Aerosol and Air Quality Research, 17: 2010–2026, 2017

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2017.06.0211

Part II: $PM_{2.5}$ and Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in the Ambient Air of Northern China

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ABSTRACT

During 2014–2016, this study investigated the atmospheric $PM_{2.5}$, R_M, $PM_{2.5}/PM_{10}$, PCDD/Fs-WHO_{2005}-TEQ, and $PM_{2.5}$-bound total-PCDD/Fs-WHO_{2005}-TEQ content of 22 cities in northern China. In general, the more highly industrialized cities had higher $PM_{2.5}$ concentrations. The lowest three-year average concentrations of $PM_{2.5}$ occurred at Lhasa and Qiqihar, and were 25.2 and 36.7 µg m^{-3}, respectively, while the highest concentrations of $PM_{2.5}$ occurred at Baoding and Shijiazhuang, and were 106 and 102 µg m^{-3}, respectively. From 2015 to 2016, the $PM_{2.5}$ concentrations of most cities decreased, but those of several others (Shijiazhuang, Taiyuan, Yinchuan, Lhasa, Sinning, Urumqi, Weinan and Xian) increased, suggesting that the air quality of these was still not well controlled. The average of R_M values of the 22 cities were 7.2, 6.5, and 6.1 in 2014, 2015 and 2016, respectively, which means the $PM_{2.5}$ concentrations in northern China were much higher than the WHO air quality regulated standard (10 µg m^{-3}). A city with a higher $PM_{2.5}$ concentration always had a higher $PM_{2.5}/PM_{10}$ ratio. Among the 22 cities, the six highest three-year averages of total-PCDD/Fs-WHO_{2005}-TEQ concentrations were 0.107, 0.102, 0.095,0.092,0.085 and 0.077 pg-WHO_{2005}-TEQ m^{-3} in Shijiazhuang, Baoding, Zhengzhou, Jinan, Linyi and Xian, respectively; the six lowest three-year averages of total-PCDD/Fs-WHO_{2005}-TEQ concentrations were 0.036, 0.037, 0.045, 0.055, 0.056 and 0.060 pg-WHO_{2005}-TEQ m^{-3} in Qiqihar, Lhasa, Dalian, Harbin, Changchun and Hohhot, respectively. The $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of 12 cities (six cities with higher $PM_{2.5}$ concentration and six with lower $PM_{2.5}$ concentration), during 2014, ranged between 0.444 and 1.000 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.672 ng-WHO_{2005}-TEQ g^{-1}. The $PM_{2.5}$ concentrations, R_M values and PCDD/Fs-WHO_{2005}-TEQ concentrations in the cities of northern China are higher than those in the south, indicating that the air quality in the north is worse than in the south. The results of this study provide a theoretical basis for proposing air pollution control strategies and improving the atmospheric environment in China.

Keywords: $PM_{2.5}$; PCDD/Fs; $PM_{2.5}/PM_{10}$ratio; TEQ; Northern China; Cities.

INTRODUCTION

Due to its harmful properties, the issue of atmospheric $PM_{2.5}$ has received much more attention in recent years. $PM_{2.5}$ is also known as fine particles, which refers to the particulate matter (PM) with aerodynamic diameters of less than 2.5 µm. The characteristics of PM are connected to this size (including physical characteristics such as columnar optical depth, size distribution, single scattering albedo, refractive index, and), while the chemical composition has a direct influence the related toxicity (NASA Facts, 2005; Kahn *et al.*, 2009; Rosenfeld *et al.*, 2014; Cheng *et al.*, 2010). A length of 2.5 µm is equal to 3.6% of the diameter of a human hair, and this means that PM can be easily inhaled deeply in human lungs and directly penetrate the pulmonary alveolar cells, thus mobbing into the blood circulatory system (Yang *et al.*, 2017). $PM_{2.5}$ can absorb toxic constituents, and have many adverse health effects due to its higher surface/mass ratio compared to other PM (WHO, 1999; EPA, 2015). A large amount of research shows that individuals exposed to high levels of $PM_{2.5}$ have

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a significantly higher risk of cardiovascular and pulmonary disease than others. In addition, $PM_{2.5}$ can interfere with atmospheric visibility by scattering and/or absorbing solar light, which impacts the amount and spectral distribution of the incoming solar radiation, and so the Earth's radiation budget (Kalaiarasan *et al*., 2017; Qi *et al*., 2017; Haywood and Boucher, 2000; Ramanathan *et al*., 2001; Kanniah and Yaso, 2010).

Some studies have shown the relationship between $PM_{2.5}/PM_{10}$ and carbon content, such as organic carbon (OC) and elemental carbon (EC), and other chemical indexes, like water-soluble ions, and elements including Be, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, Hg and Pb. A study carried out in southern Taiwan showed that As, Se, Cd, Sb, Hg were the most enriched in $PM_{2.5}$ (Kuo *et al*., 2007). Another study at a city in a semi-arid location in northeast China found that the mass level of $PM_{2.5}$ corresponded to dust storm events, and was much higher in dust storm periods than otherwise. The total mass concentration of nine kinds of water-soluble ions (SO_4^{2-}, NO_3^-, Cl^-, F^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) accounted about 17% of the $PM_{2.5}$ mass in the region (Shen *et al*., 2011). In the city of Tianjin, the concentrations of OC and EC in $PM_{2.5}$ and PM_{10} were all relatively higher in both winter and fall and lower in summer and spring, the similar conclusions have been obtained in a study of the indoor and outdoor carbonaceous pollution in Shaanxi, China (Gu *et al*., 2010; Zhu *et al*., 2010). A study of contamination of polycyclic aromatic hydrocarbons bound to $PM_{10}/PM_{2.5}$ in Xiamen, China, showed that between December 20 and 29, 2004, the concentrations of total 16 polycyclic aromatic hydrocarbons were 5.20–28.1 ng m^{-3} in PM_{10} and 3.04–11.3 ng m^{-3} in $PM_{2.5}$. The concentrations of OC and EC were 9.83–15.6 µg m^{-3} and 1.41–4.73 µg m^{-3}, respectively, as associated with PM_{10}. This suggests that the highest concentration of Σ16PAHs always occurred at areas with factories or traffic, and thus industry, vehicles and coal combustion are the major contributors of PAHs in the ambient air in Xiamen (Wang *et al*., 2007).

Atmospheric aerosols that include $PM_{2.5}$ matters refer to small liquid and solid particles suspended in the air (Wilson *et al*., 2002), and these can be emitted from multiple sources and have complex compositions. Sources such as industrial activities, energy production, construction, urban waste treatment and vehicle exhausts constitute anthropogenic sources, while dust storms, volcanic and oceans activities are natural sources. Due to the characteristics of ambient aerosols, they can significantly affect air quality, the global environment and human health.

Toxic substances such as dioxins, polycyclic aromatic hydrocarbons and heavy metals often attached to $PM_{2.5}$. Polychlorinated dibenzo-*p*-dioxin and dibenzofurans (PCDD/Fs) are unintentional by-products of combustion processes and many industrial activities, such as waste incineration, metal production, power and heating facilities and chemical manufacture processes. After being emitted from combustion sources, they are distributed in both gas and particle phases in the atmosphere. Climate change and increasing climate variability have the potential to influence the emission, distribution and degradation of PCDD/Fs (Bogdal *et al*., 2010; Chi *et al*., 2014, 2015). Factors that have been proposed by the United Nations Environment Programme as the key ones influencing the environmental fate of transportation of PCDD/Fs include the re-evaporation of secondary sources, wind, precipitation, ocean currents, the melting of ice caps and mountains glaciers at the poles, the frequency of extreme weather events, the degradation and transportation of POPs, environmental partitioning and biotic transportation (UNEP/AMAP, 2011; Chi *et al*., 2016).

Many studies have been carried on $PM_{2.5}$, PCDD/Fs, and the relationships between them. The foci of previous works can be classified into several categories: $PM_{2.5}$ emission sources, regional distribution, health impacts, analysis and simulation of concentration and chemical compositions. With the industrial production rate developing very rapidly in China, air pollution has become one of the main problems the country faces. In recent years, haze episodes have occurred frequently in northern Chinese cities, such as Beijing, Tianjin, and Hebei (Lang *et al*., 2012). Some researchers focused on the $PM_{2.5}$ pollution circumstance in recent years of Chinese special sites. A study measured the $PM_{2.5}$ concentration in ambient air of three sites in Shijiazhuang during 18–22 January 2016, aimed to understand the chemical characteristics and potential source regions of $PM_{2.5}$ in Shijiazhuang, China. The results showed that the potential sources of $PM_{2.5}$ in Shijiazhuang were from the Beijing-Tianjin region and Shandong Province (Chen *et al*., 2017). Other researchers investigated the long-term trend (2000–2015) of $PM_{2.5}$ and found that it generally decreased from 2000 to 2015, with the changes seen in the most polluted season (winter) being related to emission control effects and meteorological conditions (Lang *et al*., 2017). As one of the largest cities in China, Beijing suffers from a great number of air pollution problems, and haze incidents have been occurring more frequently in recent years (Zheng *et al*., 2014). A study showed that the sum of the concentrations of the seventeen 2,3,7,8-PCDD/Fs that were examined, and those of the concentrations of the thirteen 2,3,7,8-PBDD/Fs that were analyzed, were 1499–2799 fg m^{-3} (95.4–175.4 fg I-TEQ m^{-3}) and 1171–2424 fg m^{-3} (42.2–109.3 fg-I-TEQ m^{-3}), respectively. Moreover, the PXDD/Fs were mainly (90%) in the particulate phase in the ambient air samples collected in a suburban part of Beijing. The fraction of the total PXDD/Fs concentration that was in the particulate phase increased as the particle size decreased. More than 80% of the total PXDD/F concentrations were in the form of particles of d_{ae} < 2.5 µm (Zhang *et al*., 2015).

In light of these earlier work, the objectives of this study were to investigate the (1) $PM_{2.5}$ concentrations, (2) R_M ratio, (3) $PM_{2.5}/PM_{10}$ ratio, (4) total-PCDD/F-WHO_{2005}-TEQ concentrations, and (5) $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in the ambient air of northern China. The results of this study are expected to provide useful information to help in finding more efficient strategies to improve the air quality in China.

METHODS

This study examined a total of 22 cities belonging to 14 provinces and two municipalities in northern China. These cities were Harbin and Qiqihar in Heilongjiang province, Changchun in Jilin, Dalian and Shenyang in Liaoning, Shijiazhuang and Baoding in Hebei, Hohhot in Inner Mongolia, Jinan and Linyi in Shandong, Taiyuan in Shanxi, Yinchuan in Ningxia, Lanzhou in Gansu, and Lhasa in Tibet, along with the municipalities of Beijing and Tianjin. Both the $PM_{2.5}$ and PM_{10} concentrations in each city were obtained from the website of Air Quality Monitoring Stations (The Real Atmosphere Network).

Since PCDD/Fs exist in both particle and gas phases, the partitioning fraction will affect the atmospheric deposition processes. The partitioning between the two phases is highly dependent on their vapor pressure and the ambient temperature. The gas-particle partitioning is described by the following equation (Yamasaki *et al.*, 1982; Pankow, 1987; Pankow and Bidleman, 1991, 1992):

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

where K_p: temperature-dependent partitioning constant ($m^3\ \mu g^{-1}$);
TSP: concentration of total suspended PM ($\mu g\ m^{-3}$), as calculated by the factor $TSP/PM_{10} = 1.24$ (Sheu *et al.*, 1996);
F: concentration of the compound 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^o, gives

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

where P_L^o: subcooled liquid vapor pressure (Pa);
m_r: slope of the plot of log Kp vs. log P_L^o, –1.29;
b_r: y-intercept in the plot of log Kp vs. log P_L^o, –7.2 (Chao *et al.*, 2004).

The P_L^o used with PCDD/Fs was corrected by earlier researchers, although the correction has since been re-developed

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

where RI: gas chromatographic retention indexes (GC-RI), referring to Hale *et al.* (1985) and Donnelly *et al.* (1987);
T: ambient temperature (K) (Hung *et al.*, 2002).

The cities chosen in this study are either provincial capitals or other densely populated cities. We evaluated each concentration of the 17 PCDD/Fs congeners by using its proportion in the total PCDD/F mass concentration, which is deduced from the atmospheric concentration of PCDD/Fs during four seasons at commercial areas in Taiwan (Wang *et al.*, 2017). The total PCDD/F concentration is the average of the results of using two formulas that demonstrate the relationship between PM_{10} values and total PCDD/F mass concentrations. One was presented by Huang *et al.* (2011) shown below as Eq. (4) and the other was presented by Lee *et al.* (2016) shown as Eq. (5). The correlation coefficients of the two equations are as high as 0.9438 and 0.9855, respectively.

$$y = 0.0472 + 0.0138x \quad (4)$$

$$y = 0.0117x - 0.021 \quad (5)$$

where y: total PCDD/Fs concentration ($pg\ m^{-3}$);
x: PM_{10} concentration ($\mu g\ m^{-3}$).

To obtain the toxicity equivalent (TEQ), the concentrations of PCDD/F congeners are multiplied by their respective TEF values. There are two TEF schemes, the International Toxicity Equivalent (I-TEF) and World Health Organization TEF (WHO_{2005}-TEF) (Cheruiyot *et al.*, 2016). In this study, the WHO_{2005}-TEF was used to calculate TEQ concentration, as this is the stricter scheme. The total TEQ of PCDD/Fs is the sum of all the individual TEQ.

RESULTS AND DISCUSSION

$PM_{2.5}$ Concentration

The $PM_{2.5}$ concentration reflects the air pollutant status of a region. In addition, the $PM_{2.5}$ concentration influences both air visibility and human health. The corresponding $PM_{2.5}$ concentrations during the period between 2014–2016 in 22 cities of northern China are shown in Tables 1(A)–1(D). Among the 22 cities, the lowest annual average $PM_{2.5}$ concentrations during 2014–2016 were all found to be at Lhasa in Tibet and Qiqihar in Heilongjian, which had concentrations of 23.5 and 37.5 $\mu g\ m^{-3}$, 24.6 and 37.2 $\mu g\ m^{-3}$ and 27.4 and 35.3 $\mu g\ m^{-3}$, in 2014, 2015, and 2016, respectively. While during 2014 and 2015, the highest annual average $PM_{2.5}$ concentrations both occurred in Baoding City in Hebei, with the levels of 120 and 106 $\mu g\ m^{-3}$, respectively; however in 2016, the highest annual average $PM_{2.5}$ concentration was 98.5 $\mu g\ m^{-3}$ at Shijiazhuang City in Hebei. During 2014–2016, the lowest three-year average concentrations of $PM_{2.5}$ occurred at Lhasa and Qiqihar, and were 25.2 and 36.6 $\mu g\ m^{-3}$, respectively. While the highest concentrations of three-year average $PM_{2.5}$ occurred at Shijiazhuang and Baoding City, as 102 and 106 $\mu g\ m^{-3}$, respectively.

The six cities with the highest three-year average $PM_{2.5}$ concentrations (HAC) among the 22 cities were Baoding, Shijiazhuang, Zhengzhou, Jinan, Beijing and Linyi, with a range between 79.1 and 106 $\mu g\ m^{-3}$, and an average of 89.7 $\mu g\ m^{-3}$. The six cities with the lowest three-year average $PM_{2.5}$ concentrations (LAC) among the 22 cities were Lhasa, Qiqihar, Hohhot, Dalian, Yinchuan and Sinning, with a range between 25.2 and 52.5 $\mu g\ m^{-3}$, and an average of 41.6 $\mu g\ m^{-3}$. These results show that the six cities with the highest three-year average $PM_{2.5}$ concentrations (HAC) levels were approximately 2.2 times higher than that the LAC. The six cities with the highest $PM_{2.5}$ concentrations were Wuhan, Hefei, Chengdu, Luzhou, Changsha and Nanjing, with $PM_{2.5}$ concentrations between 47.8 and 80.5 $\mu g\ m^{-3}$, and an average of 64.4 $\mu g\ m^{-3}$. The six cities with the lowest $PM_{2.5}$ concentrations were Sanya, Haikou, Quanzhou, Fuzhou,

Table 1(A). Atmospheric $PM_{2.5}$ concentration in various cities (2014–2016) (Unit: µg m^{-3}).

Province	Month	Heilongjiang		Jilin	Liaoning	
City		Harbin	Qiqihar	Changchun	Dalian	Shenyang
2014	Jan.	123	57.1	89.0	70.7	85.7
	Feb.	114	65.5	84.8	71.2	96.3
	Mar.	56.4	37.8	68.0	68.3	78.4
	Apr.	48.6	26.6	55.0	56.1	64.7
	May.	27.7	14.7	33.7	38.1	47.6
	June	29.7	14.9	38.0	41.5	47.7
	July	48.9	41.7	52.7	45.7	53.1
	Aug.	28.7	18.4	36.9	41.9	48.6
	Sep.	21.8	13.8	26.3	28.9	42.8
	Oct.	120	44.7	141	48.5	93.9
	Nov.	145	68.7	81.0	69.3	107
	Dec.	98.8	46.2	79.4	47.9	82.3
Range		21.8–145	14.7–69.0	26.3–141	28.9–71.0	42.8–107
Mean		71.9	37.5	65.5	52.3	70.7
2015	Jan.	120	51.2	104	54.7	110
	Feb.	113	43.7	83.6	59.4	100
	Mar.	62.8	40.7	54.3	55.7	72.2
	Apr.	51.5	29.0	52.7	46.1	66.2
	May.	27.2	13.8	37.1	31.1	42.9
	June	33.6	15.9	38.0	31.5	37.5
	July	31.0	17.7	38.1	24.9	36.7
	Aug.	24.0	17.0	27.9	35.5	31.8
	Sep.	22.0	16.6	28.1	23.6	36.9
	Oct.	56.6	37.5	78.5	34.3	68.5
	Nov.	148	83.9	126	93.1	146
	Dec.	145	79.5	104	82.4	107
Range		22.0–148	13.8–83.9	27.9–104	22.4–93.1	31.8–146
Mean		69.6	37.2	64.4	47.7	71.3
2016	Jan.	96.1	42.6	73.6	53.5	77.2
	Feb.	66.7	43.0	45.7	38.9	54.3
	Mar.	55.2	43.2	59.7	47.5	66.5
	Apr.	40.2	31.0	34.5	45.7	44.6
	May.	27.0	23.7	28.1	37.7	38.8
	June	24.5	18.1	29.7	30.4	35.8
	July	24.4	22.3	27.0	24.5	37.8
	Aug.	22.9	17.0	25.4	18.8	30.6
	Sep.	23.3	20.3	27.5	27.7	37.7
	Oct.	42.7	33.1	51.2	31.2	52.2
	Nov.	99.5	61.9	71.4	49.4	73.6
	Dec.	91.6	67.1	80.1	58.5	95.6
Range		22.9–99.5	17.0–67.1	25.4–80.1	18.8–58.5	30.6–95.6
Mean		51.2	35.3	46.2	38.7	53.7

Kunming and Shenzhen, with levels between 13.6 and 32.9 µg m^{-3}, and an average of 26.0 µg m^{-3} (Tang *et al.*, 2017). The average $PM_{2.5}$ concentrations of the six northern cities with highest and lowest $PM_{2.5}$ concentration are 1.39 and 1.60 times higher than those seen in the south, respectively.

The high $PM_{2.5}$ concentrations in Shijiazhuang and Baoding City may be due to them both being highly industrialized, with a lot of coal and other fossil fuel combustion for heating. In Hebei province, there are large amount of steel, building materials, petrochemical and power production facilities, which all increase $PM_{2.5}$ emissions. In addition, the north wind in winter and spring caused by terrain conditions is not conducive to the diffusion of regional pollutants. Moreover, the south wind in summer and autumn blocks the southwest air stream of Hebei Province, which has a great influence on the air quality of this area. A related study pointed that the low temperature of a region will work against vertical ventilation and prevent the dispersion of contaminants (Tang *et al.*, 2017). The temperature in the north is lower than in the south, especially in winter, and this produces a greater need to burn coal for heating, which thus could make the air quality worse in the north. Among the 22 cities in northern China examined in this study, the lowest $PM_{2.5}$ concentrations were all found to be at Lhasa, with the

Table 1(B). Atmospheric $PM_{2.5}$ concentration in various cities (2014–2016) (Unit: μg m^{-3}).

Province	Month	Beijing	Tianjin	Hebei		Inner Mongolia
City				Shijiazhuang	Baoding	Hohhot
2014	Jan.	93.8	113	212	202	61.3
	Feb.	148	96.3	189	175	59.7
	Mar.	93.6	112	138	128	44.9
	Apr.	88.6	84.6	107	87.2	40.3
	May.	61.1	69.8	76.3	69.3	37.0
	June	54.4	57.9	84.6	71.9	26.4
	July	89.1	75.5	93.3	88.0	31.6
	Aug.	62.3	56.9	72.8	75.4	25.0
	Sep.	65.4	55.8	68.5	81.5	28.8
	Oct.	119	99.0	145	148	50.6
	Nov.	86.2	111	118	149	51.4
	Dec.	57.8	107	116	168	70.7
Range		54.4–148	55.8–113	68.5–212	69.3–202	25.0–70.7
Mean		84.9	86.6	118	120	44.0
2015	Jan.	96.5	100	146	191	59.1
	Feb.	93.0	79.5	112	159	51.2
	Mar.	85.6	72.4	93.4	110	37.6
	Apr.	70.8	64.4	77.1	80.9	27.4
	May.	55.6	51.3	59.7	64.6	25.3
	June	60.2	59.3	63.1	61.0	21.3
	July	61.1	48.6	68.8	66.6	27.0
	Aug.	44.9	49.9	62.8	71.0	20.8
	Sep.	49.9	44.2	42.3	62.1	26.5
	Oct.	74.1	53.9	49.3	81.5	38.9
	Nov.	119	88.2	120	112	75.7
	Dec.	152	125	163	214	102
Range		44.9–151	44.2–125	42.3–163	61.0–214	21.3–102
Mean		80.2	69.7	88.1	106	42.7
2016	Jan.	67.9	73.8	130.8	149.3	59.7
	Feb.	43.5	50.2	71.4	83.4	46.5
	Mar.	92.8	80.9	82.3	76.7	48.7
	Apr.	68.8	63.5	60.3	68.5	28.4
	May.	53.6	50.6	54.2	65.3	33.2
	June	59.4	53.5	45.6	54.3	23.7
	July	69.1	52.7	65.7	71.0	23.6
	Aug.	46.6	40.8	25.2	37.7	17.9
	Sep.	54.6	52.4	84.2	69.7	24.1
	Oct.	84.6	63.7	116.1	96.1	37.3
	Nov.	99.5	104.3	169.3	142.6	73.2
	Dec.	132.7	135	276.3	190.6	65.4
Range		43.5–132.7	50.2–104.3	25.2–276.3	37.7–190.6	17.9–65.4
Mean		72.8	68.5	98.5	92.1	40.1

concentrations of 23.5, 24.6, and 27.4 μg m^{-3}, in 2014, 2015, and 2016, respectively. While in southern China, the lowest $PM_{2.5}$ concentrations were all found to be at Sanya, with the concentrations of 18.5, 17.1, and 21.5 μg m^{-3}, in 2014, 2015, and 2016, respectively. The concentrations seen in Lhasa were 1.3, 1.4 and 1.3 times higher than those in Sanya in 2014, 2015, and 2016, respectively. As mentioned above, the highest $PM_{2.5}$ concentrations were found to be at Baoding, with levels of 120 and 106 μg m^{-3} in 2014 and 2015, respectively, while during 2016 the highest levels were at Shijiazhuang, with a $PM_{2.5}$ concentration of 98.5 μg m^{-3}. However, in southern China, the highest $PM_{2.5}$ concentrations were at Wuhan, with levels of 80.5 and 68.8 μg m^{-3}, respectively; in contrast, in 2016 the highest level was 64.0 μg m^{-3} in Luzhou (Tang *et al.*, 2017). These results show that in 2014, 2015, and 2016, the average $PM_{2.5}$ concentration of these six cities in the north were 1.50, 1.54 and 1.54 times higher than those of south, respectively. It can also be seen that the lowest $PM_{2.5}$ concentrations in southern China were much lower than those in northern China.

As for the three-year average $PM_{2.5}$ concentration, the highest three-year concentration in northern China was found to be in Baoding, at 106 μg m^{-3}, with the lowest at 25.2 μg m^{-3}, in Lhasa. The highest three-year average $PM_{2.5}$

Table 1(C). Atmospheric $PM_{2.5}$ concentration in various cities (2014–2016) (Unit: μg m^{-3}).

Province	Month	Shandong		Shanxi	Ningxia	Gansu	Tibet
City		Jinan	Linyi	Taiyuan	Yinchuan	Lanzhou	Lhasa
2014	Jan.	138	172	85.5	64.2	80.4	24.3
	Feb.	115	98.5	104	66.6	65.7	23.4
	Mar.	88.1	111	61.0	47.1	51.5	21.3
	Apr.	90.6	89.8	62.5	42.8	53.7	29.2
	May.	66.2	85.3	51.1	28.3	51.6	25.2
	June	75.6	75.1	45.8	27.2	50.7	20.6
	July	71.0	53.9	49.0	35.4	44.4	13.7
	Aug.	72.4	55.7	42.2	38.5	45.3	15.3
	Sep.	80.1	46.5	47.1	34.8	45.1	15.7
	Oct.	80.6	75.9	95.0	46.3	50.2	23.4
	Nov.	100.8	120.4	91.3	70.4	67.2	32.4
	Dec.	84.3	130.3	84.8	59.0	81.7	38.0
Range		66.2–138	46.5–172	42.2–104	27.2–71.0	44.4–82.0	13.7–38.0
Mean		88.6	92.9	68.3	46.7	57.3	23.5
2015	Jan.	112.8	125	83.3	81.7	63.9	23.6
	Feb.	95.0	99.6	65.1	48.4	51.5	22.0
	Mar.	81.1	87.0	60.3	39.0	51.1	17.8
	Apr.	76.7	67.6	51.7	38.4	45.0	21.8
	May.	74.3	55.9	49.7	33.5	44.8	24.4
	June	73.3	57.5	48.2	35.2	39.5	19.4
	July	75.3	43.6	42.9	44.8	44.4	23.6
	Aug.	57.3	41.0	39.0	42.4	37.0	19.7
	Sep.	69.2	45.1	44.9	32.6	37.0	18.1
	Oct.	90.6	94.0	58.4	33.8	41.4	29.0
	Nov.	114	83.2	75.1	76.2	66.0	36.2
	Dec.	158	129	105	73.7	79.3	39.8
Range		69.2–158	41.0–129	39.0–105	32.6–81.7	37.0–79.3	18.1–40.0
Mean		89.8	77.4	60.3	48.3	50.1	24.6
2016	Jan.	126.1	131.5	63.5	73.3	65.3	28.1
	Feb.	81.2	87.2	39.3	70.2	64.0	20.6
	Mar.	80.3	83.3	64.4	64.1	48.3	24.7
	Apr.	82.1	79.7	47.2	46.0	37.4	32.1
	May.	57.8	53.5	48.9	48.7	46.5	25.7
	June	51.2	38.1	45.3	35.8	39.6	18.9
	July	61.0	31.4	47.3	30.5	32.5	16.1
	Aug.	41.9	24.6	36.5	28.7	31.5	19.4
	Sep.	60.0	48.4	48.5	30.9	42.1	16.7
	Oct.	57.9	43.0	69.8	51.6	46.4	22.7
	Nov.	79.0	72.7	134.4	74.0	93.9	47.6
	Dec.	125.8	110.7	150.2	103.3	97.3	55.9
Range		41.9–126.1	24.6–131.5	36.5–150.2	28.7–103.3	31.5–97.3	16.1–55.9
Mean		75.4	67.0	66.3	54.8	53.7	27.4

concentration in southern China was found to be in Wuhan, at concentration of 68.8 μg m^{-3}, which is approximately 65% lower than that of Baoding. It can be seen that the $PM_{2.5}$ concentrations in northern China are much higher than those in southern China. One of the reasons for this is that the northern part of China is colder, and thus coal is burned for heat for a longer period of time, with the lower temperature also preventing pollutant diffusion, thus making the $PM_{2.5}$ concentration higher for the whole year.

Comparing the annual average $PM_{2.5}$ concentrations in the 22 northern cities during 2014–2016, it can be found that the concentration was highest in 2014, followed by 2015, and lowest in 2016. As for any reductions, Shijiazhuang was the city with the highest decrease in the $PM_{2.5}$ concentration between 2014 and 2015, falling from 118 to 88.1 μg m^{-3}, falling at a rate of 25.3%. While the $PM_{2.5}$ concentration in Shijiazhuang increasing to 98.5 μg m^{-3} in 2016, at a rate of 11.8%. The annual average $PM_{2.5}$ concentrations of the 22 cities in this study in 2014, 2015 and 2016 were 71.5, 64.8, and 61.4 μg m^{-3}, with the relative standard deviations (RSD) being 33.0%, 32.3% and 29.9%, respectively. The results also showed that the annual average concentration of $PM_{2.5}$ in 2015 decreased by 7.0 μg m^{-3} (9.8%) compared to 2014, while for 2016, it decreased by

3.3 μg m^{-3} (5.1%) compared to 2015. This means during these three years the $PM_{2.5}$ levels in the atmosphere have fallen, although still high. The air quality has thus improved, but there is still much more work to do here.

With regard to 2015 and 2016, the $PM_{2.5}$ concentrations of most cities decreased in the latter year. For example, that in Baoding decreased from 106 to 92.1 μg m^{-3}, while in Zhengzhou it fell from 96.1 to 78.5 μg m^{-3}, in Jinan from 89.8 to 75.4 μg m^{-3}, in Beijing from 80.2t o 72.8 μg m^{-3}, in Linyi from 77.3to 67.0 μg m^{-3}, and in Nanyang from 73.6 to 60.9 μg m^{-3}. Moreover, some cities saw an increase from 2015 to 2016. For example, the level in Shijiazhuang increased from 88.1 to 98.5 μg m^{-3}, that in Taiyuan from 60.3 to 66.3 μg m^{-3}, in Yinchuan from 48.3 to 54.8 μg m^{-3}, in Lanzhou from 50.1 to 53.7 μg m^{-3}, in Lhasa from 24.6 to 27.4 μg m^{-3}, in Sinning from 47.8 to 48.7 μg m^{-3}, in Urumqi from 64.6 to 73.1 μg m^{-3} , in Weinan from 58.8 to 76.0 μg m^{-3}, and in Xian from 57.8 to 71.4 μg m^{-3}. It can thus be noted that the Beijing-Tianjin-Hebei region still had a serious air pollution problem in 2016, although the $PM_{2.5}$ level in 2016 was still lower than 2015 and 2014.

With regard to the four seasons, defined as spring (March, April, May), summer (June, July, August), fall (September, October, November) and winter (January, February and December), the $PM_{2.5}$ concentration was always highest in winter and in summer. For example, during 2016, the average $PM_{2.5}$ concentrations in Shijiazhuang were 65.6, 45.5, 123 and 160 μg m^{-3} in spring, summer, fall and winter, respectively. In Baoding, the average $PM_{2.5}$ concentrations were 70.2, 54.3, 103 and 141 μg m^{-3}, respectively. In Beijing, they were 71.7, 58.4, 79.6 and 81.4 μg m^{-3}, respectively; while in Xian they were 53.8, 39.5, 49.5 and 101 μg m^{-3}. These results may be because the temperatures were lower in these areas in winter and higher in summer, and the transport of atmospheric contaminants through the vertical current was hindered by the cold air during the former.

The monthly average $PM_{2.5}$ concentrations in 2014 and 2015 showed obvious increases in December. For example, the level in Harbin increased from 98.8 to 145 μg m^{-3}, that in Qiqihar from 46.2 to 79.5 μg m^{-3}, in Changchun from 79.4 to 104 μg m^{-3}, Dalian from 47.9 to 82.4 μg m^{-3}, Shenyang from 82.3 to 107 μg m^{-3}, Tianjin from 107 to 125 μg m^{-3}, Shijiazhuang from 116 to 163 μg m^{-3}, Baoding from 168 to 214 μg m^{-3}, Linyi from 84.3 to 158 μg m^{-3}, Yinchuan from 84.8 to 150 μg m^{-3}, Lanzhou from 59 to 73.7 μg m^{-3}, Tibet from 38 to 39.8 μg m^{-3}, Sinning from 69 to 76.2 μg m^{-3}, Urumqi from 140 to 147 μg m^{-3}, Zhengzhou from 94.9 to 142 μg m^{-3}, Nanyang from 91.8 to 103 μg m^{-3}, Xian from 66.6 to 111 μg m^{-3}, and Weinan from 54.3 to 144 μg m^{-3}. This was due to a lower average temperature in December 2015 compared than December 2014, which further reduced vertical atmospheric convection. These results were also found by Tang *et al.* (2017).

It should be noted that among the 22 cities in northern China examined in this study, most saw remarkable declines in their $PM_{2.5}$ concentrations in October 2015 compared with the same period in 2014. This may be due to the various memorial activities that took place with regard to the Anti-Japanese War and World War II, which took place from August 20 to September 3, 2015. During these activities, China's environmental protection administration took powerful action to stop the operation of factories and so improved the air quality of the Beijing-Tianjin-Hebei region, thus enabling to reach the best quality observed during the time examined in this work.

The R_M of Atmospheric $PM_{2.5}$

The World Health Organization (WHO) standard was chosen, when the R_M of different cities was examined, and this is 10 μg m^{-3}. The R_M value is the ratio of $PM_{2.5}$ concentration to limitation, which can intuitively suggest the air pollution conditions. Comparisons of the R_M in 22 cities for the years 2014, 2015 and 2016, are shown in Figs. 1(A)–1(C), respectively. These reveal that in both 2014 and 2015 the highest R_M values occurred in Baoding, at 12.0 and 10.6, respectively. However, for 2016, the level at Baoding decreased to 9.21, and Shijiazhuang rose to first place with a level of 9.85. Over the three years, Lhasa always had the lowest R_M values among the 22 cities, at 2.35, 2.46 and 2.74 in 2014, 2015 and 2016, respectively. It can be seen from the figures that the top six R_M among the 22 cities for each year ranged between 7.91 and 10.6, and averaged 8.97. This suggests that the average $PM_{2.5}$ concentration in this period was as high as 8.97 times the WHO air quality standard. Moreover, the six lowest R_M values in three years loaded were in the range between 2.52 and 5.25, and averaged 4.21. When comparing the figures seen in 2014–2016, the average R_M values were 7.15, 6.48 and 6.14 in 2014, 2015 and 2016, respectively. The fall in the R_M value during 2015 and 2016 suggests that the $PM_{2.5}$ concentrations in these cities in northern China slowly decreased, but were still high, and most cities would have to fall by at least 80% to meet the WHO standard, at which levels they are considered harmlessness to human health.

The R_M values for southern China for the years 2014, 2015 and 2016 were 5.2, 4.3 and 4.1, respectively. Compared with northern China, the average R_M values of southern China were much lower. However, the ambient air quality for the whole of China has a long way to go to meet the WHO standard, although it has slowly improved. Moreover, the air pollution situation is better in the south than in the north, and in 2016 than in 2015. However, in order to meet the WHO standard for $PM_{2.5}$, both the north and south need to make substantial changes.

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

The $PM_{2.5}/PM_{10}$ ratio reflects the proportion of PM in the ambient air, and thus the impact of the air on human health. Due to its small particle diameter, $PM_{2.5}$ has a long standing time and propagation distance, and thus more significant impacts on the environment and human health. During 2014, the six highest annual averages of the $PM_{2.5}/PM_{10}$ concentration ratios were 0.72 in Beijing, 0.64 in Tianjin, 0.61 in Harbin, 0.61 in Dalian, 0.60 in Zhengzhou and 0.58 in Shenyang; in 2015, the six highest annual averages of the $PM_{2.5}/PM_{10}$ concentration ratios were 0.81 in Beijing, 0.63 in Harbin, 0.62 in Shenyang, 0.60 in Changchun, 0.59 in Baoding and 0.58 in Tianjin; and in 2016, the six

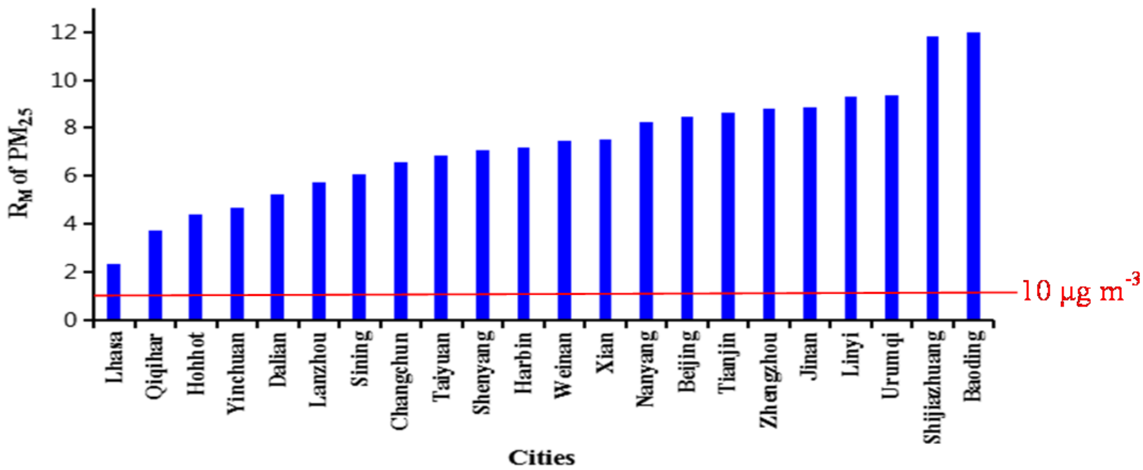

Fig. 1(A). The R_M of atmospheric $PM_{2.5}$ in various cities (2014).

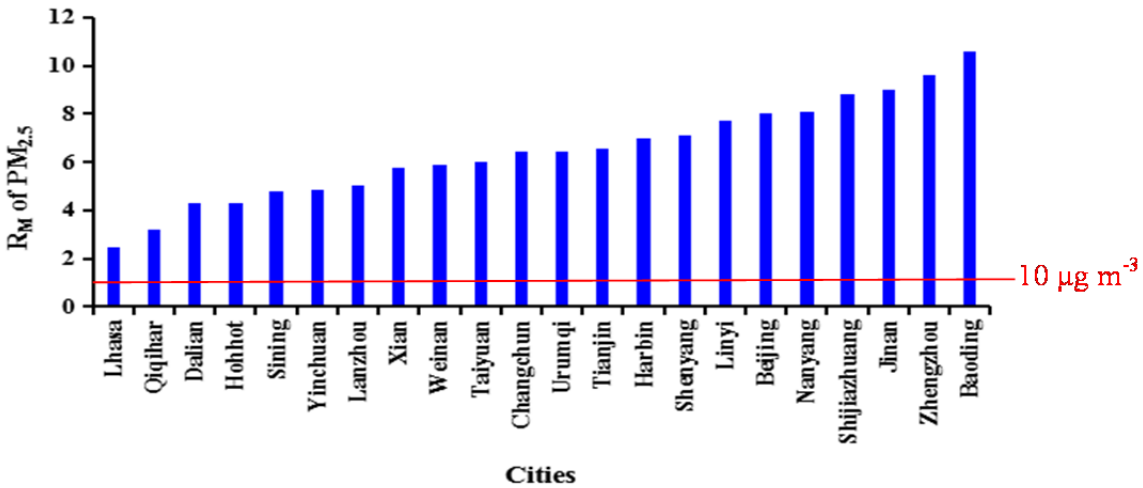

Fig. 1(B). The R_M of atmospheric $PM_{2.5}$ in various cities (2015).

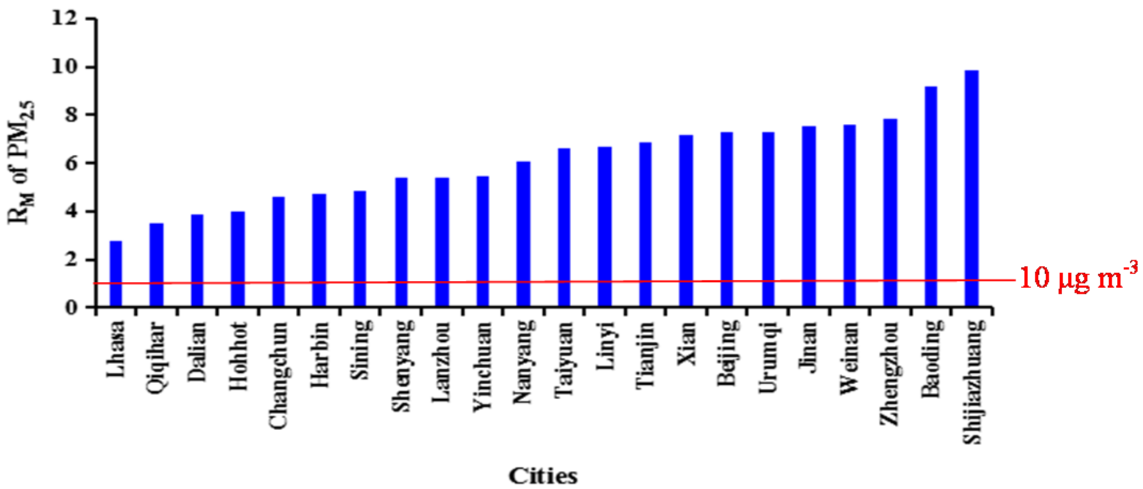

Fig. 1(C). The R_M of atmospheric $PM_{2.5}$ in various cities (2016).

highest annual averages of the $PM_{2.5}/PM_{10}$ concentration ratios were 0.76 in Beijing, 0.65 in Harbin, 0.64 in Tianjin, 0.62 in Baoding, 0.58 in Changchun and 0.58 in Dalian.

In 2014, the six lowest annual averages of the $PM_{2.5}/PM_{10}$ concentration ratios were 0.38 in Hohhot, 0.41 in Urumqi, 0.42 in Lhasa, 0.45 in Yinchuan, 0.49 in Xian and 0.50 in

Lanzhou; in 2015, the six lowest annual averages of the $PM_{2.5}/PM_{10}$ concentration ratios were 0.39 in Hohhot, 0.43 in Lhasa, 0.43 in Lanzhou, 0.43 in Yinchuan, 0.45 in Xian and 0.46 in Urumqi; and in 2016, these were 0.35 in Lhasa, 0.41 in Lanzhou, 0.43 in Hohhot, 0.44 in Sinning, 0.48 in Yinchuan and 0.50 in Xian. The three-year averages of the $PM_{2.5}/PM_{10}$concentration ratios in northern China were in the range of 0.40 and 0.76, and averaged 0.54.

It was found that, in general, a city with a high $PM_{2.5}$ concentration always had a high $PM_{2.5}/PM_{10}$ ratio, too. The top six cities in northern China with the highest three-year average $PM_{2.5}$ concentrations were Baoding, Shijiazhuang, Zhengzhou, Jinan, Beijing and Linyi, which an average $PM_{2.5}$ concentration of 89.7 µg m^{-3}, with an average $PM_{2.5}/PM_{10}$ ratio of 0.59. However, during 2014–2016, among the 22 cities examined in this work, the six with the lowest three-year average $PM_{2.5}$ concentrations were Lhasa, Qiqihar, Hohhot, Dalian, Yinchuan and Sinning City, which had an average $PM_{2.5}$ concentration of 42.1 µg m^{-3}, and with an average $PM_{2.5}/PM_{10}$ ratio of 0.48. A higher the $PM_{2.5}/PM_{10}$ ratio means the PM in the ambient air are more harmful to people.

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

In 2014, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in different cities ranged between 0.017 and 0.205 pg-WHO_{2005}-TEQ m^{-3}, and averaged 0.075 pg-WHO_{2005}-TEQ m^{-3} (Tables 2(A)–2(D)). The lowest annual average total-PCDD/Fs-WHO_{2005} -TEQ concentration was 0.033 pg-WHO_{2005}-TEQ m^{-3} in Lhasa; however, the highest was 0.121 pg-WHO_{2005}-TEQ m^{-3} in Baoding, which was 3.67 times higher than that in Lhasa.

In 2015, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in different cities ranged from 0.018 to 0.170 pg-WHO_{2005}-TEQ m^{-3}, and averaged 0.068 pg-WHO_{2005}-TEQ m^{-3} (Tables 2(A)–2(D)). The lowest annual average total-PCDD/Fs-WHO_{2005}-TEQ concentration was 0.033 pg-WHO_{2005}-TEQ m^{-3} in Lhasa; however, the highest was 0.100 pg-WHO_{2005} -TEQ m^{-3} in Baoding, which was 3.03 times higher than that in Lhasa.

In 2016, the total PCDD/Fs-WHO_{2005}-TEQ concentrations in different cities ranged between 0.019 and 0.261 pg-WHO_{2005}-TEQ m^{-3}, and averaged 0.066 pg-WHO_{2005}-TEQ m^{-3} (Tables 2(A)–2(D)). The lowest annual average total-PCDD/Fs-WHO_{2005}-TEQ concentration was 0.036 pg-WHO_{2005}-TEQ m^{-3} in Qiqihar. In contrast, the highest was 0.119 pg-WHO_{2005}-TEQ m^{-3} in Shijiazhuang, which was 3.31 times higher than that in Qiqihar.

As for the six cities with the lowest three-year average total-PCDD/Fs-WHO_{2005}-TEQ concentrations (LAC), the figures were 0.036, 0.037, 0.045, 0.055, 0.056 and 0.060 pg-WHO_{2005}-TEQ m^{-3} for Qiqihar, Lhasa, Dalian, Harbin, Changchun and Hohhot, respectively, and averaged 0.048 pg-WHO_{2005}-TEQ m^{-3}. However, the top six cities with the highest three-year average of total-PCDD/Fs-WHO_{2005}-TEQ concentrations (HAC) during 2014–2016 had figures of 0.107, 0.102, 0.095,0.092, 0.085 and 0.077 pg-WHO_{2005}-TEQ m^{-3} for Shijiazhuang, Baoding, Zhengzhou, Jinan, Linyi and Xian, respectively, and averaged 0.092 pg-WHO_{2005}-TEQ m^{-3}. It can thus be seen that the HAC was approximately 1.92 times higher than the LAC.

For comparison, the highest three-year average total-PCDD/Fs-WHO_{2005}-TEQ concentration in southern China occurred in Chengdu, which was 0.067 pg-WHO_{2005}-TEQ m^{-3}, but in the north the highest was for Shijiazhuang, with a level nearly 1.60 times that of Chengdu, at 0.107 pg-WHO_{2005}-TEQ m^{-3}. The lowest three-year average total-PCDD/Fs-WHO_{2005}-TEQ concentration in the south was 0.020 pg-WHO_{2005}-TEQ m^{-3} in Sanya, while in the north it was 1.8 times higher in Qiqihar, at 0.036 pg-WHO_{2005}-TEQ m^{-3}. By comparing the highest and lowest values, it can be seen that the total-PCDD/Fs-WHO_{2005}-TEQ concentrations in northern China were much higher than those in the south.

Tang *et al.* (2017) reported that one of the reasons why the southern cities with higher latitudes (like Wuhan and Hefei) have a relatively high total-PCDD/Fs-WHO_{2005}-TEQ concentrations compared to other cities in the south is because they are located downstream of the air pollution current flowing from the northern cities, and may be affected by the long-range transport of pollutants from these (Tang *et al.*, 2017). Wuhan and Hefei are in Hubei and Anhui provinces, respectively, and located along the edges of Henan and Shaanxi provinces. Moreover, the three-year average of total-PCDD/Fs-WHO_{2005}-TEQ concentration in Zhengzhou (in Henan province) was 0.095 pg-WHO_{2005}-TEQ m^{-3}, while in Xian (in Shaanxi province) it was 0.077 pg-WHO_{2005}-TEQ m^{-3}, both figures being at the higher end among the 22 northern cities. This proves that the air quality of cities at higher latitudes of southern China was affected by the northern cities, which had higher levels of atmospheric pollutants.

Both Baoding (0.121 pg-WHO_{2005}-TEQ m^{-3}) and Shijiazhuang (0.119 pg-WHO_{2005}-TEQ m^{-3}) had higher total PCDD/Fs-WHO_{2005}-TEQ concentrations than industrial areas (0.096 pg I-TEQ Nm^{-3}) in Taiwan (Wang *et al.*, 2010). This demonstrates that the total PCDD/Fs-WHO_{2005}-TEQ concentration is correlated with the strength of industrial activities in specific areas. It is thus obvious that the total-PCDD/F-WHO_{2005}-TEQ concentrations in the industrial areas were much higher than those in urban or rural areas.

As shown by Eduljee and Dyke (1996), industrial processes in the UK contributed 535–955 g total-PCDD/Fs I-TEQ yr^{-1}, which was approximately 90% of the total PCDD/Fs-I-TEQ emission inventory. The same study indicated that local industrial activity was always a key factor impacting the air quality level of the total-PCDD/Fs-I-TEQ concentration.

The highest monthly average total-PCDD/Fs-WHO_{2005}-TEQ concentrations occurred during January, 2014, at Shijiazhuang, and during January, 2015, at Baoding (averaging 0.205 and 0.170 pg-WHO_{2005}-TEQ m^{-3}, respectively), while for 2016, it occurred during October in Shijiazhuang, with the total-PCDD/Fs-WHO_{2005}-TEQ concentration of 0.261 pg-WHO_{2005}-TEQ m^{-3}. However, the lowest monthly average concentration for 2014 occurred in July, (0.017 pg WHO_{2005}-TEQ m^{-3}) in Lhasa, while in 2015 and 2016 it occurred during June in Qiqihar and during August at Dalian (0.018 and 0.019 pg WHO_{2005}-TEQ m^{-3}, respectively). With regard to the seasonal variation, during

Table 2(A). Total-PCDD/Fs-WHO_{2005}-TEQ concentration in various cities (2014–2016) (Unit: pg-WHO_{2005}-TEQ m^{-3}).

Province	Month	Heilongjiang		Jilin	Liaoning	
City		Harbin	Qiqihar	Changchun	Dalian	Shenyang
2014	Jan.	0.092	0.046	0.072	0.060	0.074
	Feb.	0.091	0.053	0.071	0.057	0.080
	Mar.	0.057	0.040	0.071	0.066	0.077
	Apr.	0.062	0.043	0.077	0.063	0.077
	May.	0.033	0.026	0.046	0.049	0.057
	June	0.032	0.024	0.043	0.035	0.042
	July	0.038	0.034	0.050	0.036	0.048
	Aug.	0.028	0.021	0.042	0.033	0.047
	Sep.	0.030	0.020	0.043	0.032	0.051
	Oct.	0.097	0.042	0.127	0.050	0.091
	Nov.	0.118	0.058	0.082	0.065	0.106
	Dec.	0.079	0.035	0.060	0.045	0.075
Range		0.028–0.118	0.020–0.058	0.042–0.127	0.032–0.066	0.042–0.106
Mean		0.063	0.037	0.065	0.049	0.069
2015	Jan.	0.087	0.040	0.079	0.046	0.087
	Feb.	0.087	0.042	0.067	0.061	0.081
	Mar.	0.062	0.041	0.062	0.062	0.075
	Apr.	0.061	0.044	0.067	0.052	0.078
	May.	0.038	0.021	0.061	0.042	0.055
	June	0.030	0.018	0.036	0.031	0.033
	July	0.027	0.022	0.038	0.026	0.034
	Aug.	0.021	0.025	0.027	0.032	0.030
	Sep.	0.030	0.029	0.041	0.030	0.044
	Oct.	0.051	0.040	0.069	0.042	0.064
	Nov.	0.108	0.065	0.091	0.074	0.107
	Dec.	0.105	0.054	0.073	0.063	0.081
Range		0.027–0.122	0.029–0.064	0.034–0.091	0.029–0.074	0.038–0.105
Mean		0.059	0.037	0.059	0.047	0.064
2016	Jan.	0.067	0.032	0.061	0.049	0.067
	Feb.	0.051	0.033	0.043	0.037	0.052
	Mar.	0.057	0.044	0.072	0.055	0.086
	Apr.	0.052	0.041	0.057	0.063	0.064
	May.	0.036	0.046	0.041	0.043	0.053
	June	0.021	0.021	0.028	0.025	0.034
	July	0.022	0.026	0.026	0.021	0.032
	Aug.	0.023	0.024	0.027	0.019	0.029
	Sep.	0.024	0.029	0.033	0.029	0.040
	Oct.	0.037	0.036	0.048	0.035	0.051
	Nov.	0.065	0.048	0.054	0.048	0.065
	Dec.	0.063	0.046	0.056	0.047	0.075
Range		0.021–0.067	0.021–0.048	0.027–0.072	0.019–0.049	0.029–0.075
Mean		0.043	0.036	0.045	0.039	0.054

2014–2016, the average total-PCDD/Fs-WHO_{2005}-TEQ concentrations for these 22 cities in northern China were 0.076, 0.044, 0.071 and 0.087 pg WHO_{2005}-TEQ m^{-3} in spring, summer, fall and winter, respectively. It was found that the total-PCDD/Fs-WHO_{2005}-TEQ concentrations in winter (averaged 0.087 pg WHO_{2005}-TEQ m^{-3}) were much higher than in summer (0.044 pg WHO_{2005}-TEQ m^{-3}). Comparing the monthly total-PCDD/Fs-WHO_{2005}-TEQ concentrations with the atmospheric $PM_{2.5}$ levels showed that both variables were closely related to each other. In general, the higher the level of $PM_{2.5}$, the higher the total-PCDD/Fs-WHO_{2005}-TEQ concentration. Therefore, a reduction in $PM_{2.5}$ emissions is of great importance for the control of total-PCDD/Fs-WHO_{2005}-TEQ level in the ambient air.

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

In 2014, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content in per gram $PM_{2.5}$ (abbreviated as PCDD/Fs-TEQ content) ranged between 1.00 and 0.444 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.672 ng-WHO_{2005}-TEQ g^{-1} (Tables 3(A)–3(B)). The six cities with the lowest $PM_{2.5}$ concentration were Hohhot, Lhasa, Yinchuan, Changchun, Sinning and Dalian, with levels of 1.00, 0.855, 0.762, 0.661, 0.688 and 0.570 ng-WHO_{2005}-TEQ g^{-1}, respectively, and an average of

Table 2(B). Total-PCDD/Fs-WHO_{2005}-TEQ concentration in various cities (2014–2016) (Unit: pg-WHO_{2005}-TEQ m^{-3}).

Province	Month	Beijing	Tianjin	Hebei		Inner Mongolia
City				Shijiazhuang	Baoding	Hohhot
2014	Jan.	0.069	0.101	0.205	0.180	0.069
	Feb.	0.087	0.077	0.164	0.153	0.055
	Mar.	0.082	0.103	0.152	0.150	0.083
	Apr.	0.086	0.091	0.123	0.120	0.084
	May.	0.070	0.079	0.111	0.114	0.079
	June	0.039	0.049	0.078	0.071	0.045
	July	0.052	0.059	0.078	0.074	0.052
	Aug.	0.046	0.052	0.064	0.068	0.046
	Sep.	0.054	0.053	0.069	0.086	0.055
	Oct.	0.090	0.089	0.144	0.157	0.087
	Nov.	0.078	0.095	0.117	0.149	0.074
	Dec.	0.054	0.087	0.114	0.128	0.073
Range		0.039–0.090	0.049–0.103	0.064–0.205	0.068–0.180	0.045–0.087
Mean		0.067	0.078	0.118	0.121	0.067
2015	Jan.	0.067	0.091	0.125	0.170	0.067
	Feb.	0.064	0.082	0.097	0.145	0.069
	Mar.	0.087	0.098	0.109	0.135	0.073
	Apr.	0.070	0.065	0.089	0.087	0.057
	May.	0.057	0.054	0.064	0.068	0.058
	June	0.037	0.048	0.059	0.058	0.038
	July	0.038	0.046	0.059	0.058	0.043
	Aug.	0.035	0.049	0.060	0.067	0.040
	Sep.	0.036	0.046	0.053	0.066	0.044
	Oct.	0.053	0.061	0.064	0.088	0.063
	Nov.	0.052	0.082	0.102	0.095	0.076
	Dec.	0.097	0.106	0.132	0.164	0.089
Range		0.035–0.097	0.046–0.106	0.053–0.132	0.058–0.170	0.038–0.089
Mean		0.058	0.069	0.084	0.100	0.060
2016	Jan.	0.049	0.070	0.109	0.116	0.058
	Feb.	0.034	0.048	0.063	0.066	0.049
	Mar.	0.081	0.088	0.103	0.094	0.079
	Apr.	0.070	0.076	0.079	0.095	0.063
	May.	0.054	0.055	0.063	0.077	0.071
	June	0.036	0.040	0.044	0.051	0.031
	July	0.039	0.037	0.201	0.055	0.031
	Aug.	0.031	0.034	0.030	0.030	0.026
	Sep.	0.045	0.051	0.089	0.072	0.037
	Oct.	0.062	0.055	0.261	0.091	0.055
	Nov.	0.082	0.091	0.170	0.132	0.093
	Dec.	0.089	0.099	0.213	0.136	0.060
Range		0.031–0.089	0.034–0.099	0.030–0.261	0.030–0.136	0.026–0.093
Mean		0.056	0.062	0.119	0.085	0.054

0.762 ng-WHO_{2005}-TEQ g^{-1}. However, the six cities with the highest $PM_{2.5}$ concentrations in 2014 were Shijiazhuang, Urumqi, Zhengzhou, Tianjin, Beijing, and Harbin, at 0.587, 0.859, 0.444, 0.501, 0.463 and 0.568 ng-WHO_{2005}-TEQ g^{-1}, respectively, and an average of 0.582 ng-WHO_{2005}-TEQ g^{-1}. It was found that the LAC (average PCDD/Fs-TEQ content of the six cities with the lowest average $PM_{2.5}$ concentrations) was approximately 1.31 times higher than that of the HAC (average PCDD/Fs-TEQ content of the six cities with the highest average $PM_{2.5}$ concentrations). It is interesting that in these results the $PM_{2.5}$ level in the ambient air is not correlated with the PCDD/Fs-TEQ content. For example, Shijiazhuang City has the highest $PM_{2.5}$ concentration (118 µg m^{-3}) of 12 selected cities, but the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content (0.587 ng-WHO_{2005}-TEQ g^{-1}) was only ranked seventh, lower than the levels seen in of Hohhot, Urumqi, Lhasa, Sinning, Yinchuan and Changchun, which had $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents of 1.002, 0.927, 0.855, 0.766, 0.762 and 0.858 ng-WHO_{2005}-TEQ g^{-1}, respectively.

As for the seasonal variation, for the six cities with the highest $PM_{2.5}$ concentrations, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents for Shijiazhuang in spring, summer, fall and winter were 0.717, 0.292, 0.605 and

Table 2(B). Total-PCDD/Fs-WHO_{2005}-TEQ concentration in various cities (2014–2016) (Unit: pg-WHO_{2005}-TEQ m^{-3}).

Province	Month	Beijing	Tianjin	Hebei		Inner Mongolia
City				Shijiazhuang	Baoding	Hohhot
2014	Jan.	0.069	0.101	0.205	0.180	0.069
	Feb.	0.087	0.077	0.164	0.153	0.055
	Mar.	0.082	0.103	0.152	0.150	0.083
	Apr.	0.086	0.091	0.123	0.120	0.084
	May.	0.070	0.079	0.111	0.114	0.079
	June	0.039	0.049	0.078	0.071	0.045
	July	0.052	0.059	0.078	0.074	0.052
	Aug.	0.046	0.052	0.064	0.068	0.046
	Sep.	0.054	0.053	0.069	0.086	0.055
	Oct.	0.090	0.089	0.144	0.157	0.087
	Nov.	0.078	0.095	0.117	0.149	0.074
	Dec.	0.054	0.087	0.114	0.128	0.073
Range		0.039–0.090	0.049–0.103	0.064–0.205	0.068–0.180	0.045–0.087
Mean		0.067	0.078	0.118	0.121	0.067
2015	Jan.	0.067	0.091	0.125	0.170	0.067
	Feb.	0.064	0.082	0.097	0.145	0.069
	Mar.	0.087	0.098	0.109	0.135	0.073
	Apr.	0.070	0.065	0.089	0.087	0.057
	May.	0.057	0.054	0.064	0.068	0.058
	June	0.037	0.048	0.059	0.058	0.038
	July	0.038	0.046	0.059	0.058	0.043
	Aug.	0.035	0.049	0.060	0.067	0.040
	Sep.	0.036	0.046	0.053	0.066	0.044
	Oct.	0.053	0.061	0.064	0.088	0.063
	Nov.	0.052	0.082	0.102	0.095	0.076
	Dec.	0.097	0.106	0.132	0.164	0.089
Range		0.035–0.097	0.046–0.106	0.053–0.132	0.058–0.170	0.038–0.089
Mean		0.058	0.069	0.084	0.100	0.060
2016	Jan.	0.049	0.070	0.109	0.116	0.058
	Feb.	0.034	0.048	0.063	0.066	0.049
	Mar.	0.081	0.088	0.103	0.094	0.079
	Apr.	0.070	0.076	0.079	0.095	0.063
	May.	0.054	0.055	0.063	0.077	0.071
	June	0.036	0.040	0.044	0.051	0.031
	July	0.039	0.037	0.201	0.055	0.031
	Aug.	0.031	0.034	0.030	0.030	0.026
	Sep.	0.045	0.051	0.089	0.072	0.037
	Oct.	0.062	0.055	0.261	0.091	0.055
	Nov.	0.082	0.091	0.170	0.132	0.093
	Dec.	0.089	0.099	0.213	0.136	0.060
Range		0.031–0.089	0.034–0.099	0.030–0.261	0.030–0.136	0.026–0.093
Mean		0.056	0.062	0.119	0.085	0.054

0.736 ng-WHO_{2005}-TEQ g^{-1}, respectively; for Urumqi they were 1.280, 0.663, 1.065 and 0.700 ng-WHO_{2005}-TEQ g^{-1}, respectively; for Zhengzhou they were 0.593, 0235, 0.511 and 0.435 ng-WHO_{2005}-TEQ g^{-1}, respectively; for Tianjin they were 0.587, 0.250, 0.519 and 0.647 ng-WHO_{2005}-TEQ g^{-1}, respectively; for Beijing they were 0.602, 0.185, 0.486 and 0.580 ng-WHO_{2005}-TEQ g^{-1}, respectively; for Harbin they were 0.739, 0.294, 0.614 and 0.624 ng-WHO_{2005}-TEQ g^{-1}, respectively; with averages of 0.753, 0.320, 0.633 and 0.620 ng-WHO_{2005}-TEQ g^{-1}, respectively. Because the average temperature in summer (25.5°C) was much higher than those in spring (14.2°C), fall (12.4°C) or winter (–4.2°C), greater amounts of PCDD/Fs were evaporated from the particles to the gaseous phase, resulting in lower $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ contents.

A city in the north, Urumqi, shown one interesting phenomenon with regard to its $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content. In 2014, the two highes monthly PCDD/Fs-TEQ contents occurred in April (1.56 ng-WHO_{2005}-TEQ g^{-1}) and May (1.47 ng-WHO_{2005}-TEQ g^{-1}) in spring, while the third and the fourrth highest were in October (1.20 ng-WHO_{2005}-TEQ g^{-1}) and November (1.02 ng-WHO_{2005}-TEQ g^{-1}), in fall. The lowest PCDD/Fs-TEQ contents were in February (0.684 ng-WHO_{2005}-TEQ g^{-1}),

Table 2(C). Total-PCDD/Fs-WHO_{2005}-TEQ concentration in various cities (2014–2016) (Unit: pg-WHO_{2005}-TEQ m^{-3}).

Province	Month	Shandong		Shanxi	Ningxia	Gansu	Tibet
City		Jinan	Linyi	Taiyuan	Yinchuan	Lanzhou	Lhasa
2014	Jan.	0.138	0.141	0.077	0.066	0.077	0.030
	Feb.	0.107	0.077	0.085	0.065	0.061	0.043
	Mar.	0.116	0.124	0.084	0.079	0.088	0.031
	Apr.	0.113	0.107	0.086	0.073	0.087	0.046
	May.	0.101	0.110	0.079	0.057	0.082	0.037
	June	0.071	0.077	0.049	0.033	0.051	0.026
	July	0.065	0.065	0.054	0.045	0.046	0.017
	Aug.	0.070	0.073	0.043	0.040	0.048	0.019
	Sep.	0.088	0.073	0.056	0.048	0.055	0.023
	Oct.	0.102	0.090	0.108	0.062	0.057	0.034
	Nov.	0.115	0.119	0.091	0.075	0.074	0.046
	Dec.	0.101	0.125	0.084	0.068	0.083	0.043
Range		0.065–0.138	0.065–0.141	0.043–0.108	0.033–0.079	0.046–0.088	0.017–0.046
Mean		0.099	0.098	0.075	0.059	0.067	0.033
2015	Jan.	0.118	0.107	0.072	0.087	0.066	0.030
	Feb.	0.097	0.095	0.058	0.066	0.061	0.024
	Mar.	0.110	0.101	0.068	0.075	0.091	0.030
	Apr.	0.094	0.083	0.069	0.069	0.079	0.030
	May.	0.092	0.078	0.066	0.066	0.074	0.036
	June	0.072	0.062	0.052	0.054	0.058	0.024
	July	0.069	0.052	0.044	0.058	0.049	0.029
	Aug.	0.051	0.050	0.045	0.049	0.050	0.024
	Sep.	0.078	0.061	0.053	0.043	0.051	0.029
	Oct.	0.106	0.107	0.075	0.047	0.064	0.044
	Nov.	0.097	0.082	0.075	0.078	0.081	0.051
	Dec.	0.130	0.121	0.094	0.075	0.080	0.051
Range		0.051–0.130	0.050–0.121	0.044–0.094	0.043–0.087	0.049–0.091	0.024–0.051
Mean		0.093	0.083	0.064	0.064	0.067	0.033
2016	Jan.	0.108	0.121	0.060	0.065	0.068	0.035
	Feb.	0.079	0.083	0.040	0.070	0.091	0.037
	Mar.	0.109	0.099	0.080	0.088	0.085	0.044
	Apr.	0.120	0.105	0.075	0.063	0.067	0.057
	May.	0.077	0.072	0.071	0.083	0.069	0.040
	June	0.054	0.049	0.052	0.042	0.050	0.024
	July	0.057	0.040	0.044	0.038	0.044	0.023
	Aug.	0.046	0.032	0.038	0.034	0.041	0.033
	Sep.	0.080	0.059	0.065	0.043	0.061	0.030
	Oct.	0.072	0.051	0.082	0.064	0.070	0.049
	Nov.	0.092	0.080	0.145	0.097	0.161	0.088
	Dec.	0.114	0.098	0.116	0.095	0.112	0.088
Range		0.046–0.120	0.032–0.121	0.038–0.145	0.034–0.097	0.041–0.161	0.023–0.088
Mean		0.084	0.074	0.072	0.065	0.076	0.046

December (0.689 ng-WHO_{2005}-TEQ g^{-1}) and January (0.728 ng-WHO_{2005}-TEQ g^{-1}), all in winter. This means that the highest PCDD/Fs-TEQ content in Urumqi did not occur in winter, but in spring and fall. These results may be because of the local atmospheric and geographic conditions in Urumqi, which has mountains surrounding it in three directions, and thus the atmospheric dispersion is always hindered. In addition, Urumqi uses coal-fired heating in winter, spring and fall. As such, the high level of fossil fuel combustion in Urumqi caused not only high levels of atmospheric $PM_{2.5}$, but also of total PCDD/Fs-WHO_{2005}-TEQ. This was also seen in other, similar cities in northern China.

CONCLUSIONS

The results of this study can be summarized, as follows:

1. Among the 22 cities in northern China examined in this work, those that are more highly industrialized had higher atmospheric $PM_{2.5}$ concentrations. During 2014–2016, the lowest annual average concentrations of $PM_{2.5}$ were in Lhasa and Qiqihar, and were 23.5 and 37.5 µg m^{-3} in 2014, 24.6 and 37.2 µg m^{-3} in 2015 and 27.4 and 35.3 µg m^{-3} in 2016, respectively. In contrast, the highest concentrations of $PM_{2.5}$ occurred in Baoding and Shijiazhuang, and were 120 and 118 µg m^{-3} in 2014, 106

Table 2(D). Total-PCDD/Fs-WHO$_{2005}$-TEQ concentration in various cities (2014–2016) (Unit: pg-WHO$_{2005}$-TEQ m^{-3}).

Province	Month	Qinghai	Sinkiang	Henan		Shaanxi	
City		Sinning	Urumqi	Zhengzhou	Nanyang	Xian	Weinan
2014	Jan.	0.087	0.104	0.107	0.172	0.128	0.128
	Feb.	0.083	0.069	0.079	0.080	0.135	0.125
	Mar.	0.094	0.094	0.094	0.090	0.111	0.100
	Apr.	0.092	0.106	0.083	0.061	0.088	0.075
	May.	0.077	0.096	0.082	0.096	0.088	0.086
	June	0.043	0.058	0.058	0.059	0.051	0.052
	July	0.040	0.064	0.055	0.034	0.045	0.038
	Aug.	0.044	0.073	0.052	0.042	0.049	0.041
	Sep.	0.052	0.097	0.060	0.045	0.047	0.044
	Oct.	0.056	0.088	0.104	0.087	0.093	0.088
	Nov.	0.066	0.068	0.117	0.092	0.095	0.079
	Dec.	0.067	0.121	0.113	0.095	0.085	0.051
Range		0.040–0.094	0.058–0.121	0.052–0.117	0.034–0.172	0.045–0.135	0.038–0.128
Mean		0.067	0.086	0.084	0.080	0.085	0.075
2015	Jan.	0.063	0.094	0.132	0.105	0.105	0.079
	Feb.	0.065	0.116	0.110	0.104	0.083	0.072
	Mar.	0.089	0.069	0.116	0.088	0.078	0.072
	Apr.	0.067	0.075	0.104	0.086	0.063	0.062
	May.	0.061	0.075	0.091	0.085	0.060	0.061
	June	0.039	0.049	0.076	0.061	0.043	0.039
	July	0.038	0.061	0.065	0.056	0.051	0.042
	Aug.	0.043	0.050	0.058	0.053	0.053	0.039
	Sep.	0.040	0.047	0.081	0.062	0.052	0.055
	Oct.	0.055	0.068	0.091	0.087	0.094	0.055
	Nov.	0.075	0.068	0.090	0.054	0.077	0.062
	Dec.	0.076	0.123	0.133	0.096	0.118	0.129
Range		0.038–0.089	0.047–0.123	0.058–0.133	0.053–0.105	0.043–0.118	0.039–0.129
Mean		0.059	0.075	0.096	0.078	0.073	0.064
2016	Jan.	0.063	0.143	0.132	0.109	0.116	0.113
	Feb.	0.075	0.160	0.084	0.085	0.082	0.071
	Mar.	0.092	0.077	0.120	0.097	0.111	0.116
	Apr.	0.059	0.049	0.110	0.079	0.082	0.086
	May.	0.061	0.040	0.074	0.055	0.062	0.061
	June	0.044	0.035	0.055	0.038	0.040	0.047
	July	0.041	0.034	0.039	0.030	0.031	0.038
	Aug.	0.040	0.040	0.035	0.034	0.037	0.040
	Sep.	0.047	0.047	0.070	0.055	0.064	0.070
	Oct.	0.058	0.042	0.054	0.037	0.060	0.065
	Nov.	0.106	0.067	0.107	0.081	0.135	0.136
	Dec.	0.085	0.103	0.119	0.106	0.131	0.125
Range		0.040–0.106	0.034–0.160	0.035–0.132	0.030–0.109	0.031–0.135	0.038–0.136
Mean		0.064	0.070	0.083	0.067	0.079	0.081

and 88.1 μg m^{-3} in 2015 and 92.1 and 98.5 μg m^{-3} in 2016, respectively.

2. With regard to the annual average concentrations of $PM_{2.5}$ in the 22 cities, those in 2016 were the lowest (ranging between 27.4 and 98.5 μg m^{-3}, with an average of 61.4 μg m^{-3}), followed by 2015 (ranging between 24.6 and 106 μg m^{-3}, with an average of 64.8 μg m^{-3}), and the highest in 2014 (ranging between 23.5 and 120 μg m^{-3}, with an average of 71.5 μg m^{-3}). Overall, from 2014 to 2016, the atmospheric $PM_{2.5}$ decreased in northern China.
3. When comparing the $PM_{2.5}$ concentration in 2015 with those in 2016, it was found that the levels in most of the cities decreased, but there were still some cities in which they increased. For example, the values in Shijiazhuang rose from 88.1 to 98.5 μg m^{-3}, in Taiyuan from 60.3 to 66.3 μg m^{-3}, in Yinchuan from 48.3 to 54.8 μg m^{-3}, in Lanzhou from 50.1 to 53.7 μg m^{-3}, in Lhasa from 24.6 to 27.4 μg m^{-3}, in Sinning from 47.8 to 48.7 μg m^{-3}, in Urumqi from 64.6 to 73.1 μg m^{-3} , in Weinan from 58.8 to 76 μg m^{-3}, and in Xian from 57.8 to 71.4 μg m^{-3}. This means that in 2015 and 2016 these cities all still had poor air pollution control.
4. With regard to monthly averages, during December the

Table 3(A). $PM_{2.5}$-bound total PCDD/Fs-WHO$_{2005}$-TEQ content of the six cities with the highest $PM_{2.5}$ concentration (2014) (Unit: ng-WHO$_{2005}$-TEQ g^{-1}).

Province	Month	Hebei	Xinjiang	Henan	Tianjin	Beijing	Harbin
City		Shijiazhuang	Urumqi	Zhengzhou			
2014	Jan.	0.762	0.728	0.417	0.696	0.570	0.596
	Feb.	0.681	0.684	0.339	0.614	0.457	0.640
	Mar.	0.790	0.809	0.719	0.650	0.601	0.782
	Apr.	0.713	1.56	0.599	0.625	0.552	0.842
	May.	0.648	1.47	0.462	0.487	0.654	0.594
	June	0.330	0.699	0.208	0.262	0.200	0.328
	July	0.263	0.601	0.231	0.216	0.149	0.249
	Aug.	0.284	0.688	0.265	0.272	0.206	0.306
	Sep.	0.442	0.975	0.389	0.376	0.327	0.602
	Oct.	0.648	1.20	0.526	0.554	0.478	0.599
	Nov.	0.724	1.02	0.618	0.627	0.652	0.642
	Dec.	0.764	0.689	0.550	0.631	0.713	0.636
Range		0.263–0.764	0.601–1.56	0.208–0.719	0.216–0.696	0.149–0.713	0.249–0.782
Mean		0.587	0.927	0.444	0.501	0.463	0.568

Table 3(B). PCDD/Fs-WHO$_{2005}$-TEQ content of the six cities with the lowest $PM_{2.5}$ concentrations (2014) (Unit: ng-WHO$_{2005}$-TEQ g^{-1}).

Province	Month	Inner Mongolia	Tibet	Ningxia	Jilin	Qinghai	Liaoning
City		Hohhot	Lhasa	Yinchuan	Changchun	Sining	Dalian
2014	Jan.	0.893	0.968	0.801	0.605	0.834	0.651
	Feb.	0.72	1.33	0.763	0.667	0.817	0.616
	Mar.	1.39	0.989	1.20	0.794	1.18	0.694
	Apr.	1.37	1.02	1.04	0.931	0.969	0.689
	May.	1.26	0.752	0.970	0.715	0.955	0.625
	June	0.729	0.800	0.386	0.412	0.513	0.292
	July	0.659	0.459	0.400	0.336	0.472	0.224
	Aug.	0.816	0.530	0.396	0.410	0.509	0.209
	Sep.	1.05	0.664	0.629	0.800	0.660	0.357
	Oct.	1.19	0.906	0.838	0.658	0.801	0.575
	Nov.	1.12	0.998	0.808	0.779	0.705	0.653
	Dec.	0.826	0.847	0.911	0.790	0.776	0.722
Range		0.659–1.39	0.459–1.33	0.386–1.20	0.336–0.931	0.472–1.18	0.209–0.722
Mean		1.00	0.855	0.762	0.658	0.766	0.526

$PM_{2.5}$ concentrations in most cities saw obvious increases: the concentrations of $PM_{2.5}$ in Harbin increased from 98.8 to 145 µg m^{-3}, in Qiqihar from 46.2 to 79.5 µg m^{-3}, in Changchun from 79.4 to 104 µg m^{-3}, in Dalian from 47.9 to 82.4 µg m^{-3}, Shenyang from 82.3 to 107 µg m^{-3}, Tianjin from 107 to 125 µg m^{-3}, Shijiazhuang from 116 to 163 µg m^{-3}, Baoding from 168 to 214 µg m^{-3}, Linyi from 84.3 to 158 µg m^{-3}, Yinchuan from 84.8 to 150 µg m^{-3}, Lanzhou from 59 to 73.7 µg m^{-3}, Tibet from 38 to 39.8 µg m^{-3}, Sinning from 69 to 76.2 µg m^{-3}, Urumqi from 140 to 147 µg m^{-3}, Zhengzhou from 94.9 to 142 µg m^{-3}, Nanyang from 91.8 to 103 µg m^{-3}, Xian from 66.6 to 111 µg m^{-3}, and Weinan from 54.3 to 144 µg m^{-3}. However, in the other months these cities concentrations fell.

5. The six R_M for each year were between 7.91 to 10.6, and averaged 8.97; this means that the average $PM_{2.5}$ concentrations were up to 8.97 times higher than the WHO air quality standard. However, during 2014–2016, the six lowest R_M were in the range between 2.52 to 5.25, and averaged 4.21. In addition, when we compared 2014–2016, the average R_M values were 7.2, 6.5 and 6.1 in 2014, 2015 and 2016, respectively.
6. In general, a city with a higher $PM_{2.5}$ concentration always had a higher $PM_{2.5}/PM_{10}$ ratio. During 2014–2016, the six cities with the highest $PM_{2.5}$ concentrations were Baoding, Shijiazhuang, Zhengzhou, Jinan, Beijing and Linyi, were the $PM_{2.5}$ concentration averaged 89.7 µg m^{-3} and the $PM_{2.5}/PM_{10}$ ratio averaged 0.59. However, the six cities with the lowest $PM_{2.5}$ concentrations were Lhasa, Qiqihar, Hohhot, Dalian, Yinchuan and Sinning, were the $PM_{2.5}$ concentration averaged 42.1 µg m^{-3} and the $PM_{2.5}/PM_{10}$ ratio averaged 0.48.
7. During 2014–2016, the results showed that the six highest three-year average total-PCDD/Fs-WHO$_{2005}$-TEQ concentrations were 0.107, 0.102, 0.095, 0.092, 0.085 and 0.077 pg-WHO$_{2005}$-TEQ m^{-3} in Shijiazhuang, Baoding, Zhengzhou, Jinan, Linyi and Xian, respectively. However, the six lowest three-year averages of total-PCDD/Fs-

WHO_{2005}-TEQ concentrations were 0.036, 0.037, 0.045, 0.055, 0.056 and 0.060 pg-WHO_{2005}-TEQ m^{-3} in Qiqihar, Lhasa, Dalian, Harbin, Changchun and Hohhot, respectively.

8. As for the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content, during 2014, the $PM_{2.5}$-bound total PCDD/Fs-WHO_{2005}-TEQ content of 12 cities (the six with the highest $PM_{2.5}$ concentrations, and six with the lowest $PM_{2.5}$ concentrations) ranged between 1.00 and 0.444 ng-WHO_{2005}-TEQ g^{-1} and averaged 0.672 ng-WHO_{2005}-TEQ g^{-1}. The six cities with the lowest $PM_{2.5}$ concentrations were Hohhot, Lhasa, Yinchuan, Changchun, Sinning and Dalian, at 1.00, 0.855, 0.762, 0.661, 0.688 and 0.570 ng-WHO_{2005}-TEQ g^{-1}, respectively. The six cities with the highest $PM_{2.5}$ concentrations in 2014 were Shijiazhuang, Urumqi, Zhengzhou, Tianjin, Beijing, and Harbin, at 0.587, 0.859, 0.444, 0.501, 0.463 and 0.568 ng-WHO_{2005}-TEQ g^{-1}, respectively.
9. This study presents the results of a systematic analysis of $PM_{2.5}$ and PCDD/Fs levels in northern China, and thus provides a theoretical basis for proposing better air pollution control strategies and improving the atmospheric environment in China.

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Received for review, June 22, 2017
Revised, July 17, 2017
Accepted, July 18, 2017