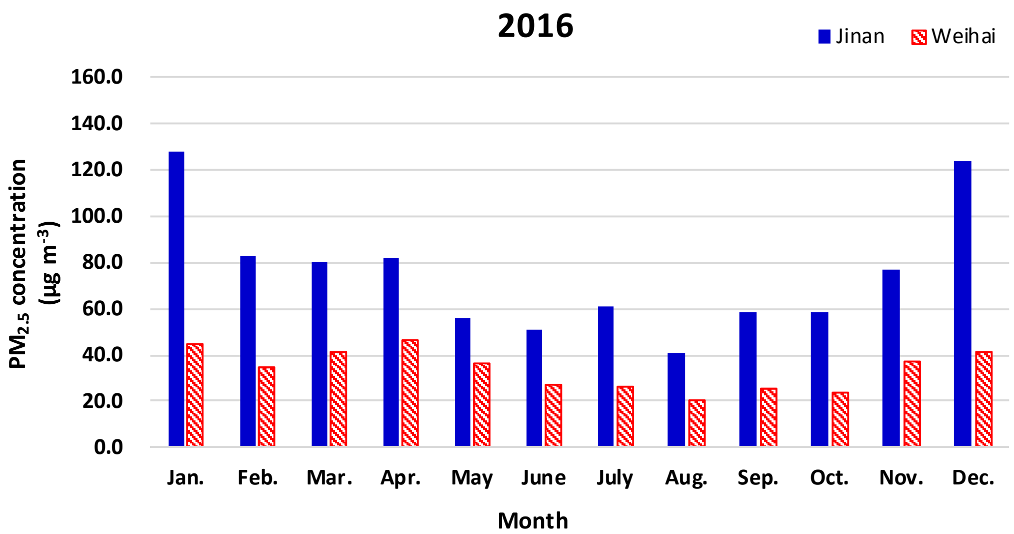

Fig. 1(b). Monthly average atmospheric PM $_{2.5}$ concentration in Jinan and Weihai during 2016.

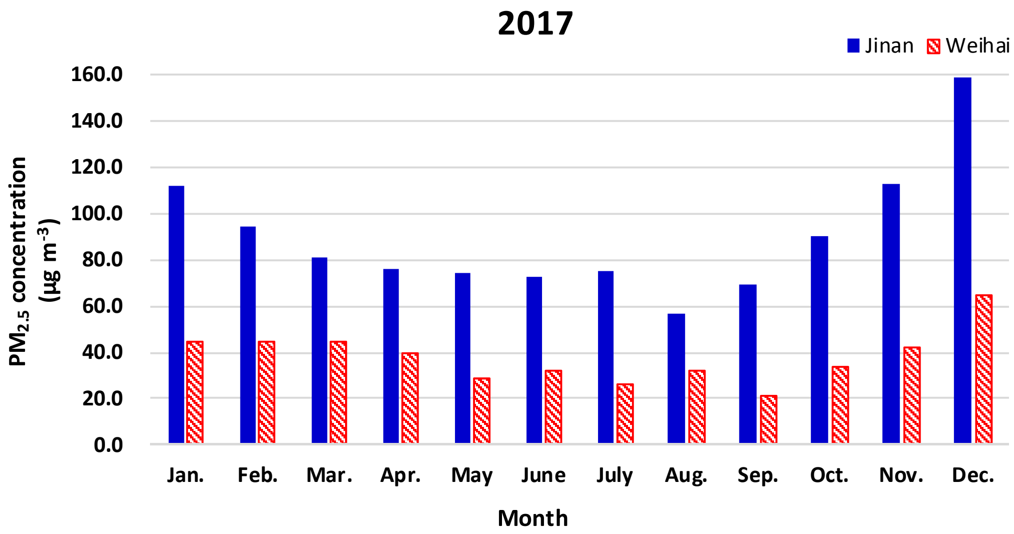

Fig. 1(c). Monthly average atmospheric PM $_{2.5}$ concentration in Jinan and Weihai during 2017.

on three sides and is affected by the ocean. It shows the characteristics of maritime climate such as spring cold, summer warm, autumn air turbulence, winter low temperature, relatively small temperature difference between day and night, long frost-free period, high wind and humidity.

In terms of seasonal changes, the four seasons are divided into spring (March, April, May), summer (June, July, August), autumn (September, October, November), and winter (January, February and December) (Zhao *et al.*, 2018). In Jinan, the average concentrations of $PM_{2.5}$ in the four seasons of 2017 were 61.3, 46.0, 49.3 and 99.3 µg m^{-3}, respectively; those in 2016 were 72.7, 51.0, 64.3, and 112 µg m^{-3}, respectively; and those in 2015 were 77.0, 68.3, 90.7, and 122 µg m^{-3}, respectively. In Weihai, the average concentrations of $PM_{2.5}$ in the four seasons of 2017 were 35.3, 21.3, 20.3, and 35.3 µg m^{-3}, respectively; those in 2016 were 41.0, 24.3, 28.3, and 39.7 µg m^{-3}, respectively, and those in 2015 were 37.0, 30.0, 32.0, and 50.7 µg m^{-3}, respectively. According to the data, we can see that the concentration of $PM_{2.5}$ varies seasonally, where summer is the lowest, and winter is the highest. The three-year average of $PM_{2.5}$ concentrations in Jinan in summer (55.1 µg m^{-3}) was 21.6% lower than that in spring (70.3 µg m^{-3}), 19.1% lower than that in autumn (68.1 µg m^{-3}), 50.4% lower than that in winter (111 µg m^{-3}). The average value of Weihai in summer (25.2 µg m^{-3}) was 33.3% lower than that in spring (37.8 µg m^{-3}), 6.3% lower than that in autumn (26.9 µg m^{-3}), and 39.9% lower than that in winter (41.9 µg m^{-3}). In the winter, so a large amount of coal is burned, a large amount of soot and waste heat are released, and the inversion of the atmosphere in the vertical direction causes the vertical movement of the air in the atmosphere to be limited, so the stability of the atmosphere is not conducive to the diffusion of pollutants. The summer temperature is higher, and the vertical airflow is more intense, which is conducive to the diffusion of $PM_{2.5}$ in the air. Therefore, it is necessary to reduce the burning of fossil fuels such as coal, use clean energy, elevate the automobile exhaust gas treatment technology, and reduce the concentration of $PM_{2.5}$ in the ambient air.

Total PCDD/Fs-WHO$_{2005}$-TEQ Concentration in Ambient Air

Figs. 2(a), 2(b) and 2(c) show the concentrations of total PCDD/Fs-WHO$_{2005}$-TEQ in the ambient air of Jinan and Weihai during the years 2015 and 2017, respectively. Eqs. (1) and (2) can be used to calculate total PCDD/F concentration based on the PM_{10} concentration.

In Jinan, the total PCDD/Fs-WHO$_{2005}$-TEQ concentrations in 2015 ranged between 0.0511 and 0.130 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0955 pg-WHO$_{2005}$-TEQ m^{-3}, and those was in the range of 0.0449–0.136 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0871 pg-WHO$_{2005}$-TEQ m^{-3} in 2016, during 2017 the total PCDD/Fs-WHO$_{2005}$-TEQ concentrations ranged between 0.0398 and 0.121 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0773 pg-WHO$_{2005}$-TEQ m^{-3}. In Weihai, the total PCDD/Fs-WHO$_{2005}$-TEQ concentrations ranged between 0.0255 and 0.104 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0462 pg-WHO$_{2005}$-TEQ m^{-3} in 2015. They ranged between 0.0186 and 0.0797 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0381 pg-WHO$_{2005}$-TEQ m^{-3} in 2016 and ranged between 0.0155 and 0.0578 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0344 pg-WHO$_{2005}$-TEQ m^{-3} in 2017. Comparing with previous studies, the total PCDD/Fs-WHO$_{2005}$-TEQ concentrations in Weihai were similar to Kaohsiung area, which was ranged 0.0210 between 0.0770 pg-WHO$_{2005}$-TEQ m^{-3} and averaged 0.0480 pg WHO$_{2005}$-TEQ m^{-3} in 2014 (Lee *et al.*, 2016). The decrease in the total PCDD/Fs-WHO$_{2005}$-TEQ concentration year by year may be due to the fact that general awareness of environmental protection has gradually strengthened, and the control of air pollution has achieved specific effects. However, it is necessary to continue to implement energy conservation and emission reduction. It can be seen that the total PCDD/Fs-WHO$_{2005}$-TEQ concentration in Weihai is much lower than that in Jinan, indicating that maritime climate regulation contributes to the spread of pollutants.

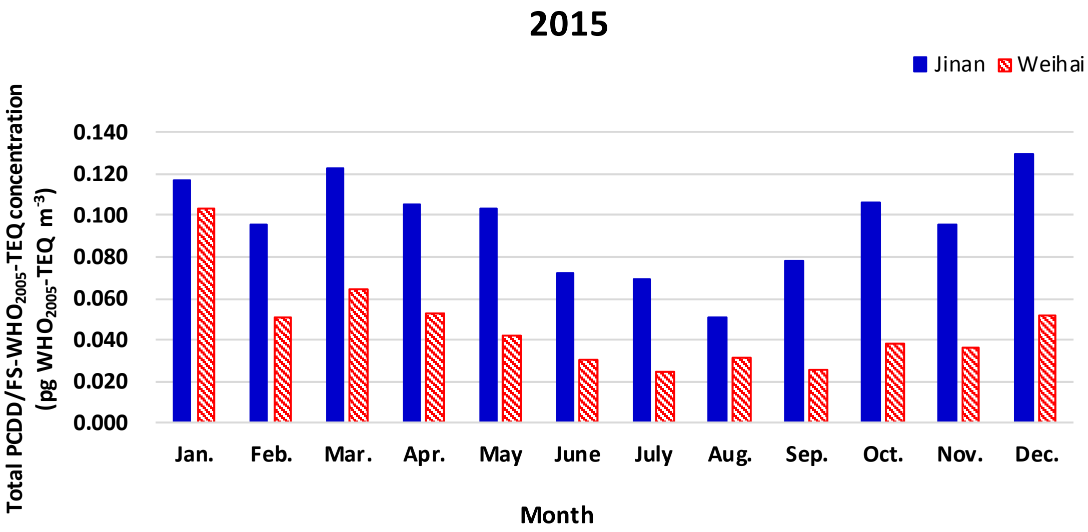

Fig. 2(a). Monthly average total PCDD/Fs-WHO_{2005}-TEQ concentration in Jinan and Weihai during 2015.

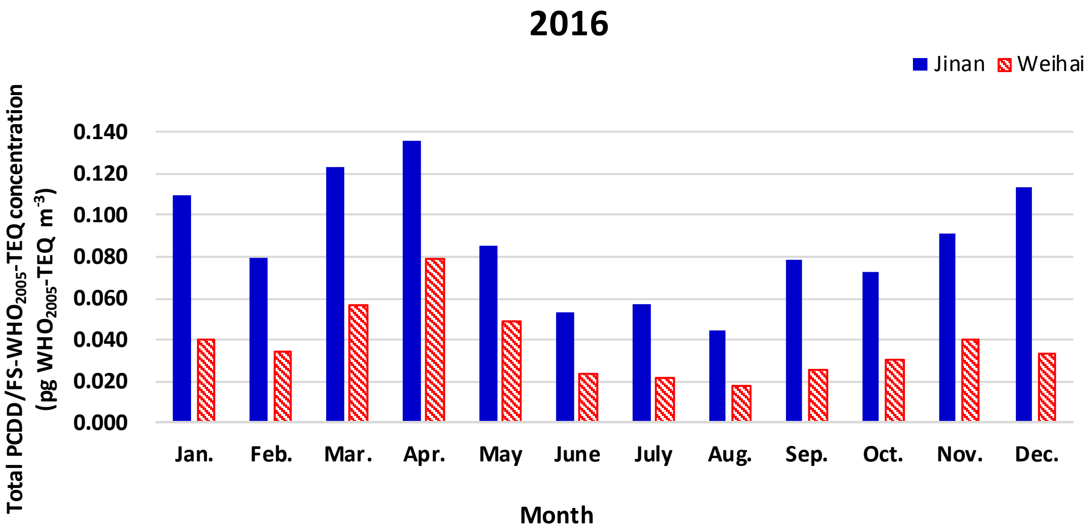

Fig. 2(b). Monthly average total PCDD/Fs-WHO_{2005}-TEQ concentration in Jinan and Weihai during 2016.

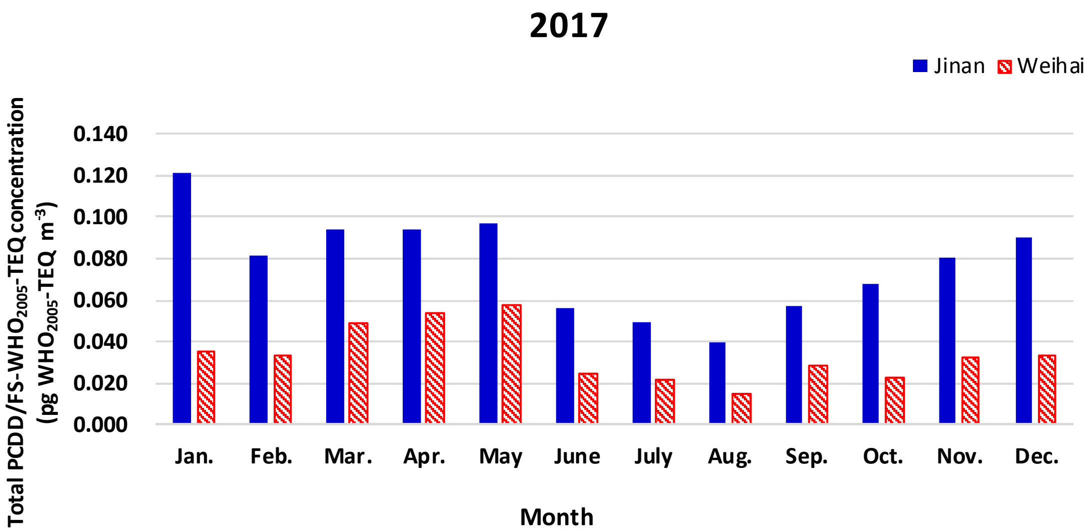

Fig. 2(c). Monthly average total PCDD/Fs-WHO_{2005}-TEQ concentration in Jinan and Weihai during 2017.

The changes in spring, summer, autumn, and winter were studied, where for Jinan, the average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the four seasons of 2015 were 0.110, 0.0641, 0.0934, and 0.103 pg-WHO_{2005}-TEQ m^{-3}; and those in 2016 were 0.115, 0.0518, 0.0807, and 0.101 pg-WHO_{2005}-TEQ m^{-3}. Those in 2017 were 0.0947, 0.0484, 0.0686, and 0.0977 pg-WHO_{2005}-TEQ m^{-3}. For Weihai, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in the four seasons of 2015 were 0.0534, 0.0293, 0.0336, and 0.0687 pg-WHO_{2005}-TEQ m^{-3}; and those in 2016 were 0.0620, 0.0217, 0.0322, and 0.0364 pg-WHO_{2005}-TEQ m^{-3}. Those in 2017 were 0.0538, 0.0210, 0.0283, and 0.0344 pg-WHO_{2005}-TEQ m^{-3}. It can be seen that the total PCDD/Fs-WHO_{2005}-TEQ concentrations in both Jinan and Weihai were the lowest in summer, which is consistent with the study of Wuhu and Bengbu (Wang *et al.*, 2018).

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

Figs. 3(a), 3(b), and 3(c) shows the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in Jinan and Weihai from 2015 to 2017.

In Jinan, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content ranged from 0.266 to 1.08 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.623 ng-WHO_{2005}-TEQ g^{-1} in 2015, ranged between 0.269 and 1.08 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.654 ng-WHO_{2005}-TEQ g^{-1} in 2016, ranged between 0.238 and 1.05 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.665 ng-WHO_{2005}-TEQ g^{-1} in 2017. As for Weihai, the content ranged between 0.0968 and 0.693 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.313 ng-WHO_{2005}-TEQ g^{-1} in 2015. In 2016, the level of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ in Weihai ranged between 0.147 and 1.37 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.629 ng-WHO_{2005}-TEQ g^{-1}. In 2017, those were between 0.137 and 0.943 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.591 ng-WHO_{2005}-TEQ g^{-1}.

In 2015, the lowest four months of $PM_{2.5}$ concentration occurred in May (74.0 µg m^{-3}), June (73.0 µg m^{-3}), August (57.0 µg m^{-3}), and September (69.0 µg m^{-3}) in Jinan, accompanied by the lowest four months of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, which was 0.342,

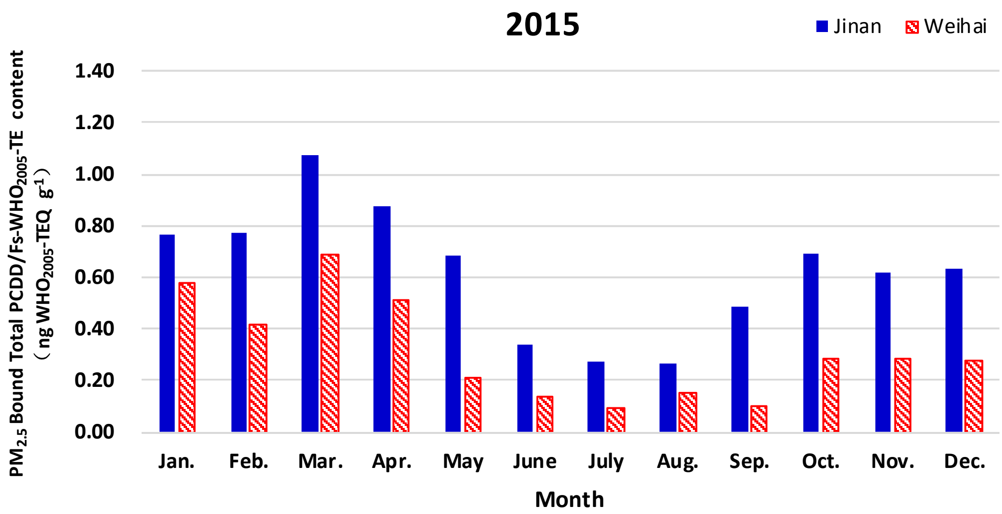

Fig. 3(a). $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content (ng-WHO_{2005}-TEQ g^{-1}) of Jinan and Weihai during 2015.

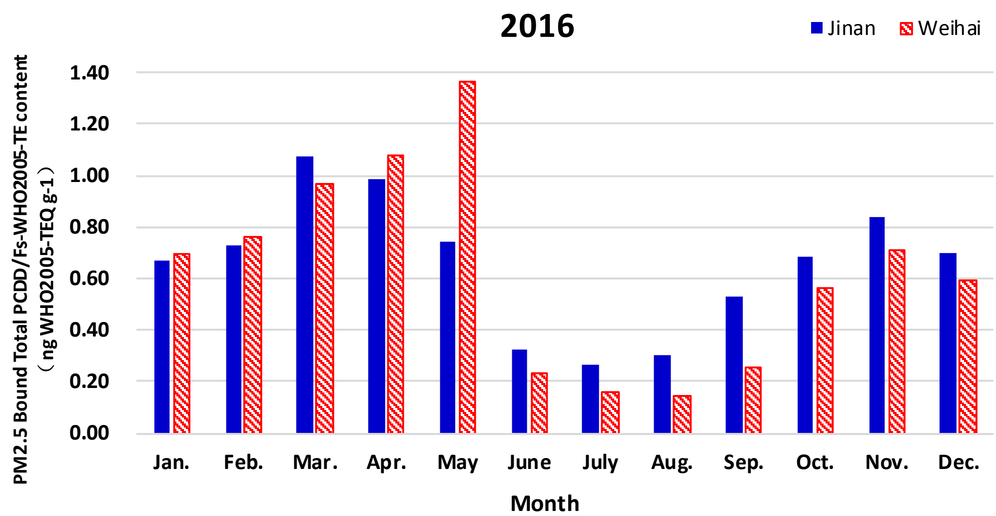

Fig. 3(b). $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content (ng-WHO_{2005}-TEQ g^{-1}) of Jinan and Weihai during 2016.

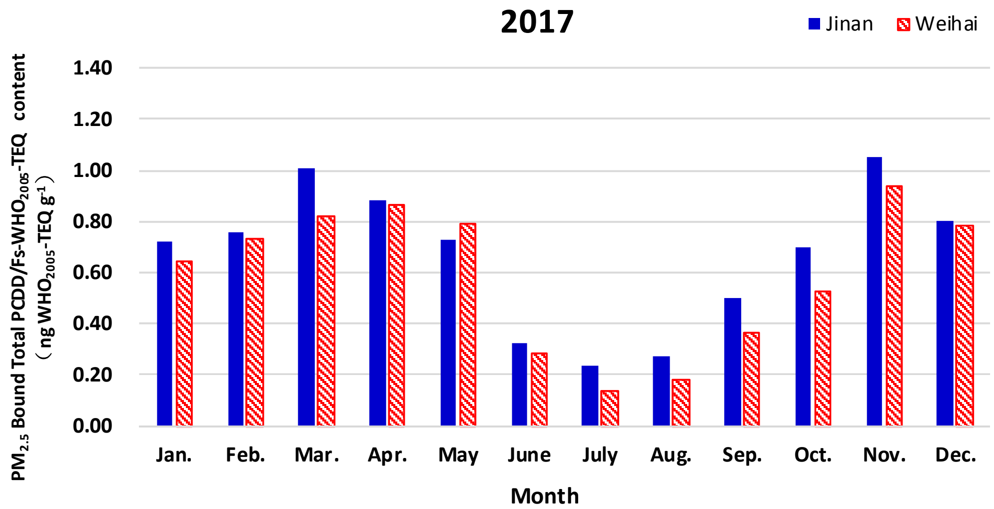

Fig. 3(c). $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content (ng-WHO_{2005}-TEQ g^{-1}) of Jinan and Weihai during 2017.

0.276, 0.266 and 0.484 ng-WHO_{2005}-TEQ g^{-1} in June, July, August, and September; and the lowest four months of $PM_{2.5}$ concentration occurred in May (28.0 µg m^{-3}), June (32.0 µg m^{-3}), July (26.0 µg m^{-3}), and September (21.0 µg m^{-3}) in Weihai, accompanied by the lowest four months of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of 0.137, 0.0968, 0.156 and 0.102 ng-WHO_{2005}-TEQ g^{-1} in June, July, August and September, respectively. However in 2016, the lowest four months of $PM_{2.5}$ concentration occurred in May (56.0 µg m^{-3}), June (51.0 µg m^{-3}), August (41.0 µg m^{-3}), and September (58.0 µg m^{-3}) in Jinan, and in June (27.0 µg m^{-3}), August (20.0 µg m^{-3}), September (25.0 µg m^{-3}) and October (23.0 µg m^{-3}) in Weihai. However, the lowest four months of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content occurred in June, July, August, and September in both Jinan and Weihai, and the values were 0.322, 0.269, 0.300, 0.529 ng-WHO_{2005}-TEQ g^{-1} and 0.234, 0.164, 0.147, 0.258 ng-WHO_{2005}-TEQ g^{-1}, respectively. In 2017, the lowest four months of $PM_{2.5}$ concentration occurred in June (53.0 µg m^{-3}), July (48.0 µg m^{-3}), August (37.0 µg m^{-3}), and September (38.0 µg m^{-3}) in Jinan, and in August (14.0 µg m^{-3}), September (21.0 µg m^{-3}), October (18.0 µg m^{-3}), and November (22.0 µg m^{-3}) in Weihai. However, the lowest four months of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content occurred in June, July, August, and September in both Jinan and Weihai, for which the levels were 0.328, 0.238, 0.273, 0.497 ng-WHO_{2005}-TEQ g^{-1} and 0.290, 0.137, 0.184, 0.370 ng-WHO_{2005}-TEQ g^{-1}, respectively. Comparing $PM_{2.5}$ concentrations with $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, it can be seen that there is no close relationship between the two trends, concurring with similar results in previous studies (Xing *et al.*, 2017; Wang *et al.*, 2018).

In terms of seasonal changes, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in the four seasons (spring, summer, fall, and winter) of 2015 was 0.878, 0.294, 0.598, and 0.724 ng-WHO_{2005}-TEQ g^{-1} in Jinan, and in 2016, it was 0.935, 0.297, 0.686, and 0.699 ng-WHO_{2005}-TEQ g^{-1}. In 2017, it was 0.871, 0.280, 0.749, and 0.761 ng-WHO_{2005}-TEQ g^{-1}. For Weihai, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in the four seasons of 2015 was 0.474, 0.130, 0.224, and 0.424 ng-WHO_{2005}-TEQ g^{-1}, while in 2016, it was 1.14, 0.182, 0.513, and 0.684 ng-WHO_{2005}-TEQ g^{-1}, and in 2017, it was 0.827, 0.204, 0.613, and 0.722 ng-WHO_{2005}-TEQ g^{-1}. It can be seen that the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in Jinan and Weihai was the lowest in summer. This was because the ambient temperature in summer was higher, so the PCDD/Fs combined with the particles were more evaporated to the gas phase. However, in the cold season, the low temperature is not conducive to the PCDD/Fs combing with the particles.

Gas-Particle Partitioning of PCDD/Fs

The gas-particle partitioning of PCDD/Fs plays an important role in the efficiency of atmospheric removal by wet and dry deposition (Bidleman *et al.*, 2000; Cheruiyot *et al.*, 2015, 2016; Redfern *et al.*, 2017). The vapor pressure, temperature, PCDD/F concentrations, and the ambient air particulate concentration are also important factors (Hoff *et al.*, 1996). The gas-particle partition was calculated from the atmospheric date using Eqs. (3), (4), and (5). Figs. 4(a), 4(b), and 4(c) shows the seasonal gas-particle partitioning of total PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Jinan and Weihai from 2015 to 2017. In Jinan, the three-year average temperatures in the four seasons were 17.1, 27.0, 16.0, and 2.67°C during 2015–2017. In the seasonal changes of gas-particle partitioning, the average fractions of gas phase total PCDD/Fs-WHO_{2005}-TEQ concentrations in the four seasons were 26.1%, 63.8%, 30.5% and 4.62%. In Weihai, the seasonal average temperatures were 15.2, 25.8, 15.9, and 2.36°C in the four seasons from 2015 to 2017; the average fractions of gas phase total PCDD/Fs-WHO_{2005}-TEQ concentrations in the four seasons were 30.7%, 74.5%, 44.4%, and 8.41%, respectively. Comparing the three-year gas-particle partitioning of PCDD/Fs in Jinan and Weihai, it can be seen that the gas phase total PCDD/Fs-WHO_{2005}-TEQ seasonal trends in the two cities were consistent, which was the highest in summer and the lowest in winter. Previous studies have reached similar conclusions, indicating that gas phase PCDD/Fs have

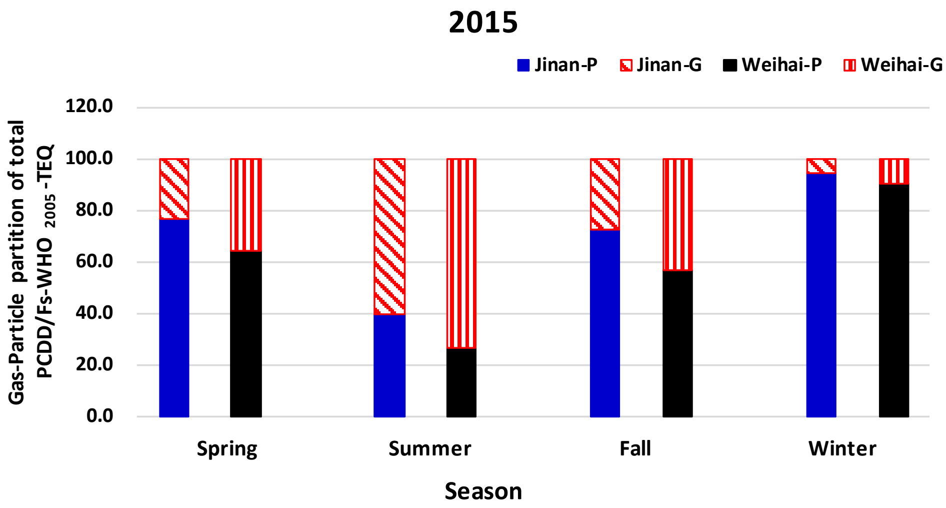

Fig. 4(a). Seasonal variations of gas-particle partition of total PCDD/Fs-WHO$_{2005}$-TEQ in the ambient air in Jinan and Weihai during 2015.

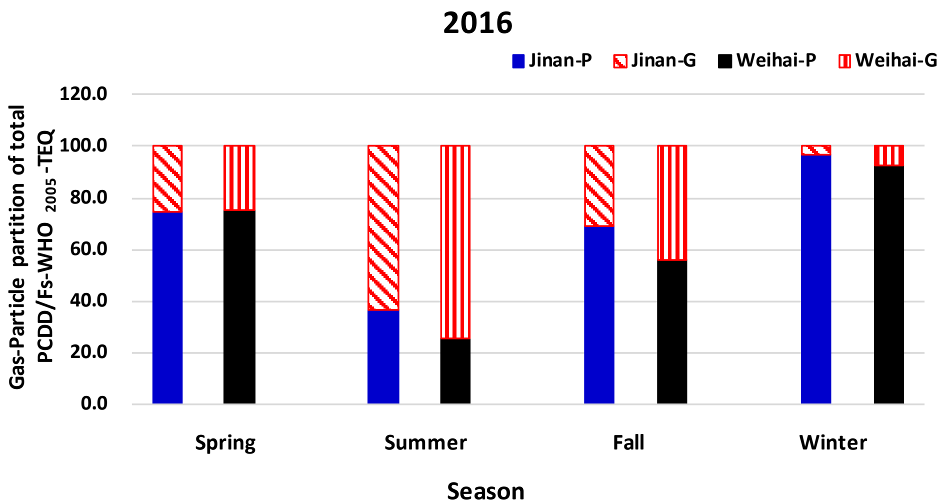

Fig. 4(b). Seasonal variations of gas-particle partition of total PCDD/Fs-WHO$_{2005}$-TEQ in the ambient air in Jinan and Weihai during 2016.

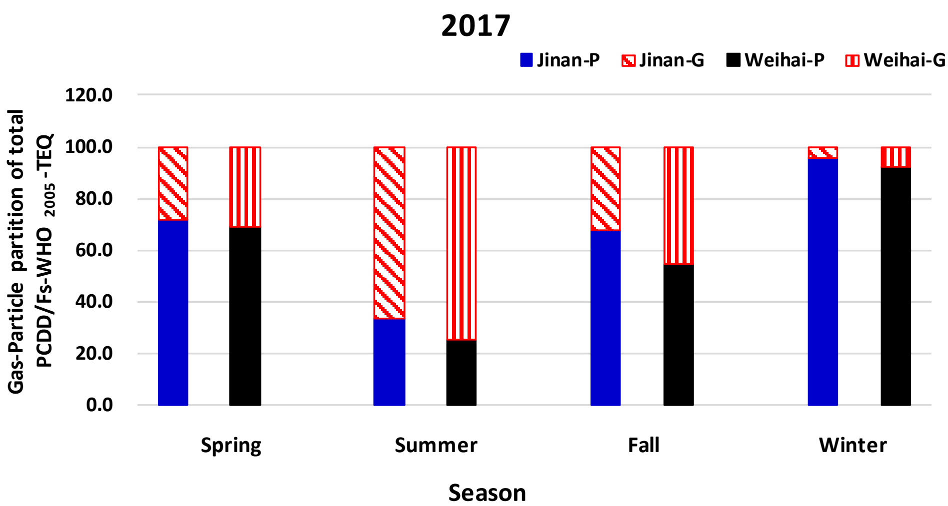

Fig. 4(c). Seasonal variations of gas-particle partition of total PCDD/Fs-WHO$_{2005}$-TEQ in the ambient air in Jinan and Weihai during 2017.

higher fraction in summer than in winter (Wang *et al.*, 2010; Huang *et al.*, 2011; Lee *et al.*, 2016; Wang *et al.*, 2018; Zhao *et al.*, 2018). This may be due to the fact that lower molecular weight PCDD/Fs usually have a higher vapor pressure (Wang *et al*, 2010; Huang *et al*, 2011a). When the temperature is high in summer, the proportion of PCDD/Fs in the gas phase rises and when the temperature is low in winter, some PCDD/Fs in the gas phase are transferred to the particle phase, so the fraction of PCDD/Fs in the gas phase increases with the increases in temperature.

Wet Deposition

Wet deposition removes particle-phase and vapor-phase PCDD/Fs from the atmosphere through rainfall, cloud droplets, and snow (Lee *et al.*, 2016). Fig. 5(a) shows the monthly average wet deposition fluxes of total PCDD/Fs-WHO-TEQ for Jinan and Weihai in 2016. The monthly average wet deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Jinan and Weihai ranged between 0 and 245 and between 11.6 and 105 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. The annual total wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ for Jinan and Weihai were 1288 and 483 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ respectively. It can be seen that the annual wet deposition fluxes of Jinan in 2016 were approximately 2.67 times higher than those in Weihai. Compared with the other cities in China, the annual wet deposition fluxes were varied in Wuhu (1538 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), Shijiazhuang (622 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$), and Guangzhou (570 pgWHO_{2005}-TEQ m^{-2} $year^{-1}$) (Chen *et al.*, 2017; Zhu *et al.*, 2017a, b; Wang *et al.*, 2018). Based on monthly $PM_{2.5}$ concentrations and rainfall data, it can be seen that the values for $PM_{2.5}$ concentration and rainfall were 74.9 μg m^{-3} and 1008 mm in Jinan, respectively, and were 33.3 μg m^{-3} and 535 mm in Weihai. However, the higher $PM_{2.5}$ concentrations with less rainfall occurred in Shijiazhuang (88.0 μg m^{-3} and 535 mm), and the lower $PM_{2.5}$ concentrations accompanied by more rainfall in Guangzhou (38.9 μg m^{-3} and 2234 mm) have resulted in a lower wet deposition flux. (Wang *et al.*, 2018). These results indicate that $PM_{2.5}$ and rainfall are related to wet deposition but are not the main influencing factors. PM_{10}, snow, temperature, and wind speed are also some of the key factors affecting the total PCDD/Fs-WHO_{2005}-TEQ wet deposition in environment (Wang *et al.*, 2010; Huang *et al.*, 2011b; Suryani *et al.*, 2015; Zhu *et al.*, 2017).

In 2016, the highest monthly rainfall in Jinan was 421 mm in August. The highest monthly wet deposition flux was 329 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in August, and lower concentration of $PM_{2.5}$ was 41.0 μg m^{-3} in August. The lowest monthly rainfall in Jinan was 0 mm in March. The lowest monthly wet deposition flux was 0 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in March, and the concentration of $PM_{2.5}$ was 80.0 μg m^{-3} in March. As for Weihai, the monthly rainfall was 39.4 mm in April. The highest monthly wet deposition flux was 106 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in April and the highest concentration of $PM_{2.5}$ was 46.0 μg m^{-3} in April; lower monthly rainfall was 28.6 mm in June. The lowest monthly wet deposition flux was 11.6 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ in June, and the lower concentration of $PM_{2.5}$ was 27.0 μg m^{-3} in June. It can be seen that rainfall and $PM_{2.5}$ concentrations may be the key factors influencing the wet deposition flux, the maximum and minimum wet deposition fluxes occurred in the month with the highest and lowest rainfall respectively, which means that rainfall has the greatest impact on wet deposition (Suryani *et al.*, 2015). It can be speculated that the higher rainfall, the greater the wet deposition will be. It can also be inferred that the greater the rainfall, the greater the wet deposition flux, and possibly the lower the $PM_{2.5}$ concentration will be in the atmosphere.

In terms of seasonal changes, the seasonal average wet deposition fluxes and rainfall for the four seasons were 154, 737, 195, 203 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ and 58.0, 819, 76.7, 44.3 mm in Jinan, respectively. In Weihai, the seasonal average wet deposition fluxes and rainfall for the four seasons were 214, 58.5, 84.2, and 127 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ and 115, 215, 114, 92.1 mm, respectively. The wet deposition flux is always lowest in winter, and previous studies have shown that rainfall and rainy days may affect wet deposition flux rather than temperature (Lee *et al.*, 2016).

According to the monthly rainfall and monthly wet deposition fluxes, the total PCDD/Fs-WHO_{2005}-TEQ concentration in the rain can be calculated using the wet deposition fluxes divided by rainfall intensity (Wang *et al.*, 2018). Fig. 5(b) shows the monthly average total PCDD/Fs-WHO_{2005}-TEQ concentrations in the rain in Jinan and Weihai in 2016. During 2016, the monthly average total PCDD/Fs-WHO_{2005}-TEQ concentration in the rain of Jinan and Weihai were ranged between 0.781 and 4.58 and between 0.221 and 2.68 pg WHO_{2005}-TEQ L^{-1}, respectively; and averaged 2.82 and 1.16 pg WHO_{2005}-TEQ L^{-1}, respectively. In terms of seasonal changes, the average concentration of total PCDD/Fs-WHO_{2005}-TEQ in the rain in Jinan and Weihai were 3.77, 0.94, 2.47, 4.09 and 2.03, 0.31, 0.86, 1.42 pg WHO_{2005}-TEQ L^{-1} in the four seasons (spring, summer, fall, and winter), respectively. It can be seen that the lowest value occured in summer and that the highest value occured in winter. This is because there is more rain in summer. Due to the dilution effect, in summer, the more amount of rainfall reached the ground, the lower PCDD/Fs concentration in rainwater.

Fig. 5(c) shows the scavenging ratios (Stot) for Jinan City and Weihai City in 2016. It can be seen that Jinan and Weihai had similar trends of Stot in 2016. The Stot values for Jinan and Weihai ranged between 23420 and 41340 and between 17810 and 40520, respectively. The average Stot values for Jinan and Weihai in 2016 were 33080 and 30870, respectively. The minimum and maximum values of scavenging ratio in two cities occurred in winter and summer, respectively. It can be seen that the scavenging ratio (Stot) is inversely related to temperature (Chen *et al.*, 2017).

Regression Analysis

A regression analysis is a statistical analysis method that

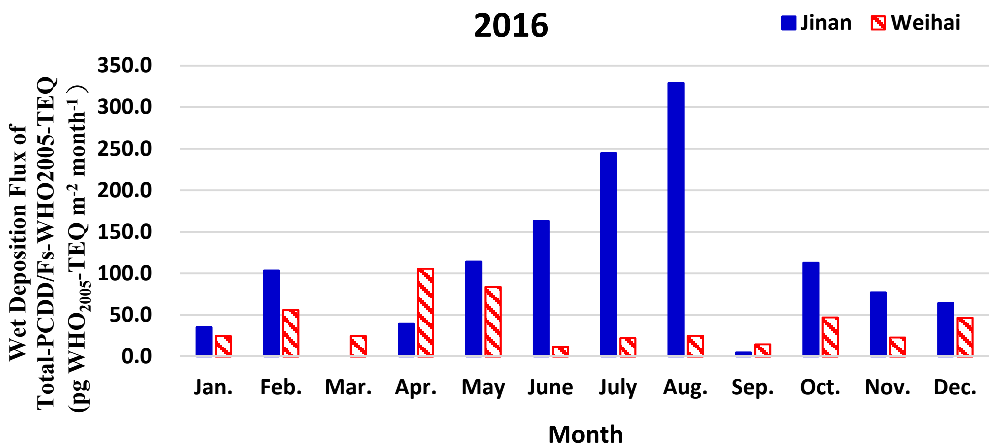

Fig. 5(a). Monthly average wet deposition flux of total PCDD/Fs-WHO_{2005}-TEQ during 2016.

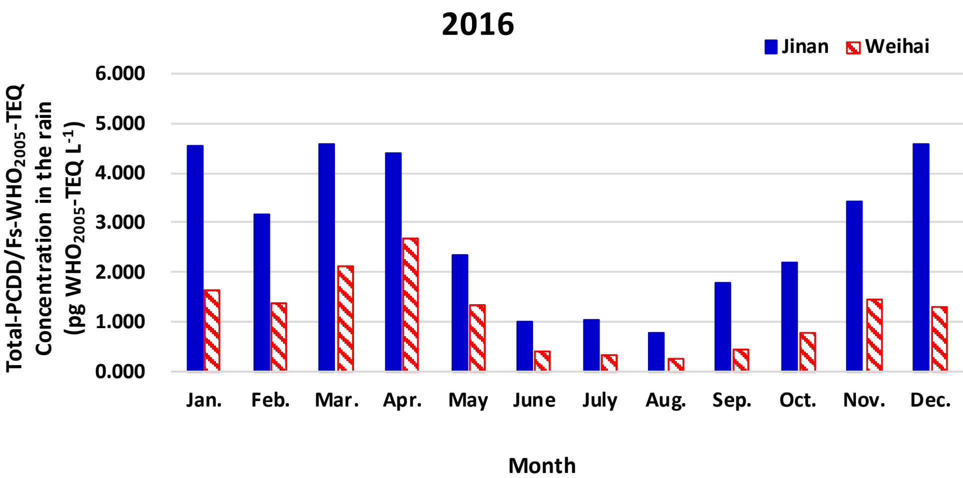

Fig. 5(b). Monthly average concentration of total PCDD/Fs-WHO_{2005}-TEQ in the rain during 2016.

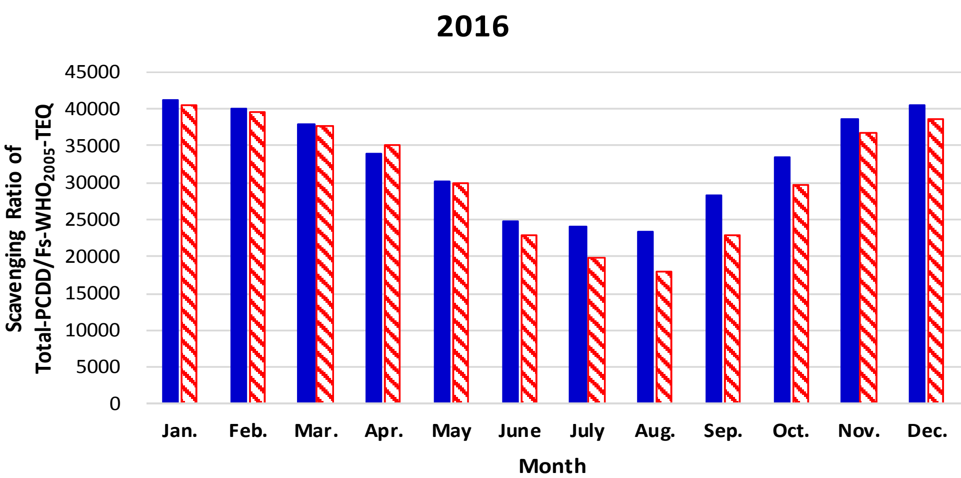

Fig. 5(c). Monthly average scavenging ratio of total PCDD/Fs-WHO_{2005}-TEQ during 2016.

determines the quantitative relationship between two or more variables. During 2015–2017, the daily AQI values for Jinan and Weihai were collected from the website. In Figs. 6(a)–6(c), the daily AQI of Jinan and Weihai is taken as the X axis, and the total concentration of PCDD/Fs-WHO_{2005}-TEQ is taken as the Y axis. A linear regression analysis plot with 95% confidence was established. The three-year (2015, 2016, and 2017) regression equations were

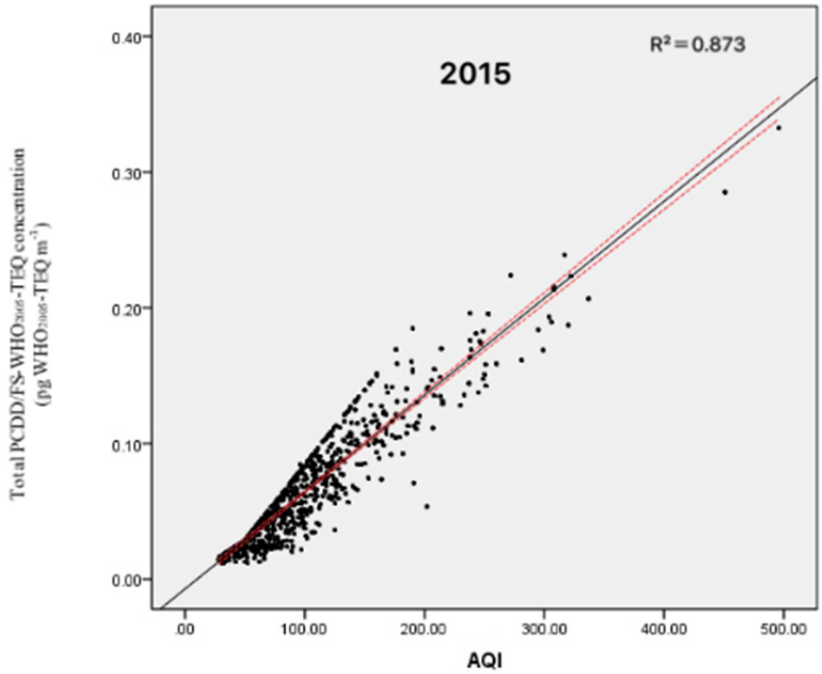

Fig. 6(a). Correlation between AQI and total PCDD/Fs-WHO_{2005}-TEQ concentration in ambient air during 2015.

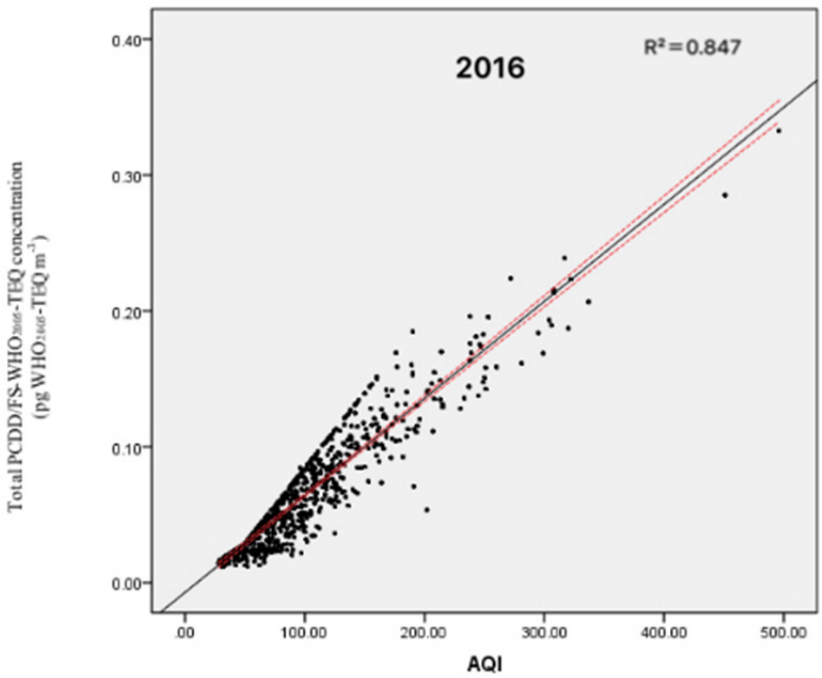

Fig. 6(b). Correlation between AQI and total PCDD/Fs-WHO_{2005}-TEQ concentration in ambient air during 2016.

$Y = -7.09 \times 10^{-3} + 7.14 \times 10^{-4}X$, $Y = -9.27 \times 10^{-3} + 7.42 \times 10^{-4}X$, and $Y = -7.80 \times 10^{-3} + 6.79 \times 10^{-4}X$, respectively. The total concentration of PCDD/Fs-WHO_{2005}-TEQ increased with the increase of AQI, showing a positive correlation. The R^2 values of three years were 0.873, 0.847, and 0.744, respectively. Therefore, knowing the AQI value, the total concentration of PCDD/Fs-WHO_{2005}-TEQ in the ambient air can be roughly estimated using the regression equation (Zhao *et al.*, 2018). The AQI values of Jinan and Weihai were mostly between 30.0 and 250, and the corresponding total PCDD/Fs-WHO_{2005}-TEQ concentration was about 0.0195–0.185 pg-WHO_{2005}-TEQ m^{-3} in 2015. The AQI values were mostly between 30.0 and 200 in 2016. The data distribution trend for 2017 was similar to those of 2015 and 2016. However, the value of AQI in 2017 was mainly concentrated between 32.0 and 150, which indicates

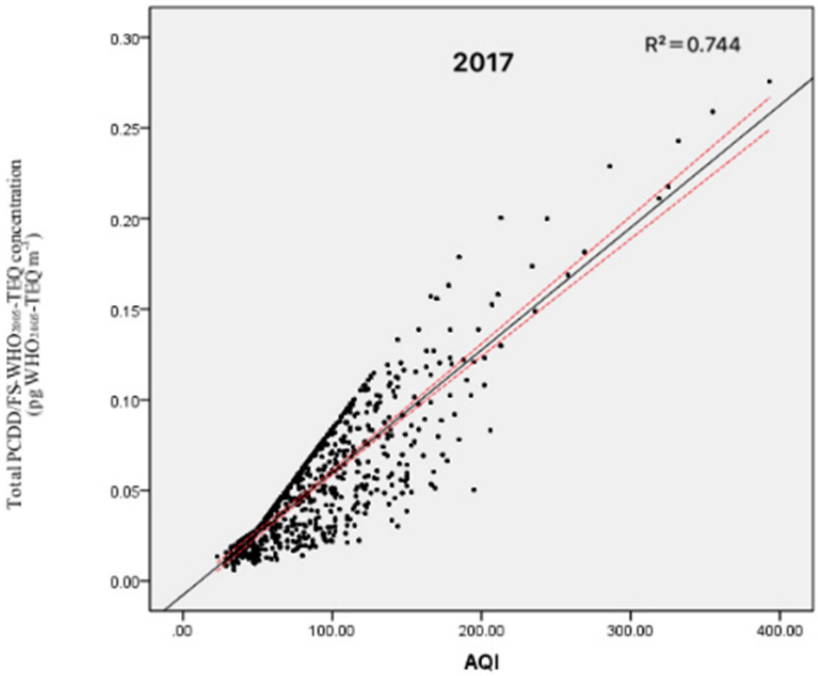

Fig. 6(c). Correlation between AQI and total PCDD/Fs-WHO_{2005}-TEQ concentration in ambient air during 2017.

that the numbers of days with good air quality was higher, indicating that the air quality in 2017 was better than those of 2015 and 2016.

CONCLUSION

1. The average concentration of $PM_{2.5}$ in Weihai was 32.9 µg m^{-3} in the three years (2015, 2016, and 2017) under investigation, and the $PM_{2.5}$ level was much lower than that in Jinan. It was also much lower than Bengbu (61.4 µg m^{-3}). This may be due to Weihai is located in the eastern part of Shandong Province, surrounded by the sea on three sides and is affected by the ocean. It shows the characteristics of maritime climate such as spring cold, summer cool, autumn air turbulence, winter low temperature, small temperature difference between day and night, long frost-free period, high wind and humidity.
2. The annual decrease in the total PCDD/Fs-WHO_{2005}-TEQ concentration year by year in Jinan and Weihai may be due to the fact that general awareness of environmental protection has gradually strengthened, and the control of air pollution has achieved certain effects. However, it is necessary to continue to implement energy conservation and emission reduction. It can be seen that the total PCDD/Fs-WHO_{2005}-TEQ concentration in Weihai was much lower than that in Jinan during the study period, indicating that maritime climate regulation contributes to the spread of pollutants.
3. Comparing $PM_{2.5}$ concentrations with $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, it can be seen that there is no close relationship between the two trends, which concurs with similar results in previous studies. It can be seen that the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents in Jinan and Weihai is the lowest in summer. This was because the ambient temperature in summer was higher, so the PCDD/Fs combined with the particles were more evaporated to the gas phase. However, in the cold season, the low temperature is not conducive to the PCDD/Fs combing with the particles.
4. The gas phase PCDD/Fs had higher scores in summer than in winter. This may be due to the fact that lower molecular weight PCDD/Fs usually have a higher vapor pressure. When the temperature is high in summer, the proportion of PCDD/Fs in the gas phase rises, and when the temperature is low in winter, some PCDD/Fs in the gas phase are transferred to the particle phase, so the fraction of PCDD/Fs in the gas phase increases with the increases in temperature.
5. $PM_{2.5}$ and rainfall are related to wet deposition but are not the main influencing factors. PM_{10}, snow, temperature, and wind speed are also some of the key factors affecting the total PCDD/Fs-WHO_{2005}-TEQ wet deposition in environment. The maximum and minimum wet deposition fluxes occurred in the month with the highest and lowest rainfall, respectively, which means that rainfall has the greatest impact on wet deposition. The greater the rainfall, the greater the wet deposition flux, and the lower the $PM_{2.5}$ concentration in the atmosphere. The wet deposition flux is always lowest in winter, and studies have shown that rainfall and rainy days may affect wet deposition flux rather than temperature. In summer, the more rainfall that is in the atmosphere, the lower the PCDD/Fs concentrations will be in rainwater. The average Stot for Jinan and Weihai in 2016 were 33080 and 30870, respectively. The minimum and maximum values of scavenging ratio in

two cities occurred in winter and summer. It can be seen that the scavenging ratio (Stot) is inversely related to temperature.

6. The total concentration of PCDD/Fs-WHO_{2005}-TEQ increased with the increase of AQI, showing a positive correlation. The R^2 values for the three years were 0.873, 0.847, and 0.744, respectively. Therefore, knowing the AQI value, the total concentration of PCDD/Fs-WHO_{2005}-TEQ in the ambient air can be roughly estimated using the regression equation.

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