



## Spatial and temporal variation of particulate matter and gaseous pollutants in China during 2014–2016



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### HIGHLIGHTS

- All of the pollutants except O<sub>3</sub> decreased slightly during 2014–2016.
- All of the pollutants except O<sub>3</sub> displayed the highest levels in the winter.
- PM<sub>10</sub> is a major pollutant affecting the air quality of China.

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### ABSTRACT

China is experiencing severe air pollution due to rapid economic development and accelerated urbanization. High-resolution temporal and spatial air pollution data are imperative to understand the physical and chemical processes affecting air quality of China. The data of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, CO, NO<sub>2</sub>, and O<sub>3</sub> in 187 Chinese cities during January 2014 and November 2016 were collected to uncover the spatial and temporal variation of the pollutants in China. The annual mean concentrations of PM<sub>2.5</sub> exceeded the Grade I standard of Chinese Ambient Air Quality (CAAQS) for all of the cities except several cities in Hainan, and more than 100 cities exceeded the CAAQS Grade II standard. The concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, CO, and NO<sub>2</sub> decreased from 2014 to 2016, whereas the O<sub>3</sub> level increased dramatically during this period. The concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, CO and NO<sub>2</sub> exhibited the highest levels in winter and the lowest in summer, and evidently decreased from 2014 to 2016, whereas the O<sub>3</sub> concentration peaked in spring and summer, and dramatically increased from 2014 to 2016. The non-attainment ratios were highest in winters, while high pollution days were also frequently observed in the Southeast region in autumn and in the Northwest region in spring. Pearson correlation analysis indicated that all of the pollutants exhibited significant correlation one another. PM<sub>10</sub> was a major pollutant affecting the air quality of China in all of the seasons. Both SO<sub>2</sub> and NO<sub>2</sub> exerted significantly adverse effects on the air quality in spring and autumn, but CO played an important role on the air quality in winter. O<sub>3</sub> was found to be the dominant species among the pollutants affecting the air quality in summer, suggesting that photochemical O<sub>3</sub> formation should be paid more attention to improve the air quality in summer. The results of geographical weight regression (GWR) showed that more significant correlations among the pollutants and the highest air quality index (AQI) appeared in the south of China. The impacts of PM<sub>10</sub> and NO<sub>2</sub> on the air quality increased from the east to the west of China, while SO<sub>2</sub> and O<sub>3</sub> exhibited the opposite variation. The data presented herein supplied an important support for the future source apportionment and intra- and inter-regional transport modeling of pollutants.

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## 1. Introduction

Worse air quality of urban sites has become an increasingly concerned issue due to its adverse effects on human health and climate change. The increasing health risk is associated with the elevation of particulate matter (PM) such as PM<sub>2.5</sub> and PM<sub>10</sub>. As a typical inhalable particle, the increase of PM<sub>2.5</sub> is closely associated with the occurrence of respiratory and cardio-vascular diseases (Araujo, 2011; Kim et al., 2015). Besides, PM plays significant roles on climate change (Paasonen et al., 2013). Some aerosol particles probably absorb or scatter the solar radiation, thereby affecting the global climate (Anenberg et al., 2012). On the other hand, they can act as cloud condensation nuclei (CCN) to govern heat transfer properties in the atmosphere, consequently altering cloud formation process and rainfall patterns (Carslaw et al., 2013; Kalkavouras et al., 2017). Moreover, regional transport of PM<sub>2.5</sub> has been regarded as a major cause of severe haze in some cities (Li et al., 2015a,b). Apart from the effects of PM, gaseous pollutants also play significant roles on human health and environment. For example, the accumulation of many gaseous pollutants including CO, SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub> increased the susceptibility to respiratory diseases and reduced the lung fraction, thereby leading to hematological problems and cancer. In recent years, severe haze pollution has appeared in many regions of China, such as Jing-Jin-Ji, Yangtze River Delta (YRD), and Pearl River Delta (PRD), all of which could be associated with the accumulation of gaseous pollutants. SO<sub>2</sub> and NO<sub>2</sub> could be transformed to SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> via gas-to-particle process under the condition of high relative humidity (RH), which has been confirmed to cause severe haze (Huang et al., 2011a,b; Niu et al., 2016). Moreover, some gaseous pollutants such as NO<sub>x</sub> could be potential precursors to photo-oxidants as ozone in ambient air, resulting in fog-haze pollutions (Zhou et al., 2014). Although series of control measures have been adopted by Chinese governments to alleviate atmospheric pollution, a lot of fog-haze episodes still come up frequently in the metropolitans of China, such as Beijing and Shanghai. Therefore, to investigate the spatial-temporal variations of both PM and gaseous pollutants is essential to understand current air pollution status in China, to assess the effects of control measures applied by local and central governments, and to provide scientific judgment on haze governance.

A growing body of studies about temporal and spatial variations of the pollutants have been reported (Johansson et al., 2007; Karar et al., 2006; San Martini et al., 2015). Chen et al. (2016) analyzed the spatial and diurnal variation of PM<sub>2.5</sub> in Nanjing using complete ensemble empirical mode decomposition with adaptive noise (CEEMDAN) method and elucidated the relationship between PM<sub>2.5</sub> concentrations and meteorological factors. Wu et al. (2015) predicted the spatial-temporal variation of the concentrations of the atmospheric pollutants using land use regression method and concluded that PM<sub>2.5</sub> showed more remarkably temporal variation than spatial variation. Spatial and temporal variation of multiple areas and the pollutants have also been reported recently. Wang et al. (2014a,b) studied the spatial and temporal variation of six criteria pollutants in 31 provincial capital cities of China and observed that the concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, CO and SO<sub>2</sub> peaked at the cities located in the North region, followed by those in the West and the East regions. In addition, Liu et al. (2016a,b,c) investigated the spatial and temporal variation of the air pollutants, API, and mortality in 120 cities of China for one year and observed significant clustering of API, the concentrations of the air pollutants and mortalities in the northwest of China. Chai et al. (2014) analyzed the annual variation of the gaseous pollutants in 26 cities of China and concluded that China was still faced an arduous challenge. On the basis of such conclusion, they thus

suggested that more efforts should be taken to control air pollution. Although some studies on the spatial distribution of the pollutants in a large scale range have been reported, the pollutant levels in dozens of the cities can not accurately reflect the spatial distribution of the pollutants in China. The methods of spatial statistics and geo-statistics (e.g., ordinary Kriging interpolation and inverse distance weight) should be applied to evaluate the spatial-temporal variation of the ambient pollutants. To the best of our knowledge, hardly any studies have concerned about ambient pollutants covered the data of multi observation stations and assessed the spatial variation of the pollutants in China through spatial interpolation methods.

Herein, the officially released data of PM<sub>2.5</sub> and PM<sub>10</sub> and four gaseous pollutants (CO, SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub>) at 187 cities of China during the period of January 2014–November 2016 were collected in order to quantify the pollutant levels accurately in China, as well as spatial and temporal variation trend. The aim of this study is (1) to understand the correlation of six pollutants and determine the dominant factor for the air quality index, (2) to decipher the seasonal variation of six pollutants and identify key factors, and (3) to investigate the inter-annual variability of the pollutants and assess the effects of control measures adopted by governments on the pollutant levels in recent years. The quantification of the spatial and temporal variation of six criteria pollutants will raise the awareness of authorities about the air quality in China, provide a more comprehensive understanding of current situation of air quality, and obtain appropriate strategies to promote environmental protection.

## 2. Methods

Three-year long ambient monitoring data of PM<sub>2.5</sub>, PM<sub>10</sub>, CO, SO<sub>2</sub>, NO<sub>2</sub>, and 8-h O<sub>3</sub> in 187 cities were analyzed to assess the air quality status in China. The real-time hourly concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, CO, SO<sub>2</sub>, NO<sub>2</sub>, and 8-h O<sub>3</sub> at 187 cities were downloaded from the website of China air quality monitoring platform (<http://www.aqistudy.cn/>). Hourly air quality data of six pollutants at individual monitoring site for major cities have been published through the website from January 2013. To date, the monitoring sites have covered Mainland China. This data is imperative to supply more detailed information about the current air quality situations in China. The data of six criteria pollutants at 187 cities from January 1 st, 2014 to November 15 th, 2016 were collected because many monitoring sites had not been established before 2014. All of the data were supplied by the national air quality monitoring sites located in each city. The monitoring sites have been designed as a mixture of urban and background sites, including most of the sites in urban area, and a few of sites in suburban and rural areas as the background sites. The mean concentrations of the species were calculated on the basis of the data supplied by the monitoring sites in each city.

The automated monitoring systems were installed to determine the concentrations of the gaseous pollutants including SO<sub>2</sub>, NO<sub>2</sub>, CO and O<sub>3</sub> at each site. The continuous monitoring system of PM<sub>2.5</sub> and PM<sub>10</sub> is composed of the sample collection unit, the sample measurement unit, the data collection and transport unit, and other accessory equipment. SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub> were measured using the ultraviolet fluorescence method (TEI, Model 43i from Thermo Fisher Scientific Inc., USA), chemiluminescence method (TEI Model 42i from Thermo Fisher Scientific Inc., USA), and the UV-spectrophotometry method (TEI model 49i from Thermo Fisher Scientific Inc., USA), respectively. CO was measured using the non-dispersive infrared absorption method and the gas filter correlation infrared absorption method (TEI, Model 48i from Thermo Fisher Scientific Inc., USA).

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