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Sensitivity Analysis of Atmospheric $PM_{2.5}$-Bound Content and Dry Deposition of Total PCDD/Fs-TEQ: In the Case of Xiamen and Zhangzhou, China

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ABSTRACT

This study investigated atmospheric $PM_{2.5}$ concentration, $PM_{2.5}/PM_{10}$ ratio, total PCDD/Fs concentration, PCDD/F phase distribution, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, and dry deposition of PCDD/Fs for Xiamen and Zhangzhou Cities during 2015–2017, and sensitivity analysis of both atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content and dry deposition of PCDD/Fs in these two cities. During 2015–2017, the three-year average concentration of $PM_{2.5}$ in Xiamen was 27.6 µg m^{-3}, while that of Zhangzhou was 33.9 µg m^{-3}; this level is still higher than the WHO annual $PM_{2.5}$ standard (10.0 µg m^{-3}). In addition, the summer $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in Xiamen and Zhangzhou Cities was 0.131 ng-WHO_{2005}-TEQ g^{-1} and 0.161 ng-WHO_{2005}-TEQ g^{-1}. And it is lower than the average of the other three seasons. In Xiamen, the average monthly dry deposition flux in these three years was 322 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, while that of Zhangzhou was 378 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, respectively. Sensitivity analysis of atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content showed that the most sensitive parameters are total PCDD/F mass concentration and PM_{10} concentration, followed by atmospheric temperature and $PM_{2.5}$ concentration; in addition, the sensitivity analysis of atmospheric dry deposition is similar to those of atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content. The results of this study provide useful information for better understanding $PM_{2.5}$, particle-bound PCDD/Fs content and PCDD/Fs dry deposition in the ambient air of urban cities.

Keywords: $PM_{2.5}$; PM_{10}; PCDD/Fs; Phase distribution; Dry deposition; Sensitivity analysis.

INTRODUCTION

Particulate matter (PM), polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) in the ambient air have precipitated great public concern because studies have shown that they are significantly associated with the incidence of lung and heart disease (Ito *et al*., 2006; Wang *et al*., 2018). Particulate matter (PM) is a suspension of solid or liquid particles in the form of a type of an aerosol in the atmosphere (Ghosh *et al*., 2014). PM can be divided into TSP (ranging from 0 to 100 µm), PM_{10} (ranging from 0 to10 µm) and $PM_{2.5}$ (ranging from 0 to 2.5 µm) according to the aerodynamic diameters (Lu *et al*., 2016; Tang *et al*., 2017; Wang *et al*., 2017). The sources of PM are generally divided into natural or man-made, of which natural sources are forest burning, sandstorms and volcanic eruptions, while industrial activities, automobile exhaust and construction are the main anthropogenic sources (Bilos *et al*., 2001; Kong *et al*., 2014; Alghamdi *et al*., 2015). PM in ambient air is an aggregate of organic and inorganic carbon, mineral elements, nitrates, ammonium, sulfates and so on (Zhu *et al*., 2017). Previous studies of PM in the atmosphere have shown that it not only adversely affects air quality or even global climate, but also affects human health due to the toxicity of its particle (Chen *et al*., 2014; Huang *et al*., 2014; Wang *et al*., 2014; Liu *et al*., 2016).

We all know that PCDD/Fs is persistent organic pollutants (POPs) and semi-volatile organic compounds (SOCs), which can be transported over long distances and interact in the environment for a long time (Wu *et al*., 2009b; Chen *et al*., 2014; Lee *et al*., 2016; Redfern *et al*., 2017). They can also bioaccumulate in adipose tissue, and their effects are magnified in the food chain (Shih *et al*., 2009). PCDD/Fs

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are extremely dangerous chemicals, and their main routes of enter the human body are ingestion, inhalation and skin contact (Chen *et al*., 2010), Thereby causing damage to the immune system and even leading to cancer (Lin *et al*., 2010; Chi *et al*., 2011). The main ways in which PCDD/Fs are released into the environment are the combustion process and some industrial activities such as metal smelting processes and waste incineration (Wang *et al*., 2003; Hsieh *et al*., 2009; Chuang *et al*., 2010, 2011). PCDD/Fs are complex mixtures of different congeners, and 17 of these have been proved to be very toxic, with the 2,3,7,8 positions attached by chlorine atoms being the most toxic, where the toxicities are estimated by the toxicity equivalents (TEQ) (Cheruiyot, *et al*., 2016). After being discharged from the combustion equipment, PCDD/Fs are distributed in the gas phase and in the particle phase in the atmosphere (Chen *et al*., 2011a). Many studies have shown that the gas-particle partitioning of PCDD/Fs is determined by vapor pressure and ambient temperature, as well as other parameters (Wu *et al*., 2009a; Wang *et al*., 2010; Cheruiyot *et al*., 2015). As the temperature rises, more PCDD/Fs volatilizes into gas (Oh *et al*., 2001). Chemical and photochemical reactions can degrade PCDD/Fs, but their removal depends mainly on atmospheric deposition (Chi *et al*., 2009; Wu *et al*., 2009a; Huang *et al*., 2011a; Mi *et al*., 2012).

Dry deposition is one of the major routes by which air pollutants enter the ecosystem (Mi *et al*., 2012). Dry deposition of PCDD/Fs is the sum of the gas phase sedimentation and particle phase sedimentation, while the methods by which dry deposition occurs includes turbulent diffusion, sedimentation, inertial forces, electrical migration and diffusion electrophoresis (Zhu *et al*., 2017). The factors governing the dry deposition process include wind, temperature, humidity, and the size and shape of the particles, as well as the surface characteristics of both the particles and sink media (Chandra *et al*., 2015).

This study investigated the $PM_{2.5}$ concentrations, $PM_{2.5}/PM_{10}$ ratios, PCDD/F concentrations, gas-particle partitioning, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, and dry deposition of total PCDD/Fs-WHO_{2005}-TEQ. In addition, a sensitivity analysis of both $PM_{2.5}$-bound content and dry deposition of total PCDD/Fs-WHO_{2005}-TEQ was also conducted and discussed.

METHODS

Two cities, Xiamen (24°23′N, 117°53′E) and Zhangzhou (23°48′N, 117°34′E) in Fujian province, China, were evaluated in this study. Monthly average concentrations of $PM_{2.5}$ and PM_{10} from two urban air quality monitoring stations and statistical yearbooks, as well as monthly temperatures and precipitation. The total PCDD/F concentration was simulated using a regression analysis of the PM_{10} concentration. Tang *et al.* (2017) reported that there is a high correlation between PM_{10} values and total PCDD/F mass concentrations (Wang *et al*., 2018). included the following two regression equations:

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

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

Y_1, Y_2: total PCDD/F concentration (pg m^{-3});
x: PM_{10} concentration in ambient air (μg m^{-3}).
The total PCDD/F concentration value is the average of Y_1 and Y_2.

Gas-Particle Partitioning

The PCDD/F concentrations in the gas and particle phases, respectively, were calculated using a gas-particle partitioning model, as in 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, which was multiplied by the 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:

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

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 $b_r = -7.2$ and with $R^2 = 0.94$. These values were used in this study to establish the partitioning constant (K_p) for PCDD/Fs.

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

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

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

Atmospheric Dry Deposition of PCDD/Fs

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

$$F_{d,t} = F_{d,g} + F_{d,p} \quad (6)$$

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

where, $F_{d,t}$ is the dry deposition flux of total PCDD/Fs contributed by both the gas- and particle-phases, and, $F_{d,g}$

and $F_{d,p}$ are the PCDD/Fs dry deposition flux contributed by the gas and particle phases, respectively. C_p and C_g are the calculated concentrations of PCDD/Fs in the particle and gas phases. C_T is the measured concentration of total PCDD/Fs in the ambient air; $V_{d,T}$ is the dry deposition velocity of total PCDD/Fs, and $V_{d,g}$ and $V_{d,p}$ are the dry deposition velocities of the gas- and particle-phases, respectively.

In this study, the mean dry deposition velocity of total PCDD/Fs ($V_{d,T}$= 0.42 cm s^{-1}) was as proposed by Shih *et al*. (2006). Due to the lack of measured data for PCDD/Fs, a selected value (0.010 cm s^{-1}) for the gas-phase PAH dry deposition velocity, as proposed by Sheu *et al*. (1996) and used by Lee *et al*. (1996) was used in this study.

RESULTS AND DISCUSSION

$PM_{2.5}$ Concentration

The $PM_{2.5}$ concentration not only has a significant correlation with air visibility and human health, but can also reflect the PCDD/F concentration in a given region. The monthly average $PM_{2.5}$ concentrations in the ambient air of Xiamen and Zhangzhou in 2015 and 2017 are shown in Figs. 1(a), 1(b) and 1(c), respectively. In the case of Xiamen, the three-year data indicated that the lowest concentration of $PM_{2.5}$ occurred in 2017, and the monthly

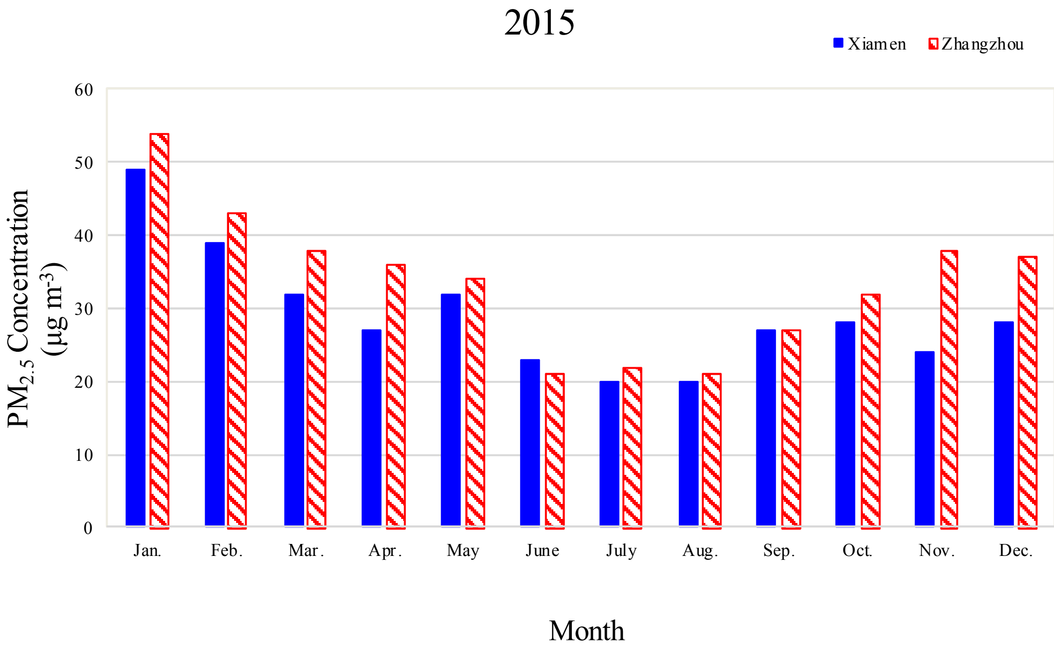

Fig. 1(a). Monthly average atmospheric $PM_{2.5}$ concentration in Xiamen and Zhangzhou during 2015.

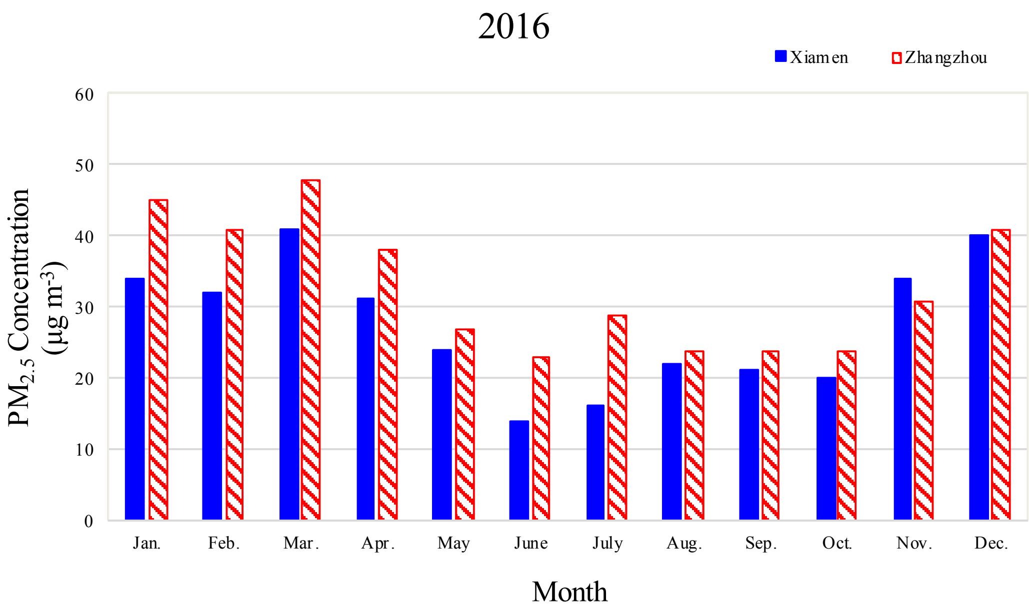

Fig. 1(b). Monthly average atmospheric $PM_{2.5}$ concentration in Xiamen and Zhangzhou during 2016.

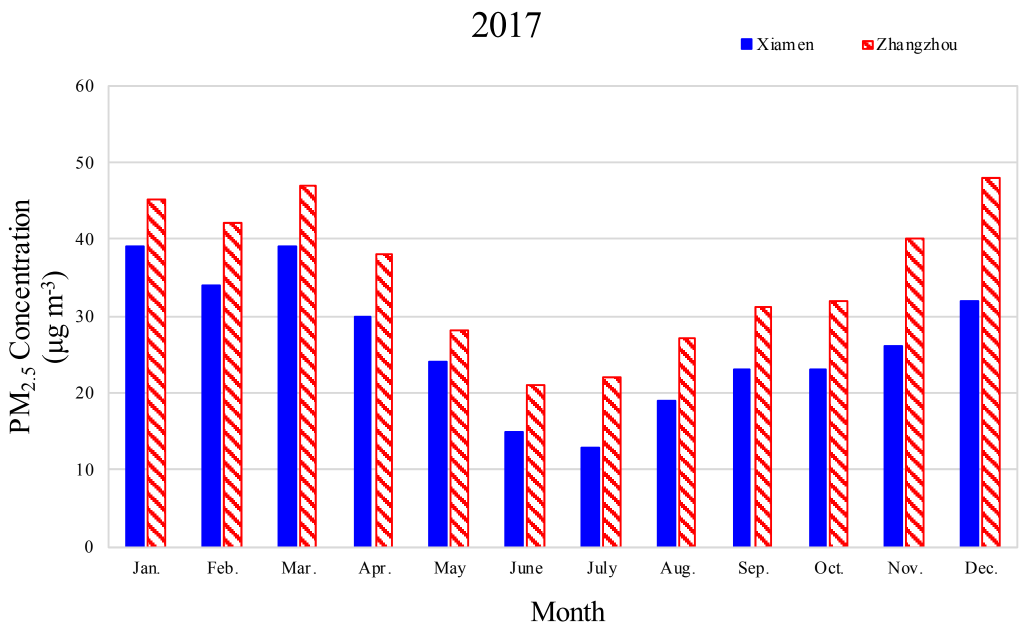

Fig. 1(c). Monthly average atmospheric $PM_{2.5}$ concentration in Xiamen and Zhangzhou during 2017.

average $PM_{2.5}$ concentration ranged from 13.0 to 39.0 µg m^{-3} and with an average of 26.4 µg m^{-3}. During 2016, the range was between 14.0 and 41.0 µg m^{-3} and with an average of 27.4 µg m^{-3}. In 2015, the monthly average $PM_{2.5}$ concentration ranged between 20.0 and 49.0 µg m^{-3}, with an average of 29.1 µg m^{-3}. Comparing the annual average $PM_{2.5}$ concentration, we can infer that the highest concentration occurred in 2015, and the lowest occurred in 2017. Compared with the average concentrations from 2015 to 2017, $PM_{2.5}$ concentration was reduced by approximately 10.2%. In general, Xiamen's three-year average $PM_{2.5}$ concentrations ranged from 13.0 to 49.0 µg m^{-3}, with an average of 27.6 µg m^{-3}. The concentration of $PM_{2.5}$ was less than that of cities in central and northern China such as Handan and Bengbu (Wang *et al.*, 2018; Zhao *et al.*, 2018). This is because the economic pillar of Xiamen is mainly two major industries - electronics and machinery, followed by a third service-based industry, which does not emit very much pollution. Xiamen itself is surrounded by the sea, and its forest coverage is high (45%), which also resulted in a lower $PM_{2.5}$ concentration. It can also be seen that even if the air quality in Xiamen significantly improves, the $PM_{2.5}$ concentration in Xiamen will still be higher than the WHO air quality regulated standard (10 µg m^{-3}). Therefore there are still gaps from the international standards, and efforts should still be made to improve the air quality.

Zhangzhou (Figs. 1(a), 1(b) and 1(c)), is a coastal city, and its main economic pillar is the cultivation and processing of agricultural products. Therefore, the pollution emitted into the atmosphere is much less than that of the industrial cities in central and northeastern China (Handan and Bengbu). The average monthly $PM_{2.5}$ concentration ranges between 21.0 and 48.0 µg m^{-3} and averaged 35.1 µg m^{-3} during 2017; in 2016, it ranged from 23.0 to 48.0 µg m^{-3}, with an average value of 32.9 µg m^{-3}; in 2015, the average $PM_{2.5}$ concentration ranged from 21.0 to 54.0 µg m^{-3}, with an average of 33.6 µg m^{-3}. These results show that from 2015 to 2016, the annual average concentration of $PM_{2.5}$ decreased from 33.6 µg m^{-3} to 32.9 µg m^{-3}, which was a decrease of approximately 2.1%. From 2016 to 2017, the annual average concentration of $PM_{2.5}$ increased from 32.9 µg m^{-3} to 35.1 µg m^{-3}, an increase of approximately 6.7%. In general, the concentration of $PM_{2.5}$ within the three years for Zhangzhou was between 21.0 and 54.0 µg m^{-3}, with an average of 33.9 µg m^{-3}.

As for seasonal variations, we define January, February and December as winter and define March, April and May as spring. June, July, August are defined as summer, and September, October, November are defined as fall. In Xiamen, in 2015, the average $PM_{2.5}$ concentrations in spring, summer, fall and winter were 30.3, 21.0, 26.3 and 38.7 µg m^{-3}, and those in 2016 were 32.0, 17.3, 25.0 and 35.3 µg m^{-3}; in 2017 they were 31.0, 15.7, 24.0 and 35.0 µg m^{-3}. In Zhangzhou, in 2015, the average $PM_{2.5}$ concentrations in spring, summer, fall, and winter were 36.0, 21.3, 32.3 and 44.7 µg m^{-3}; those in 2016 were 37.7, 25.3, 26.3, and 42.3 µg m^{-3}, and the average $PM_{2.5}$ concentrations in 2017 were 37.7, 23.3, 34.3 and 45.0 µg m^{-3}. This demonstrates that $PM_{2.5}$ concentration changes with the seasons, where the $PM_{2.5}$ concentrations are highest in winter and are the lowest in summer. In Xiamen, the average $PM_{2.5}$ concentration over the three years in summer was 18.3 µg m^{-3}, and that in winter was 36.3 µg m^{-3}. The values in winter were 2.0 times higher than those in summer. As for Zhangzhou, the average value over the three years of summer was 23.3 µg m^{-3}, which it was 44.7% lower than that in winter (44.0 µg m^{-3}).

There are two main reasons for changes of in $PM_{2.5}$ concentration: pollution source emissions and meteorological conditions. Due to the effects of these two conditions, $PM_{2.5}$ concentrations have obvious seasonal variations. In winter, due to the obvious cooling of the nighttime ground radiation, the "lower atmospheric temperature" is prone to a "inversion," and the air's horizontal and vertical exchange and circulation capacities are weakened. The pollutants emitted in the air are confined to the shallow atmosphere and gradually gather into the earth. In summer, there are frequent cyclones, so, water vapor transport is good, which is conducive to precipitation. Frequent rainfall and windy weather are conducive to the diffusion and removal of $PM_{2.5}$. Therefore, the $PM_{2.5}$ concentration in the summer is the lowest. Xiamen and Zhangzhou are both coastal cities, and there is not much industrial pollution. So, the atmospheric environment in these two cities is better than that of cities in central and northern China.

$PM_{2.5}/PM_{10}$ Ratio

The $PM_{2.5}/PM_{10}$ ratio can reflect the contribution of fine particulates to ambient air pollutants, thus reflecting air pollution. The monthly mean values of $PM_{2.5}/PM_{10}$ in the ambient air around Xiamen and Zhangzhou are shown in Figs. 2(a), 2(b), and 2(c).

The ratio of $PM_{2.5}/PM_{10}$ in Xiamen each month in 2015 was between 0.48 and 0.70, with an average of 0.58. In 2016, the $PM_{2.5}/PM_{10}$ ratio was between 0.43 and 0.69, with an average of 0.56. The $PM_{2.5}/PM_{10}$ ratio for each

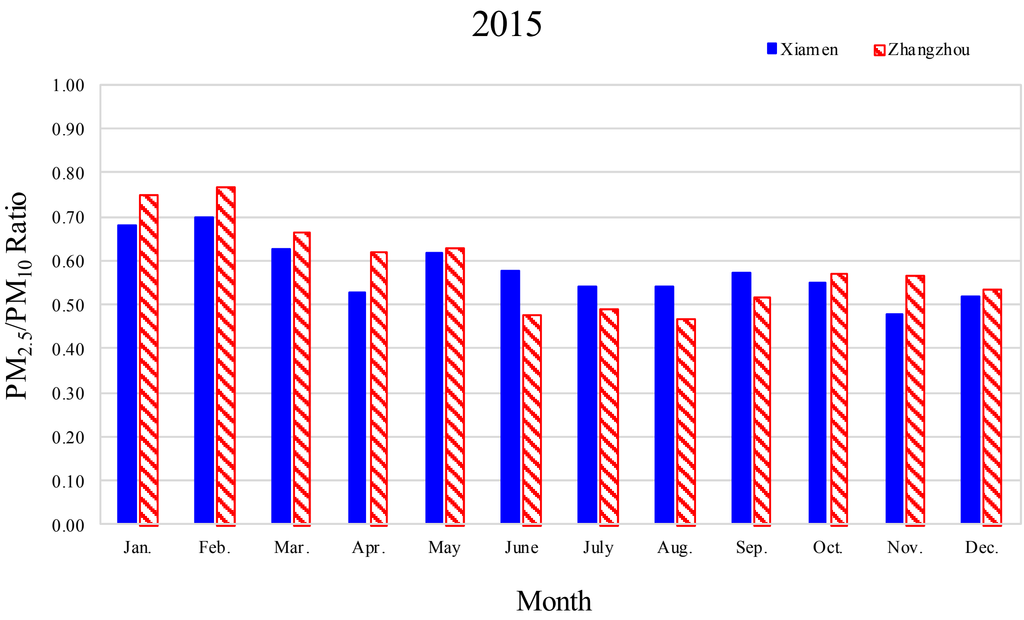

Fig. 2(a). Monthly Average $PM_{2.5}$ /PM_{10} Ratio in Xiamen and Zhangzhou during 2015.

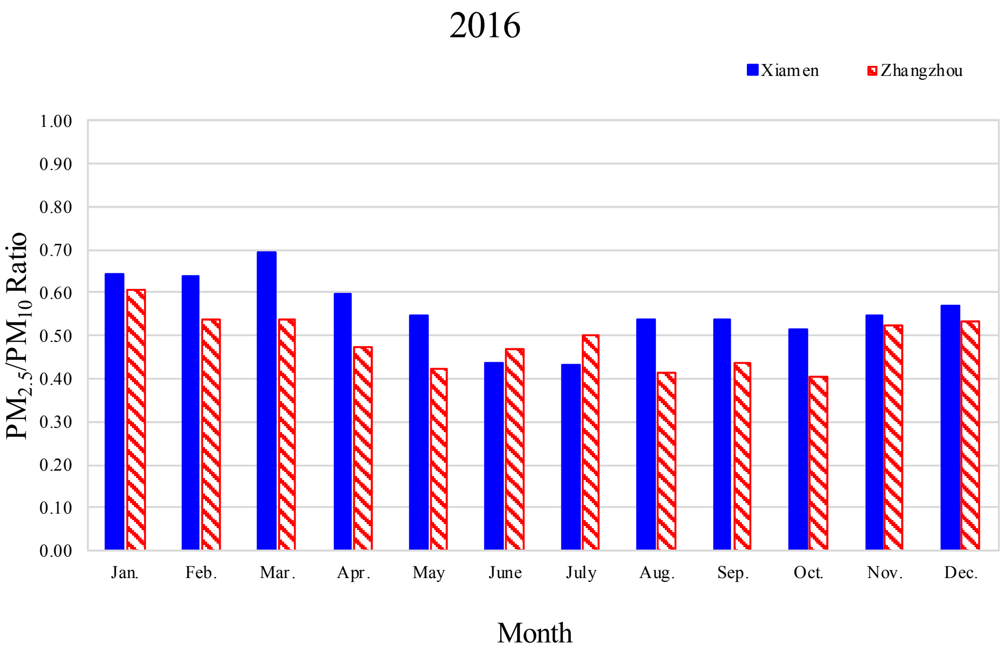

Fig. 2(b). Monthly Average $PM_{2.5}$ /PM_{10} Ratio in Xiamen and Zhangzhou during 2016.

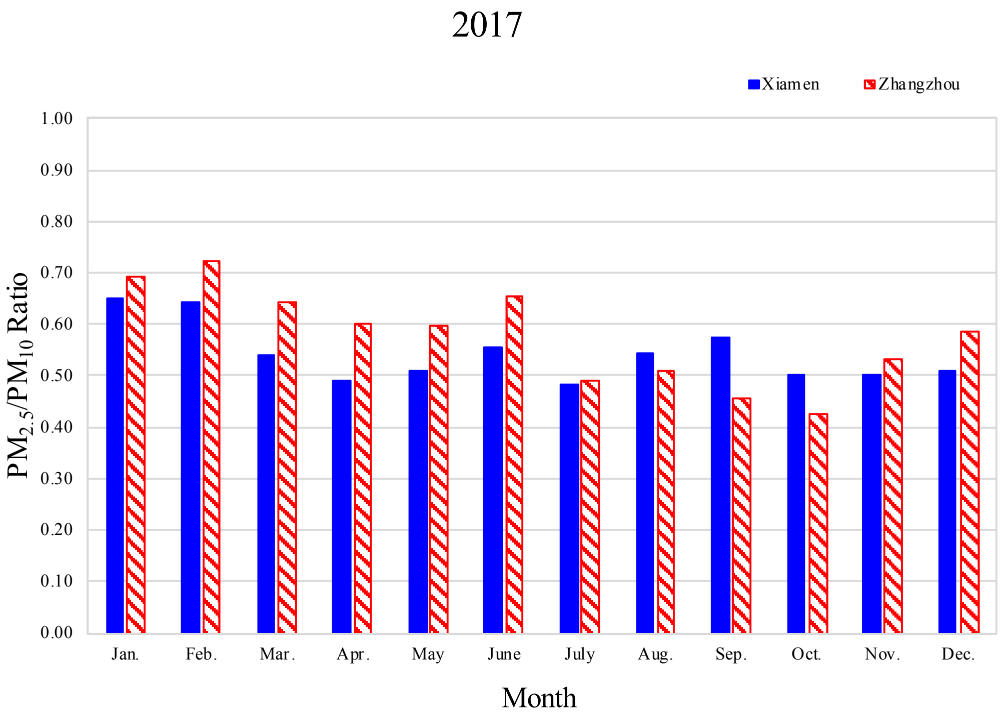

Fig. 2(c). Monthly Average $PM_{2.5}$ /PM_{10} Ratio in Xiamen and Zhangzhou during 2017.

month in 2017 was between 0.48 and 0.65. The average value was 0.54. The value of $PM_{2.5}$/PM_{10} per month in Zhangzhou in 2015 was between 0.47 and 0.77, with an average of 0.59. In 2016, the monthly $PM_{2.5}$/PM_{10} ratio was between 0.41 and 0.61, with an average of 0.49. The $PM_{2.5}$/PM_{10} ratio each month in 2017 was between 0.43 and 0.72, and the average value was 0.58. In general, the $PM_{2.5}$/PM_{10} ratio in Xiamen during the three years under consideration ranged from 0.43 to 0.70, with an average of 0.56. The three-year $PM_{2.5}$/PM_{10} ratio in Zhangzhou was between 0.43 and 0.77 with an average of 0.55.

In Xiamen, the three months with the highest $PM_{2.5}$/PM_{10} ratios in 2015 were January, February, and March. Their averages were 0.68, 0.70, and 0.63, respectively. In 2016, the months with the highest average $PM_{2.5}$/PM_{10} ratio were 0.64 in January and February and 0.69 in March; the highest average for the three-month period for the $PM_{2.5}$/PM_{10} ratio in 2017 was 0.65 in January; in February, it was 0.64, and in September, it was 0.58. However, in 2015, the lowest average $PM_{2.5}$/PM_{10} concentration ratio for the three months was April, and in November and December, the values were 0.53, 0.48, and 0.52, respectively; in 2016, the three lowest average $PM_{2.5}$/PM_{10} months were June (0.44), July (0.43) and October (0.51). In 2017, the lowest four months average $PM_{2.5}$/PM_{10} ratios were 0.49 and 0.48 in July, and 0.50 in October and November.

In Zhangzhou, the three months with the highest $PM_{2.5}$/PM_{10} ratio in 2015 were January (0.75), February (0.77) and March (0.67); In 2016, the highest three-month $PM_{2.5}$/PM_{10} ratio average was 0.61 in January and 0.54 in February and March; in 2017, the three months with the highest $PM_{2.5}$/PM_{10} ratios were January, February, and June, for which the averages were 0.69, 0.72, 0.66, respectively. In contrast, the three months with the lowest $PM_{2.5}$/PM_{10} concentration ratios in 2015 were June (0.48), July (0.49), and August (0.47). In 2016, the three months with the lowest average $PM_{2.5}$/PM_{10} concentration ratios were May, August, and October, with values of 0.42, 0.41, and 0.41, respectively. The average value of $PM_{2.5}$/PM_{10} for the lowest three months of 2017 were 0.49 in July, 0.46 in September and 0.43 in October.

In general, the increase in the $PM_{2.5}$/PM_{10} ratio was found to be accompanied by higher $PM_{2.5}$ concentrations. This conclusion mainly proves that $PM_{2.5}$ is the main component in atmospheric particles, and previous research (Tang *et al.*, 2017) has also concurred with this result. These atmospheric particles are mainly attributed to the conversion of gas particles, undergoing coagulation and flocculation processes, and agglomerating $PM_{2.5}$.

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

The concentrations of PCDD/Fs-WHO$_{2005}$-TEQ in the atmospheric environment in Xiamen and Zhangzhou from 2015 to 2017 are shown in Figs. 3(a), 3(b), and 3(c). In 2015, the monthly PCDD/Fs-WHO$_{2005}$-TEQ average concentrations in Xiamen ranged from 0.020 to 0.041 pg-WHO$_{2005}$-TEQ m^{-3}, with an average of 0.030 pg-WHO$_{2005}$-TEQ m^{-3}. In January, the highest concentration was 0.041 pg-WHO$_{2005}$-TEQ m^{-3}, while the lowest was in July and August at 0.020 pg-WHO$_{2005}$-TEQ m^{-3}. In 2016, the monthly concentration of PCDD/Fs-WHO$_{2005}$-TEQ ranged between 0.017–0.040 pg-WHO$_{2005}$-TEQ m^{-3}, with an average of 0.029 pg-WHO$_{2005}$-TEQ m^{-3}; the highest concentration was in March and December (0.040 pg-WHO$_{2005}$-TEQ m^{-3}), while the lowest was in June (0.017 pg-WHO$_{2005}$-TEQ m^{-3}).

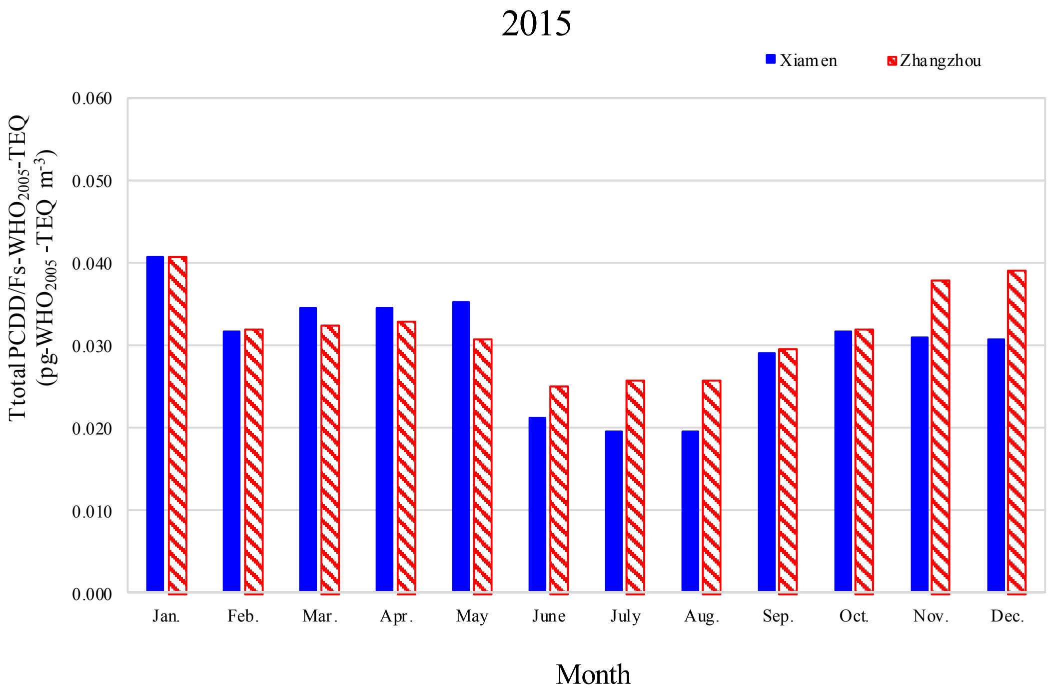

Fig. 3(a). Monthly total PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Xiamen and Zhangzhou during 2015.

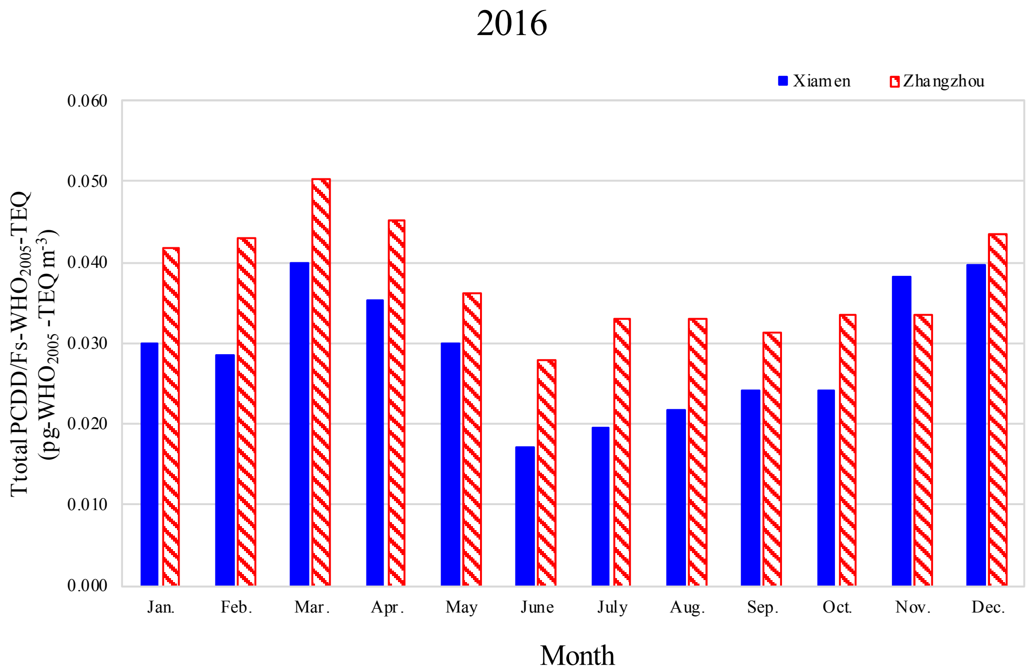

Fig. 3(b). Monthly total PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Xiamen and Zhangzhou during 2016.

In 2017, the average monthly PCDD/Fs-WHO_{2005}-TEQ concentration was 0.014–0.048 pg-WHO_{2005}-TEQ m^{-3}, with an average of 0.030 pg-WHO_{2005}-TEQ m^{-3}. The highest concentration was in March (0.048 pg-WHO_{2005}-TEQ m^{-3}), while June and July had the lowest (0.014 pg-WHO_{2005}-TEQ m^{-3}).

In Zhangzhou in 2015, the monthly mean concentration of PCDD/Fs-WHO_{2005}-TEQ ranged from 0.025 to 0.041 pg-WHO_{2005}-TEQ m^{-3}, with an average of 0.032 pg-WHO_{2005}-TEQ m^{-3}. The highest concentration was in January was 0.041 pg-WHO_{2005}-TEQ m^{-3}, while June had the lowest (0.025 pg-WHO_{2005}-TEQ m^{-3}). In 2016, the monthly average total PCDD/Fs-WHO_{2005}-TEQ concentrations ranged from 0.028 to 0.050 pg-WHO_{2005}-TEQ m^{-3}, with an average of 0.038 pg-WHO_{2005}-TEQ m^{-3}. The highest concentration was in March (0.050 pg-WHO_{2005}-TEQ m^{-3}); the lowest concentration was in June (0.028 pg-WHO_{2005}-TEQ m^{-3}). In 2017, the monthly average total PCDD/Fs-WHO_{2005}-TEQ

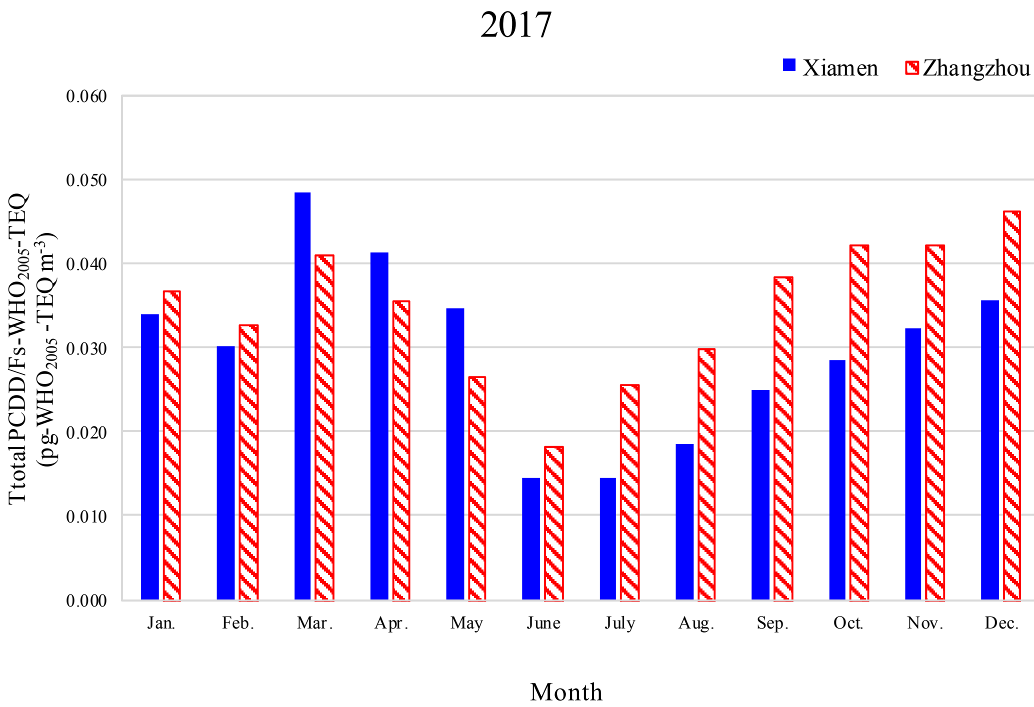

Fig. 3(c). Monthly total PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Xiamen and Zhangzhou during 2017.

concentration ranged between 0.018 and 0.064 pg-WHO_{2005}-TEQ m^{-3}, and the average value was 0.035 pg-WHO_{2005}-TEQ m^{-3}. The highest concentration (0.046 pg-WHO_{2005}-TEQ m^{-3}) occurred in December, with the lowest concentration occurring (0.018 pg-WHO_{2005}-TEQ m^{-3}) in June.

Regarding the seasonal changes in Xiamen, in 2015, the PCDD/Fs-WHO_{2005}-TEQ concentrations in the spring, summer, autumn and winter were 0.035, 0.020, 0.031 and 0.035 pg-WHO_{2005}-TEQ m^{-3}, respectively. In 2016, the PCDD/Fs-WHO_{2005}-TEQ concentrations in the spring, summer, autumn and winter were 0.035, 0.020, 0.029, and 0.033 pg-WHO_{2005}-TEQ m^{-3}; The concentrations of PCDD/Fs-WHO_{2005}-TEQ in spring, summer, autumn, and winter in 2017 were 0.041, 0.016, 0.029, and 0.033 pg-WHO_{2005}-TEQ, respectively. In Zhangzhou, the PCDD/Fs-WHO_{2005}-TEQ concentrations in spring, summer, autumn, and winter were 0.032, 0.026, 0.033 and 0.037 pg-WHO_{2005}-TEQ m^{-3} in 2015; In 2016, the concentrations in spring, summer, autumn and winter were 0.044, 0.031, 0.033 and 0.037 pg-WHO_{2005}-TEQ m^{-3}, respectively, and the concentrations of PCDD/Fs-WHO_{2005}-TEQ in 2017 were 0.035, 0.025, 0.041 and 0.039 pg-WHO_{2005}-TEQ m^{-3}.

These results indicate that the lowest PCDD/Fs-WHO_{2005}-TEQ concentrations in both cities occurred in summer, which means that the level of particulate matter will affect total PCDD/Fs-WHO_{2005}-TEQ concentrations. Therefore, higher PCDD/Fs-WHO_{2005}-TEQ concentrations are always accompanied by higher concentrations of particulate matter. Therefore, controlling the source of PM emissions subsequently leads to a reduction in the level of environmental dioxins.

Gas-Particle Partitioning of PCDD/Fs

The gas-particle partitioning of PCDD/Fs plays an important role in dry and wet atmospheric deposition. Several factors are important here, including ambient temperature, PCDD/F concentration, atmospheric pressure, and atmospheric particulate concentration (Hoff *et al.*, 1996). The calculation of the gas-particle partitioning is based on the meteorological data and the Eqs. (3), (4) and (5), as well as the seasonal total PCDD/Fs-WHO_{2005}-TEQ in the atmosphere of Xiamen and Zhangzhou during the period from 2015–2017, for which the gas-particle partitioning is shown in Figs. 4(a), 4(b) and 4(c).

In Xiamen, in 2015, the seasonal mean temperatures in spring, summer, autumn, and winter were 21.3, 28.7, 24.5, and 15.2°C, respectively. In 2016, the seasonal mean temperatures in spring, summer, autumn, and winter were 20.8, 29.1, 24.8, and 14.4°C. In 2017, the average temperatures in spring, summer, autumn and winter were 21.1, 28.7, 25.3, and 15.4°C, respectively. As for the seasonal changes in gas partitioning, in 2015, the fractions of gas phase total PCDD/Fs-WHO_{2005}-TEQ meteorological concentrations in spring, summer, autumn and winter were accounted for 40.0%, 70.0%, 53.3% and 26.7%, respectively. In 2016, the gas partitioning of PCDD/Fs-WHO_{2005}-TEQ in spring, summer, autumn, and winter were 43.3%, 70.0%, 53.3% and 23.3%, respectively; the gas partitioning of total PCDD/Fs-WHO_{2005}-TEQ in the spring, summer, autumn and winter of 2017 were 40.0%, 70.0%, 56.7% and 30.0%, respectively.

In Zhangzhou, the average seasonal temperatures in spring, summer, autumn, and winter were 22.3, 29.7, 25.2 and 15.7°C, respectively; in 2016, the seasonal mean temperatures in spring, summer, autumn, and winter were

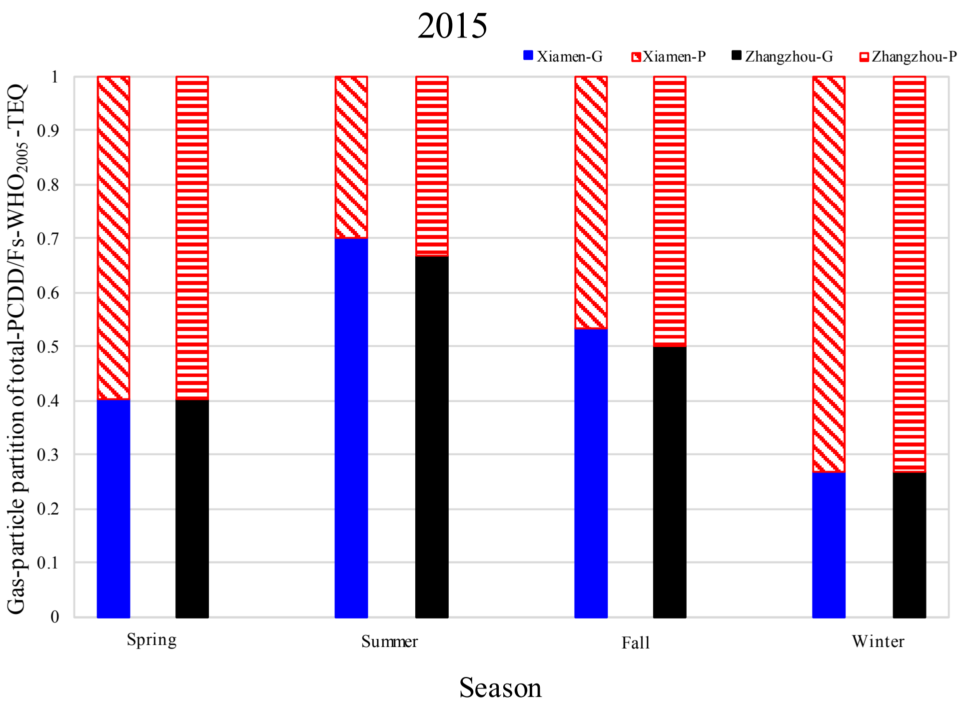

Fig. 4(a). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Xiamen and Zhangzhou during 2015.

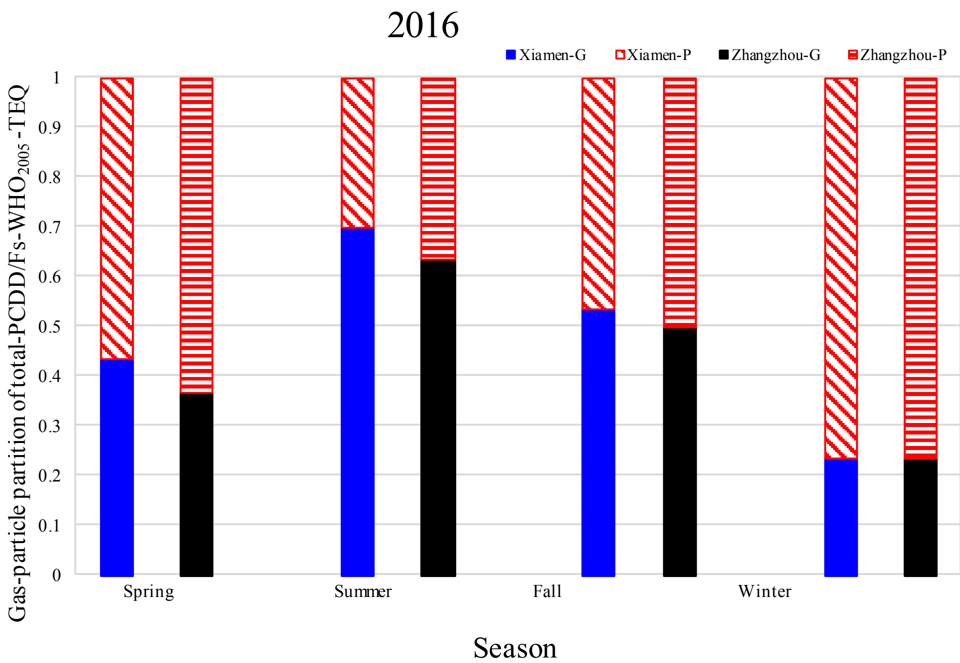

Fig. 4(b). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO_{2005}-TEQ in the ambient air in Xiamen and Zhangzhou during 2016.

21.9, 30.2, 25.4°C and 15.1°C, respectively; and in 2017, the average temperatures in spring, summer, autumn, and winter were 21.9, 29.6, 26.2, and 16.2°C, respectively. As for the seasonal changes in gas distribution, in 2015, the fractions of gas phase total PCDD/Fs-WHO_{2005}-TEQ concentrations in spring, summer, autumn, and winter were accounted for 40.0%, 66.7%, 50.0%, and 26.7%, respectively; in 2016, the fractions of gas phase total PCDD/Fs-WHO_{2005}-TEQ in spring, summer, fall and winter were 36.7%, 63.3%, 50.0% and 23.3%, respectively; in 2017, the fractions of gas phase total PCDD/Fs-WHO_{2005}-TEQ were 43.3%, 70.0%, 50.0% and 30.0% in spring, summer, fall and winter, respectively.

These results indicate that the gas phase partitioning of

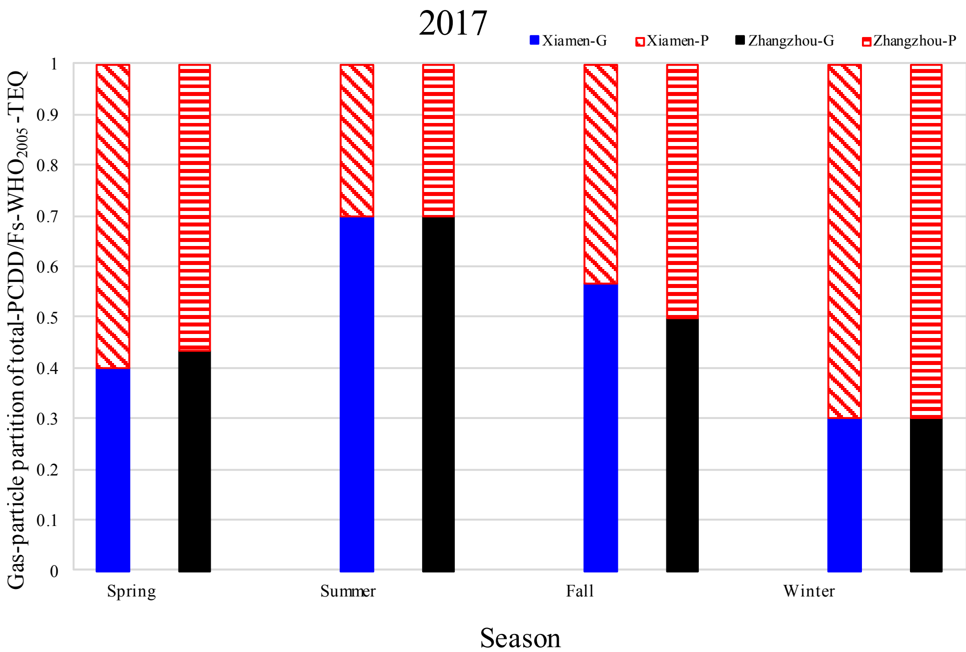

Fig. 4(c). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO$_{2005}$-TEQ in the ambient air in Xiamen and Zhangzhou during 2017.

PCDD/Fs in summer was higher than that in winter, which may have been due to the fact that lower molecular weight PCDD/Fs usually have higher vapor pressure. With an increase in the ambient temperature, the proportion of PCDD/Fs in the gas phase also increases. The temperature decreases, and some PCDD/Fs are exchanged and transferred to the particle phase. As a result, lower molecular weight PCDD/Fs mainly exist in the gas phase, and the fraction of gas phase PCDD/Fs increases with increases temperature.

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

The monthly mean values of $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content in Xiamen and Zhangzhou are shown in Figs. 5(a), 5(b), and 5(c).

For Xiamen, in 2015, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content ranged from 0.128 to 0.499 ng-WHO$_{2005}$-TEQ g^{-1}, with an average of 0.320 ng-WHO$_{2005}$-TEQ g^{-1}. In 2016, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content ranged between 0.138 and 0.505, with an average of 0.320 ng-WHO$_{2005}$-TEQ g^{-1}. In 2017, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content ranged between 0.113 and 0.654 ng-WHO$_{2005}$-TEQ g^{-1} with an average of 0.341 ng-WHO$_{2005}$-TEQ g^{-1}. The average of $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content was 0.327 ng-WHO$_{2005}$-TEQ g^{-1} for the three years from 2015 through 2017. As for Zhangzhou, in 2015, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content was between 0.153 and 0.508 ng-WHO$_{2005}$-TEQ g^{-1}, with an average of 0.279 ng-WHO$_{2005}$-TEQ g^{-1}. In 2016, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content was 0.163–0.591 ng-WHO$_{2005}$-TEQ g^{-1}, with an average of 0.363 ng-WHO$_{2005}$-TEQ g^{-1}. The content in 2017 ranged between 0.112 and 0.492 ng-WHO$_{2005}$-TEQ g^{-1}, and the mean value was 0.295 ng-WHO$_{2005}$-TEQ g^{-1}. The average $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content was 0.312 ng-WHO$_{2005}$-TEQ g^{-1} for the three years from 2015 through 2017, which was 4.2% less than Xiamen.

For the seasonal variations in 2015 in Xiamen, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content in spring, summer, fall, and winter was 0.416, 0.131, 0.281, and 0.45 ng-WHO$_{2005}$-TEQ g^{-1}, and the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ contents in spring, summer, fall, and winter was 0.406, 0.144, 0.262, and 0.469 ng-WHO$_{2005}$-TEQ g^{-1}, respectively, in 2016. In 2017, they were 0.513, 0.117, 0.265, and 0.469 ng-WHO$_{2005}$-TEQ g^{-1}. In Zhangzhou, the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ contents for the spring, summer, autumn, and winter in 2015 was 0.278, 0.162, 0.257, and 0.421 ng-WHO$_{2005}$-TEQ g^{-1}, respectively. In 2016, it was 0.427, 0.183, 0.305, and 0.536 ng-WHO$_{2005}$-TEQ g^{-1}. The $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content in spring, summer, fall, and winter was 0.307, 0.139, 0.311, and 0.421 ng-WHO$_{2005}$-TEQ g^{-1} in 2017.

The results showed that the average of $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content in summer was lower than the other three seasons. This is because the average ambient temperature in summer is higher than the other three seasons, and more PCDD/Fs combine with the particles and evaporate into the gas phase, so the $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ contents is decreases.

Dry Deposition

Dry deposition fluxes of PCDD/Fs were calculated based on Eqs. (6) and (7). The terms C_g and C_p were determined based on the gas-particle partitioning shown in the previous section, after when the unknown $V_{d,p}$ could be calculated, and the monthly dry deposition fluxes in Zhangzhou and Xiamen from 2015 to 2017 are shown in Figs. 6(a), 6(b), and 6(c)

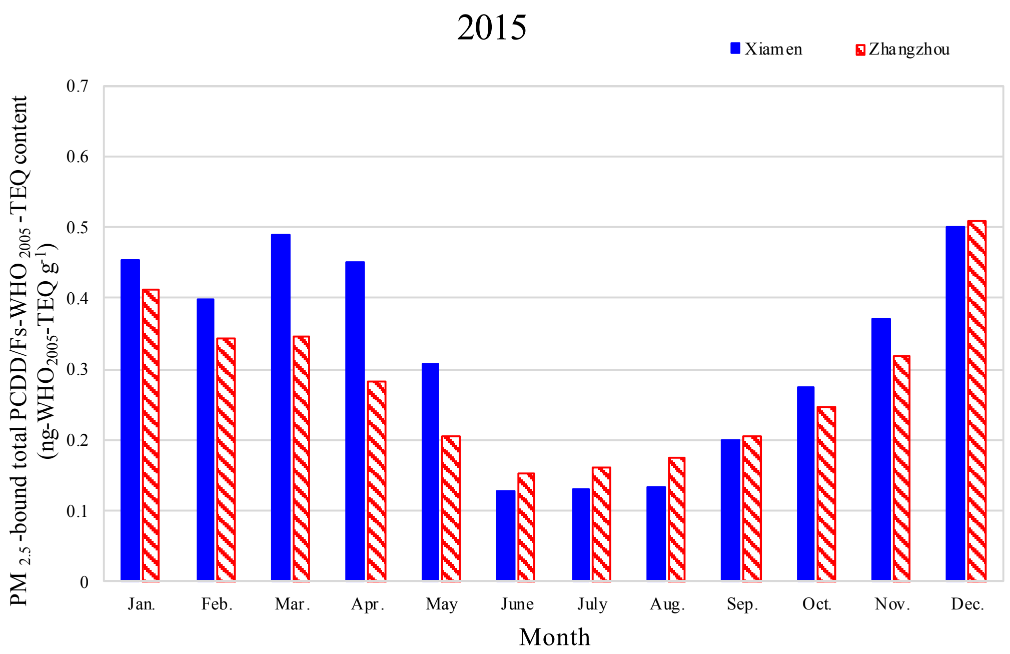

Fig. 5(a). $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of Xiamen and Zhangzhou during 2015.

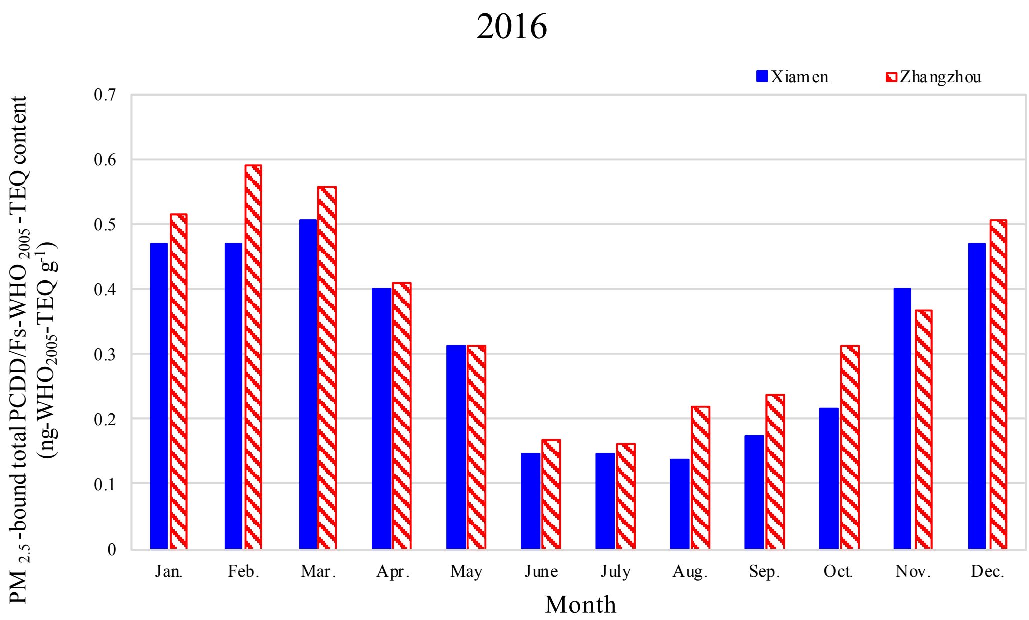

Fig. 5(b). $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of Xiamen and Zhangzhou during 2016.

In Xiamen, the average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in 2015 ranged between 214 and 443 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 3914 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which was about 2.96 times lower than that of the same year in Handan (11590 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhao *et al.*, 2018). The maximum monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ of 443 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ occurred in January, and the lowest value 214 pg WHO_{2005}-TEQ m^{-2} $Month^{-1}$, occurred in July and August, which two times lower than the highest value. In 2016, the monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ ranged between 186 and 434 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 3791 pg WHO_{2005}-TEQ m^{-2} $years^{-1}$. The dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in March of 434 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ was the maximum monthly average dry deposition flux for the year, for which the lowest level occurred in July (186 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), which was about 57.1% lower than that in March. During 2017, the monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ ranged from 158 to 527 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the

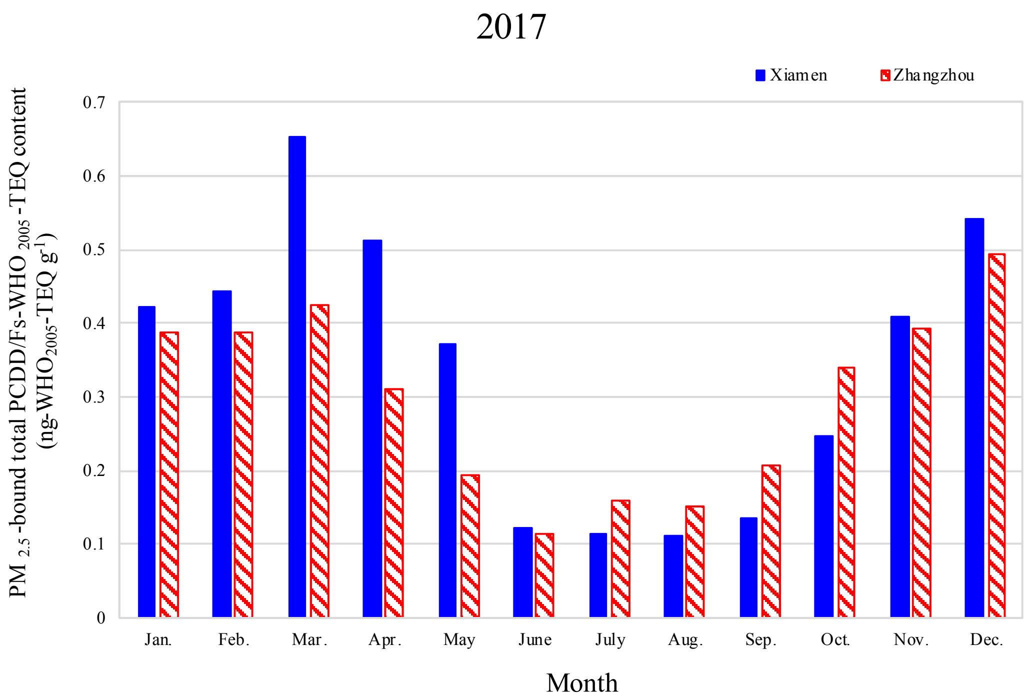

Fig. 5(c). $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of Xiamen and Zhangzhou during 2017.

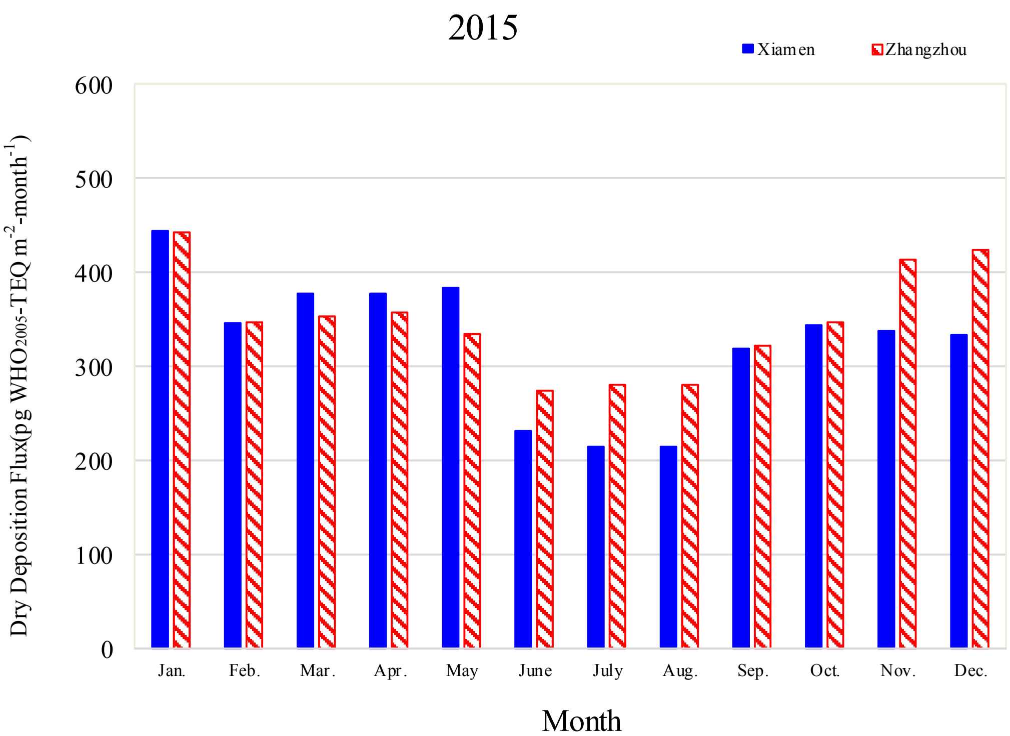

Fig. 6(a). Monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen and Zhangzhou in 2015.

annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 3886 pg WHO_{2005}-TEQ m^{-2} $years^{-1}$. The maximum monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ of 527 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ occurred in March, and lowest level of dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ occurred in June and July, with a value of 158 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$.

As for Zhangzhou, in 2015, the monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ ranged from 273 to 443 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 4168 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, which about 56.9% lower than that of in Kaifeng in the same year (9680 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhao *et al.*, 2018). The average

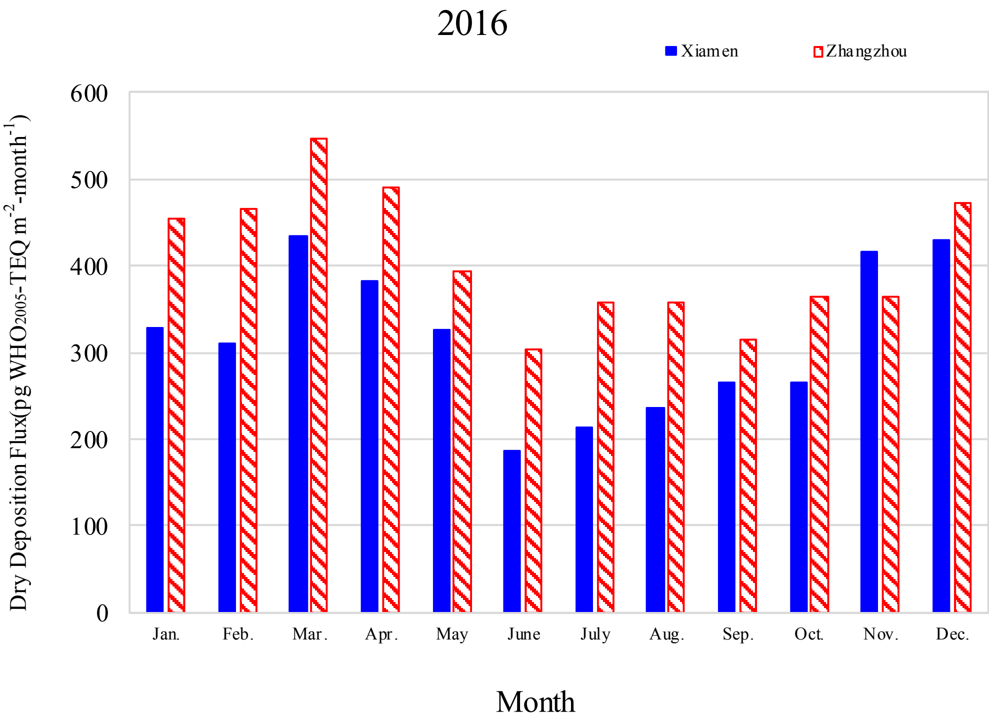

Fig. 6(b). Monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen and Zhangzhou in 2016.

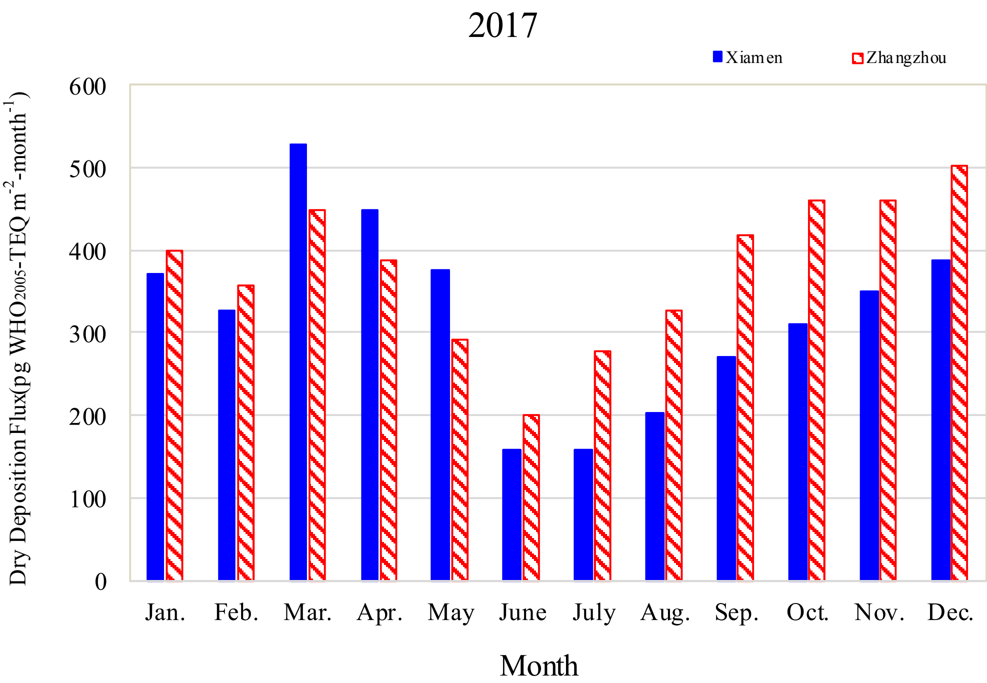

Fig. 6(c). Monthly average dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen and Zhangzhou in 2017.

dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in January (443 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) was the highest, for which the level ratio was at its lowest level in June (273 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) and was approximately 38.3% lower than that in January. In 2016, the monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ ranged from 303 to 546 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$. The annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Zhangzhou was 4889 pg WHO_{2005}-TEQ m^{-2} $years^{-1}$, which was about two times higher than the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Guangzhou in 2014 (2470 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) (Zhu *et al.*, 2017). The maximum monthly mean dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ (546 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) occurred in March, which was about 1.8 times higher than the value in June

(303 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) which was the lowest level. In 2017, the monthly average dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in December was at a maximum of 503 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, the minimum appeared in June at 200 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$, and the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ was 4538 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$.

The highest dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ at Xiamen WHO_{2005}-TEQ m^{-2} $month^{-1}$ occurred in March 2016 (434 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) and 2017 (527 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$) and in January 2015 (443 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$). The lowest dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ occurred in June and July of 2017(158 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$), in June 2016 (186 pg WHO_{2005}-TEQ m^{-2} $month^{-1)}$, and in July and August 2015 (214 pg WHO_{2005}-TEQ m^{-2} $Month^{-1}$). The highest dry deposition fluxes in Zhangzhou occurred in January 2015, in March 2016 and in December 2017, while the lowest dry deposition fluxes occurred in June every year.

Sensitivity Analysis

As sensitivity analysis can provide a basis for confirming some important parameters for $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents. For example, in this study, it was taken into account that ambient temperature, $PM_{2.5}$ concentration, PM_{10} concentration, and total PCDD/F mass concentration may affect $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents. In the case of Xiamen, sensitivity analyses were carried out depending on the ambient air temperature = 22.3°C, PM_{10} = 50.0 µg m^{-3}, $PM_{2.5}$ =27.0 µg m^{-3}, and total-PCDD/F mass concentration = 0.68 pg m^{-3}. The parametric sensitivity for the atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in Xiamen and Zhangzhou are shown in Figs. 7 and 8. The parametric sensitivity for the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen and Zhangzhou are shown in Figs. 9 and 10.

P: initial value of parameters;

ΔP: increase or reduction in parameters;

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

ΔS: response in each of the parameters.

The sensitivity analysis demonstrated that the most two sensitive parameters for atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents were total PCDD/F mass concentration and PM_{10}. In regard to the total PCDD/F mass concentration parameter, when ΔP/P was changed from 0% to +60%, ΔS/S responded from 0% to +31%. However, when ΔP/P was changed from +60% to +100%, ΔS/S responded from +31.2% to 6.82%. This may be because PCDD/F mass concentrations are the root cause of total PCDD/Fs-WHO_{2005}-TEQ, so a change in PCDD/F mass concentration has a significant effect on $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents. When the impact of the rate of change reaches its peak, the impact on $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ decreases accordingly.

It can also be seen in Fig. 7, that influence of the PM_{10} concentration on the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content is more complicated than is the case for parameters. When ΔP/P was changed from 0% to +20%, ΔS/S responded from 0% to +27%. When ΔP/P was changed from +20% to +40%, ΔS/S decreased from +27% to –0.63%.

The sensitivity analysis indicated that air temperature has an effect on atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents, where ΔP/P was increased from 0% to +40%, ΔS/S responded from 0% to –96%. However, a decrease in air temperature has an effect on the atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content. When ΔP/P was decreased from 0% to –40%, ΔS/S also decreased 0% from –27%. Temperature influenced the atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content by altering the gas-particle partitioning of PCDD/Fs, where the high molecular weight PCDD/Fs make a significant contribution to the total PCDD/F mass concentration and mainly exist in the particle phase. When the air temperature was increasing, as more particles bound to PCDD/Fs evaporated to the gas phase, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content was significantly reduced. When the temperature is relatively low, PCDD/Fs mostly remain in the particle phase, and when the temperature reduced, the rest of the gas phase PCDD/Fs change into the particle phase.

The decrease in the $PM_{2.5}$ concentration was negatively correlated with atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents, and there are two stages for the change of the effect of $PM_{2.5}$ concentration: when ΔP/P changed from –60% to +40%, ΔS/S becomes –100% to +17%, but when ΔP/P changed from +40% to +100%, the ΔS/S response was from +17% to –34%. The effect of $PM_{2.5}$ concentration on the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content was mainly represented by particle-bound PCDD/F. The lower $PM_{2.5}$ concentration means that the atmospheric stability will be better, which is conducive to the diffusion and migration of air pollutants, so the PCDD/F also has a significant decline. The $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content is the total PCDD/Fs-WHO_{2005}-TEQ divided by $PM_{2.5}$ concentration, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content also decreased slowly. Since $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content is the ratio of the total PCDD/Fs-WHO_{2005}-TEQ concentration calculated by dividing by $PM_{2.5}$, the higher $PM_{2.5}$ concentration is always accompanied by lower air temperature. The ratio of $PM_{2.5}/PM_{10}$ is high, which means that the rate of increase of $PM_{2.5}$ concentration is greater than that of PM_{10}, but also higher than that of PM_{10}. The increase in the concentrations of PCDD/F and the rate of $PM_{2.5}$ concentration is greater than that of total PCDD/Fs-WHO_{2005}-TEQ concentration, therefore $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content decreases when the $PM_{2.5}$ concentration continues to rise.

In Zhangzhou, sensitivity analyses based on the values of ambient air temperature = 23.1°C, PM_{10} = 58.0 µg m^{-3}, $PM_{2.5}$ = 32.0 µg m^{-3} and total-PCDD/F mass concentration = 0.75 pg m^{-3}. The parametric sensitivity for the atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents of Zhangzhou is shown in Fig. 8.

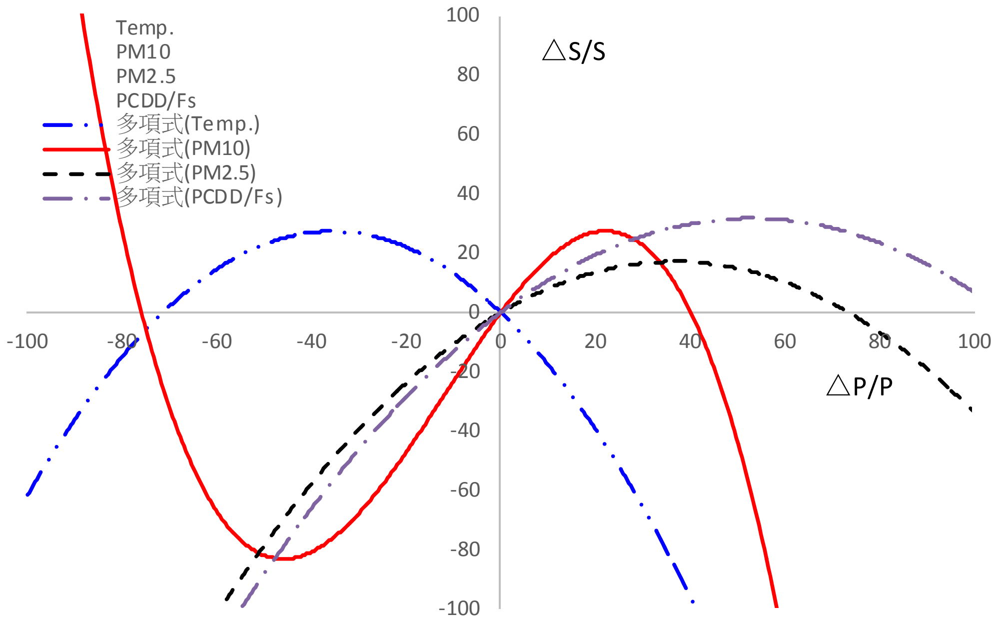

Fig. 7. Sensitivity analysis for the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of Xiamen.

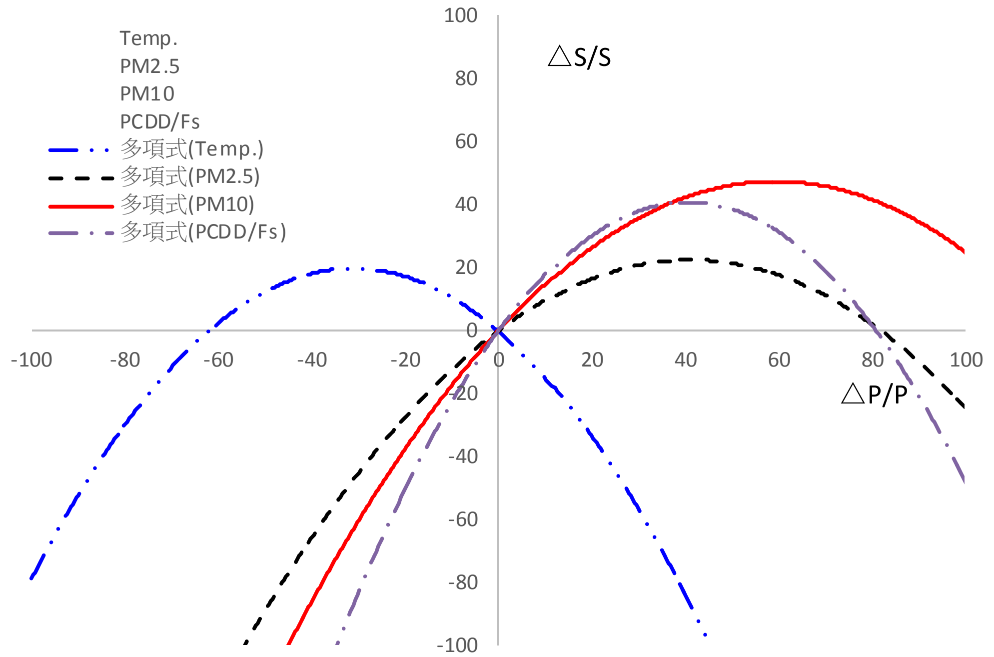

Fig. 8. Sensitivity analysis for the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of Zhangzhou.

The sensitivity analysis demonstrated that the most sensitive parameters for atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents was PM_{10}. In regard to the parameter of total PCDD/Fs concentration, when ΔP/P was increased from 0% to +40%, ΔS/S responded from 0% to +41%. This may be because PCDD/F are the root cause of total PCDD/Fs-WHO_{2005}-TEQ, so the change in PCDD/Fs mass concentration has a significant effect on $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents.

It can also be seen in Fig. 8, that influence of the PM_{10} concentration on the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content is very close to the total PCDD/F concentration. When ΔP/P was changed from 0% to +60%, ΔS/S responded from 0% to +47.5%. This reveals that the PM_{10} concentration also has a close relationship with the total PCDD/F concentration. Their fitting curves basically coincide.

The sensitivity analysis indicated that air temperature has an effect on atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents and can be divided into two stages, when ΔP/P was reduced from –100% to –40%, ΔS/S reflected from –78.7% to 18.2%. However, when ΔP/P was

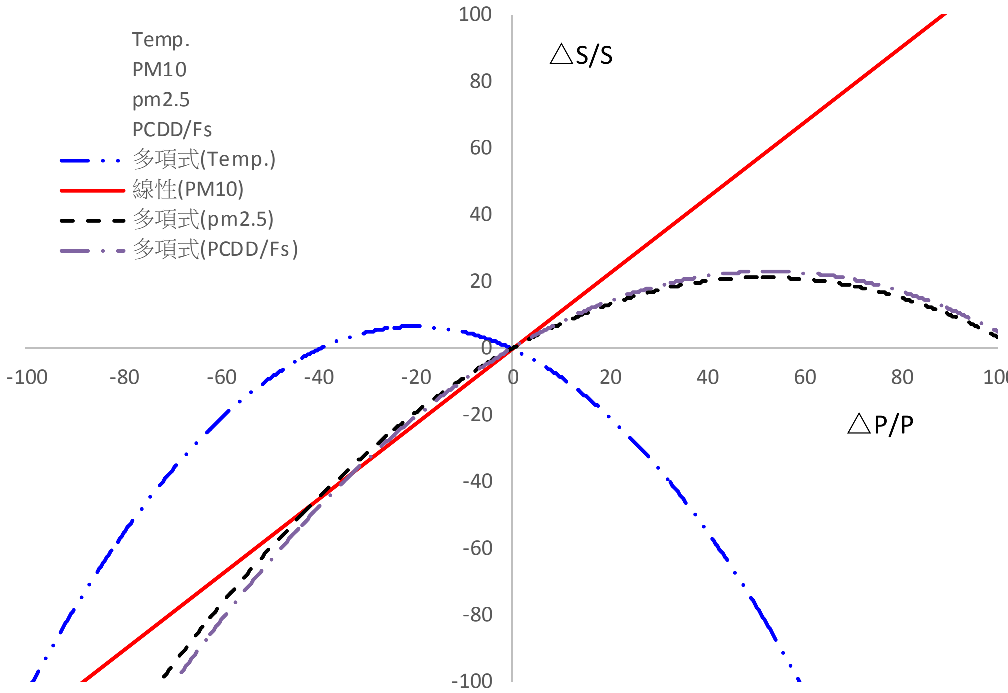

Fig. 9. Sensitivity analysis for the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen.

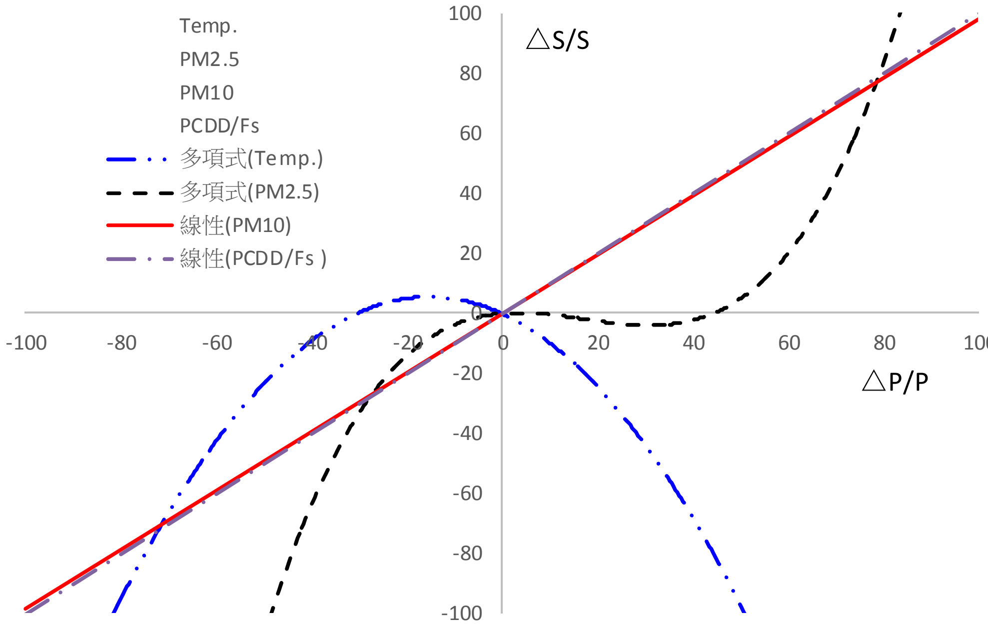

Fig. 10. Sensitivity analysis for the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ in Zhangzhou.

increased from –40% from 40%, ΔS/S decreased 18.2% to –84.4%. The temperature influenced the atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents by changing the gas-particle partitioning of PCDD/Fs, the high molecular weight PCDD/F contributes a lot to the total PCDD/F mass concentration, mainly in the particle phase. When the air temperature was increasing, as more particles bound to PCDD/Fs evaporated to the gas phase, $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content was significantly reduced.

The decrease in the $PM_{2.5}$ concentrations was negatively correlated or positively correlated with the atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, when ΔP/P was decreased from 0% to –40%, ΔS/S reflected from 0% to –65.8%. There are two stages can be divided in influence of increasing $PM_{2.5}$: when ΔP/P was increased from 0% to +40%, ΔS/S reflected from 0% to +22.6%, respectively, but when ΔP/P was increased from +40% to +100%, ΔS/S reflected from +20% to +10% conversely.

The results of the sensitivity analysis showed that the sensitivity of atmospheric $PM_{2.5}$-bound total PCDD/Fs-

WHO_{2005}-TEQ contents to total PCDD/F mass concentration and PM_{10} concentration was very close and high, followed by air temperature and $PM_{2.5}$ concentration.

In Xiamen, $PM_{2.5}$ and PCDD/F mass concentration have similar sensitivity for the dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ. The influence of PCDD/F mass concentration can be divided into two parts: When ΔP/P was decreased from 0% to –60%, ΔS/S also decreased from 0% to –77%; when ΔP/P was increased from 0% to +60%, ΔS/S reflected from 0% to +20.8%, but ΔP/P was increased from +60% to +80%, ΔS/S decreased from 20.8% to 15.3%, respectively. As for PCDD/F mass concentration, when ΔP/P was increased 0% to +60%, ΔS/S reflected from 0% to +22.7%, ΔP/P was increased +60% to +100%, ΔS/S reduced from +22.7% to +5.09%. The parameter of the air temperature has a significant effect on dry deposition flux, can be divide into two stages: when ΔP/P was increased –80% to –20%, ΔS/S also increased from –55.6% to +6.7%, when ΔP/P was altering –20% to +40%, ΔS/S reflected from +6.7% to –54.2%. When the temperature is below 17.0°C, this parameter has a positive influence on dry deposition flux, when the temperature is higher than 17.0°C, the air temperature has negative correlation with the dry deposition of total PCDD/Fs-WHO_{2005}-TEQ. As for PM_{10}, it can be seen that the dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ is most sensitive to PM_{10} concentration.

In the case of Zhangzhou, according to Fig. 8, the sensitivity analysis indicated that the effects of PM_{10} and PCDD/F mass concentration on dry deposition fluxes are very similar, both of them have a significant, positive effect on the dry deposition fluxes of total PCDD/Fs-WHO_{2005}-TEQ. The parameter of $PM_{2.5}$ concentration effect on dry deposition fluxes can be divided into two parts: When ΔP/P was increased –40% to 0%, +40% to +80%, ΔS/S reflected from –63.8% to –14%, –2.3% to +82.8%; when ΔP/P was increased 0% to +40%, ΔS/S decreased from 0% to –2.3%, respectively. As for the effects of air temperature on dry deposition is very like to the city of Xiamen, there are two parts to analyze: when ΔP/P was increased –80% to –20%, ΔS/S reflected from –95.1% to +5%; when ΔP/P was increased –20% to +40%, ΔS/S responded from +5% to –67.6%, respectively.

The above analysis indicates that the parameters most sensitive to the dry deposition of total PCDD/Fs-WHO_{2005}-TEQ are atmospheric PM_{10} concentration and PCDD/F mass concentration, followed by $PM_{2.5}$ concentration and air temperature.

CONCLUSION

1. The $PM_{2.5}$ concentration in Xiamen ranged between 13.0 and 49.0 µg m^{-3} in the three-year period under investigation, with an average value of 27.6 µg m^{-3}. The concentration of $PM_{2.5}$ in Zhangzhou during the same period was 21.0–54.0 µg m^{-3}, with an average of 33.9 µg m^{-3}. The concentration of $PM_{2.5}$ in Xiamen was lower than that in Zhangzhou. Since the two cities are close to the sea, and their heavy industry pollution is relatively low, their $PM_{2.5}$ concentrations are lower than most cities in China. About the seasonal variations, in Xiamen, the average value of the $PM_{2.5}$ concentration over the three years in the summer was 18.3 µg m^{-3}, and the average in winter was 36.3 µg m^{-3}. The values in winter were 2.0 times higher than those in summer. In Zhangzhou, the average value over the three years in summer was 23.3 µg m^{-3}, which was 44.7% lower than that in winter (44.0 µg m^{-3}).
2. The $PM_{2.5}/PM_{10}$ ratio in the three years from 2015 to 2017 in Xiamen was in the range of 0.43–0.70, with an average of 0.56. The $PM_{2.5}/PM_{10}$ ratio in the three years of Zhangzhou was between 0.43–0.77 with an average of 0.55. The high $PM_{2.5}/PM_{10}$ ratio is highly correlated with the higher $PM_{2.5}$ concentration. This shows that $PM_{2.5}$ is the main part of atmospheric particles.
3. In Xiamen, the average $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content during the summer in the study period was 0.131 ng-WHO_{2005}-TEQ g^{-1}, which was lower than the value of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content for the other three seasons; in Zhangzhou, the mean value of $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in the summer was also significantly lower than in the other three seasons. This is due to the fact that summer temperatures are higher and in summer more particles bound PCDD/Fs into vapor phase and $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content was reduced.
4. In terms of the gas partitioning of PCDD/Fs, for Xiamen, the three-year average proportion of gas phase total PCDD/Fs-WHO_{2005}-TEQ concentration in summer (70.0%) is much higher than the other three seasons. In Zhangzhou, the gas phase fraction in summer (66.7%) was the highest among the four seasons. In general, the most relevant parameter in the fraction of gas phase total PCDD/Fs-WHO_{2005}-TEQ is ambient temperature.
5. In 2015, the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen was 3914 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, and in Zhangzhou was 4168 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. In 2016, the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen (3791 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$) was about 23.5% lower than that of Zhangzhou (4889 pg WHO_{2005}-TEQ m^{-2} $years^{-1}$). In 2017, the annual dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ in Xiamen was 3886 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$, and that in Zhangzhou was 4538 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. It can be seen that the dry deposition flux in summer is lower than in the other three seasons. This is because higher temperatures cause more PCDD/Fs to evaporate into the gas phase during the summer.
6. For sensitivity analysis, we can get such a result that whether it is atmospheric $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content or dry deposition flux of total PCDD/Fs-WHO_{2005}-TEQ are most sensitive to total PCDD/F mass concentration and PM_{10} concentration, then atmospheric temperature and $PM_{2.5}$ concentration.

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