Air Qual Atmos Health (2017) 10:359–370
DOI 10.1007/s11869-016-0432-1

Spatial and seasonal variations of gaseous and particulate matter pollutants in 31 provincial capital cities, China

Daiying Yin[1] · **Suping Zhao**[2,3] · **Jianjun Qu**[1]

Received: 8 May 2016 / Accepted: 29 August 2016 / Published online: 6 September 2016

Abstract In order to know air pollution situation and their health, environmental, and climate effects, the air quality data with high temporal and spatial resolutions are essential. The spatial and seasonal variations of six criteria pollutants were investigated in 31 provincial capital cities between April 2014 and March 2015 using hourly mean air quality monitoring data, and the cities were classified by cluster analysis based on annual variations of air pollutants. The annual mean concentrations of $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm) and PM_{10} (particulate matter with aerodynamic diameter less than 10 μm) were high for all cities, which exceeded Chinese Ambient Air Quality Standards (CAAQS) Grade I standards. Only Fuzhou, Haikou, Kunming, and Lasa met Grade II standards for $PM_{2.5}$ and PM_{10}. Additionally, elevated SO_2 concentration was observed in northern cities, especially in winter. However, the seasonal variation of O_3 was opposite to other pollutants with the lowest concentrations in the winter and the highest in the summer. Winter domestic heating has significant impact on urban air quality, especially SO_2 and PM_{10}.

Keywords Spatial variation · Gaseous · Particulate pollutants · China · Cluster analysis

Introduction

Urban air pollution receives increasing attention due to its remarkable environmental, climate, and health effects (Du and Li 2016). As the largest developing country, economic growth in China over the past decades has been one of the strongest in the world history (Kan et al. 2012). However, such a rapid economic expansion is greatly driven by the use of fossil fuels such as coal, petroleum, and natural gases, which results in considerable increases of both gaseous and particulate matter pollutants in ambient air. Seventy percent of the most polluted cities in the world are located in China, and air quality of only 5 cities met World Health Organization Air Quality Guidelines in China (Bapna 2012). Many laws and regulations have been formulated by Chinese central and local governments in recent years to reduce primary emissions and improve urban air quality (GB3095-1996, GB13223-2003). Additionally, the new air quality standards were released in late February 2012 (GB3095-2012), and O_3, $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm), and CO were included in the newly revised standards. However, some air quality degradation events have been extensively reported in many Chinese cities (Chan and Yao 2008; Cheng et al. 2013; Qu et al. 2010; Chai et al. 2014; Li et al. 2014; Wang et al. 2008a, 2008b, 2014a, 2014b, 2014c, 2014d; Chen and Xie 2014), especially extreme haze-fog episodes in North China Plain (Che et al. 2009; Zhao et al. 2013; Guo et al. 2014; Zhang et al. 2014; Wang et al. 2014a, 2014b, 2014c, 2014d; Han et al. 2014; Huang et al. 2014). The reasons that caused the phenomenon were investigated using different methods such as satellite data and air quality model. Some

---

✉ Suping Zhao
zhaosp@lzb.ac.cn

[1] Key Laboratory of Desert and Desertification, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou 730000, China

[2] Key Laboratory of Land Surface Process & Climate Change in Cold & Arid Regions, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou 730000, China

[3] Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP3), Shanghai 200433, China

researchers indicated that the frequent extreme haze-fog episodes in China were caused by a large amount of emissions of trace gaseous pollutants, aerosols, and ozone precursors due to frequent human activities (Tan et al. 2016; Wang et al. 2013), while other studies showed that weather conditions such as large-scale circulation and local meteorological conditions (wind speed and direction, temperature, and relative humidity) become increasingly worse for diffusion of air pollutants due to barrier effect of Qinghai-Tibet Plateau to central and eastern China (Cao et al. 2015; Jia et al. 2015; Xu et al. 2016; Zheng et al. 2015). To understand current fine particulate matter pollution situations and its temporal and spatial variations in China, the central government decided to monitor $PM_{2.5}$ in the most polluted provincial capital cities including three key regions, i.e., the Yangtze River Delta, the Pearl River Delta, and the North China Plain. Since January 2013, the hourly real-time concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO, and O_3 for 74 major cities have been released online (http://113.108.142.147:20035/emcpublish/). These data with high resolutions can be used to study air pollution levels in some major Chinese cities and investigate formation mechanism of extreme haze events in central or eastern China using some statistical methods or numerical models (Hu et al. 2014).

The spatial–temporal distributions of air pollutants concentrations in China have been reported by some researchers based on the limited observation data (Wang et al. 2008a; Wang et al. 2008b; Han et al. 2009; Meng et al. 2009; Sun et al. 2013; Zheng et al. 2013; Chai et al. 2014; Chen and Xie 2014; Li et al. 2014; Wang et al. 2014a, 2014b; Wang et al. 2014c, 2014d; Zhang et al. 2014; Martini et al. 2015). Most of the studies are limited in one single megacity such as urban areas of Guangzhou (Li et al. 2014), Beijing (Wang et al. 2008a, 2008b) and Tianjin (Han et al. 2009), or one single air pollutant such as PM_{10} (Johansson et al. 2007; Karar et al. 2006; Qu et al. 2010), $PM_{2.5}$ (Martini et al. 2015) and NO_2 (Lewne et al. 2004). $PM_{2.5}$ mass concentration levels were assessed by Martini et al. (2015) using some statistical methods based on the air quality data from US Consulates in Beijing, Chengdu, Guangzhou, Shanghai, and Shenyang. It was found that the diurnal variations of $PM_{2.5}$ concentrations were significant across seasons and between cities. The previous studies pay much less attention to larger spatial coverage of multiple air pollutants due to scarce air quality monitoring stations. Recently, some studies were conducted on spatial and temporal variations of nationwide multiple pollutants including $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO, and O_3 based on hourly mean data published by the Ministry of Environmental Protection (MEP) of China (Chai et al. 2014; Wang et al. 2014a, 2014b, 2014c, 2014d). Their studies found that concentrations of SO_2, CO, $PM_{2.5}$, and PM_{10} were much higher in northern cities than those in southern cities while NO_2 and O_3 did not show significant differences between northern and southern cities. The number of days exceeded Chinese Ambient Air Quality Standards (CAAQS) was the highest in the winter, but highly polluted days also frequently occurred in the southeastern China during autumn due to biomass burning and in the west region during spring due to dust events. The previous studies gave a basic understanding of current air pollution situations in China, but few of them cover long-term fine particle mass concentrations data ($PM_{2.5}$, particulate matter with aerodynamic diameter less than 2.5 μm) and most of them divide pollution regions only based on geographical location of the cities.

The objective of this study is to investigate nationwide spatial and seasonal variations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO, and O_3 using the air quality data published by MEP for 31 provincial capital cities from April 2014 to March 2015. This study provides useful information regarding the status of nationwide air pollution. The paper was organized as follows. Data and methods used in this study such as monitoring systems and locations were introduced in detail in "Data and methods." Results and discussion were given in "Results and discussion" and concluded in "Conclusions."

Data and methods

The data used in this study were from China National Environmental Monitoring Center (http://113.108.142.147:20035/emcpublish/). The arrangement rules of state controlling air sampling sites, the instruments measured gaseous and particulate matter pollutants, the quality assurance, and controls of data and analytical methods such as cluster analysis were introduced in detail by Zhao et al. (2016), which can be referred to if one wanted to know that.

Results and discussion

Overview of air pollution situations in China

The Chinese capital cities experienced severe air pollution in recent years such as long-lasting haze events in North China Plain due to more primary emissions from human activities and poor diffusion conditions. To better see air pollution levels in China, the annual mean concentrations of gaseous (SO_2, NO_2, CO, and O_3) and particulate matter pollutants ($PM_{2.5}$ and PM_{10}) and the ratios of $PM_{2.5}$ to PM_{10} in the 31 provincial capital cities were summarized in Table 1. The cities were divided into two classes, i.e., one with winter heating and the other without winter heating to know effect of domestic heating on urban air quality in China. The annual mean $PM_{2.5}$ concentrations varied from 42.3 $\mu g/m^3$ (Huhehaote) to 102.1 $\mu g/m^3$ (Shijiazhuang) for the cities with home heating in winter, which were much higher than those for the cities

Table 1 Annual mean concentrations of the six criteria pollutants and the ratios of $PM_{2.5}$ to PM_{10} in the 31 capital cities

Cities	$PM_{2.5}$, μg/m^3	PM_{10}, μg/m^3	CO, mg/m^3	NO_2, μg/m^3	O_3, μg/m^3	SO_2, μg/m^3	$PM_{2.5}/PM_{10}$, %
Winter heating							
Beijing	78.0 ± 70.6	117.8 ± 87.9	1.2 ± 1.0	53.9 ± 29.3	56.5 ± 53.7	16.4 ± 18.0	61.5 ± 21.1
Changchun	65.8 ± 64.2	112.8 ± 85.8	0.9 ± 0.5	43.9 ± 22.5	52.0 ± 37.1	36.4 ± 38.1	56.1 ± 17.9
Haerbin	71.0 ± 82.7	108.2 ± 101.0	0.9 ± 0.5	51.0 ± 27.1	42.2 ± 30.5	53.5 ± 62.5	61.1 ± 17.2
Huhehaote	42.3 ± 36.2	115.5 ± 86.1	1.5 ± 1.0	42.4 ± 23.0	43.3 ± 34.6	43.4 ± 41.2	36.9 ± 16.0
Jinan	84.4 ± 49.8	166.8 ± 85.6	1.2 ± 0.7	53.8 ± 27.4	67.1 ± 54.5	61.6 ± 44.1	50.6 ± 12.8
Lanzhou	56.1 ± 35.9	120.7 ± 107.3	1.5 ± 0.9	42.9 ± 21.9	41.5 ± 28.4	24.0 ± 17.0	50.3 ± 15.0
Shenyang	74.4 ± 67.7	125.3 ± 105.7	1.1 ± 0.6	49.9 ± 25.0	55.5 ± 45.2	74.4 ± 80.2	57.3 ± 15.4
Shijiazhuang	102.1 ± 77.1	176.1 ± 106.1	1.4 ± 1.2	46.9 ± 28.7	50.2 ± 45.4	57.1 ± 48.2	55.3 ± 15.3
Taiyuan	64.5 ± 49.6	120.7 ± 70.9	1.7 ± 0.8	37.0 ± 18.0	40.5 ± 36.8	77.4 ± 82.3	50.0 ± 13.5
Tianjin	80.9 ± 62.5	134.3 ± 88.2	1.6 ± 0.9	52.2 ± 27.8	49.0 ± 45.3	39.1 ± 33.9	58.9 ± 18.1
Wulumuqi	65.3 ± 58.1	151.8 ± 102.7	1.3 ± 1.0	54.3 ± 27.3	31.1 ± 29.9	23.2 ± 18.8	44.3 ± 20.3
Xi’an	60.5 ± 38.0	130.6 ± 71.6	1.7 ± 0.7	40.8 ± 16.7	36.3 ± 36.4	25.5 ± 21.0	46.5 ± 12.3
Xining	57.3 ± 32.7	111.5 ± 86.0	1.2 ± 0.7	32.2 ± 13.9	44.7 ± 24.1	31.2 ± 29.9	55.1 ± 14.7
Yinchuan	46.7 ± 31.6	108.5 ± 86.6	1.1 ± 0.5	37.2 ± 20.7	46.1 ± 31.9	62.3 ± 75.5	44.7 ± 14.3
Zhenzhou	92.2 ± 62.2	161.7 ± 88.4	1.6 ± 0.6	51.0 ± 22.4	45.5 ± 31.0	40.1 ± 32.7	56.6 ± 17.2
No winter heating							
Changsha	68.4 ± 38.8	97.3 ± 49.4	1.0 ± 0.3	40.5 ± 20.5	45.9 ± 32.7	22.8 ± 12.0	70.8 ± 15.0
Chengdu	65.0 ± 42.4	108.7 ± 65.0	1.1 ± 0.4	50.0 ± 20.6	46.2 ± 40.2	15.6 ± 12.3	59.1 ± 8.6
Fuzhou	31.5 ± 16.6	62.1 ± 28.6	0.8 ± 0.2	29.1 ± 13.8	64.2 ± 39.6	6.5 ± 3.4	52.0 ± 14.5
Guangzhou	43.8 ± 24.2	64.6 ± 32.7	1.0 ± 0.3	44.3 ± 22.6	46.3 ± 47.0	16.0 ± 8.8	66.8 ± 10.5
Guiyang	43.7 ± 24.7	67.7 ± 35.1	0.7 ± 0.3	26.4 ± 11.8	51.6 ± 24.9	19.4 ± 17.6	64.4 ± 13.1
Haikou	22.4 ± 17.7	40.3 ± 23.4	0.7 ± 0.8	13.0 ± 7.8	41.2 ± 28.2	5.3 ± 3.4	52.7 ± 13.8
Hangzhou	60.2 ± 32.5	95.2 ± 47.9	0.9 ± 0.3	46.4 ± 20.6	56.6 ± 44.4	18.4 ± 11.9	64.1 ± 14.4
Hefei	73.1 ± 43.3	113.9 ± 61.4	1.0 ± 0.4	26.6 ± 11.9	34.2 ± 18.8	19.6 ± 9.7	65.4 ± 16.9
Kunming	30.9 ± 17.5	60.0 ± 31.9	1.1 ± 0.4	32.0 ± 14.1	47.8 ± 32.0	18.2 ± 11.9	52.7 ± 13.9
Nanchang	48.6 ± 30.8	79.6 ± 45.3	1.0 ± 0.3	30.3 ± 15.6	47.4 ± 35.1	22.4 ± 13.9	60.6 ± 15.6
Nanjing	69.2 ± 42.7	115.6 ± 63.9	0.9 ± 0.4	48.7 ± 25.2	53.4 ± 49.3	19.9 ± 13.4	60.6 ± 14.5
Nanning	46.0 ± 33.3	79.2 ± 51.4	1.0 ± 0.3	34.6 ± 19.6	46.4 ± 34.6	14.6 ± 13.4	57.6 ± 14.4
Shanghai	54.2 ± 41.3	79.0 ± 52.6	0.9 ± 0.4	45.7 ± 25.6	67.3 ± 40.6	18.5 ± 13.5	67.4 ± 17.1
Chongqing	61.6 ± 41.6	93.9 ± 54.6	1.1 ± 0.3	38.8 ± 16.1	35.4 ± 38.2	18.5 ± 9.1	63.8 ± 11.6
Wuhan	73.5 ± 43.7	115.5 ± 62.4	1.1 ± 0.5	53.0 ± 28.6	60.2 ± 54.6	27.4 ± 42.4	64.9 ± 16.5
Lasa	22.8 ± 15.6	53.3 ± 34.4	0.9 ± 0.4	21.3 ± 15.1	77.1 ± 31.6	10.3 ± 5.2	44.1 ± 13.1

without winter heating with lowest concentration of 22.4 μg/m^3 in Haikou. No cities in China were up to Chinese Ambient Air Quality Standards (CAAQS) Grade I standard (15 μg/m^3) of $PM_{2.5}$, and only four cities including Fuzhou, Haikou, Kunming, and Lasa met the CAAQS Grade II standard (35 μg/m^3). The annual mean concentrations of PM_{10} were between 40.3 μg/m^3 (Haikou) and 176.1 μg/m^3 (Shijiazhuang). PM_{10} exceeded the CAAQS Grade I standard (40 μg/m^3) in all cities and exceeded the CAAQS Grade II standard (70 μg/m^3) with the exception of part cities without winter heating including Fuzhou, Guangzhou, Guiyang, Haikou, Kunming, and Lasa. CO and SO_2 concentrations in Taiyuan were the highest among the provincial capital cities partly due to the amount mining of coal in Shanxi. SO_2 concentrations exceeded Grade I standard (20 μg/m^3) in the cities with winter heating, and the cities exceeded Grade II standard (60 μg/m^3) have domestic heating in winter. NO_2 concentration differences between the cities with winter heating and those without winter heating were less evident. NO_2 was higher in Beijing than the other cities, which may be attributed to more vehicle numbers in Beijing. No clear spatial difference was found for O_3 among the cities, which was accordance with the results of Chai et al. (2014). As a typically secondary pollutant, O_3 is formed by some photochemical reactions between NO_x and volatile organic compounds (VOC) in the atmosphere (Atkinson 2000). To some extent, the spatial patterns of O_3 were in contrast to other pollutants, such as the concentrations of $PM_{2.5}$, $PM_{10,}$ CO, NO_2, and SO_2

were low in Lasa, while O_3 (77.1 ± 31.6 μg/m^3) was the highest among 31 provincial capital cities in China.

Higher concentrations of $PM_{2.5}$, PM_{10}, CO, and SO_2 were generally observed in the cities with winter heating, indicating effect of home heating on urban air quality in the northern China. Xiao et al. (2015) analyzed the impact of winter heating on aerosol loadings over China using the MODIS-Aqua Collection 6 aerosol product from 2004 to 2012 and found that domestic heating had a large contribution to elevated $PM_{2.5}$ concentrations in more than three quarters of central and eastern China. Furthermore, some studies showed that high concentrations in the North China were due to the emissions from fossil fuel combustion, coal-based industries, and biomass burning (Chai et al. 2014; Pui et al. 2014; Qu et al. 2010; Yang et al. 2013; Zheng et al. 2005; Zhang, et al. 2009; Zhao et al. 2011).

Cluster analysis was used to classify the annual variations of each pollutant in the 31 cities as several groups with similar pollutant concentrations and variation trends within groups to better see air pollution situations in major Chinese cities. SO_2 and $PM_{2.5}$ were divided into five and three clusters, respectively, while the remaining air pollutants formed four groups each. The clustering results from annual variations of each pollutant in 31 provincial capital cities and the corresponding cities to each cluster are shown in Figs. 1 and 2, respectively. The red, gray, and blue symbols, respectively, represented severely, moderately, and slightly polluted cities in Fig. 2. The air pollution in winter was more severe than that in other seasons mainly due to more primary emissions such as domestic heating (Xiao et al. 2015), poor diffusional conditions, and shallow boundary layer in winter. Some studies revealed local meteorology largely impacted on air pollution levels with the negative correlations between meteorological factors such as temperature, relative humidity, precipitation, wind speed and mixing layer height (MLH), and air pollution index (API) (Han et al. 2009; Xu et al. 2011). High NO_x concentration in ambient air can also depress producing of O_3 and lead to low O_3 concentration near the ground surface (Han et al.

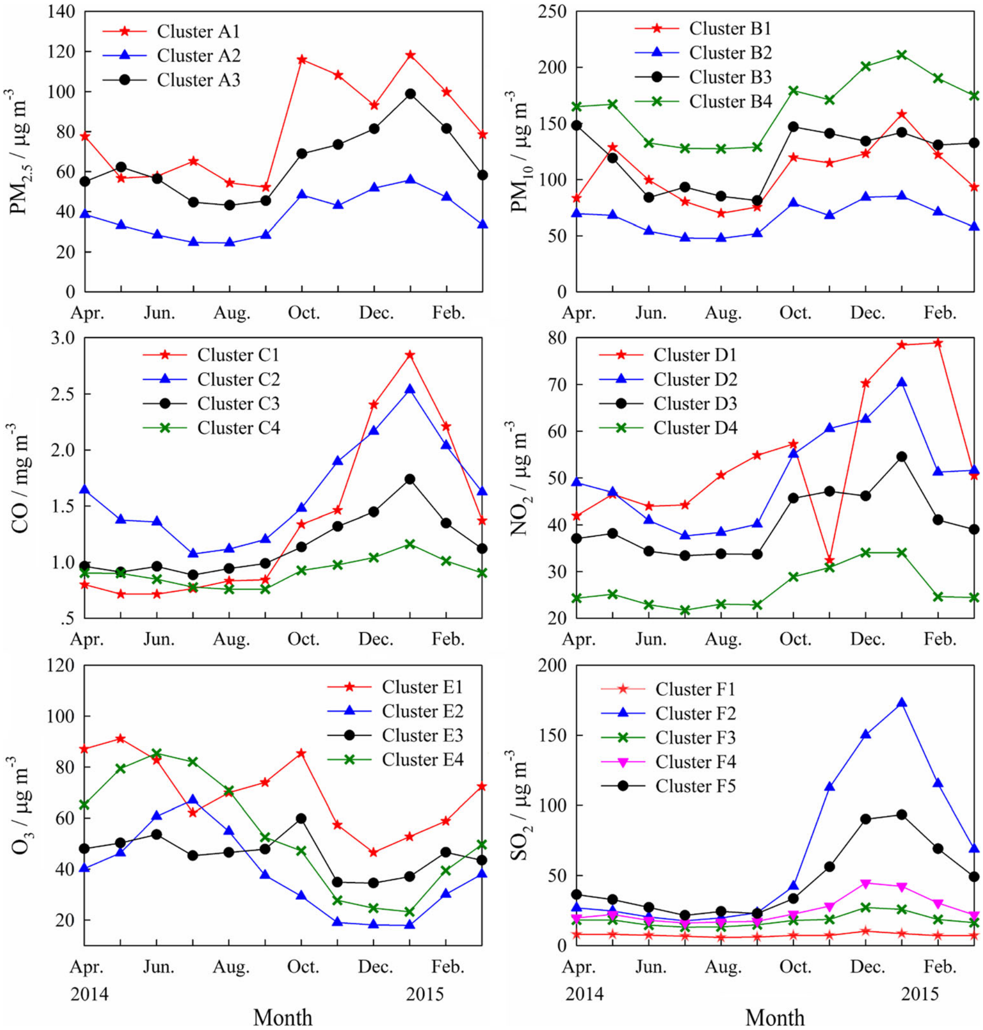


Fig. 1 Clustering results for annual variations of monthly mean concentrations of six criteria air pollutants in 31 provincial capital cities

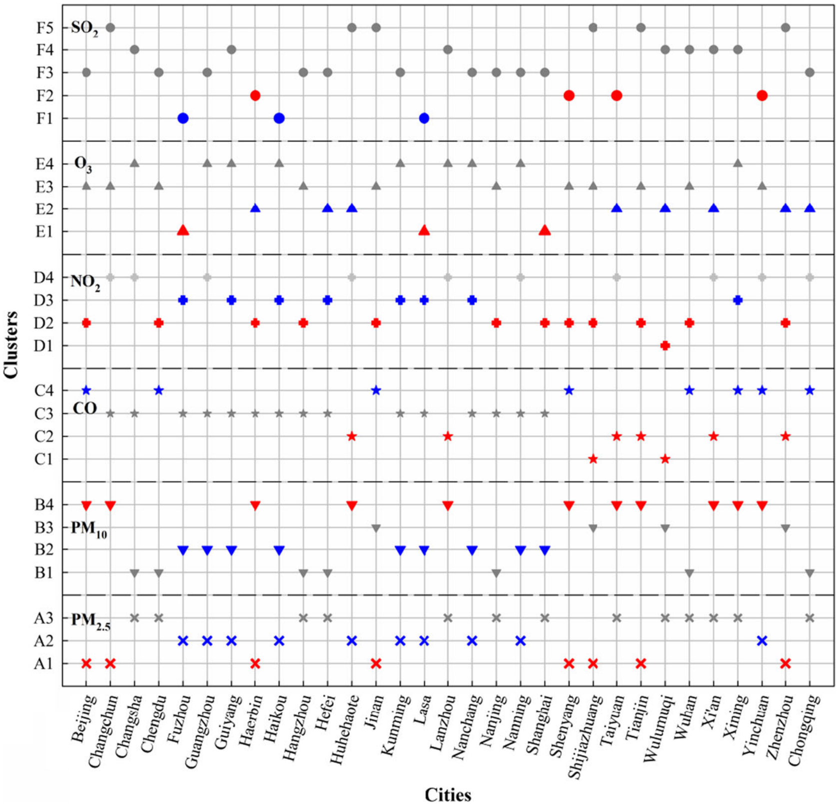
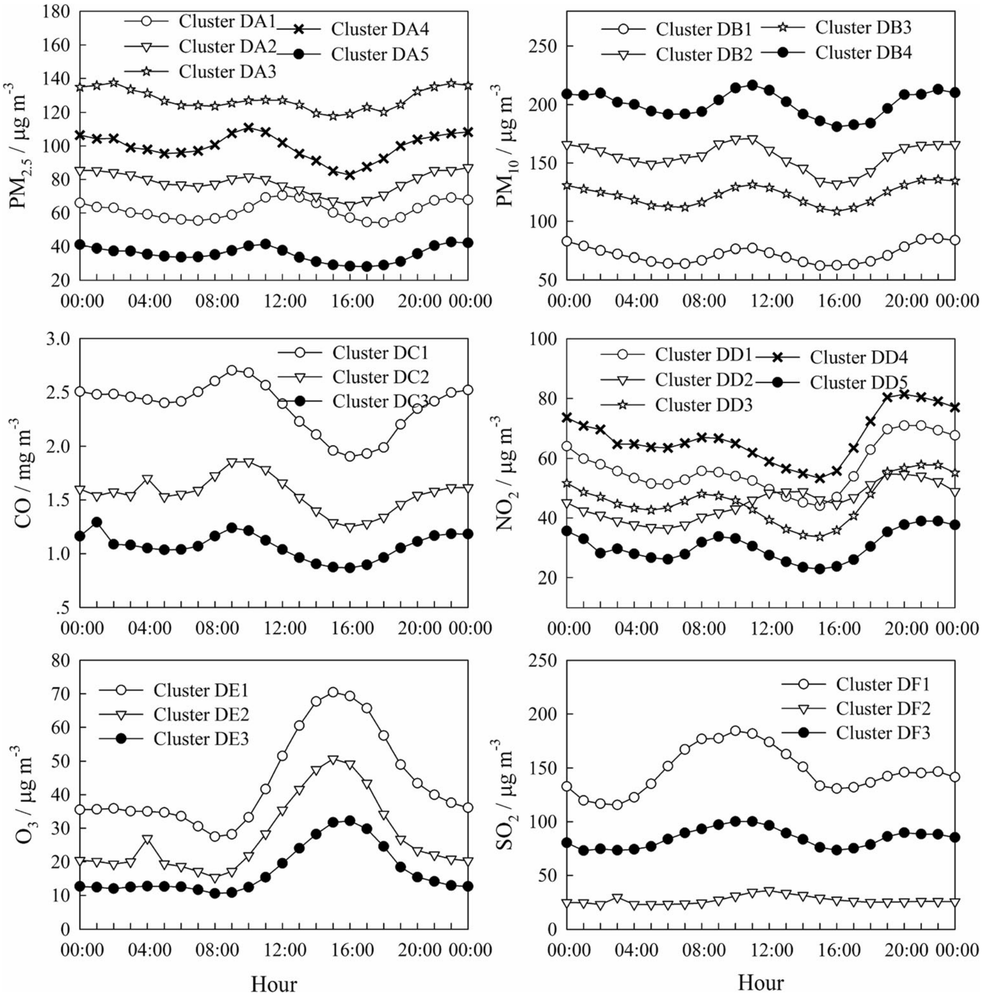


Fig. 2 The corresponding cities to each cluster in Fig. 1. The *red*, *gray*, and *blue symbols* represented severely, moderately, and slightly polluted cities, respectively

2009). The nitrogen monoxide (CO) from combustion is a major sink for O_3, and thus, the concentrations of O_3 are negatively correlated with the concentrations of all the combustion emitted pollutants.

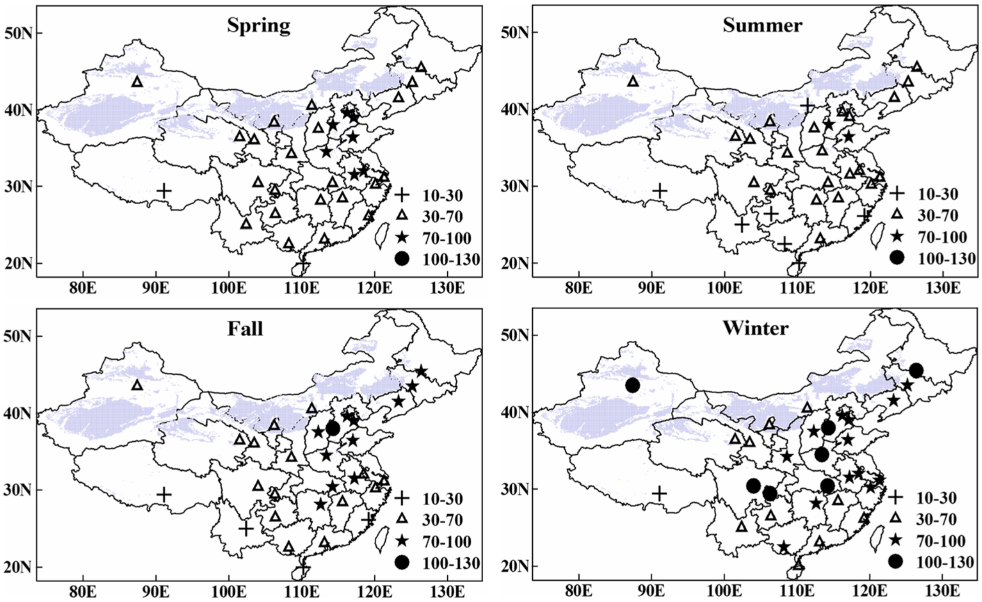
The differences between clusters for each pollutant were less in spring and summer than those in fall and winter, especially SO_2 due to more coal combustion for domestic heating in northern China and adverse diffusion conditions. The mean annual variation of $PM_{2.5}$ in northern cities (Cluster A1) was bimodal with peaks in October 2014 and January 2015, which mainly appears in North China Plain (Beijing, Jinan, Shijiazhuang, Tianjin, and Zhenzhou) and Northeast China (Changchun, Haerbin, and Shenyang), while that in southern cities (Cluster A2) was less evident with slightly high $PM_{2.5}$ in winter (see Figs. 1 and 2). The annual variations of O_3 were generally in contrast to that of other pollutants with the highest value in summer (Fig. 1). However, O_3 and $PM_{2.5}$ have significantly high values in October 2014 for the cities of North China Plain and Northeast China (Clusters A1 and E3 of Fig. 1). O_3 is formed by some photochemical reactions between NO_x and VOCs in the low atmosphere (Atkinson 2000). Liu et al. (2005) estimated that vehicular exhaust was the major contributor of VOC (on average 57.7 %), followed by painting operations, gasoline vapor, and liquefied petroleum gas (LPG) at 12.4, 11.3, and 5.8 %, respectively. The high O_3 concentration in October 2014 in North China Plain indicated that high volatile organic compounds in the atmosphere due to more motor vehicles in the two regions. Therefore, the high $PM_{2.5}$ concentration in October 2014 in northern cities (Cluster A1) may be due to more secondary organic aerosols (SOA) from chemical reactions of VOCs. The study of Chai et al. (2014) also showed that monthly variation of $PM_{2.5}$ had a weak peak in October 2011 in northern cities, which was consistent with the results from our study. High $PM_{2.5}$ concentration in January 2015 for Clusters A1 and A3 may be due to integrated effects of primary emissions such as domestic heating and adverse diffusional conditions. The annual variation of PM_{10} concentrations in northern cities also had a remarkable peak in April–May 2014 with the exception of the peaks in October 2014 and January 2015, and the variations of $PM_{2.5}$ and PM_{10} were inconsistent in April–May 2014 due to larger impact of dust events on PM_{10} than on $PM_{2.5}$ (Clusters A1 and B4) (Zhao et al. 2015c). However, the variations of $PM_{2.5}$ and PM_{10} concentrations were synchronous in southern China (Clusters A2 and B2). Furthermore, elevated CO (Clusters C1 and C2) and SO_2 (Clusters F2 and F5) concentrations all appeared in northern cities (see Fig. 2). For NO_2, Cluster D1 had significant low value in November and only Wulumuqi was assigned to the cluster. Particulate matter ($PM_{2.5}$ and PM_{10}) and other gaseous pollutants (SO_2 and CO) also had significant low values in November 2014 in Wulumuqi,

Fig. 3 Clustering results for diurnal variations of hourly mean concentrations of six criteria air pollutants in 31 provincial capital cities in the winter



especially PM_{10} and SO_2, which is in accordance with NO_2 (data not shown). Therefore, the phenomenon that Cluster D1 has significant low value in November is true and the NO_2 concentration is valid. The low concentration of five criteria air pollutants ($PM_{2.5}$, PM_{10}, SO_2, CO, and NO_2) in November 2014 in Wulumuqi was maybe related to favorable weather

Fig. 4 Spatial distribution of seasonal mean $PM_{2.5}$ mass concentrations (μg/m^3) during the study period (the *gray-shaded areas* represent desert and desertified land in China)



conditions. The precipitation was 31.6 mm in November 2014 in Wulumuqi, which was much higher than other months with the exception of April and May 2014 (data not shown). The air pollutants were partly scavenged from the atmosphere by raindrops or snow particles. Clusters F2 and F5, which mainly appears in North China Plain (Tianjin, Zhenzhou, Jinan, and Taiyuan) and Northeast China (Shenyang, Haerbin, and Changchun), had marked seasonal variation with maximal SO_2 in winter season due to domestic heating, while other clusters have much weaker seasonal variation in SO_2 concentration and mainly appears in southern cities without winter heating in those cities. The weak peak of SO_2 concentration in Clusters F1, F3, and F4 was mainly related to poor diffusional conditions in winter season in southern cities. Stagnant air with weak winds and low boundary layer height appeared frequently in winter, and thus, the air pollutants were trapped near the surface layer and local air pollution was aggravated (Zhao et al. 2015b). The above analyses indicated that SO_2 concentrations in the cities of North China Plain and Northeast China in winter season were much higher than other regions of China, indicating that there were apparent spatial feature of air pollution in China. However, the regional feature of air pollution cannot be better showed in our study due to only used the data from provincial capital cities and the distance among the cities is relatively far. Additionally, the O_3 was significantly higher in Fuzhou, Lasa, and Shanghai than that in other cities, while other primarily emitted pollutants such as SO_2 and CO were low in the three cities.

The cluster analysis was also used to divide mean diurnal variations of each air pollutant in all cities in the winter and clustering results were shownin Fig. 3. As can be seen from the figure, the diurnal variations of O_3 concentration were unimodal with a peak at 15:00~16:00 (Beijing time, hereafter the same), while other air pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, and CO) had two obvious peaks at morning and evening rush hours due to effect of on-road motor vehicles. Furthermore, it is worth noting that the air pollutants from primary emissions also had a peak in winter early morning which were comparable with or sometimes more significant than the morning and evening peaks due to stagnant weather and the shallow planetary boundary layer in winter morning causing high concentration of air pollutants near surface layer in these cities. The above analyses indicated that local adverse weather conditions such as more stagnant air and the lower planetary boundary layer in winter played a major role in winter air pollution. Additionally, the pulses of some air pollutant concentrations such as CO (Clusters DC2 and DC3) and O3 (Cluster DE2) can be seen in Fig. 3. After careful analyzing, we confirmed the phenomenon was primarily related to underlying discharges from some industries during nighttime in some cities (Zhao et al. 2016).

Spatial and seasonal variations of six criteria pollutants

$PM_{2.5}$ mass concentrations have large temporal variation with the highest in the winter (December 2014–February 2015) and the lowest in the summer (June–August 2014) for the Chinese provincial capital cities (Fig. 4), which was mainly related to more emissions and more adverse meteorological conditions in winter as source emissions and meteorological field are two major factors influencing concentrations of air pollutants. Furthermore, less emissions and favorable diffusion and dilution conditions in summer lead to low air pollution in China. The spring

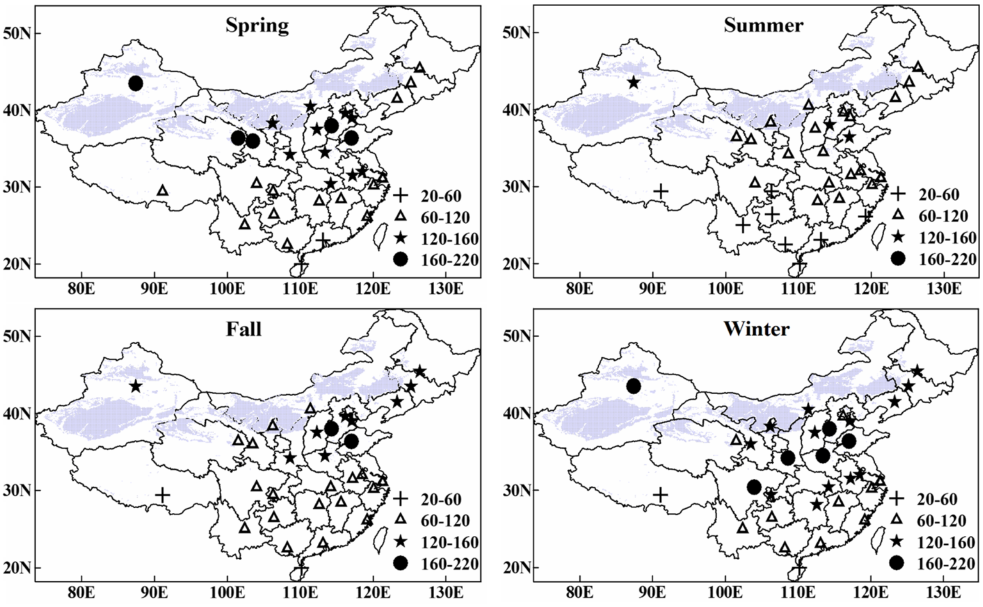


Fig. 5 Spatial distributions of seasonal mean PM_{10} mass concentrations ($\mu g/m^3$) during the study period (the *gray-shaded areas* represent desert and desertified land in China)

(April–May 2014) and fall (September–November 2014) concentrations almost were comparable, but $PM_{2.5}$ concentrations were much higher in fall than those in spring for the cities located at northeastern China probably due to biomass burning near the cities in fall. The elevated $PM_{2.5}$ in winter were caused by combined effect of both domestic heating and poor meteorological conditions for air pollution dilution and diffusion (Chai et al. 2014). The $PM_{2.5}$ mass concentrations were even higher in eastern China than those in other areas in four seasons, especially North China Plain (Beijing, Tianjin, Shijiazhuang, Jinan, and Zhenzhou) due to large urban agglomeration in NCP. Additionally, industrial sources can also lead to high $PM_{2.5}$ in the cities with heavy industry. For example, Cheng et al. (2012) indicated that about 27 % of $PM_{2.5}$ can be attributed to industrial emissions in Wuhan.

PM_{10} reached maximum in the spring in some cities of Northwestern China such as Wulumuqi, Xining, Lanzhou, and Yinchuan (Fig. 5). Dust plumes came from Asian desert areas were main pollution sources of particulate matter in northwestern cities in spring. The dust plumes, originated from Taklimakan and Gobi Deserts, were transported by strong winds to downstream cities and then affected urban air quality (Qu et al. 2010; Zhao et al. 2015a; Chen et al. 2015). The study of Ta et al. (2004) found that the "high PM_{10} episodes" in northern China were mostly due to desert dust invasions. In addition, Wang et al. (2006) studied the effects of floating dust, dust storm, and blowing dust on PM_{10} mass concentration in four north Chinese cities including Beijing, Huhehaote, Xi'an, and Lanzhou and found that floating dust had the largest contribution to PM_{10}

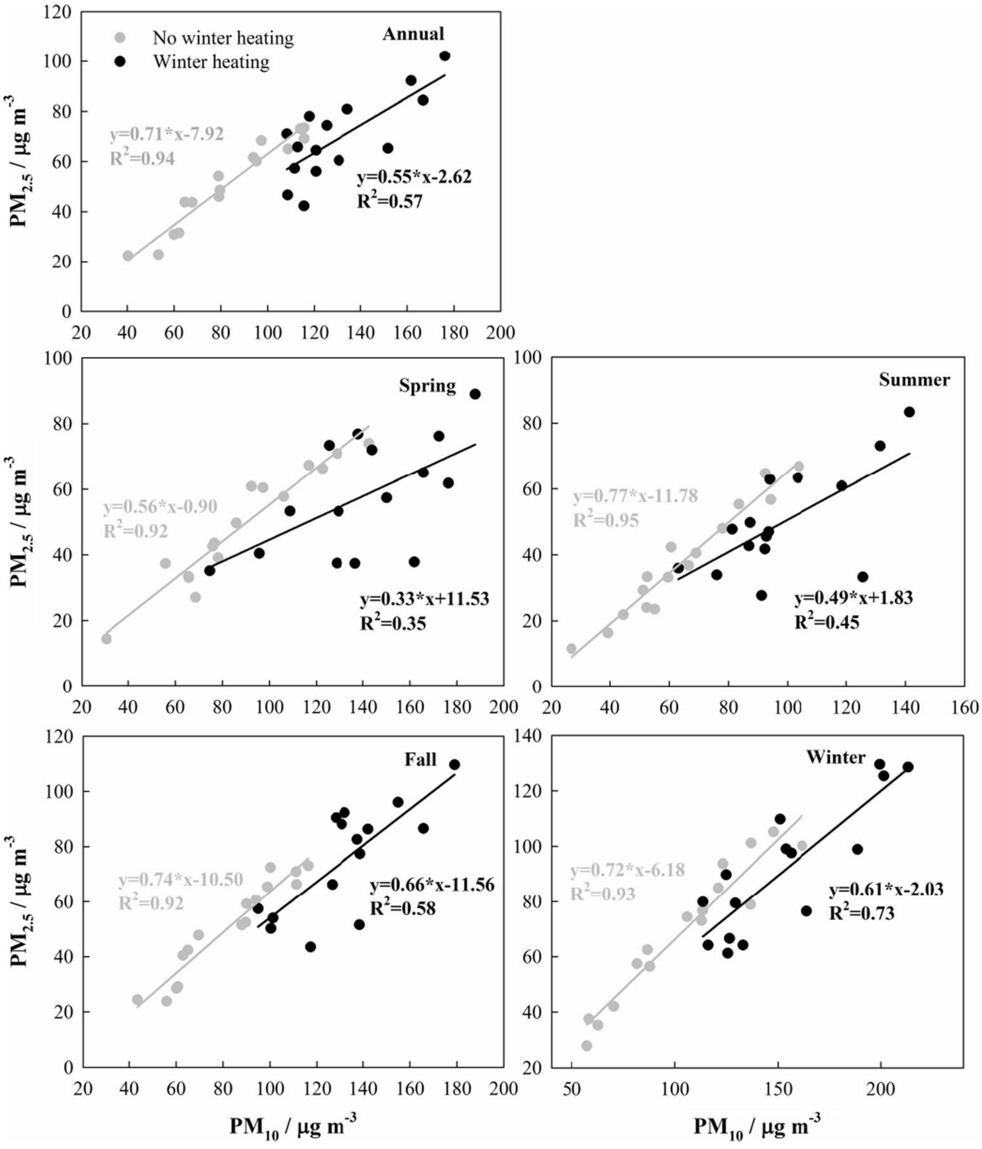


Fig. 6 Relationships between annual and seasonal mean $PM_{2.5}$ and PM_{10} concentrations for the cities with (*black dots*) or without winter heating (*gray dots*)

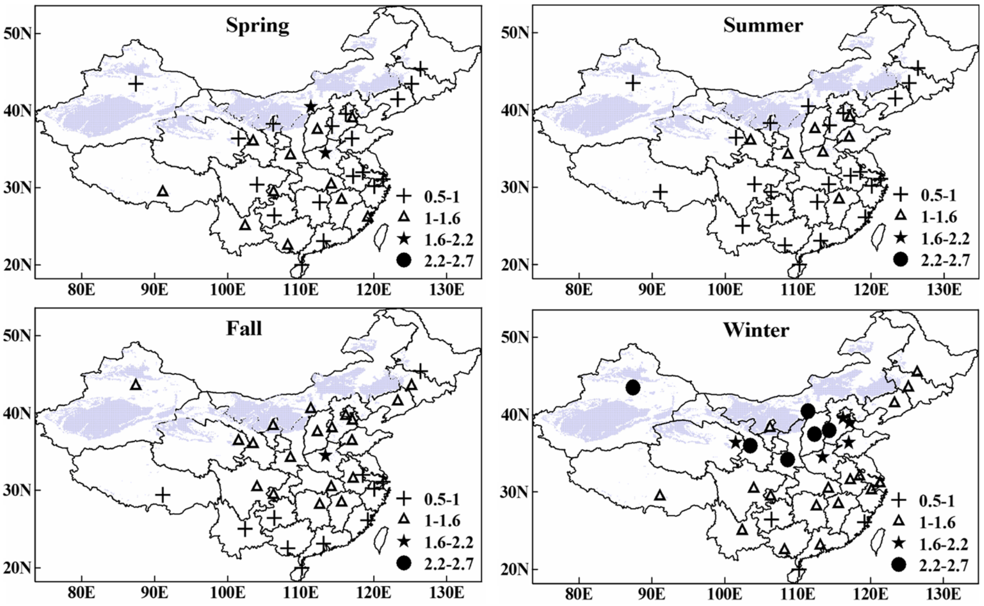


Fig. 7 Spatial distributions of seasonal mean CO concentrations (mg m^{-3}) during the study period (the *gray-shaded areas* represent desert and desertified land in China)

followed by dust storm and blowing dust. As can also be seen from Fig. 6, $PM_{2.5-10}$ (particles with size between 2.5 and 10 μm) accounted for a large proportion of PM_{10} in Northern China in spring (67 %), indicating effects of dust events and domestic heating on urban air quality. For the other cities, PM_{10} concentrations show a similar seasonal variation as $PM_{2.5}$ and especially in the southern cities. $PM_{2.5}$ concentration accounted for PM_{10} was high in Southern China (>55 %), resulting in the synchronously seasonal variations between $PM_{2.5}$ and PM_{10} mass concentrations (Fig. 6).

CO (Fig. 7), NO_2 (Fig. 8), and SO_2 (Fig. 9) also exhibited the synchronously seasonal trends with $PM_{2.5}$ mass concentrations, which reflected combined effects of weather conditions and primary emissions. Stagnant air with weak winds and low boundary layer height appeared frequently in winter, and thus, the air pollutants were trapped near the surface layer and local air pollution was aggravated (Zhao et al. 2015b). Furthermore, frequent air pollution episodes in winter were partly attributed to home coal combustion for heating (Zhang et al. 2009; Zhao et al. 2011). Furthermore, the difference of

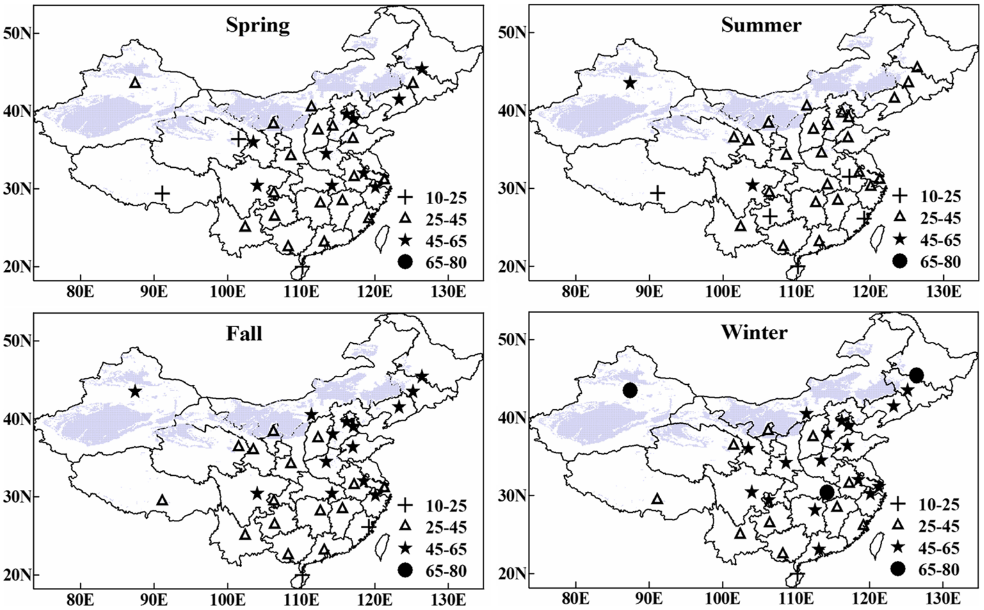


Fig. 8 Spatial distributions of seasonal mean NO_2 concentrations (μg/m^3) during the study period (the *gray-shaded areas* represent desert and desertified land in China)

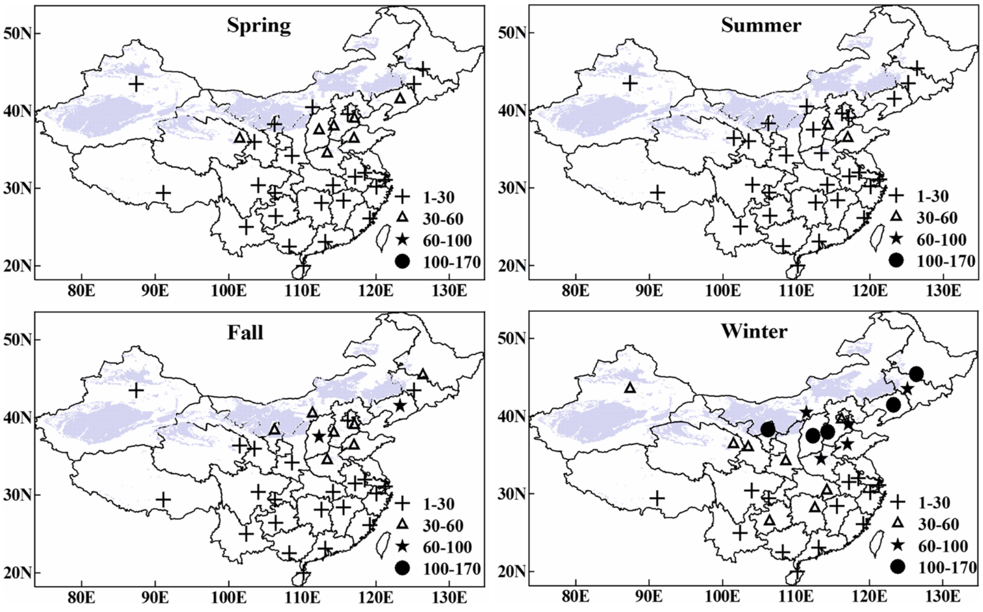


Fig. 9 Spatial distributions of seasonal mean SO_2 concentrations ($\mu g/m^3$) during the study period (the *gray-shaded areas* represent desert and desertified land in China)

SO_2 concentrations between summer and winter was more obvious than the two other gases pollutants in the Northern China, suggesting large impact of the domestic home heating on urban air quality. The domestic heating also is an important pollution source in winter and spring for the northern cities, which can lead to significant increases of some air pollutants such as SO_2 and CO. The seasonal variation of O_3 was opposite to the five other pollutants ($PM_{2.5}$, PM_{10}, CO, NO_2, and SO_2) with a minimum in the winter and a maximum in the summer (Fig. 10), which may be due to dependence of O_3 formation rate on the intensity of solar radiation (Atkinson 2000).

In the spatial scale, the air pollutants from primary emissions such as CO and SO_2 (Figs. 7 and 9) in northern China were significantly higher than those in southern China, especially North China Plain (NCP). NCP was one of the most polluted areas in China due to more local primary emissions from coal-based industries and strong regional transportation between the cities. The study by Hu et al. (2014) indicated regional characteristics of air pollution were significant in North China Plain.

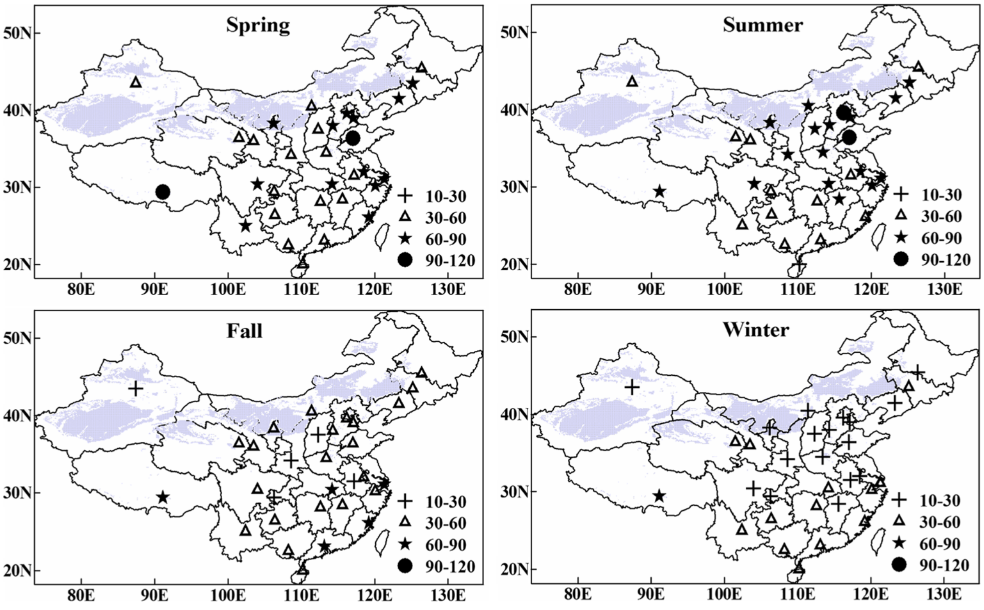


Fig. 10 Spatial distributions of seasonal mean O_3 concentrations ($\mu g/m^3$) during the study period (the *gray-shaded areas* represent desert and desertified land in China)

Conclusions

This study has reported the spatial and seasonal variations of $PM_{2.5}$, PM_{10}, CO, NO_2, SO_2, and O_3 using air quality monitoring data observed in 31 Chinese provincial cities during April 2014 to March 2015, which will help to obtain a better understanding of the current air pollution situation in China and provide useful information regarding the status of nationwide air pollution. The annual mean concentrations of $PM_{2.5}$ and PM_{10} for all cities exceeded CAAQS Grade I standard. Only Fuzhou, Haikou, Kunming, and Lasa met Grade II standard for $PM_{2.5}$ and PM_{10}. Higher SO_2 concentration was observed in northern cities and especially in winter, indicating large emissions from coal combustion and biomass burning during the heating period. Additionally, $PM_{2.5\text{-}10}$ (particles with size between 2.5 and 10 μm) accounted for a large proportion of PM_{10} in northern China in spring (67 %), indicating the effect of dust events on urban air quality in northern China. The seasonal variation of O_3 was opposite to other pollutants with the lowest concentrations in the winter and the highest in the summer.

With rapid increases of vehicle counts, urban expansion and economic development in the last two decades in China, severe air pollution problems have significantly negative effects on human health. Almost all the Chinese cities are suffering from a high concentration of primary or secondary pollutants. This suggests that China should conduct region-oriented air pollution management plans. This study only analyzed spatial and seasonal variations of six criteria pollutants in China. In order to better understand main factors influencing urban air quality, future study is necessary to investigate the associations between air quality and weather conditions and transportation of air pollutants in both intra- and inter-region.

Acknowledgments This work was financially funded by the Open Fund Program of Key Laboratory of Land Surface Process and Climate Change in Cold and Arid Regions, Chinese Academy of Sciences (No. LPCC201510), and Opening Project of Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP^3) (FDLAP16005). We were very grateful to China National Environmental Monitoring Center (http://113.108.142.147:20035/emcpublish/) for the provision of air quality monitoring data used in this study.

References

Atkinson R (2000) Atmospheric chemistry of VOCs and NO_x. Atmos Environ 34:2063–2101

Bapna M (2012) Toward an environmentally sustainable future: country environmental analysis of the People's Republic of China. 2012 The Asian Development Bank, Manila

Cao Z, Sheng L, Liu Q, Yao X, Wang W (2015) Interannual increase of regional haze-fog in North China Plain in summer by intensified easterly winds and orographic forcing. Atmos Environ 122:154–162

Chai F, Gao J, Chen Z, Wang S, Zhang Y, Zhan J, Zhang H, Yun Y, Ren C (2014) Spatial and temporal variation of particulate matter and gaseous pollutants in 26 cities in China. J Environ Sci 26:75–82

Chan CK, Yao X (2008) Air pollution in mega cities in China. Atmos Environ 42(1):1–42

Che H, Zhang X, Li Y, Zhou Z, Qu JJ, Hao X (2009) Haze trends over the capital cities of 31 provinces in China, 1981-2005. Theor Appl Climatol 97:235–242

Chen Y, Xie S (2014) Characteristics and formation mechanism of a heavy air pollution episode caused by biomass burning in Chengdu, Southwest China. Sci Total Environ 473–474:507–517

Chen J, Zhao SP, Yin DY, Qu JJ (2015) Effect of dust process on air quality in cities of Northern China. J Desert Res 35(2):423–430

Cheng H, Wang Z, Feng J, Chen H, Zhang F, Liu J (2012) Carbonaceous species composition and source apportionment of $PM_{2.5}$ in urban atmosphere of Wuhan. Ecol Environ Sci 21:1574–1579

Cheng Z, Jiang J, Fajardo O, Wang S, Hao J (2013) Characteristics and health impacts of particulate matter pollution in China (2001-2011). Atmos Environ 65:186–194

Du YJ, Li TT (2016) Assessment of health-based economic costs linked to fine particulate ($PM_{2.5}$) pollution: a case study of haze during January 2013 in Beijing, China. Air Qual Atmos Health 9(4):439–445

Guo S, Hu M, Zamora ML, Peng J, Shang D, Zheng J, Du Z, Wu Z, Shao M, Zeng L, Molina MJ, Zhang R (2014) Elucidating severe urban haze formation in China. Proc Natl Acad Sci U S A 111(49):17373–17378. doi:10.1073/pnas.1419604111

Han S, Bian H, Tie X, Xie Y, Sun M, Liu A (2009) Impact of nocturnal planetary boundary layer on urban air pollutants: measurements from a 250-m tower over Tianjin, China. J Hazard Mater 162:264–269

Han S, Wu J, Zhang Y, Cai Z, Feng Y, Yao Q, Li X, Liu Y, Zhang M (2014) Characteristics and formation mechanism of a winter haze-fog episode in Tianjin, China. Atmos Environ 98:323–330

Hu J, Wang Y, Ying Q, Zhang H (2014) Spatial and temporal variability of $PM_{2.5}$ and PM_{10} over the North China Plain and the Yangtze River Delta, China. Atmos Environ 95:598–609

Huang RJ, Zhang YL, Bozzetti C, Ho KF, Cao JJ, Han YM, Daellenbach KR, Slowik JG, Platt SM, Canonaco F, Zotter P, Wolf R, Pieber SM, Bruns EA, Crippa M, Ciarelli G, Piazzalunga A, Schwikowski M, Abbaszade G, Schnelle-Kreis J, Zimmermann R, An ZS, Szidat S, Baltensperger U, El Haddad I, Prevot ASH (2014) High secondary aerosol contribution to particulate pollution during haze events in China. Nature 514(7521):218–222

Jia B, Wang Y, Yao Y, Xie Y (2015) A new indicator on the impact of large-scale circulation on wintertime particulate matter pollution over China. Atmos Chem Phys 15:11919–11929

Johansson C, Norman M, Gidhagen L (2007) Spatial & temporal variations of PM10 and particle number concentrations in urban air. Environ Monit Assess 127:477–487

Kan H, Chen R, Tong S (2012) Ambient air pollution, climate change, and population health in China. Environ Int 42:10–19

Karar K, Gupta AK, Kumar A, Biswas AK (2006) Seasonal variations of PM_{10} and TSP in residential and industrial sites in an urban area of Kolkata, India. Environ Monit Assess 118(1-3):369–381

Lewne M, Cyrys J, Meliefste K, Hoek G, Brauer M, Fischer P, Gehring U, Heinrich J, Brunekreef B, Bellander T (2004) Spatial variation in nitrogen dioxide in three European areas. Sci Total Environ 332:217–230

Li L, Qian J, Ou CQ, Zhou YX, Guo C, Guo Y (2014) Spatial and temporal analysis of air pollution index and its timescale-dependent relationship with meteorological factors in Guangzhou, China, 2001-2011. Environ Pollut 190:75–81

Liu Y, Shao M, Zhang J, Fu L, Lu S (2005) Distributions and source apportionment of ambient volatile organic compounds in Beijing city, China. J Environ Sci Heal 40:1843–1860

Martini FMS, Hasenkopf CA, Roberts DC (2015) Statistical analysis of PM2.5 observations from diplomatic facilities in China. Atmos Environ 110:174–185

Meng ZY, Xu XB, Yan P, Ding GA, Tang J, Lin WL, Xu XD, Wang SF (2009) Characteristics of trace gaseous pollutants at a regional background station in Northern China. Atmos Chem Phys 9:927–936

Pui DYH, Chen SC, Zuo Z (2014) $PM_{2.5}$ in China: measurements, sources, visibility and health effects, and mitigation. Particuology 13:1–26

Qu WJ, Arimoto R, Zhang XY, Zhao CH, Wang YQ, Sheng LF, Fu G (2010) Spatial distribution and interannual variation of surface PM_{10} concentrations over eighty-six Chinese cities. Atmos Chem Phys 10:5641–5662

Sun Y, Zhou X, Wai K, Yuan Q, Xu Z, Zhou S, Qi Q, Wang W (2013) Simultaneous measurement of particulate and gaseous pollutants in an urban city in North China plain during the heating period: implication of source contribution. Atmos Res 134:24–34

Ta W, Wang T, Xiao H, Zhu X, Xiao Z (2004) Gaseous and particulate air pollution in the Lanzhou Valley, China. Sci Total Environ 320:163–176

Tan JH, Duan JC, Zhen NJ, He KB, Hao JM (2016) Chemical characteristics and source of size-fractionated atmospheric particle in haze episode in Beijing. Atmos Res 167:24–33

Wang S, Yuan W, Shang K (2006) The impacts of different kinds of dust events on PM_{10} pollution in northern China. Atmos Environ 40:7975–7982

Wang W, Ren L, Zhang Y, Chen J, Liu H, Bao L, Fan S, Tang D (2008a) Aircraft measurements of gaseous pollutants and particulate matter over Pearl River Delta in China. Atmos Environ 42:6187–6202

Wang W, Chai FH, Zhang K, Wang SL, Chen YZ, Wang XZ, Yang YQ (2008b) Study on ambient air quality in Beijing for the summer 2008 Olympic Games. Air Qual Atmos Health 1(1):31–36

Wang ZB, Hu M, Wu ZJ, Yue DL (2013) Long-term measurements of particle number size distributions and the relationships with air mass history and source apportionment in the summer of Beijing. Atmos Chem Phys 13:10159–10170

Wang LT, Wei Z, Yang J, Zhang Y, Zhang FF, Su J, Meng CC, Zhang Q (2014a) The 2013 severe haze over southern Hebei, China: model evaluation, source apportionment, and policy implications. Atmos Chem Phys 14:3151–3173

Wang S, Xing J, Zhao B, Jang C, Hao J (2014b) Effectiveness of national air pollution control policies on the air quality in metropolitan areas of China. J Environ Sci 26:13–22

Wang X, Chen J, Cheng T, Zhang R, Wang X (2014c) Particle number concentration, size distribution and chemical composition during haze and photochemical smog episodes in Shanghai. J Environ Sci 26:1894–1902

Wang Y, Ying Q, Hu J, Zhang H (2014d) Spatial and temporal variations of six criteria air pollutants in 31 provincial capital cities in China during 2013–2014. Environ Int 73:413–422

Xiao Q, Ma Z, Li S, Liu Y (2015) The impact of winter heating on air pollution in China. Plos One 10(1):e0117311. doi:10.1371/journal.pone.0117311

Xu WY, Zhao CS, Ran L, Deng ZZ, Liu PF, Ma N, Lin WL, Xu XB, Yan P, He X, Yu J, Liang WD, Chen LL (2011) Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in the North China Plain. Atmos Chem Phys 11:4353–4369

Xu X, Zhao T, Liu F, Gong SL, Kristovich D, Lu C, Guo Y, Cheng X, Wang Y, Ding G (2016) Climate modulation of the Tibetan Plateau on haze in China. Atmos Chem Phys 16:1365–1375

Yang L, Cheng S, Wang X, Nie W, Xu P, Gao X, Yuan C, Wang W (2013) Source identification and health impact of $PM_{2.5}$ in a heavily polluted urban atmosphere in China. Atmos Environ 75:265–269

Zhang Q, Streets DG, Carmichael GR, He KB, Huo H, Kannari A, Klimont Z, Park IS, Reddy S, Fu JS, Chen D, Duan L, Lei Y, Wang LT, Yao ZL (2009) Asian emissions in 2006 for the NASA INTEX-B mission. Atmos Chem Phys 9:5131–5153

Zhang J, Chen J, Yang L, Sui X, Yao L, Zheng L, Wen L, Xu C, Wang W (2014) Indoor $PM_{2.5}$ and its chemical composition during a heavy haze-fog episode at Jinan, China. Atmos Environ 99:641–649

Zhao Y, Nielsen CP, Lei Y, McElroy MB, Hao J (2011) Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China. Atmos Chem Phys 11:2295–2308

Zhao XJ, Zhao PS, Xu J, Meng W, Pu WW, Dong F, He D, Shi QF (2013) Analysis of a winter regional haze event and its formation mechanism in the North China Plain. Atmos Chem Phys 13(11):5685–5696

Zhao SP, Yin DY, Qu JJ (2015a) Identifying sources of dust based on CALIPSO, MODIS satellite data and backward trajectory model. Atmos Pollut Res 6(1):36–44

Zhao SP, Yu Y, Yin DY, He JJ (2015b) Meteorological dependence of particle number concentrations in an urban area of complex terrain, Northwestern China. Atmos Res 164–165:304–317

Zhao SP, Yu Y, Xia DS, Yin DY, He JJ, Liu N, Li F (2015c) Urban particle size distributions during two contrasting dust events originating from Taklimakan and Gobi Deserts. Environ Pollut 207:107–122

Zhao SP, Yu Y, Yin DY, He JJ, Liu N, Qu JJ, Xiao JH (2016) Annual and diurnal variations of gaseous and particulate pollutants in 31 provincial capital cities based on in situ air quality monitoring data from China national environmental monitoring center. Environ Int 86:92–106

Zheng M, Salmon LG, Schauer JJ, Zeng L, Kiang CS, Zhang Y, Cass G (2005) Seasonal trends in $PM_{2.5}$ source contributions in Beijing, China. Atmos Environ 39:3967–3976

Zheng S, Wang M, Wang S, Tao Y, Shang K (2013) Short-term effects of gaseous pollutants and particulate matter on daily hospital admissions for Cardio-cerebrovascular disease in Lanzhou: evidence from a heavily polluted city in China. Int J Environ Res Public Health 10:462–477

Zheng XY, Fu YF, Yang YJ, Liu GS (2015) Impact of atmospheric circulations on aerosol distributions in autumn over eastern China: observational evidence. Atmos Chem Phys 15:12115–12138