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Particulate Matter Distributions in China during a Winter Period with Frequent Pollution Episodes (January 2013)

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ABSTRACT

Particulate matter distributions in China during January 2013 were analyzed using hourly $PM_{2.5}$ and PM_{10} concentrations from 74 cities. Five haze episodes occurred in this month. Both $PM_{2.5}$ and PM_{10} concentrations increased rapidly at the beginning of January 2013 and remained at high levels throughout the month with monthly average values of 128.7 and 184.4 μg/m^3, respectively. On January 12th, the most polluted day in this month, 13 cites were severely polluted with daily average $PM_{2.5}$ concentrations greater than 300 μg/m^3, and 18 cities were heavily polluted with daily average $PM_{2.5}$ concentrations between 200 and 300 μg/m^3. These episodes often occurred in a large spatial domain with the North China Plain as the most polluted area, including Jing-Jin-Ji area (Beijing, Tianjin, and Heibei provinces). Both $PM_{2.5}$ and PM_{10} had good correlations with ambient CO, NO_2, and SO_2 concentrations. High PM concentrations often occurred at low wind speeds and high relative humidity. In addition, PM levels in January 2013 were compared with those from other international cities.

Keywords: Haze; Pollution Episode; $PM_{2.5}$; PM_{10}; China.

INTRODUCTION

Along with fast economic development, especially the rapid increase of fossil fuel consumption, electricity generation, and number of motor vehicles, air pollution has become a severe environmental problem in China (Wang and Hao, 2012). Among different air pollutants, atmospheric particulate matter (PM), especially those with an aerodynamic diameter of 2.5 μm or less ($PM_{2.5}$), has drawn significant attention. Atmospheric PM plays an important role in urban and regional air pollution (Querol *et al.*, 2004; Shimadera *et al.*, 2013), visibility reduction (Appel *et al.*, 1985; Wang *et al.*, 2012a), and global climate change (Booth *et al.*, 2012; Randles *et al.*, 2013). They can also cause serious adverse health effects. The associations between exposures to fine particles and mortality and morbidity were widely discussed in the past decade (Pope and Dockery, 2006; Wong *et al.*, 2008; Anenberg *et al.*, 2010; Walsh, 2014). Recent studies in China reported increasing incidence rates for cardiovascular and respiratory diseases and intensive care visits due to PM exposure (Kan *et al.*, 2012; Cheng *et al.*, 2013; Yang *et al.*, 2013; Zhang *et al.*, 2013). In 2010, ambient particulate matter pollution ($PM_{2.5}$) has become the fourth leading risk factor for disability-adjusted life-years in China (Yang *et al.*, 2013).

Many studies investigated $PM_{2.5}$ pollution in megacities or regions in China, such as Beijing (He *et al.*, 2001), Shanghai (Wang *et al.*, 2006), the Yangtze River Delta Region (Wang *et al.*, 2012b), and the Pearl River Delta Region (Peng *et al.*, 2011). Since PM pollution in China is frequently occurring as regional events, it is important to characterize nation-wide $PM_{2.5}$ concentrations simultaneously. A few studies have analyzed $PM_{2.5}$ pollution in a large spatial domain in China based on literature reviews (Yang *et al.*, 2011a), ground level $PM_{2.5}$ measurements in 14 cities (Cao *et al.*, 2012), aerosol optical depth (AOD) data from ground sites (Wang *et al.*, 2011) or satellite observations (He *et al.*, 2012). However, these studies have limitations when characterizing nation-wide PM pollution levels, e.g., lack of data from well compared measuring methods, insufficient number of sites, high uncertainties when deriving $PM_{2.5}$ concentrations from AOD data.

Since June 2000, the Ministry of Environmental Protection of China (MEP) started to publish a daily air pollutant index (API), an integrated index calculated using daily concentrations of SO_2, NO_2, and PM_{10} (particulate matter with an aerodynamic diameter of 10 μm or less). API data has been used to estimate PM_{10} concentrations in 86 Chinese cities and to analyze the long-term variation of PM_{10} across China (Yang, 2009; Qu *et al.*, 2010; Cheng *et al.*, 2013). In

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2012, China released a new ambient air quality standard including $PM_{2.5}$ as a pollutant for the first time (details are given in the Supplementary Information (SI)). Following that, 74 Chinese cities including Beijing and Shanghai started to monitor hourly concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO, and O_3. Since January 1st, 2013, these data have been available to the public online in real time. By February 2014, the number of cities increased to 190. For the first time, there are hourly $PM_{2.5}$ concentrations in many Chinese cities. The new national monitoring network was on line in time to capture the pollution characteristics in January 2013 when frequent pollution episodes occurred across China. These episodes attracted significant attention from all over the world. They also helped to promote the release of Ten Air Pollution Prevention and Control Measures by the State Council of China with the focus on $PM_{2.5}$.

The aim of this study is to characterize pollution episodes occurred in January 2013 across China. We investigated $PM_{2.5}$ and PM_{10} pollution in 74 Chinese cities during this month using the data from the national monitoring network. Spatial distributions of $PM_{2.5}$ pollution in China were examined. We compared PM levels in January with data from the last decade and with PM concentrations in other international cities during January 2013 and in the past. Correlations between PM concentrations and gaseous pollutants and meteorological parameters were also studied.

DATA SOURCES

$PM_{2.5}$, PM_{10}, NO_2, SO_2, CO, and O_3 hourly concentrations at 484 monitoring sites in 73 cities (without Beijing, the list is given in SI) from December 24th 2012 to January 31th 2013 were downloaded from the China National Urban Air Quality Real-time Publishing Platform (http://113.108.142.147:20035/emcpublish/) supported by the MEP. For a given city, data from all sites were averaged to represent its average pollution level. Data from different cities were added to this publishing platform over time. The first city was added on December 15th. By December 24th, data from 73 cities (without Beijing) had been added in this platform. On January 1st 2013, this platform was officially introduced to the public. Since the data from Beijing was not included in this platform until January 17th, data from Beijing Air Quality Automatic Monitoring System (http://zx.bjmemc.com.cn/) supported by the Beijing Municipal Environmental Monitoring Center (BMEMC) were used instead to characterize the air quality during January 2013. Since September 2012, the Beijing monitoring system publishes hourly concentrations of the above six pollutants at 35 sites including the 12 sites later used by the national publishing platform. For some reason, the hourly $PM_{2.5}$ data in Beijing from January 1st to January 12th were not collected. For this period, $PM_{2.5}$ data released by the U.S. Embassy Beijing Air Quality Monitor (http://beijing.usembassy-china.org.cn/070109air.html) were used to complement the analysis. Before the end of 2012, daily or hourly PM_{10} concentration measured by the government was not available to the public. The air pollution index (API) of Beijing from July 2000 to December 2012 was downloaded from the National Daily Air Quality Report (http://datacenter.mep.gov.cn/) supported by the MEP. Daily PM_{10} concentrations were then estimated from the API value when PM_{10} was the primary pollutant (see SI for more information). The same method has been used previously to study PM_{10} pollution in Chinese cities (e.g., Qu *et al.*, 2010; Cheng *et al.*, 2013). Together with the PM_{10} data from the BMEMC, the long-term variation of PM_{10} in Beijing from July 2000 to December 2013 can be analyzed.

The Tapered Element Oscillating Microbalance (TEOM) and the Automatic Beta Radiation Attenuation Monitor are two kinds of instruments used at these sites to measure $PM_{2.5}$ and PM_{10}. Their calibration and data quality control are supported by China National Environmental Monitoring Center. The same dataset is also used for air quality compliance practices in China. Beijing's $PM_{2.5}$ data from the monitoring system and those from the U.S. Embassy in Beijing show good agreement (Fig. S-1 in the SI).

Daily $PM_{2.5}$ and PM_{10} concentrations in London in January 2013 were downloaded from the London Air Quality Network (http://www.londonair.org.uk/london/asp/datadownload.asp). Average $PM_{2.5}$ and PM_{10} concentrations in London were determined from 26 monitoring sites with both $PM_{2.5}$ and PM_{10} data, including 12 roadside sites, 6 urban background sites, 3 curbside sites, 3 suburban sites, and 2 industrial sites. Daily $PM_{2.5}$ and PM_{10} concentrations in Los Angles (LA) in January 2013 were downloaded from the U.S. EPA Air Data site (http://www.epa.gov/airquality/airdata/ad_data.html), which publishes data for 4 sites in LA. Average concentrations in LA were determined from these sites.

Meteorological data including wind speed and relative humidity for 74 cities were downloaded from the China Meteorological Data Sharing Service System (http://cdc.cma.gov.cn/home.do) supported by China Meteorological Administration.

RESULTS AND DISCUSSION

Overview of PM Pollution in January 2013

In January 2013, severe pollution episodes with high particulate matter concentrations occurred in China. As shown in Fig. 1, both $PM_{2.5}$ and PM_{10} concentrations increased rapidly at the beginning of January 2013 and remained at a high level throughout the month. The same patterns were observed for other pollutants such as NO_2, SO_2, and CO (Fig. S-2). Five pollution episodes can be identified from the temporal profiles of these five pollutants, i.e., January 7th to 8th (peak on 8th), 10th to 16th (peak on 12th), 18th to 19th (peak on 19th), 21th to 23th (peak on 22th), and 27th to 30th (peak on 28th). As a result of these episodes, the average $PM_{2.5}$ and PM_{10} concentrations in 74 cities during January were 128.7 and 184.4 μg/m^3, respectively. For comparison, the average $PM_{2.5}$ concentrations in London and Los Angeles during the same month were 17.5 and 15.1 μg/m^3, respectively. Among these episodes, the one from 10th to 16th was the heaviest and lasted the longest. During these seven days, the average $PM_{2.5}$ concentration in 74 cities was 159.2 μg/m^3. 36 sites from 24 cities (there

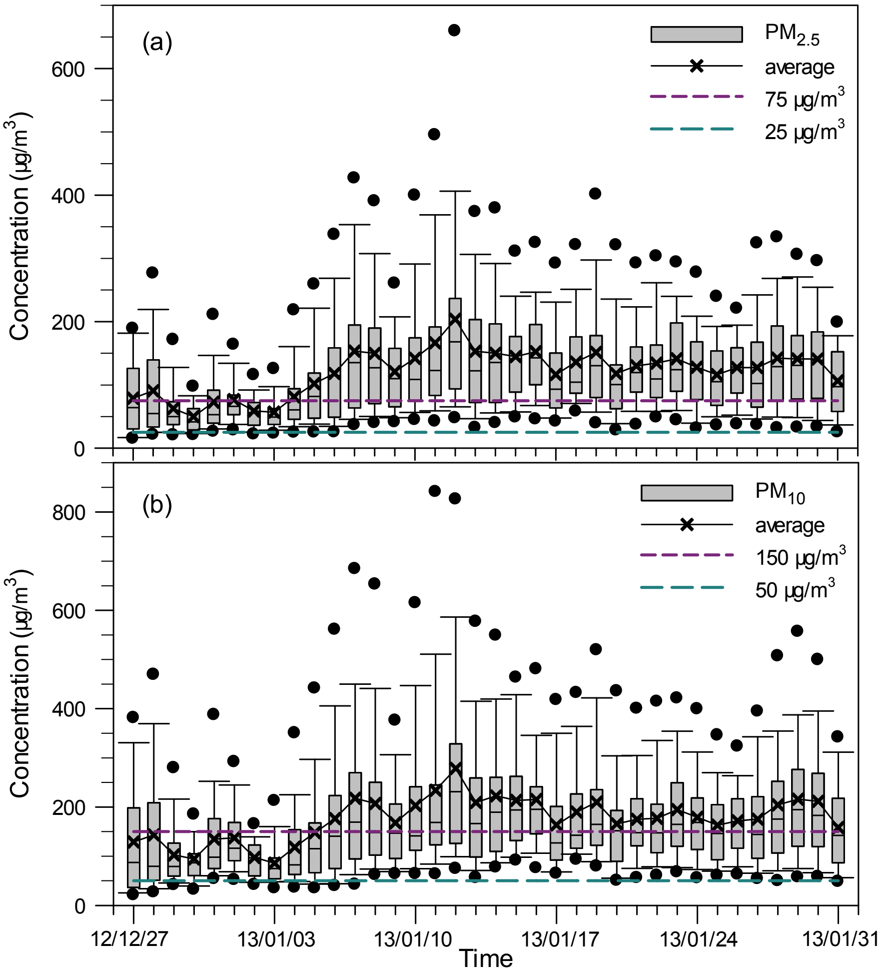

Fig. 1. Statistical results for daily average (a) $PM_{2.5}$ and (b) PM_{10} concentrations in 74 cities. The short dash lines and long dash lines are the class II values in the new China air quality standard (CNAAQS 2012) and the WHO guideline (AQG 2005), respectively. Class II values in CNAAQS apply for residential, commercial, cultural, industrial, and rural areas. In a single box plot, the central rectangle spans the first quartile to the third quartile. The segment inside the rectangle shows the median. The whiskers above and below the box show the locations of the 10th and 90th percentiles. The points above and below the whiskers show the 5th and 95th percentiles.

are 164 sites in total for these cities) reported hourly $PM_{2.5}$ concentration higher than 900 µg/m^3 and 125 monitoring sites from 43 cities reported hourly $PM_{2.5}$ concentration higher than 500 µg/m^3. On January 12th, the most polluted day in this month, 14 cities were severely polluted with daily average $PM_{2.5}$ value greater than 300 µg/m^3 and 18 cities were heavily polluted with daily average $PM_{2.5}$ concentration between 200 and 300 µg/m^3. This large-scale regional event happened in middle and eastern China. The most polluted cities were Langfang, Xingtai, Baoding, and Shijiazhuang which are all in Hebei province. Their daily $PM_{2.5}$ levels on January 12th were 718.1, 695.3, 666.8, and 654.6 µg/m^3, respectively.

$PM_{2.5}$ pollution in China showed significantly regional characteristics with the North China Plain, including Jing-Jin-Ji area (Beijing, Tianjin, and Heibei province), being the most polluted area (Fig. 2 and Table 1). Fig. 2(a) indicates that a large spatial domain in China was simultaneously experiencing severe $PM_{2.5}$ pollution on 12th January, the most polluted day during the month. Hebei was the most polluted province in China during January 2013. It is noticed that two cities (Zhangjiakou and Chengde) in the north of the Jing-Jin-Ji area reported $PM_{2.5}$ concentrations of 50–75 µg/m^3 (light green dots, Fig. 2(a)) whereas the other cities in the Jing-Jin-Ji area reported much higher $PM_{2.5}$ concentrations. Zhangjiakou and Chengde are much less developed. In addition, the Jundushan Mountain and the Yanshan Mountain prevent pollutants transporting from other developed area including Beijing. According to the monthly average $PM_{2.5}$ concentration in January 2013 (Fig. 2(b) and Table S1), the most polluted six cities (Xingtai, Shijiazhuang, Handan, Baoding, Hengshui, and Langfang) and the 8th most polluted city (Tangshan) are all in Hebei province. The 7th, 9th, and 10th most polluted cities are Jinan (Shandong province), Zhengzhou (Henan province), and Xi'an (Shaanxi province), respectively. Similar regional distributions of air pollution in China was reported previously using other methods. A study based on visibility variation from 1975 to 2005 identified

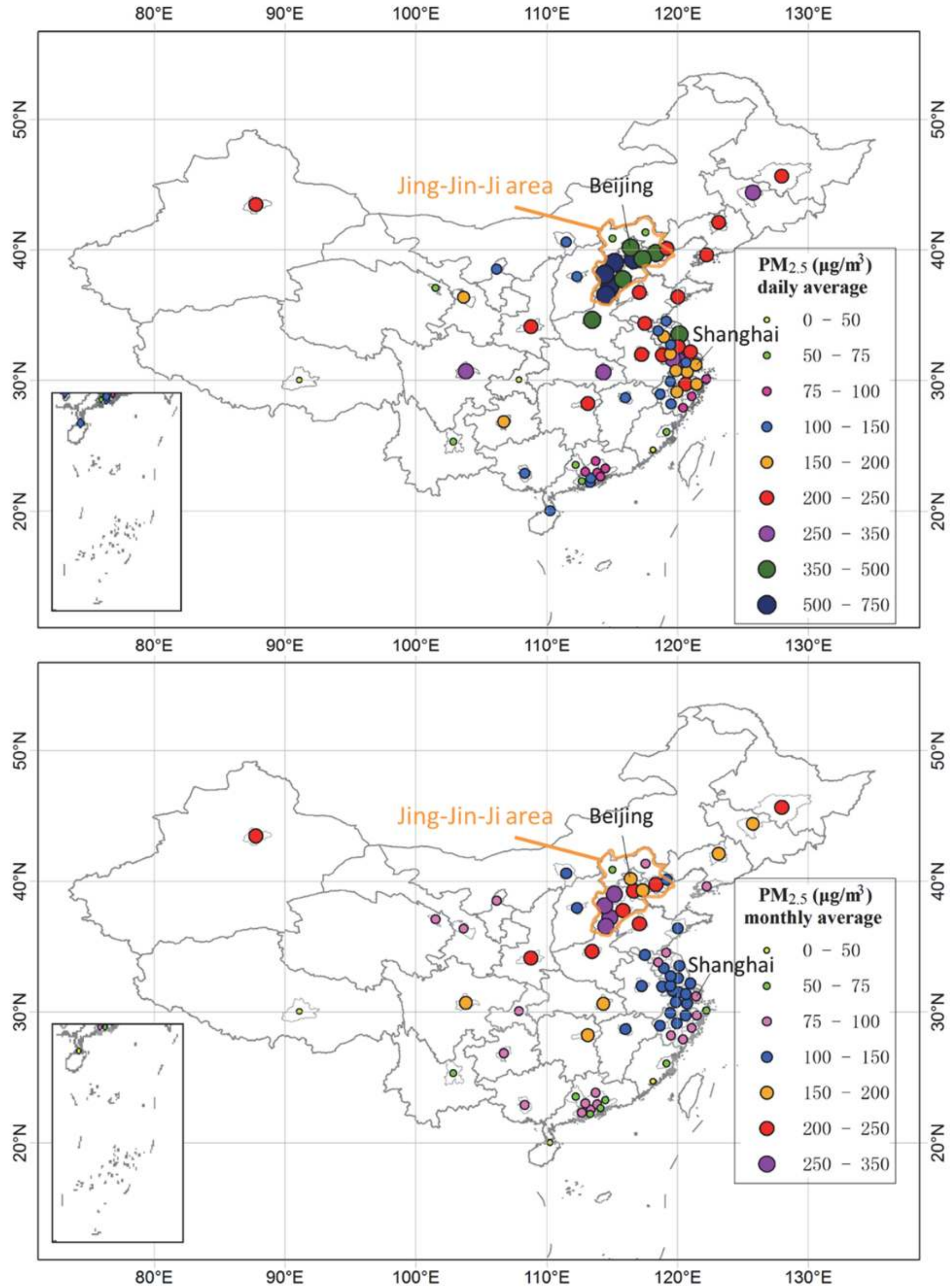

Fig. 2. $PM_{2.5}$ concentrations in 74 Chinese cities: (a) daily average on 12th January 2013 and (b) monthly average in January 2013. Locations of 74 cities and their monthly average concentrations of six pollutants are given in Table S-1.

Table 1. Monthly average concentrations of six pollutants at different regions in China.

	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	SO_2 (μg/m^3)	NO_2 (μg/m^3)	CO (mg/m^3)	O_3 (μg/m^3)
Jing-Jin-Ji Area	195	303	161	78	3.6	27
North China Plain*	168	254	124	69	3.0	32
Yangtze River Detla	115	152	50	66	1.6	31
Pearl River Detla	74	102	29	57	1.4	47
Other area	117	164	74	55	1.9	34

* The Jing-Jin-Ji Area is included in North China Plain.

middle and eastern China as one of the four major haze areas in China (Zhang *et al.*, 2012). PM_{10} measurement data from the national monitoring network (Qu *et al.*, 2010; Cheng *et al.*, 2013; Wang *et al.*, 2013) and modeled results on PM_{10} and $PM_{2.5}$ (Chen *et al.*, 2013; Shimadera *et al.*, 2013) reported consistent results with this study based on $PM_{2.5}$ data from the national monitoring network, i.e., PM pollution in middle and eastern China is severer than in any other area in China. $PM_{2.5}$ concentration is typically highest in winter due to the unfavorable meteorological conditions and emissions from winter heating in northern China (He *et al.*, 2001; Duan *et al.*, 2006; Jahn *et al.*, 2011; Zhao *et al.*, 2011).

The average $PM_{2.5}$ concentration in 14 Chinese cities during January 2003 was found to be 161.7 µg/m^3 (Cao *et al.*, 2012), which is 26% higher than that of 74 Chinese cities in January 2013. However, significantly more public attention occurred during the pollution episodes in January 2013 due to their large spatial coverage, high frequency, long lasting time, and high peak hourly $PM_{2.5}$ concentrations. In January 2013, $PM_{2.5}$ concentrations in 74 cities frequently violated air quality standards. Daily and annual average $PM_{2.5}$ concentrations were included in the new China National Ambient Air Quality Standard (CNAAQS, 2012) and the World Health Organization Air Quality Guideline (AQG, 2005). On January 12th, the most polluted day in this month, 63 of the 74 cities had daily average $PM_{2.5}$ concentrations greater than 75 µg/m^3 (CNNAQS, 2012). On January 3rd, the cleanest day in this month, 61 of the 74 cities had daily average $PM_{2.5}$ concentrations meeting CNAAQS (< 75 µg/m^3), while only 5 cities met AQG (< 25 µg/m^3). From January 5th to 31st, the percentage of cities with daily $PM_{2.5}$ concentration satisfying CNNAQS was in the range of 12% to 39%. During the same period, the number of cities with daily $PM_{2.5}$ concentrations meeting AQG was no more than 3. During the severest episode (Jan. 10th to 16th), over 70% of the 74 cities did not meet CNNAQS. Hourly $PM_{2.5}$ concentrations even reached 1000 µg/m^3 in eleven heavily polluted cities including Shijiazhuang, Xingtai, Xi'an, and Wuhan.

Comparison with Other International Cities and Historical Data

Beginning in the last century, air pollution has affected urban environments all over the world. The most well-known air pollution events include the 1952 London Great Smog and the Los Angles Photochemical Smog. After many years of battling air pollution, these two cities have improved their air quality significantly. While daily $PM_{2.5}$ and PM_{10} concentrations in London and LA were no more than 35 µg/m^3 in January 2013, those in Chinese cities were much higher. Fig. 3 shows monthly average PM concentrations in Beijing, Shanghai, and Xingtai (the most polluted city in January 2013) comparing to London and Los Angles. $PM_{2.5}$ in Xingtai, 324.2 µg/m^3, was 18 and 21 times higher than those in London and LA, respectively. $PM_{2.5}$ in Beijing was 9 and 11 times higher than those in London and LA, respectively, while $PM_{2.5}$ in Beijing in 1992–1993 was only 5 times higher than that in Los Angeles in 1986 (Zhang and Friedlander 2000). In January 2013, Shanghai had lower PM pollution than Beijing. Its monthly

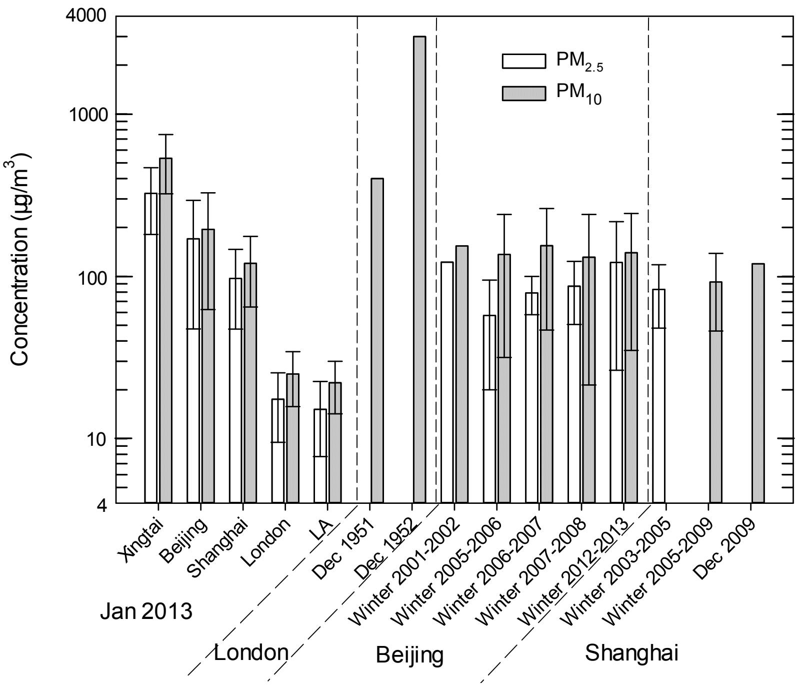

Fig. 3. PM concentrations in different cities during different periods. The whiskers show the standard deviation of the data. Beijing's PM concentrations for January 2013 and winter of 2012 to 2013 were calculated based on data sets described in previous section. Additional data sources include Beijing: winter 2001–2002 (Duan *et al.*, 2006) and winter 2005–2008 (Yu *et al.*, 2011); Shanghai: winter 2003–2005 (Wang *et al.*, 2006), winter 2005–2009 (Zhao *et al.*, 2011), and Dec 2009 (Wang *et al.*, 2012b).

average $PM_{2.5}$ concentration was 5 and 6 times higher than those in London and LA, respectively.

Historical PM data in London, Beijing, and Shanghai were also included in Fig. 3. The maximum daily PM_{10} concentration during the London Great Smog was estimated to be 4460 µg/m^3 and the monthly average PM_{10} level in December 1952 was approximately 3000 µg/m^3 (see SI for more information and references). PM_{10} concentrations in Chinese cities were much lower than this level. However, it should be noted that the PM_{10} level in London in December 1951, one year prior to the Great Smog event, was only 430 µg/m^3 (Authority, 2002), approximately 2 times higher than PM_{10} levels in Beijing in January 2013. PM_{10} concentration in Xingtai was 534.8 µg/m^3, even higher than that in London in December 1951. Compared to previously reported Beijing winter PM levels in the last decade, $PM_{2.5}$ in Beijing in January 2013 was higher. Continuous $PM_{2.5}$ monitoring data collected since 2012 shows a significantly increasing trend with an average increasing rate of 1.48 µg/(m^3·month) (Fig. 4(a)). $PM_{2.5}$ in Shanghai in January 2013 was at a comparable level with those in other periods. This indicates that in Chinese cities like Beijing and Shanghai $PM_{2.5}$ pollution did not improve during the past decade.

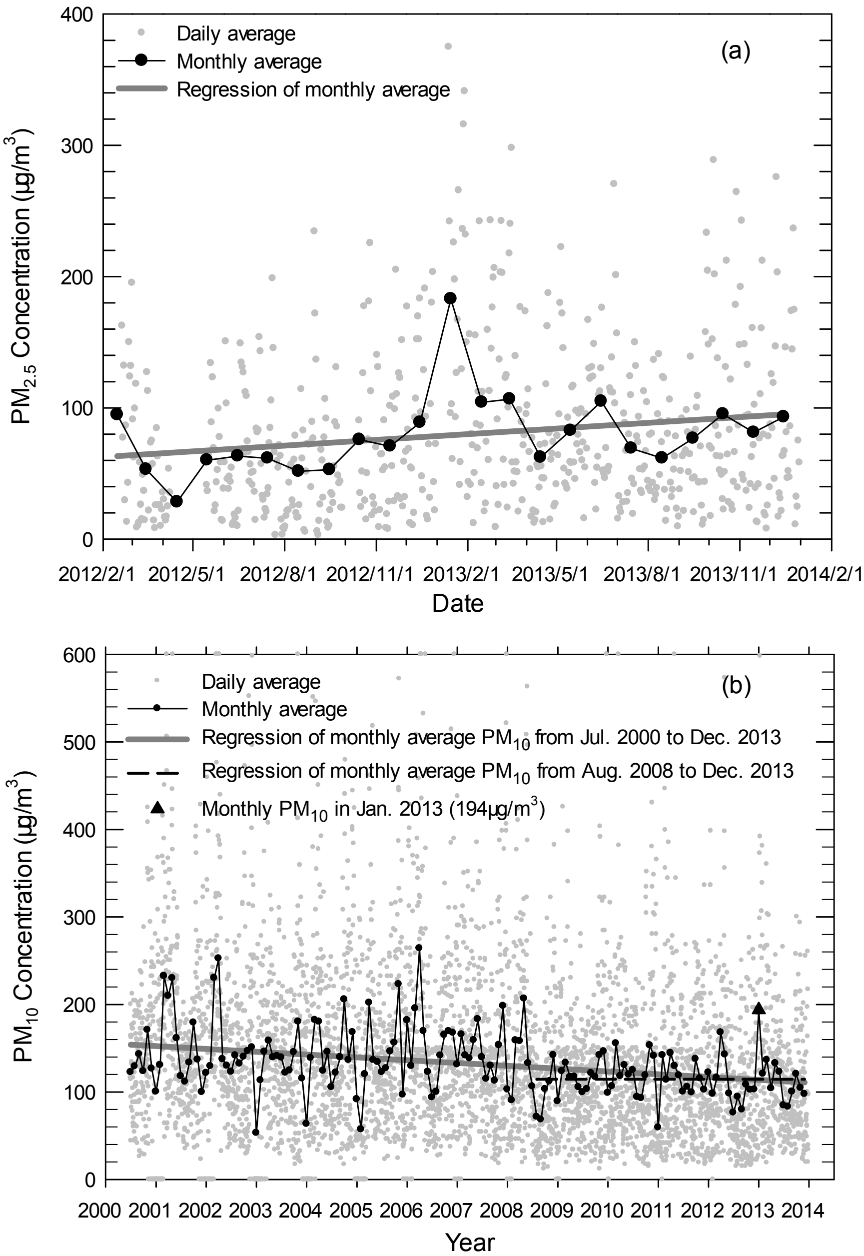

Fig. 4. (a) $PM_{2.5}$ concentrations in Beijing from February 2012 to December 2013. Before October 2012, only $PM_{2.5}$ concentrations at Chegongzhuang site are available. After that, $PM_{2.5}$ concentrations were averaged from all 35 sites in Beijing. (b) PM_{10} concentrations in Beijing from July 2000 to December 2013. Before 2013, the PM_{10} levels were calculated using the API data.

To further explore PM_{10} pollution trends, we took Beijing as an example. Long-term PM_{10} concentrations from the national monitoring network is shown in Fig. 4(b). During the period from July 2000 to December 2013, January 2013 was not the month with the highest PM_{10} concentration. There are twelve months with average PM_{10} concentrations higher than January 2013. Three of them occurred in the winter or autumn season, i.e., October 2004, November 2005, and December 2007, while others occurred in March, April, or May which were mainly due to sand storms. The last time the monthly average PM_{10} level was higher than January 2013 is May 2008. Linear regression from July 2000 to December 2013 indicates that PM_{10} in Beijing was slowly decreasing at a rate of 0.27 $\mu g/(m^3 \cdot month)$ with fluctuations. This is attributed to control measures for ensuring good air quality during the 2008 Beijing Olympic Games (Wang *et al.*, 2010; Yang *et al.*, 2011b). Accordingly, PM_{10} concentrations in the summer of 2008 were at a relatively low level. However, if the linear regression is taken from August 2008 to December 2013, it shows that PM_{10} in Beijing was relatively stable during this period. The monthly average PM_{10} concentration in January 2013 was 194 ± 132 $\mu g/m^3$, while the highest level in 2007 was 198 ± 134 $\mu g/m^3$ which occurred in December. Similar variations in the trend occurred in other Chinese cities. Based on the same PM_{10} data set from the national monitoring network, it was found that most of the 86 Chinese cities had annual PM_{10} concentrations continually decreasing from 2001 to 2011 (Cheng *et al.*, 2013; Wang *et al.*, 2013). Wang *et al.* (2013) pointed out that the reduction of annual PM_{10} concentration from 2001 to 2005 was relatively fast. While after 2006, the decreasing rate slowed down and even showed a slight increase in 2010 compared to 2009. In addition, some Chinese cities favored by tourists and cities with rapid economically development showed an opposite increasing trend.

Analysis of Pollution Episodes

During smog events in China, PM concentrations often increased together with gaseous pollutants such as CO, SO_2, and NO_2. The latter two also serve as the main gaseous precursors for secondary inorganic aerosol. For instance, sulfate, nitrate, and ammonium together accounted for approximately 55% of $PM_{2.5}$ mass concentration in Beijing during January 8^{th} to 14^{th}, 2013 (Cao *et al.*, 2014). PM showed positive correlations with SO_2, NO_2, and CO based on hourly averaged data (Table 2), indicating that Chinese cities have significant emissions from both coal combustion and vehicles. For Beijing in which most large industries have been moved out and the remaining ones face tight standards, PM had much stronger correlation with CO and NO_2 than with SO_2. Vehicle emissions in Beijing were reported to contribute approximately 71%–85% of ambient CO concentrations (Hao *et al.*, 2000) and 67%–71% of ambient NO_x concentrations (Wang *et al.*, 2009). These findings indicate that in Beijing vehicle emissions play an important role in pollution episodes. A recent study reported that the coexistence of NO_x and SO_2 leads to rapid conversion of SO_2 to sulfate, the decrease of SO_2 and the increase of PM during episodes in January 2013 (He *et al.*, 2014), which may also explain the stronger correlations between PM and NO_2. It should be noted that the correlation between PM and gaseous precursors (SO_2 and NO_2) is affected by relative humidity (Table 3). High relative humidity enhances the aqueous phase conversion of gas precursors. Therefore, the fitted slope between PM and precursors decreased remarkably with increasing relative humidity. In contrast, the correlation between $PM_{2.5}$ and CO was not affected by relative humidity.

Wind speed and relative humidity are two meteorological parameters that show positive correlation with PM concentrations. We took Beijing as an example again and

Table 2. Correlation coefficients (r) for five pollutants based on hourly concentrations in January 2013.

	74 cities				Beijing			
	PM_{10}	SO_2	NO_2	CO	PM_{10}	SO_2	NO_2	CO
$PM_{2.5}$	0.921	0.585	0.663	0.738	0.986	0.609	0.900	0.936
PM_{10}		0.652	0.647	0.764		0.577	0.899	0.913

Table 3. Correlation coefficients (r) between $PM_{2.5}$ and gaseous pollutants at different relative humidities (RH).

	RH	Beijing		Tianjin		Shijiazhuang		Shanghai	
		r^2	slope	r^2	slope	r^2	slope	r^2	slope
NO_2-$PM_{2.5}$	< 50%	0.695	0.343	0.801	0.388	0.821	0.208	0.639	0.324
	50–80%	0.830	0.233	0.758	0.247	0.568	0.153	0.490	0.318
	80–90%	0.622	0.201	0.824	0.213	0.333	0.108	0.496	0.282
	> 90%	0.813	0.183	0.762	0.205			0.496	0.249
SO_2-$PM_{2.5}$	< 50%	0.624	0.372	0.898	1.219	0.755	0.878	0.606	0.288
	50–80%	0.586	0.261	0.517	0.505	0.278	0.329	0.522	0.276
	80–90%	0.345	0.218	0.616	0.461	0.046	0.191	0.582	0.212
	> 90%	0.193	0.087	0.361	0.390			0.539	0.246
CO-$PM_{2.5}$	< 50%	0.836	0.013	0.829	0.019	0.925	0.014	0.837	0.006
	50–80%	0.874	0.015	0.690	0.013	0.598	0.010	0.731	0.007
	80–90%	0.615	0.013	0.783	0.014	0.236	0.006	0.657	0.010
	> 90%	0.802	0.011	0.528	0.014			0.504	0.010

presented daily $PM_{2.5}$ and PM_{10} concentrations for January 2013 and other winter periods as a function of daily average wind speed and relative humidity (Fig. 5). Both $PM_{2.5}$ and PM_{10} increase with decreasing wind speed and increasing relative humidity. Low wind speed means unfavorable atmospheric dilution and dispersion conditions that lead to the accumulation of pollutants. High relative humidity can contribute to particle growth through water uptake and aqueous redox chemistry (e.g., the oxidation of sulfur dioxide to sulfate). The monthly average wind speed in January 2013 was the lowest in the past ten years while the monthly average relative humidity in January 2013 was the highest in the past ten years. During the five pollution episodes in January 2013, the average $PM_{2.5}$ concentration was 147.9 µg/m^3 with an average wind speed of 1.90 m/s and an average relative humidity of 72.9%. In February 2013, $PM_{2.5}$ concentrations were dramatically reduced to 104 µg/m^3 when higher wind speeds (2.27 m/s) and lower relative humidities (50.9%) occurred. Similar relationships between PM and these two meteorological conditions had been previously observed (e.g., He *et al.*, 2001; Sun *et al.*, 2013). At high wind speeds, high PM concentrations, especially PM_{10}, may also occur which is mainly due to fugitive dust. Wind-blown dust can increase PM_{10} concentrations significantly (Feng *et al.*, 2011). For instance, during the dust storm in Beijing on April 6th, 2000, PM_{10} concentrations reached a level as high as 1500 µg/m^3 and the storm lasted for 14 hours (Xie *et al.*, 2005). Dust was reported to account for ~20% of $PM_{2.5}$ in Chinese cities (Cao *et al.*, 2012). Its contribution to $PM_{2.5}$ increased to ~42% when frequent sandstorms invaded Beijing in April 2001 (Yang *et al.*, 2011a).

The pollution processes in most middle and east China locations are similar during wintertime. Meteorological conditions with low wind speeds, high relative humidities, and low temperatures occur frequently. Domestic heating in winter generates much more atmospheric pollutants than in summertime. The absence of wind leads to the accumulation of pollutants. The temperature inversion with a warm air layer overlying a cold air at ground level also prevents pollutants from good dispersion. Both gas phase and aqueous phase chemical reactions form secondary pollutants. Accordingly, concentrations of both primary and secondary pollutants increase significantly. During haze episodes, PM concentrations typically increase by a factor of 3–5. On January 3rd, the cleanest day in January 2013, average $PM_{2.5}$ and PM_{10} concentrations for 74 cities were 59.9 and 86.5 µg/m^3, respectively, while on January 12th, they increased by a factor of 3.4 and 3.2, respectively. This is consistent with previous reports that $PM_{2.5}$ concentrations can be 3 times higher on smoggy days than clear days in north China (Li *et al.*, 2011). However, these pollutants were usually cleared out by a strong wind and/or precipitation.

CONCLUSION

We characterized PM pollution in China during January 2013 using data from the national monitoring network. Five pollution episodes occurred during this month. Monthly average $PM_{2.5}$ and PM_{10} concentrations in 74 cities were 128.7 and 184.4 µg/m^3, respectively. On January 12th, the most polluted day in this month, 13 cites were severely polluted and 18 cities were heavily polluted. $PM_{2.5}$ pollution in China showed significant regional characteristics with the North China Plain, include Jing-Jin-Ji area (Beijing,

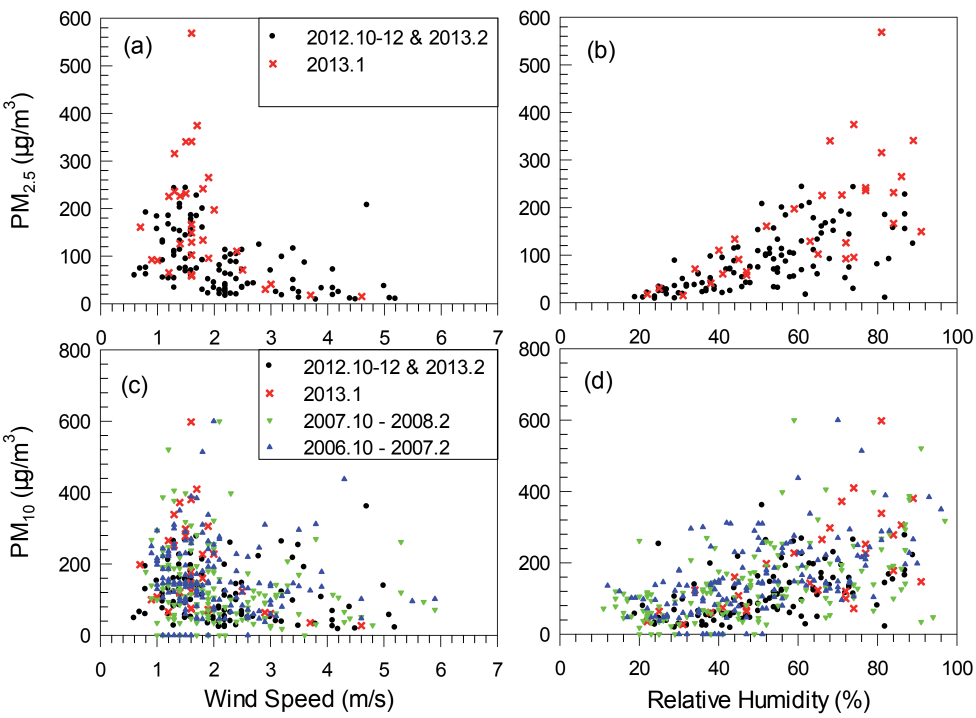

Fig. 5. The correlations between (a) $PM_{2.5}$ and wind speed, (b) $PM_{2.5}$ and relative humidity, (c) PM_{10} and wind speed, (d) PM_{10} and relative. Daily average data in Beijing for different months were shown here as an example.

Tianjin, and Heibei province), being the most polluted area. $PM_{2.5}$ and PM_{10} are strongly correlated with NO_2, SO_2, and CO, indicating that both vehicle emissions and coal combustion contribute to these pollution episodes. This is consistent with the high nitrate and sulfate observed in fine particles. High $PM_{2.5}$ and PM_{10} concentrations often occurred at low wind speed and high relative humidity. PM pollution levels in Chinese cities were lower than that during the 1952 London Great Smog, but were much higher than London and LA during January 2013. PM_{10} concentrations in Xingtai, the most polluted city in January 2013, were even higher than that in London in December 1951, one year prior to the London Great Smog. Though average PM_{10} concentration in China showed a decreasing trend during the last decade, the rate of decrease was slowing down. $PM_{2.5}$ pollution did not improve in cities like Beijing and Shanghai during the last decade. In Beijing, $PM_{2.5}$ even showed a tendency to increase. With the release of the Ten Air Pollution Prevention and Control Measures by the State Council of China in 2013, stricter air pollution control measures will be implemented and lower $PM_{2.5}$ level might be anticipated in the future.

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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Supplementary Information

"Particulate Matter Distributions in China during a Winter Period with Frequent Pollution Episodes (January 2013)"

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1. Comparison of $PM_{2.5}$ data from the BMEMC and those from the U.S. Embassy in Beijing

As shown in Figure S-1, daily $PM_{2.5}$ concentrations reported by the U.S. Embassy in Beijing correlates very well with those published by Beijing Municipal Environmental monitoring Center (BMEMC). $PM_{2.5}$ daily concentration from the U.S. Embassy is ~25% higher than that from BMEMC. This is possibly attributed to sampling locations. The former was measured at an urban site inside the U.S. Embassy while data from BMEMC is the average of 35 sites including urban, rural, roadside, and background sites. The slope is used to calculate the $PM_{2.5}$ level in Beijing during January 1st to January 12th.

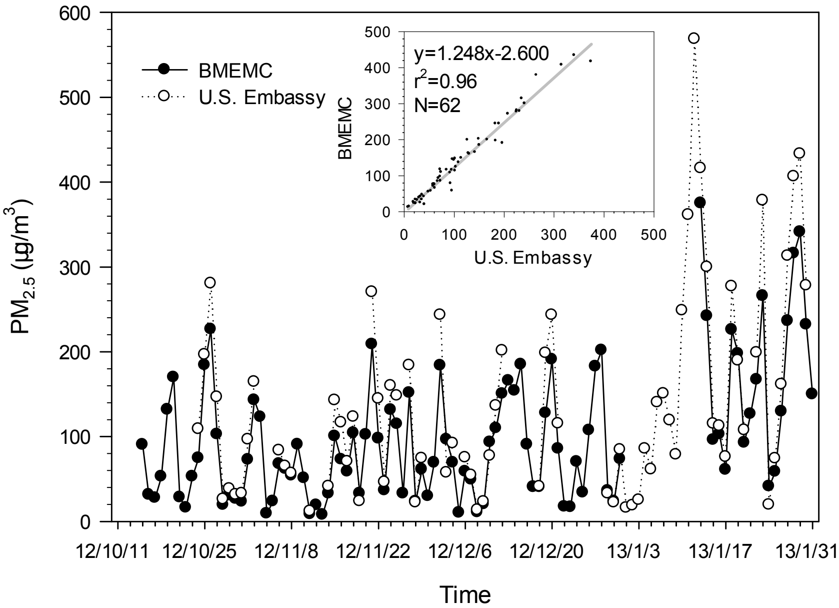

Figure S-1. Comparison of daily $PM_{2.5}$ from the BMEMC and those the U.S. Embassy in Beijing

2. Estimation of daily PM_{10} from the API value

For the days when PM_{10} was reported as the primary pollutant, daily PM_{10} concentrations were derived from the API using the following equation:

$$C = \left[(I - I_{low})/(I_{high} - I_{low})\right] \times \left((C_{high} - C_{low})\right) + C_{low} \qquad \text{(S-1)}$$

where C is the concentration of PM_{10}, I is the reported API values. I_{low} and I_{high} represent API grading limits that are lower and larger than I, respectively; C_{high} and C_{low} denote the PM_{10} concentrations corresponding to I_{high} and I_{low}, respectively.

3. List of 74 national cities and their pollutant concentrations

Table S-1. List of 74 national cities in this study and their monthly average concentrations of six pollutants in January 2013. NCP(JJJ) means the Jing-Jin-Ji (Beijing, Tianjin, and Heibei provience) area. It is included in the NCP (North China Plain). YRD and PRD are the Yangtze River Delta and the Pearl River Delta, respectively.

City	Province	latitude	longitude	$PM_{2.5}$ μg/m³	PM_{10} μg/m³	SO_2 μg/m³	NO_2 μg/m³	CO mg/m³	O_3 μg/m³	Area	Number of sites
Beijing	Beijing	39.9	116.4	169	194	73	95	3.9	14	NCP(JJJ)	35
Baoding	Hebei	38.9	115.5	271	371	197	93	3.9	51	NCP(JJJ)	5
Cangzhou	Hebei	38.3	116.8	78	177	102	33	2.8	59	NCP(JJJ)	1
Chengde	Hebei	41.0	117.9	85	138	80	60	1.9	22	NCP(JJJ)	5
Handan	Hebei	36.6	114.5	278	424	220	90	4.4	13	NCP(JJJ)	4
Hengshui	Hebei	37.7	115.7	236	413	177	80	3.5	13	NCP(JJJ)	3
Langfang	Hebei	39.5	116.7	233	309	122	76	4.2	15	NCP(JJJ)	3
Qinhuangdao	Hebei	39.9	119.6	112	194	108	60	3.0	34	NCP(JJJ)	5
Shijiazhuang	Hebei	38.0	114.5	302	541	279	99	4.9	15	NCP(JJJ)	6
Tangshan	Hebei	39.6	118.2	223	290	196	93	4.6	29	NCP(JJJ)	6
Xingtai	Hebei	37.1	114.5	324	535	269	113	5.2	20	NCP(JJJ)	4
Zhangjiakou	Hebei	40.7	114.5	65	124	124	48	1.4	48	NCP(JJJ)	5
Tianjin	Tianjin	39.1	117.3	157	228	148	74	3.7	20	NCP(JJJ)	15
Zhengzhou	Henan	34.8	113.6	215	321	99	66	5.2	22	NCP	9
Jinan	Shandong	36.6	117.0	228	352	220	105	3.1	21	NCP	8
Qingdao	Shandong	36.1	120.4	137	183	132	64	2.0	45	NCP	9
Huaian	Jiangsu	33.6	119.0	138	194	35	45	1.4	30	NCP	5
Lianyungang	Jiangsu	34.7	119.3	88	134	75	61	1.8	32	NCP	4
Suqian	Jiangsu	34.0	118.3	89	108	54	42	1.6	69	NCP	4
Xuzhou	Jiangsu	34.3	117.2	122	186	92	67	2.9	23	NCP	7
Yancheng	Jiangsu	33.4	120.2	128	143	39	44	1.2	67	NCP	4

Nantong	Jiangsu	32.0	120.9	118	180	40	44	1.3	36	YRD,NCP	5
Taizhou	Jiangsu	32.4	119.9	134	200	53	51	1.6	33	YRD,NCP	3
Yangzhou	Jiangsu	32.4	119.4	109	151	40	55	1.5	28	YRD,NCP	4
Changzhou	Jiangsu	31.8	120.0	139	174	65	95	1.6	30	YRD	5
Nanjing	Jiangsu	32.1	118.8	136	209	74	73	2.0	28	YRD	9
Suzhou	Jiangsu	31.3	120.6	111	127	48	73	1.3	25	YRD	6
Wuxi	Jiangsu	31.6	120.3	142	156	63	60	2.3	36	YRD	6
Zhenjiang	Jiangsu	32.2	119.5	124	173	45	60	1.2	31	YRD	4
Shanghai	Shanghai	30.7	121.2	97	120	36	66	1.3	37	YRD	10
Hangzhou	Zhejiang	30.2	120.1	113	136	44	72	1.6	21	YRD	11
Huzhou	Zhejiang	30.9	120.1	134	173	37	79	1.6	24	YRD	3
Jiaxing	Zhejiang	30.8	120.7	133	142	75	99	3.2	40	YRD	3
Ningbo	Zhejiang	29.9	121.6	87	125	48	64	1.4	30	YRD	8
Shaoxing	Zhejiang	30.0	120.6	128	174	82	75	1.2	35	YRD	2
Taizhou	Zhejiang	28.6	121.4	84	119	29	57	1.5	33	YRD	3
Zhoushan	Zhejiang	30.0	122.2	51	73	19	39	0.7	34	YRD	3
Dongwan	Guangdong	23.0	113.8	80	102	41	58	1.3	48	PRD	5
Foshan	Guangdong	23.0	113.1	82	121	42	68	1.6	33	PRD	8
Guangzhou	Guangdong	23.2	113.4	82	102	23	77	1.2	31	PRD	11
Huizhou	Guangdong	23.0	114.4	65	97	19	36	1.2	61	PRD	5
Jiangmen	Guangdong	22.6	113.1	80	110	37	51	1.7	35	PRD	4
Shenzhen	Guangdong	22.6	114.2	62	95	17	53	1.5	65	PRD	11
Zhaoqing	Guangdong	23.1	112.5	65	84	29	46	1.5	43	PRD	4
Zhongshan	Guangdong	22.5	113.4	82	110	32	68	1.2	40	PRD	4
Zhuhai	Guangdong	22.3	113.5	71	97	22	59	1.3	66	PRD	4
Hefei	Anhui	31.8	117.2	142	173	41	50	1.6	28	other	10
Chongqing	Chongqing	29.6	106.5	79	167	43	43	1.1	28	other	15
Fuzhou	Fujian	26.1	119.3	52	73	8.7	43	1.0	23	other	6
Xiamen	Fujian	24.6	118.1	45	68	24	48	0.9	57	other	4
Kanzhou	Gansu	36.1	103.8	97	141	79	51	2.3	16	other	3
Nanning	Guangxi	22.8	108.3	97	123	26	47	1.5	33	other	7
Guiyang	Guizhou	26.6	106.8	86	119	82	40	1.1	32	other	10
Haikou	Hainan	20.0	110.4	48	69	8.8	24	1.0	61	other	5
Harbin	Heilongjiang	45.7	126.6	205	264	78	81	1.9	21	other	12
Wuhan	Hubei	30.6	114.2	189	214	68	81	2.0	27	other	10
Changsha	Hunan	28.2	113.0	164	179	50	73	2.0	20	other	10
Hohhot	Inner Mongolia	40.8	111.7	118	182	137	47	3.7	33	other	8
Nanchang	Jiangxi	28.7	115.9	121	181	78	57	1.5	39	other	7
Changchun	Jilin	43.8	125.3	190	249	107	69	1.9	50	other	10
Dalian	Liaoning	38.6	121.6	95	136	106	43	1.2	33	other	10
Shenyang	Liaoning	41.8	123.5	192	257	244	49	2.9	21	other	11

Yinchuan	Ningxia	38.5	106.1	99	177	196	54	2.2	32	other	6
Xining	Qinghai	36.7	101.7	98	171	73	43	2.8	18	other	4
Xi'an	Shaanxi	34.3	109.0	210	301	114	77	3.8	27	other	13
Taiyuan	Shanxi	37.9	112.5	122	184	128	40	1.7	23	other	9
Chengdu	Sichuan	30.7	104.0	168	224	53	74	2.0	28	other	8
Lasa	Tibet	29.6	91.1	38	84	70	28	1.5	87	other	6
Urumqi	Xinjiang	43.9	87.6	206	289	85	95	4.6	9.7	other	7
Kunming	Yunnan	25.0	102.7	57	113	35	48	1.8	39	other	7
Jinhua	Zhejiang	29.1	119.7	107	147	56	65	1.8	24	other	3
Lishui	Zhejiang	28.4	119.9	81	96	49	46	1.1	59	other	3
Quzhou	Zhejiang	29.0	118.9	113	147	47	51	1.2	26	other	3
Wenzhou	Zhejiang	28.0	120.7	91	131	24	66	1.0	21	other	4

4. The new China National Ambient Air Quality Standard (CNAAQS 2012) and the World Health Organization Air Quality Guideline (AQG 2005)

Table S-2 class II values for the six pollutants in CNAAQS 2012 and AQG 2005. Except for those specified, daily average concentrations are given. Class II values in CNAAQS 2012 apply for residential, commercial, cultural, industrial, and rural areas.

	CNAAQS 2012	AQG 2005
$PM_{2.5}$	75μg/m^3	25μg/m^3
PM_{10}	150μg/m^3	50μg/m^3
SO_2	150μg/m^3	20μg/m^3
NO_2	80μg/m^3	40μg/m^3 (annual mean)
CO	4mg/m^3	--
O_3	160μg/m^3 (daily maximum 8-hour O_3 concentrations)	50μg/m^3 (8-hour average)

5. Gaseous pollutants in January 2013

Figure S-2 shows the variations of four gaseous pollutants (SO_2, NO_2, CO, and O_3) during January 2013. SO_2, NO_2, and CO had similar patterns with $PM_{2.5}$ and PM_{10}. They increased rapidly at the beginning of January 2013 and remained at a high level throughout the month. However, O_3 had a different variation. As shown in Figure S-2(d), daily maximum 8-hr O_3 concentrations decreased sharply on January 5th and slowly increased throughout the month.

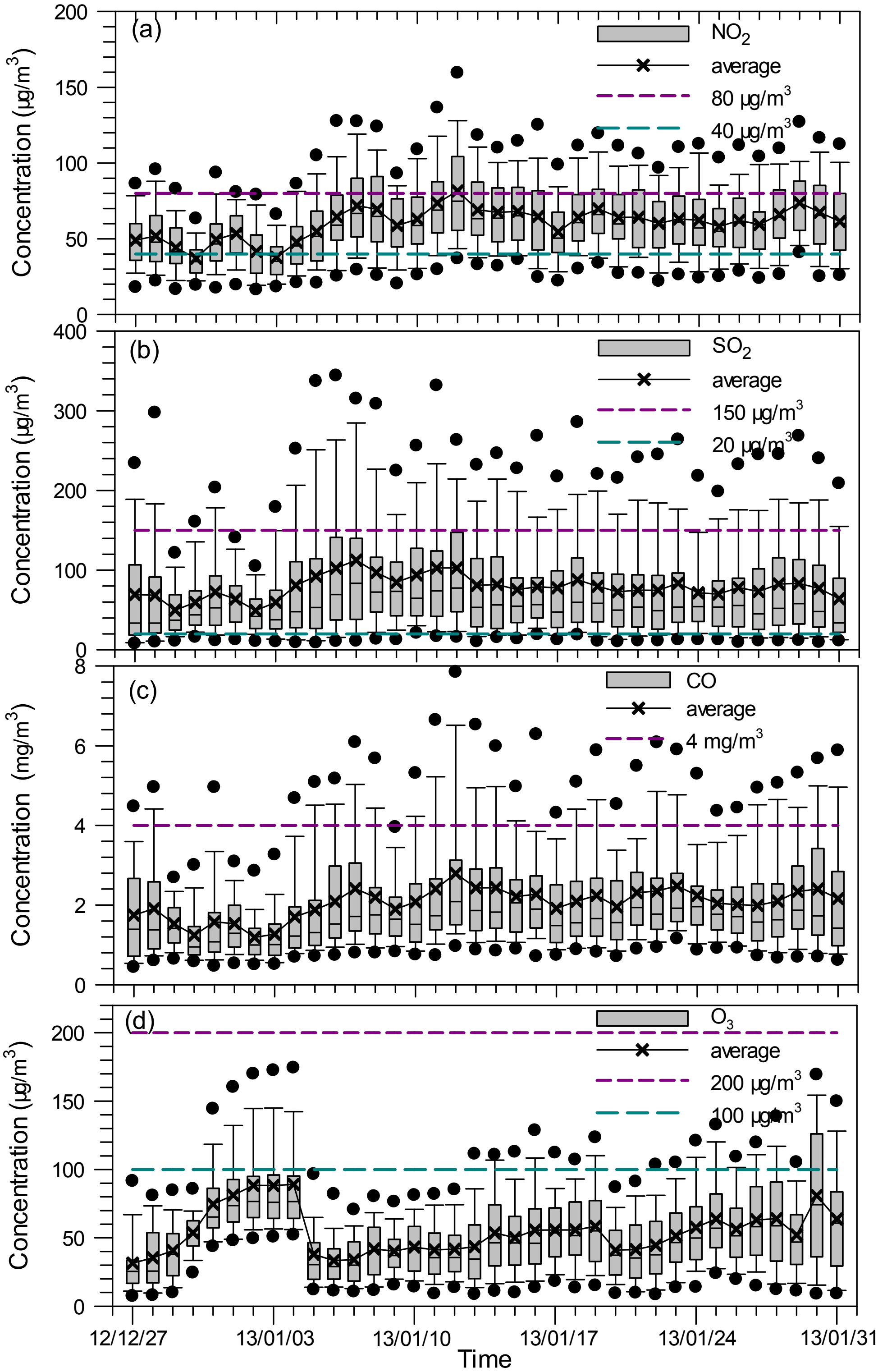

Figure S-2. Statistical result of daily average (a) NO_2, (b) SO_2, (c) CO, and (d) daily maximum 8-hr O_3 concentrations in 74 cities. The short dash lines and long dash lines are the class II values for the new China air quality standard (CNAAQS 2012) and the WHO guideline (AQG 2005), respectively. In a single box plot, the central rectangle spans the first quartile to the third quartile. The segment inside the rectangle shows the median. The whiskers above and below the box show the locations of the 10^{th} and 90^{th} percentiles. The points above and below the whiskers show the 5^{th} and 95^{th} percentiles.

6. PM_{10} Concentration during 1952 London Great Smog

At the time of London Great Smog, the concentration of black smoke (BS) rather than PM_{10} was measured. During the first few days of the event, smoke concentrations increased from 0.49 mg/m^3 to 2.46 mg/m^3, and then continued to rise to 4.46 mg/m^3 on the two most polluted days (Authority, 2002). PM_{10} concentration can be estimated from smoke concentration by assuming a ratio of PM_{10}/BS. Hoek et al. (1997) reported that most PM_{10}/BS values at 28 sites in 10 countries in Europe were between 0.5 and 1.5 and measurements in London from 1955 to 1962 showed that the average ratio of total suspended particulate (TSP) to black smoke was 2. The ratio of PM_{10} to TSP is often assumed to be 0.5. Therefore, we used the PM_{10}/BS ratio of 1 in this study to estimate PM_{10} concentrations during the London Great Smog. The same ratio was also used in examining the health effects of particulate matter exposures (Dockery and Pope 1994). The maximum PM_{10} concentration in the London Great Smog was then estimated to be 4460 μg/m^3 which is reasonable considering that the monthly average PM_{10} level in December 1952 was reported to be ~3000 μg/m^3 (Davis et al., 2002).

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