



Atmospheric PM_{2.5} and Total PCDD/Fs-WHO₂₀₀₅-TEQ Level: A Case of Handan and Kaifeng Cities, China

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ABSTRACT

From 2015 to 2017, the atmospheric PM_{2.5}, the total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations, PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, and the gas-particle partitioning of PCDD/Fs in Handan and Kaifeng were investigated. In addition, a regression analysis of the air quality index (AQI) and total PCDD/Fs-WHO₂₀₀₅-TEQ concentration was also studied. From 2015–2017, in Handan, the three-year average PM_{2.5} concentrations in spring, summer, autumn and winter were 66.9, 61.3, 74.8, and 138 $\mu\text{g m}^{-3}$, respectively and averaged 85.3 $\mu\text{g m}^{-3}$, which was 1.2 orders of magnitude higher than that in Kaifeng (71.1 $\mu\text{g m}^{-3}$). The three-year average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in spring, summer, autumn, and winter in Handan were 0.88, 0.28, 0.67, and 0.72 ng WHO₂₀₀₅-TEQ g⁻¹, respectively, and averaged 0.64 ng WHO₂₀₀₅-TEQ g⁻¹, which was 1.1 order of magnitude higher than that in Kaifeng (0.57 ng WHO₂₀₀₅-TEQ g⁻¹). The three-year average fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in spring, summer, autumn, and winter in Handan were 26.4%, 62.8%, 28.9%, and 3.34%, respectively, and averaged 30.4%. The three-year average fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in Kaifeng (34.3%) was 1.1 order of magnitude higher than that in Handan (30.4%). Positive correlations between the AQI and the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration were found and their R² values in 2015, 2016, and 2017 were 0.929, 0.921, and 0.876, respectively. Therefore, the AQI can be used to roughly predict the level of total PCDD/Fs-WHO₂₀₀₅-TEQ in ambient air through the use of a regression equation in these two cities.

Keywords: AQI; PM_{2.5}; PM₁₀; PCDD/Fs; Phase distribution; Regression analysis; TEQ.

INTRODUCTION

Atmospheric particulate matter (PM) is the general various solid and liquid particulate matter present in the atmosphere. A variety of particulate matter disperses in the air to form a relatively large, stable suspension system, the aerosol system; therefore, atmospheric particles are part of the atmospheric aerosol (Ghosh *et al.*, 2014). According to the aerodynamic diameters of PM, it can be divided into TSP (ranging from ~0 to 100 μm), PM₁₀ (ranging from ~0 to 10 μm) and PM_{2.5} (ranging from ~0 to 2.5 μm) (Chow *et al.*,

2015; Lu *et al.*, 2016). PM_{2.5} lasts a long time in ambient air and has a significant impact on human health and atmospheric visibility. Some of the particles come directly from natural sources, such as forest fires, volcanic eruptions, and marine spraying, while anthropogenic sources of PM_{2.5} are primarily from petrochemical fuel combustion, biomass burning, waste incineration, automobiles, factories, construction activities, and open burning of non-natural materials.

Primary PM were emitted into the atmosphere directly from emission sources, while the secondary aerosol was formed from precursors, such as sulfur oxides, nitrogen oxides, volatile organic compounds and ammonia, under specific environmental conditions (Lee *et al.*, 2016; Zhu *et al.*, 2017). The chemical and physical composition of PM varies significantly with the locations, climates, and seasons of the year. PM_{2.5} is also known as an inhaled lung particle. Because of its large specific surface area, PM_{2.5} easily enriches a large number of poisonous, harmful substances, stays in the atmosphere longer, and has a longer distance transportation, resulting in greater impact on human health and the atmospheric environment. The WHO set the average PM_{2.5} annual limit at 10 $\mu\text{g m}^{-3}$, increasing by 4% for every 10 $\mu\text{g m}^{-3}$ of human mortality. PM_{2.5} can be inhaled to

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cause pneumonia and dissolve into the blood and cause diseases of the heart and reproductive system (Yang *et al.*, 2017). Greater amounts of PM_{2.5} in the atmosphere can also significantly reduce atmospheric visibility.

The Air Quality Index (AQI) is based on the ambient air quality standards and pollutants that impact human health, ecology, and the environment, and make the routine monitoring of six air pollutants (PM_{2.5}, PM₁₀, SO₂, NO₂, O₃, and CO) concentrations simplified into the form of a single conceptual index value. It classifies air pollution levels and air quality conditions as suitable for representing a city's daily air quality conditions and trends. The AQI can be divided into six levels as follows: 0–50 (Good; Green Color), 51–100 (Moderate; Yellow), 101–150 (Unhealthy for Sensitive Groups; Orange), 151–200 (Unhealthy; Red), 201–300 (Very Unhealthy; Purple) and 301–500 (Hazardous; Maroon), corresponding to the six levels of air quality, the higher the AQI value, the greater the level of air pollution and the more obvious the impact on the human health. Fig. 1 shows the AQI and the corresponding color.

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) were included in the Stockholm Convention on Persistent Organic Pollutants in 2001, and was one of the first 12 persistent organic pollutants (POPs) to be controlled. The atmosphere is the main way for PCDD/Fs to migrate and transport in the environment, which is also the main way for pollution to reach land and aquatic ecosystems. Because PCDD/Fs can diffuse rapidly in the atmospheric environment, can migrate through the atmosphere to regional units, even globally, and can be exchanged through the atmosphere, water, soil and plants into the human food chain, regional monitoring of the atmospheric environment for PCDD/F pollution and investigation into its environmental impact are very important (Fiedler *et al.*, 1992; Hutzinger *et al.*, 1992). PCDD/Fs are semi-volatile organic compounds (SVOCs) and POPs that are toxic to human health and to the ecosystem (Schechter *et al.*, 2006; Cheruiyot *et al.*, 2015,

2016; Li *et al.*, 2016; Redfern *et al.*, 2017). There are 210 possible congeners of PCDD/Fs, of which only 17 with chlorine atoms attached to the 2, 3, 7, and 8 positions have been shown to be especially toxic. In particular, the 2,3,7,8-tetrachloro-dibenzo-*p*-dioxin (2,3,7,8-TCDD) is one of the most toxic chemicals (Van den Berg *et al.*, 1998; Li *et al.*, 2016). It is more than 1,000 times more toxic than potassium cyanide, which is known as the “century poison” (Li *et al.*, 2008). The International Cancer Center has classified it as a human carcinogen (IARC, 1997; U.S. Department of Health and Human Services, 2000; Paustenbach, 2002; Cole *et al.*, 2003). A large number of studies have shown that very low concentrations of PCDD/Fs can cause lethal effects in animals and can also lead immune toxicity, reproductive disorders, and endocrine function changes, the most serious irreversible teratogenic, carcinogenic, mutagenic effect (Zhou *et al.*, 2002). There is given a toxicity equivalence factor (TEF) of 1, and the other 16 congeners have TEF values set relative to this. Furthermore, these TEFs are multiplied by the corresponding concentrations of individual homologues, when then produce the toxic equivalent quantities (TEQs) (Lohmann and Jones, 1998; Van den Berg *et al.*, 1998; Cheruiyot *et al.*, 2016). Since PCDD/Fs are SVOCs, their distribution in the atmosphere between the gas-phase and the particle-phase determines their subsequent fate (Lohmann *et al.*, 2000; Suryani *et al.*, 2015). PCDD/Fs can be removed from ambient air by both dry and wet deposition (Huang *et al.*, 2011; Mi *et al.*, 2012; Redfern *et al.*, 2017).

Two industrial cities in China, Handan City and Kaifeng City, were investigated in this study. Their PM_{2.5} data from 2015 to 2017 were obtained from the Environmental Monitoring Website. The total PCDD/Fs-WHO₂₀₀₅-TEQ ambient concentrations were estimated using the equations reported in previous study, which demonstrate the relationship between PM₁₀ values and total PCDD/F concentrations. Then the gas-particle phase distribution formula was used to calculate PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content.

AQI	Levels of Health Concern	Color
0 to 50	Good	Green
51 to 100	Moderate	Yellow
101 to 150	Unhealthy for Sensitive Groups	Orange
151 to 200	Unhealthy	Red
201 to 300	Very Unhealthy	Purple
301 to 500	Hazardous	Maroon

Fig. 1. Six levels of AQI, the corresponding color and health concern (<https://www.airnow.gov/>).

A linear regression analysis of AQI for total PCDD/Fs- WHO_{2005} -TEQ in ambient air was also conducted.

METHODS

The Investigated Cities

Handan is the third largest city in Hebei Province, which is the economic center of the southern province of China. Handan is an industrial city with pillar industries for steel, coal and cement, all of which are high energy consumption and high air pollutant emission industries. Handan is located at the junction of Shanxi, Henan, Shandong, and Hebei provinces, which can be called the industrial center of the four provinces. It is also the hardest hit areas in terms of particulate matter (Ren *et al.*, 2004; Wang *et al.*, 2012; Zhao Xiujuan *et al.*, 2012). Handan is located in the western foothills of the Taihang Mountains, east of the North China Plain, and is under the influence of the southwest wind. It is vulnerable to impacts from the Yellow River valley pollution emanating from Shanxi Province (Chen *et al.*, 2008). Zhang *et al.* (2012) used online monitoring data of four months to analyze the atmospheric particulate pollution in Handan in the summer and autumn. Wei *et al.* (2014) used online monitoring data to study the characteristics of severe haze pollution in Handan City in January 2013. In recent years, there has been very little research data available for Handan.

Kaifeng City is a key city designated by the state environmental protection administration to prevent and control national air pollution because a significant amount of industrial waste gas and automobile exhaust have caused air pollution in this city. The main air pollution sources include climate and human causes of sand dust, construction dust, coal-based pollution, motor vehicle exhaust pollution, and cooking fume pollution. With the acceleration of the industrialization process and the rapid development of the tertiary industry, in recent years, the number of newly built, reconstructed, and expanded coal-fired boilers in urban areas has expanded to a total of 222, together with 386 originals, totaling 608. These boilers burn 2 million tons of coal each year, generating 430,000 tons of slag, releasing 18,000 tons of soot and 15,000 tons of sulfur dioxide into the atmosphere. Coal has become a major factor causing this area to exceed the limits for major pollutants such as sulfur dioxide and respirable particulate pollution, nearly half of the air pollution load.

Data Collection

The concentrations of $PM_{2.5}$ and PM_{10} in Handan and Kaifeng were obtained from the Environmental Monitoring Station. Both ambient air temperature and rainfall intensity in Handan and Kaifeng were obtained from the Weather Network.

Wang *et al.* (2016) evaluated the concentrations of the 17 PCDD/Fs congeners by using their individual proportions in the total PCDD/F mass concentration, which was deduced from the atmospheric concentration of PCDD/Fs during all four seasons in commercial areas in Taiwan. The total PCDD/F concentration was the average of the results of two analytical equations. One was presented by Huang *et*

al. (2011), shown as $y_1 = 0.0472 + 0.0138x$, and the other was presented by Lee *et al.* (2016), shown as $y_2 = 0.0117x - 0.021$, where y_1 and y_2 comprised total dioxin mass concentrations, and x was the concentration of PM_{10} in the atmosphere. The differences between these two regression equations ranged between 20% and 30%, which was acceptable for PCDD/Fs which were in trace amount.

Gas-Particle Partitioning

Gaseous and particulate concentrations of PCDD/Fs, respectively, were evaluated using gas-particle partitioning multiplying the total concentrations of PCDD/Fs. The gas-particle partitioning was simulated with an equation proposed by several researchers that successfully describes the gas-particle partitioning constant (Yamasaki *et al.*, 1982; Pankow, 1987; Pankow and Bidleman, 1991, 1992):

$$K_p = \frac{F/TSP}{A} \quad (1)$$

K_p : temperature-dependent partitioning constant ($m^3 \mu g^{-1}$)
TSP: concentration of total suspended particulate matter, multiplied by PM_{10} concentration with $1.24 (\mu g m^{-3})$

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

$$\log K_p = m_r \times \log P_L^0 + b_r \quad (2)$$

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 has 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 (K_p) 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)

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

RI: gas chromatographic retention indexes developed by Donnelly *et al.* (1987) and Hale *et al.* (1985).

RESULTS AND DISCUSSION

PM_{2.5} Concentrations

The monthly average $PM_{2.5}$ concentrations in the ambient air of Handan and Kaifeng during the years 2015 and 2017 are shown in Figs. 2(a), 2(b) and 2(c), respectively. In the

case of Handan, in 2015, the monthly average $PM_{2.5}$ concentrations ranged between 58.3 and 176 $\mu g m^{-3}$, with an average of 90.6 $\mu g m^{-3}$, which was a 14.5% order of magnitude lower than for Baoding City (106 $\mu g m^{-3}$) (Xing *et al.*, 2017); during 2016, the concentrations ranged from 40.2 to 226 $\mu g m^{-3}$ and averaged 79.8 $\mu g m^{-3}$, which was a 19.0% order of magnitude lower than for Shijiazhuang City (98.5 $\mu g m^{-3}$) (Xing *et al.*, 2017); in 2017, the $PM_{2.5}$ concentration ranged from 60.2 to 173 $\mu g m^{-3}$ at an average of 82.5 $\mu g m^{-3}$. During the three years under investigation (2015–2017), the highest monthly average value occurred in December (173 $\mu g m^{-3}$), and the lowest $PM_{2.5}$ concentration was in August (58.3 $\mu g m^{-3}$). As to seasonal variations, the four seasons were defined as spring (March, April, and May), summer (June, July, and August), autumn (September, October, and November) and winter (January, February and December). From 2015 to 2017, in Handan, the three-year average $PM_{2.5}$ concentrations in spring, summer, autumn, and winter were 66.9, 61.3, 74.8 and 138 $\mu g m^{-3}$, respectively. As a whole, the three-year average $PM_{2.5}$ concentration in Handan in winter (138 $\mu g m^{-3}$) was 2.3 times that in summer (61.3 $\mu g m^{-3}$).

As for Kaifeng, during 2015, the monthly average $PM_{2.5}$ concentrations ranged from 44.0 to 138 $\mu g m^{-3}$ and averaged 73.8 $\mu g m^{-3}$, which was a 23.2% order of magnitude lower than for Zhengzhou City (96.1 $\mu g m^{-3}$) (Xing *et al.*, 2017); during 2016, these concentrations ranged from 35.1 to 141 $\mu g m^{-3}$ and averaged 71.3 $\mu g m^{-3}$, which was a 9.2% order of magnitude lower than for Zhengzhou City (78.5 $\mu g m^{-3}$) (Xing *et al.*, 2017); in 2017, the monthly average $PM_{2.5}$ concentration ranged from 34.0 to 135 $\mu g m^{-3}$ with an average of 65.9 $\mu g m^{-3}$. During the three years under investigation (2015–2017), the highest monthly average value occurred in January (135 $\mu g m^{-3}$), and the lowest $PM_{2.5}$ concentration was in August (38.6 $\mu g m^{-3}$). As to seasonal variations, in Kaifeng, from 2015–2017, the three-year average $PM_{2.5}$ concentrations in spring, summer, autumn, and winter were 62.6, 41.3, 64.3 and 116 $\mu g m^{-3}$, respectively. Overall, the three-year average of $PM_{2.5}$ concentration in Kaifeng in winter (116 $\mu g m^{-3}$) was 2.8 times that in summer (41.3 $\mu g m^{-3}$).

Compared to the annual average concentrations of $PM_{2.5}$ in Handan, from 2015 (90.6 $\mu g m^{-3}$) to 2016 (79.8 $\mu g m^{-3}$), the $PM_{2.5}$ concentration decreased by 11.9%, while, that from 2016 (79.8 $\mu g m^{-3}$) to 2017 (82.5 $\mu g m^{-3}$) increased by 7.0%. In Kaifeng, between 2015 (73.8 $\mu g m^{-3}$) and 2016 (71.3 $\mu g m^{-3}$), the $PM_{2.5}$ concentration decreased by 3.4%, while that from 2016 (71.3 $\mu g m^{-3}$) to 2017 (65.9 $\mu g m^{-3}$) the $PM_{2.5}$ value decreased by 7.6%.

Figs. 2(d) and 2(e) show the monthly increase/decrease of the $PM_{2.5}$ concentration in Handan and Kaifeng, respectively. For Handan, from 2015 to 2016, most of the monthly average $PM_{2.5}$ concentrations decreased, which were 27%, 34%, 10%, 34%, 35%, 29%, 45%, 3%, and 19% for January, February, March, May, June, July, August, September, and October, respectively, while those in April, November, and December increased by 7%, 18%, and 28%, respectively. From 2016 to 2017, the decreases of the $PM_{2.5}$ concentration were 36%, 3%, 16%, and 48% for March, April, November, and December, respectively,

while those for January, February, May, June, July, August, September, and October increased by 59%, 47%, 61%, 45%, 11%, 55%, 32%, and 63% respectively. As to the seasonal changes of $PM_{2.5}$ concentration, from 2015 to 2016, $PM_{2.5}$ concentrations in spring, summer, and winter decreased by 11%, 36%, and 6% respectively, while that for autumn increased by 0.9%; from 2016 to 2017, both spring and winter levels decreased by approximately 2%, while those in summer and autumn increased by 34% and 15%, respectively.

In Kaifeng, from 2015 to 2016, the monthly average $PM_{2.5}$ concentrations decreased by 14%, 40%, 28%, 17%, 20%, and 33% for February, May, June, July, August, and October, respectively, while those in January, March, April, September, November, and December increased by 9%, 16%, 42%, 12%, 10% and 1%, respectively. From 2016 to 2017, decreased $PM_{2.5}$ concentrations were 4%, 31%, 11%, 18%, 18%, and 34% for January, April, July, September, November and December, respectively, while those for February, May, June, August, and October increased by 44%, 45%, 15%, 6%, and 27%, respectively. From 2015 to 2016, the increases in the seasonal average $PM_{2.5}$ concentrations in spring and winter were 3% and 0.3%, respectively, while the seasonal average $PM_{2.5}$ concentrations in summer and autumn decreased by 22% and 2%, respectively; from 2016 to 2017, those in summer increased by 4%, while those in spring, autumn, and winter decreased by 3%, 9% and 6%, respectively.

In 2017, the annual average $PM_{2.5}$ concentration in Handan (82.5 $\mu g m^{-3}$) and Kaifeng (65.9 $\mu g m^{-3}$) were 8.3 and 6.6 orders of magnitude higher than the WHO air quality regulated standard (10 $\mu g m^{-3}$). With the quick development of the economy and the rapid growth of industrialization, the air quality in China is becoming an increasingly important public issue. A more effective control strategy for reducing the $PM_{2.5}$ concentrations in the ambient air is really very urgent.

According to the above results, it is clear that the ambient air in Handan is more polluted by $PM_{2.5}$ than that in Kaifeng City. This is due to the fact that Handan is a highly industrialized city with large numbers of steel mills and coal-fired power plants, which generated a significant amount of air pollutants.

The monthly average $PM_{2.5}$ concentration is always highest in winter and lowest in summer. This is because the low ground temperature in winter enhances the stability of the atmosphere and thus hinders pollutant diffusion and reduces vertical dispersion in the atmosphere (Tang *et al.*, 2017; Xing *et al.*, 2017). In addition, in the winter cold season, more demand for heating and larger amounts of coal and biomass combustion produce a significant amount of $PM_{2.5}$ and other pollutants. In the summer, in both Handan and Kaifeng, because of their warm temperate semi-humid semi-arid continental monsoon climate, which makes it hot and rainy in the summer, atmospheric instability is good for vertical dispersion and benefits the proliferation of the atmospheric wet deposition necessary to remove large amounts of $PM_{2.5}$ particles from the air and reduce $PM_{2.5}$ concentrations.

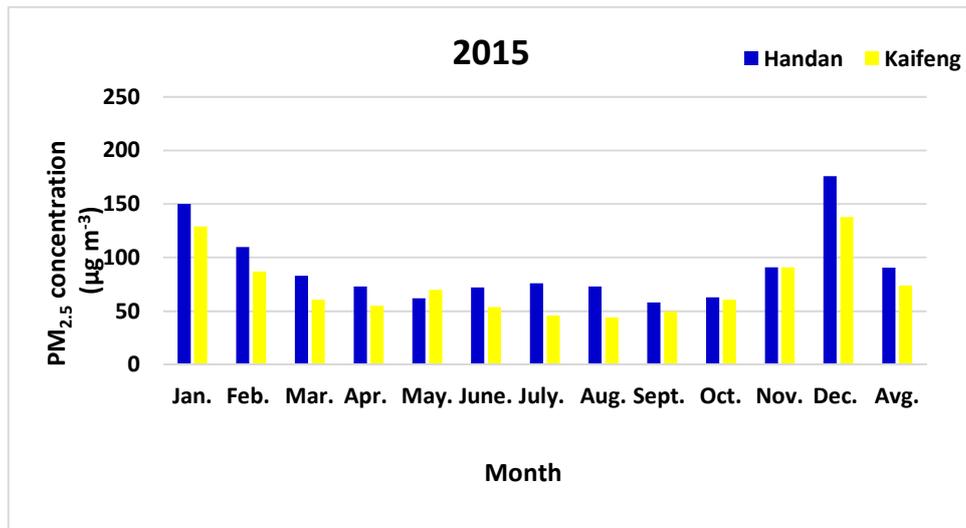


Fig. 2(a). Monthly average atmospheric PM_{2.5} concentration in Handan and Kaifeng during 2015.

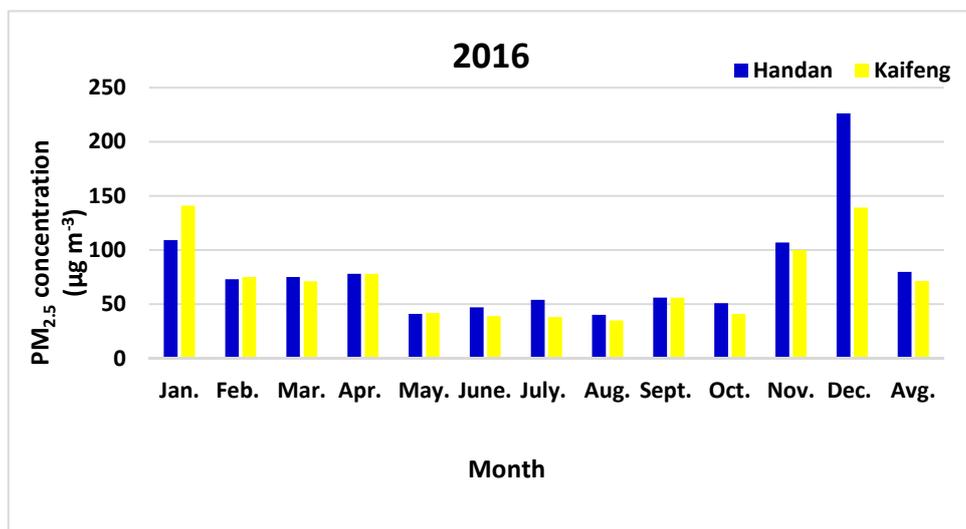


Fig. 2(b). Monthly average atmospheric PM_{2.5} concentration in Handan and Kaifeng during 2016.

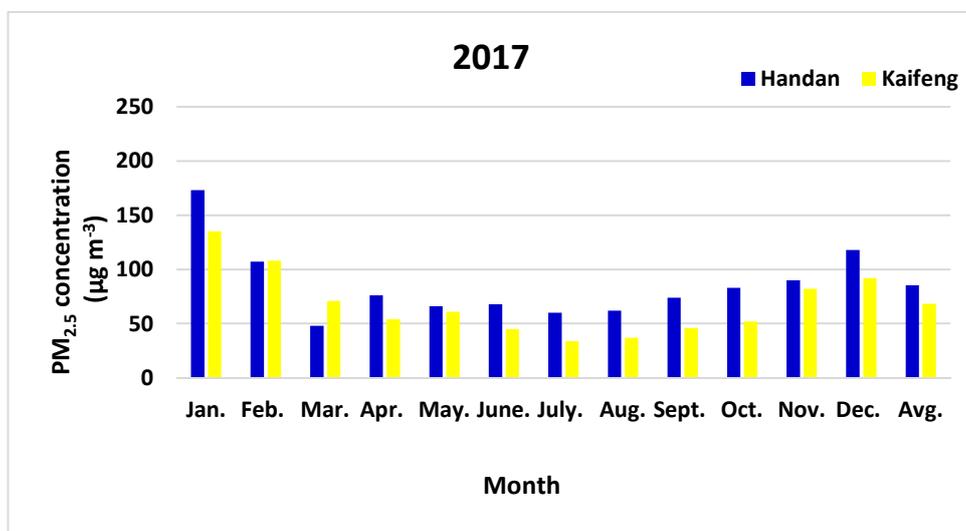


Fig. 2(c). Monthly average atmospheric PM_{2.5} concentration in Handan and Kaifeng during 2017.

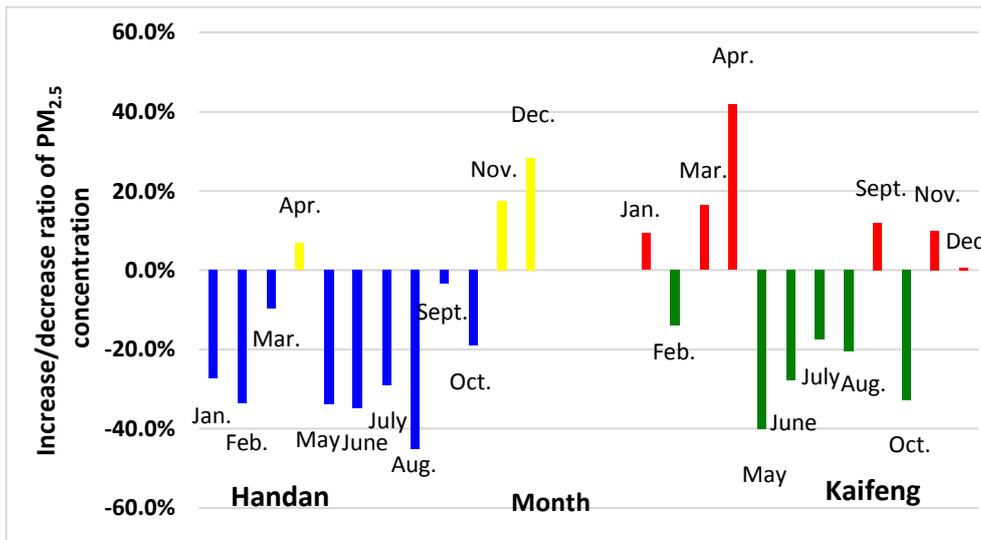


Fig. 2(d). Increase/decrease of PM_{2.5} concentration in Handan and Kaifeng during 2015–2016.

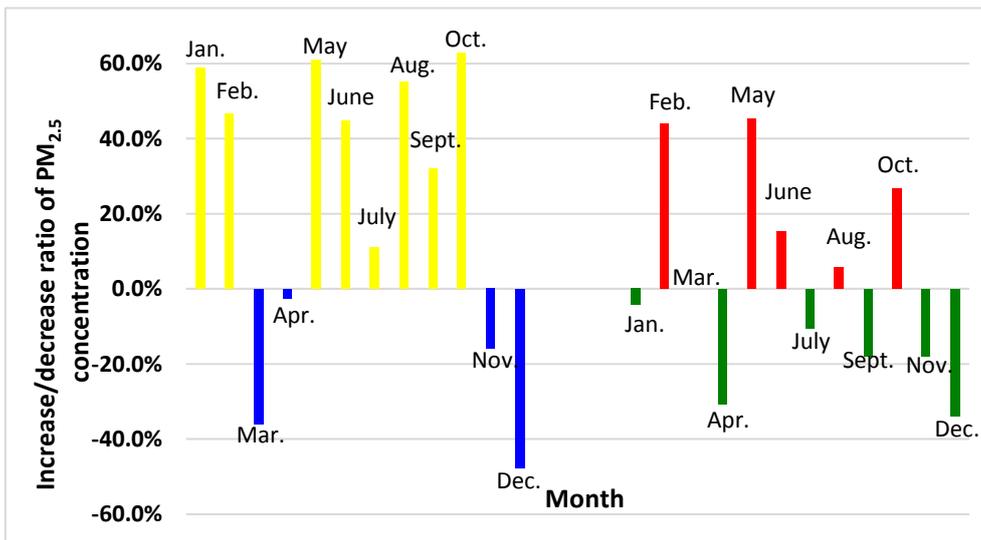


Fig. 2(e). Increase/decrease of PM_{2.5} concentration in Handan and Kaifeng during 2016–2017.

Total PCDD/Fs-WHO₂₀₀₅-TEQ Concentration in Ambient Air

From 2015 to 2017, the total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the ambient air of Handan and Kaifeng cities are shown in Fig. 3. In 2015, in Handan, the monthly average total PCDD/Fs-WHO₂₀₀₅-TEQ concentration ranged between 0.066 and 0.169 pg-WHO₂₀₀₅-TEQ m⁻³ and averaged 0.097 pg-WHO₂₀₀₅-TEQ m⁻³, which was a 3% order of magnitude lower than Baoding City (0.100 pg-WHO₂₀₀₅-TEQ m⁻³) (Xing *et al.*, 2017); December had the highest concentration (0.169 pg-WHO₂₀₀₅-TEQ m⁻³), while June had the lowest (0.066 pg-WHO₂₀₀₅-TEQ m⁻³); in 2016, the monthly average total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations ranged between 0.043 and 0.180 pg-WHO₂₀₀₅-TEQ m⁻³ and averaged 0.089 pg-WHO₂₀₀₅-TEQ m⁻³, which was a 4.7% order of magnitude higher than Baoding City (0.085 pg-WHO₂₀₀₅-TEQ m⁻³) (Xing *et al.*, 2017); December had the highest concentration (0.180 pg-WHO₂₀₀₅-TEQ m⁻³), while

August had the lowest (0.043 pg-WHO₂₀₀₅-TEQ m⁻³); in 2017, the monthly average total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations ranged from 0.045 to 0.162 pg-WHO₂₀₀₅-TEQ m⁻³ and averaged 0.092 pg-WHO₂₀₀₅-TEQ m⁻³. January had the highest concentration (0.162 pg-WHO₂₀₀₅-TEQ m⁻³), while July had the lowest (0.045 pg-WHO₂₀₀₅-TEQ m⁻³).

In 2015, in Handan, the seasonal variations in total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations were 0.099, 0.070, 0.083 and 0.136 pg-WHO₂₀₀₅-TEQ m⁻³ in spring, summer, autumn, and winter, respectively; in 2016, the concentrations were 0.099, 0.046, 0.092 and 0.118 pg-WHO₂₀₀₅-TEQ m⁻³, respectively, and in 2017, the concentrations were 0.101, 0.057, 0.085 and 0.126 pg-WHO₂₀₀₅-TEQ m⁻³, respectively. It can be seen that the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration exhibits obvious seasonal variations, where the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration is generally the largest in winter and smallest in summer.

In Kaifeng, in 2015, the monthly average total PCDD/Fs-

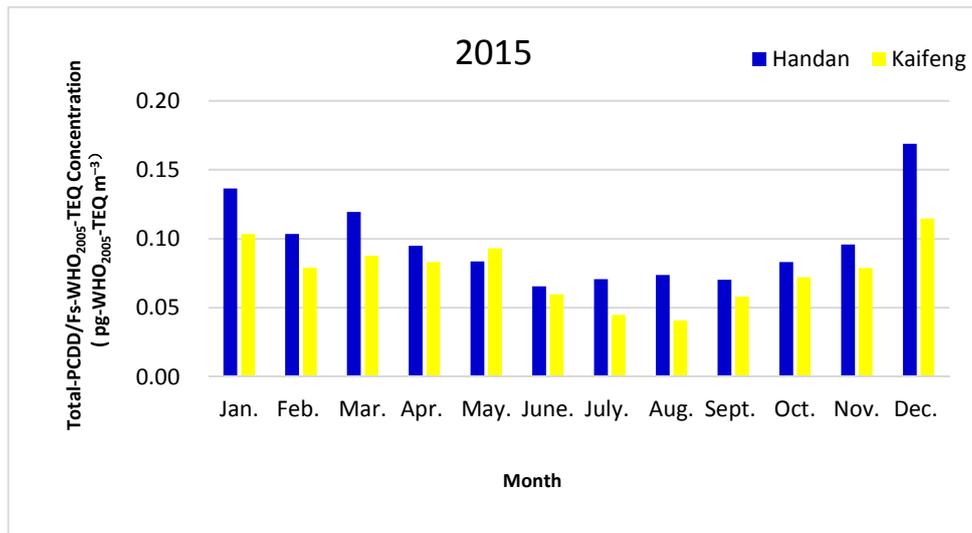


Fig. 3 (a). Monthly average total-PCDD/Fs-WHO₂₀₀₅-TEQ concentration in Handan and Kaifeng during 2015.

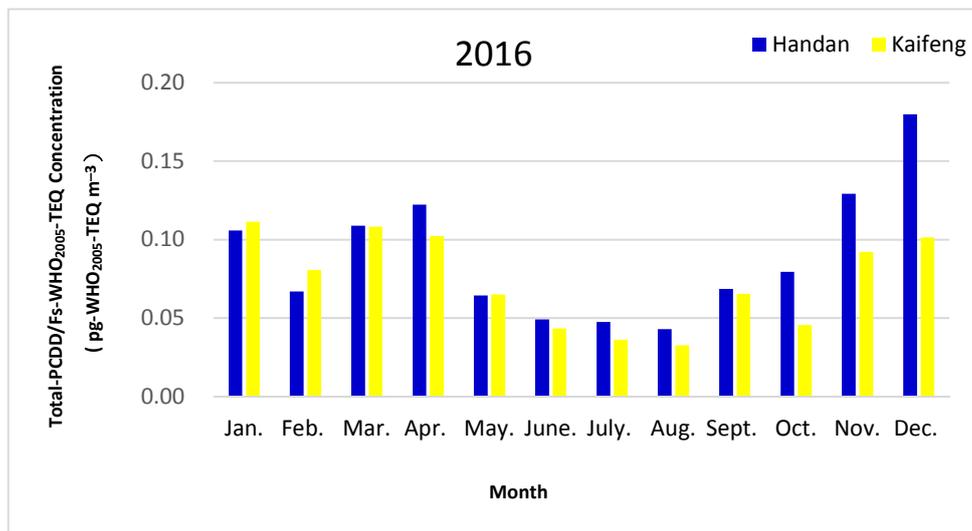


Fig. 3(b). Monthly average total-PCDD/Fs-WHO₂₀₀₅-TEQ concentration in Handan and Kaifeng during 2016.

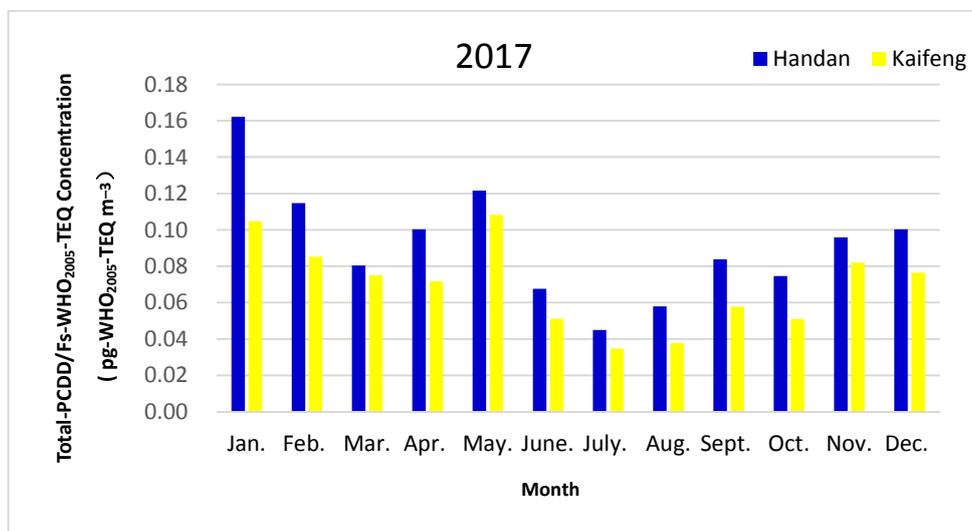


Fig. 3(c). Monthly average total-PCDD/Fs-WHO₂₀₀₅-TEQ concentration in Handan and Kaifeng during 2017.

WHO₂₀₀₅-TEQ concentrations ranged between 0.041 and 0.115 pg-WHO₂₀₀₅-TEQ m⁻³ and averaged 0.076 pg-WHO₂₀₀₅-TEQ m⁻³, which was a 20.8% order of magnitude lower than Zhengzhou City (0.096 pg-WHO₂₀₀₅-TEQ m⁻³) (Xing *et al.*, 2017); December had the highest concentration (0.115 pg-WHO₂₀₀₅-TEQ m⁻³), and August had the lowest (0.041 pg-WHO₂₀₀₅-TEQ m⁻³); in 2016, the monthly average total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations ranged from 0.033 to 0.111 pg-WHO₂₀₀₅-TEQ m⁻³ and averaged 0.074 pg-WHO₂₀₀₅-TEQ m⁻³, which was a 10.8% order of magnitude higher than Zhengzhou City (0.083 pg-WHO₂₀₀₅-TEQ m⁻³) (Xing *et al.*, 2017); January had the highest concentration (0.111 pg-WHO₂₀₀₅-TEQ m⁻³); in 2017, the monthly average total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations ranged between 0.035 and 0.108 pg-WHO₂₀₀₅-TEQ m⁻³ and averaged 0.070 pg-WHO₂₀₀₅-TEQ m⁻³. May had the highest concentration (0.108 pg-WHO₂₀₀₅-TEQ m⁻³), and July had the lowest (0.035 pg-WHO₂₀₀₅-TEQ m⁻³).

In 2015, in Kaifeng, the seasonal variations in total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations were 0.088, 0.049, 0.070 and 0.099 pg-WHO₂₀₀₅-TEQ m⁻³ in spring, summer, autumn, and winter, respectively; in 2016, they were 0.092, 0.037, 0.068 and 0.098 pg-WHO₂₀₀₅-TEQ m⁻³, respectively; and in 2017, they were 0.085, 0.041, 0.064 and 0.089 pg-WHO₂₀₀₅-TEQ m⁻³, respectively. This also verifies that the maximum value for the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration mentioned above occurs in winter, and the minimum occurs in summer.

For monthly PM_{2.5} concentrations and the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in the ambient, it was found that the higher PM_{2.5} value results in a higher total PCDD/Fs-WHO₂₀₀₅-TEQ concentration (Xing *et al.*, 2017). The concentration of the total PCDD/Fs-WHO₂₀₀₅-TEQ in Handan was found to be higher than that in Kaifeng. On the one hand, the latitude of Handan is higher than that of Kaifeng (Xing *et al.*, 2017; Tang *et al.*, 2017), and on the other hand, it is industrial. Handan is a nationally important iron and steel base and has an annual production capacity of 40 million tons. The ranking table of the PCDD/Fs emission sources in the world are presented in Lu *et al.* (2008), where it can be seen that the production processes for steel and other metals in China ranked first as emission sources of PCDD/Fs. Therefore, the total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in Handan are closely related to its high-yielding steel industry. As for the seasonal variations in total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations, the highest concentrations were in winter and the lowest were in summer, which is also related to the consumption of heating coal and atmospheric inversion that occurs in winter.

According to the figure analyses, the total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in the ambient air of Kaifeng are decreasing annually. In addition, the number of motor vehicles has been increasing in Handan. Every year, Hebei Province has added more than 3 million motor vehicles, and this number has continued to grow for the past eight years. In Handan, the number of motor vehicles in the second quarter of 2015 was 1,630,426 units; at the end of 2016, the number of vehicles was 1,665,187 units.

Quaß (2004) pointed out that, comparing the PCDD/F of European vehicle emissions with other sources of PCDD/Fs (the second largest source being boiler furnaces), although PCDD/Fs from European vehicular emissions are not the major sources of emissions, they do have a significant influence on environmental quality and contribute about 2% of the total emissions (Quaß *et al.*, 2004).

PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ content

The contribution of the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in ambient air mainly comes from the particle phase (the contribution of the particle phase ranged from 58.6% to 94.3%). Previous studies in Asian countries have shown that the total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations for the particle phase in summer account for 36.4%–71.5% of their total concentrations (Hayakawa *et al.*, 2004). More than 90% of the PCDD/F studies on the ambient environment have proposed that the PCDD/F mass is mainly concentrated in the particulate phase (Lee *et al.*, 2008). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in Handan and Kaifeng cities from 2015 to 2017 are shown in Fig. 4. In the case of Handan, the annual average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content was 0.63, 0.65 and 0.63 ng WHO₂₀₀₅-TEQ g⁻¹ in 2015, 2016, and 2017, respectively, with an average 0.64 ng WHO₂₀₀₅-TEQ g⁻¹. However, in Kaifeng, the annual average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content was 0.58, 0.58 and 0.54 ng WHO₂₀₀₅-TEQ g⁻¹ in 2015, 2016, and 2017, respectively, and averaged 0.57 ng WHO₂₀₀₅-TEQ g⁻¹, which was approximately an 11.4% order of magnitude lower than that in Handan.

In Handan, the three-year monthly average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content from January to December in order was 0.74, 0.74, 1.07, 0.83, 0.73, 0.32, 0.23, 0.31, 0.45, 0.74, 0.82, and 0.68 ng WHO₂₀₀₅-TEQ g⁻¹, respectively. The highest level occurred in March (1.07 ng WHO₂₀₀₅-TEQ g⁻¹) and was a 4.7 order of magnitude higher than that of the lowest level, which occurred in July (0.23 ng WHO₂₀₀₅-TEQ g⁻¹). In Handan, the three-year average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in spring, summer, autumn, and winter was 0.88, 0.28, and 0.67 and 0.72 ng WHO₂₀₀₅-TEQ g⁻¹, respectively, which showed that in spring (0.88 ng WHO₂₀₀₅-TEQ g⁻¹) to be a 3.1 order of magnitude higher than that in summer (0.28 ng WHO₂₀₀₅-TEQ g⁻¹).

As for Kaifeng, the three-year monthly average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content from January to December in sequence was 0.61, 0.69, 0.92, 0.79, 0.70, 0.34, 0.20, 0.24, 0.40, 0.58, 0.66, and 0.65 ng WHO₂₀₀₅-TEQ g⁻¹, respectively, which showed that the highest level occurred in March (0.92 ng WHO₂₀₀₅-TEQ g⁻¹) and was approximately a 4.6 order of magnitude higher than that of the lowest one, which occurred in July (0.20 ng WHO₂₀₀₅-TEQ g⁻¹). Seasonal variations in the three-year average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in spring, summer, autumn, and winter were 0.80, 0.26, 0.55, and 0.65 ng WHO₂₀₀₅-TEQ g⁻¹, respectively, which indicated that of spring (0.80 ng WHO₂₀₀₅-TEQ g⁻¹) to be a 3.1 order of magnitude higher than that in summer (0.26 ng WHO₂₀₀₅-TEQ g⁻¹).

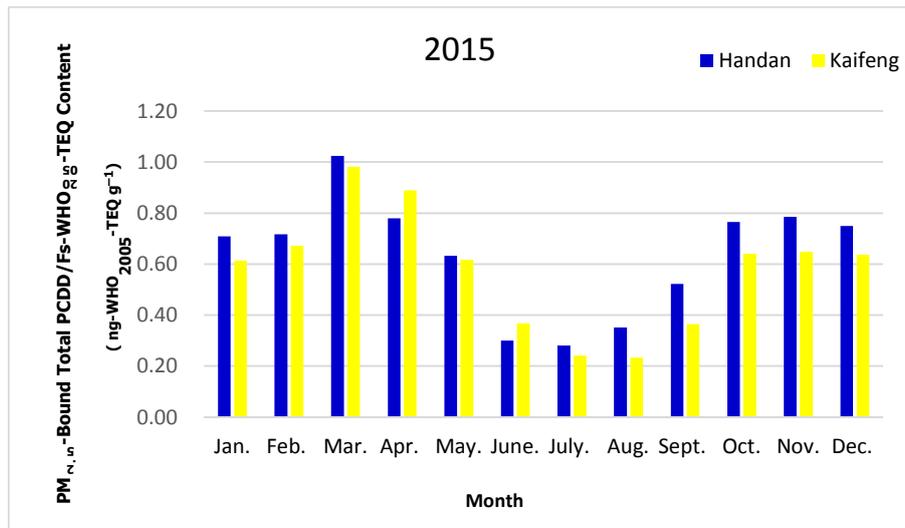


Fig. 4(a). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ Content (ng-WHO₂₀₀₅-TEQ g⁻¹) of Handan and Kaifeng during 2015.

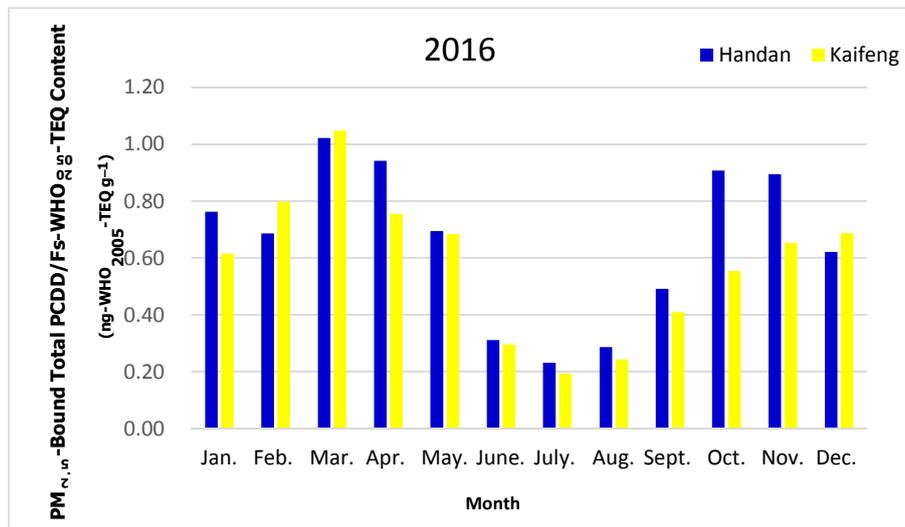


Fig. 4(b). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content (ng-WHO₂₀₀₅-TEQ g⁻¹) of Handan and Kaifeng during 2016.

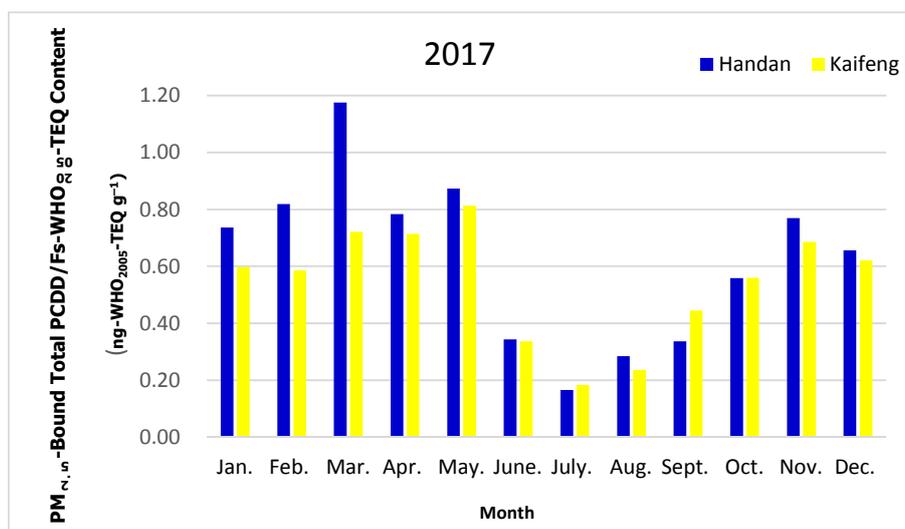


Fig. 4(c). PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content (ng-WHO₂₀₀₅-TEQ g⁻¹) of Handan and Kaifeng during 2017.

A comparison of the seasonal variations in the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content indicates that between 2015 and 2017 in Handan, the average PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in summer was approximately only 58.2%, 64.6%, and 64.5% lower than the average value of the other three seasons (spring, autumn, and winter), respectively. In Kaifeng, from 2015–2017, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in summer was approximately only 58.3%, 64.6%, and 60.4% lower than the average value of the other three seasons (spring, autumn, and winter), respectively. From 2015–2017, the average temperature in the summer (26.9°C) for the three years under investigation in Handan was much higher than that in spring (16.8°C), autumn (15.5°C), and winter (2.4°C), which was very similar in Kaifeng, where from 2015–2017, the average temperature in the summer (27.3°C) was also much higher than that in spring (17.3°C), autumn (16.2°C), and winter (4.2°C). As the temperature rose, a greater amount of PCDD/F mass evaporated from the particle to the gaseous phase, resulting in a lower PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content (Xing *et al.*, 2017).

Gas-Particle Partitioning of PCDD/Fs

The gas-particle partitioning of PCDD/F and other POPs is an important factor that influences the efficiency of atmospheric removal by wet and dry deposition (Bidleman *et al.*, 2000; Cheruiyot *et al.*, 2015, 2016; Redfern *et al.*, 2017). When PCDD/Fs are emitted into the atmosphere, they have a specific distribution between the gas and particle phases based on their concentrations, the atmospheric temperature, vapor pressure, and the ambient air particulate concentration (Hoff *et al.*, 1996). Here, the gas-particle partitioning was calculated from the atmospheric temperature using Eq. (1), Eq. (2), and Eq. (3), and the seasonal gas-particle partitioning of total PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Handan and Kaifeng from 2015–2017 is shown in Fig. 5. It can be clearly seen that the gas-particle partitioning of PCDD/F exhibits obvious seasonal variations. In Handan, from 2015–2017, the three-year average temperatures in spring, summer, autumn, and winter were 16.8, 26.9, 15.5, and 2.4°C, respectively. As for the seasonal variations in gas-particle partitioning, the average fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in spring, summer, autumn, and winter were 26.4%, 62.8%, 28.9% and 3.3%, respectively. In Kaifeng, from 2015–2017, the seasonal average temperatures were 17.3, 27.3, 16.2, and 4.2°C in spring, summer, autumn, and winter, respectively; the average fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in spring, summer, autumn, and winter were 17.3%, 27.3%, 16.2%, and 4.2%, respectively. The above results indicate that the fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ in both cities in summer were all higher than those in the gas phase in spring, autumn, and winter. A similar conclusion has been drawn in previous studies suggesting that gas phase PCDD/Fs have higher fractions in summer than in winter (Wang *et al.*, 2010; Huang *et al.*, 2011; Lee *et al.*, 2016).

Regression Analysis

A regression analysis is used to study the calculation method and theory for the specific dependency of one variable on another variable. In this study, the daily AQI values for Handan and Kaifeng cities were collected and compiled from 2015 to 2017 on the website of the Ministry of Environmental Protection of the People's Republic of China. The AQI for Handan City and Kaifeng City was taken as the X axis, and the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration was taken as the Y axis. A confidence level of 95% was set up to establish the linear regression analysis chart. The regression equations for three years, respectively, were $Y = 2.83 \times 10^{-6} + 6.96 \times 10^{-4}X$, $Y = -3 \times 10^{-3} + 7.02 \times 10^{-4}X$, and $Y = -8.85 \times 10^{-4} + 6.61 \times 10^{-4}X$, respectively. As shown in Fig. 6, there are positive correlations between the AQI and the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in ambient air. The R² values in 2015, 2016, and 2017 were 0.929, 0.921, and 0.876, respectively. The AQI can be used to roughly predict the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in ambient air through a regression equation. For example, in 2015, the AQI values in Handan City and Kaifeng City ranged from 100 to 200, the corresponding total PCDD/Fs-WHO₂₀₀₅-TEQ concentration of about 0.1 to 0.2 pg-WHO₂₀₀₅-TEQ m⁻³. It can be seen from the figure that the distribution of data in 2016 is relatively uniform. The AQI values of 200 to 300 are also distributed, indicating that there are more moderate pollution days in 2016 and air pollution is more serious. The trend of data distribution in 2017 is similar to that of 2016. However, the value of the AQI in 2017 is mostly concentrated between 100 and 150, the numbers of days with good air quality is more, the maximum AQI for both 2015 and 2016 were all close to 500 and the maximum AQI did not exceed 400 during 2017, which showed the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration was approximately 0.3 pg-WHO₂₀₀₅-TEQ m⁻³ is not more than 2015 and 2016 of the maximum, indicating that the air quality of 2017 is better than that of 2015 and 2016.

CONCLUSION

The three-year average PM_{2.5} concentration in Handan (85.3 μg m⁻³) was 1.2 times of magnitude higher than that in Kaifeng (71.1 μg m⁻³). The total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in Handan (0.093 pg-WHO₂₀₀₅-TEQ m⁻³) was 1.3 times of magnitude higher than that in Kaifeng (0.073 pg-WHO₂₀₀₅-TEQ m⁻³). This may relate to the high-yielding steel industry in Handan. The higher consumption of heating coal and atmospheric inversion that occurs in winter resulted in winter exhibited the highest PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in these two cities. The higher temperature in summer cause lower PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content compared to those in winter. Furthermore, highly positive correlations between the AQI and the total PCDD/Fs-WHO₂₀₀₅-TEQ concentration were found and the AQI can be used to roughly predict the level of total PCDD/Fs-WHO₂₀₀₅-TEQ in ambient air through the use of a regression equation in these two cities.

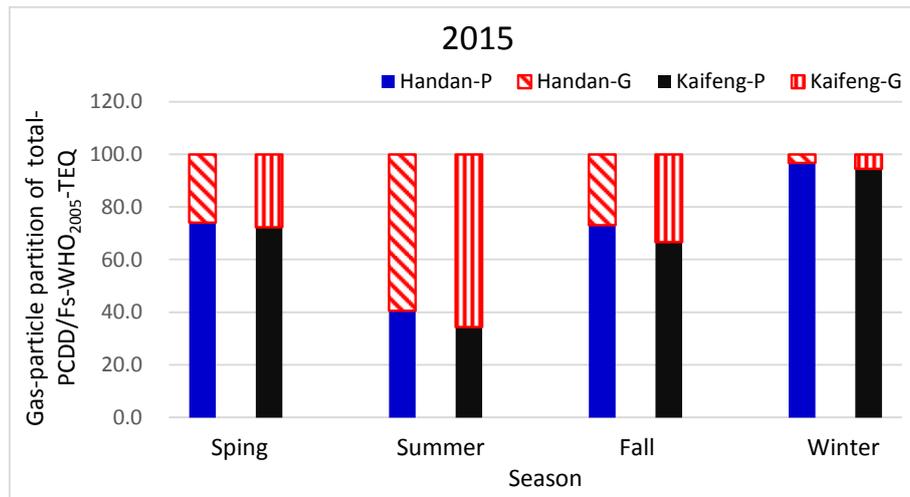


Fig. 5(a). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Handan and Kaifeng during 2015.

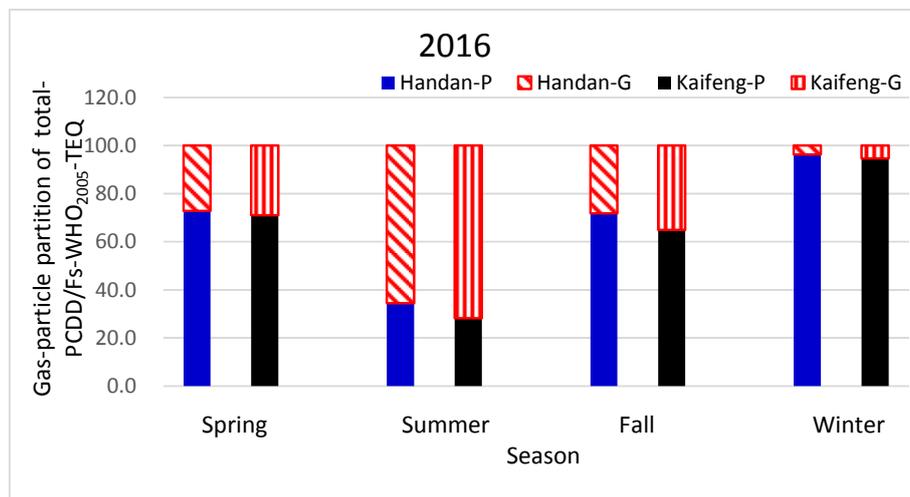


Fig. 5(b). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Handan and Kaifeng during 2016.

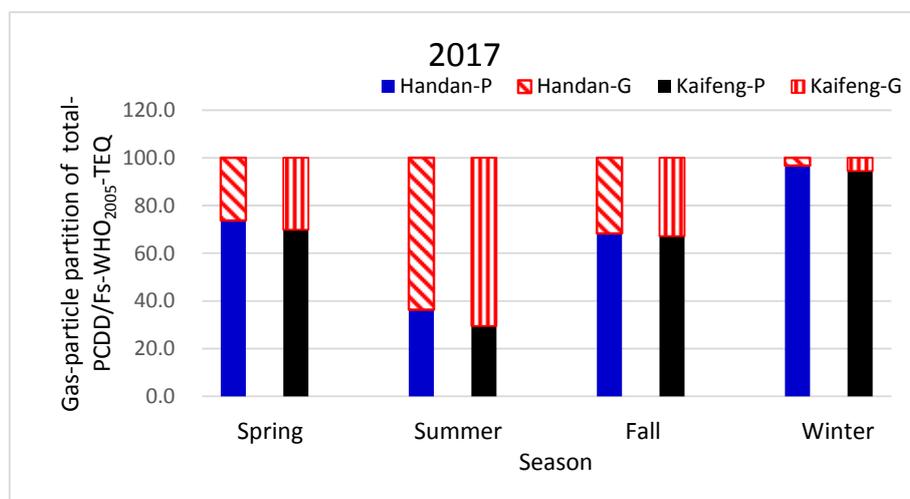


Fig. 5(c). Seasonal variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air in Handan and Kaifeng during 2017.

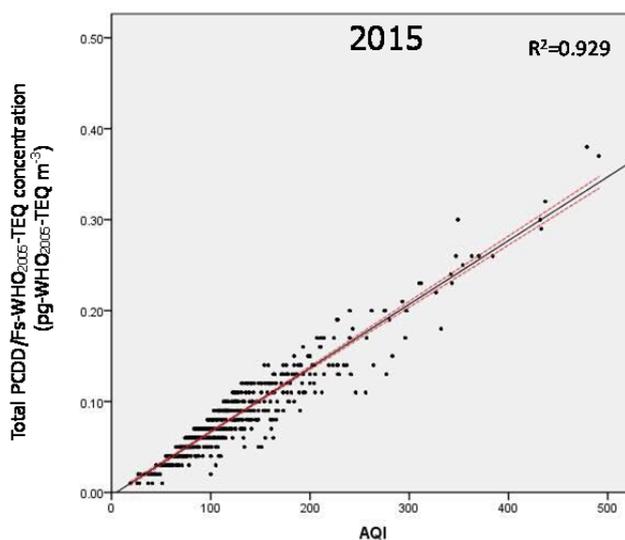


Fig. 6(a). Correlation between AQI and total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in ambient air during 2015.

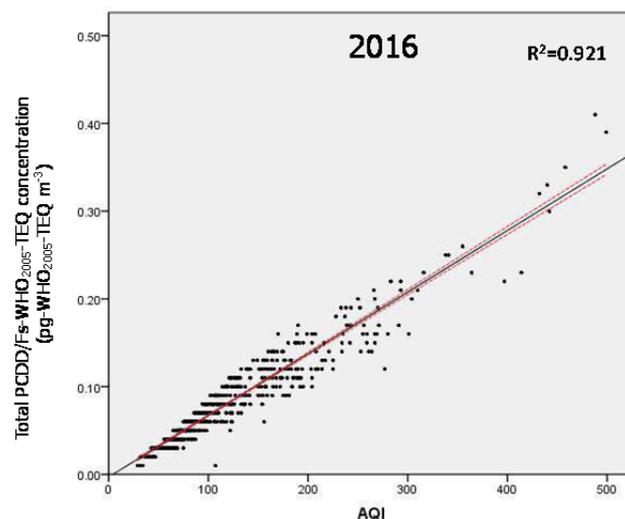


Fig. 6(b). Correlation between AQI and total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in ambient air during 2016.

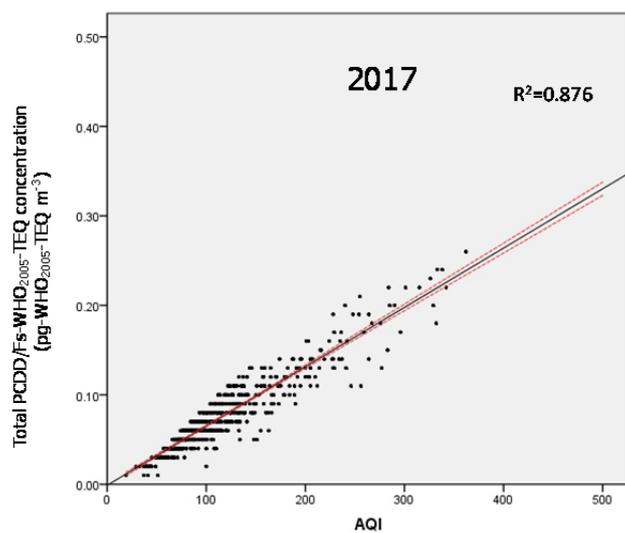


Fig. 6(c). Correlation between AQI and total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in Ambient Air during 2017.

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