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Roles of regional transport and heterogeneous reactions in the $PM_{2.5}$ increase during winter haze episodes in Beijing

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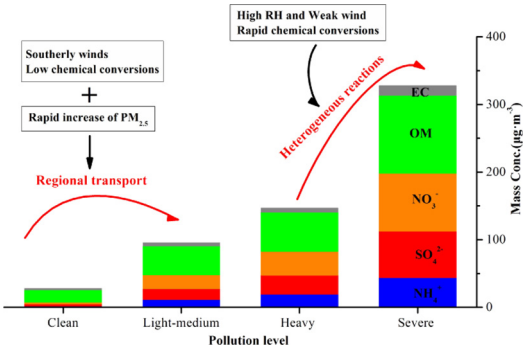
HIGHLIGHTS

- Processes leading to the increase of $PM_{2.5}$ in Beijing in winter haze episodes were determined.
- Regional transport from the south dominated the increase in the initial stage of haze occurrence.
- Heterogeneous reactions dominated the increase in the later stages of haze occurrence.

GRAPHICAL ABSTRACT



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ABSTRACT

Regional transport and chemical conversions are two major processes that lead to the severe haze pollution in China. Our observations during five haze episodes in Beijing between February 19 and March 12 of 2014 show that the two processes played different roles as $PM_{2.5}$ increased from the clean (<75 μg m^{-3}) to the light-medium pollution level (75–150 μg m-3) and to levels of heavy (150–250 μg m^{-3}) and severe (>250 μg m^{-3}) pollution. In the initial twelve hours of each episode, the $PM_{2.5}$ reached the light-medium level with an increase of approximately 120 μg m^{-3}. At the same time, the particle (~10–700 nm) number concentration also showed a distinct increase accompanied by a rapid increase in the mean diameter. A light-medium $PM_{2.5}$ occurred in the south areas prior to the haze occurrence in Beijing and the southerly winds were predominant, indicating the rapid increase of $PM_{2.5}$ in the initial stage was caused by the regional transport from the south. Subsequently, $PM_{2.5}$ elevated to the heavy and severe levels when the wind was weak, relative humidity was high and ozone concentration was low. The increase of $PM_{2.5}$ in the elevated stages was characterized by a high percentage (45% for the heavy level and 55% for the severe level) of secondary inorganic components, indicating the substantial contribution of the formation of secondary aerosols. In addition, the increases of the mean diameter (from 108 nm to 120 nm) and the total volume concentration (by 67%) are regarded as a consequence of heterogeneous reactions on the surfaces of aerosol particles because the particle number concentration remained nearly constant in these two stages. Our results indicate that, during the five winter haze episodes, the regional transport

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from the south was the major reason for the initial-stage $PM_{2.5}$ increase, while heterogeneous reactions dominated the later elevation.

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1. Introduction

In recent years, many areas in China have frequently suffered from periods of severe haze pollution (Che et al., 2015; Cheng et al., 2012; Liu et al., 2013). The pollution is characterized by frequent occurrences, high levels of $PM_{2.5}$ concentration (several hundred micrograms per cubic meter), and low visibility (several hundred meters) (Tao et al., 2014; Y.H. Wang et al., 2015). It has become an urgent environmental issue in China because of its adverse effect on air quality and public health (Lin et al., 2015; Tie et al., 2009; L.L. Wang et al., 2015; Zheng et al., 2015).

Compared to other areas in China, the north China plain has suffered from more severe air pollution in winter (Zhang et al., 2015). This is caused by a combination of the high emission of air pollutants that results from heating and frequent stable meteorological conditions in this area (Elser et al., 2016; Q.Q. Wang et al., 2015). Beijing, the capital of China, is located in the northeast part of the north China plain and surrounded by mountains in three directions except in the south. Due to its special topography, Beijing is easily affected by air pollutants from industrial activities in the south, resulting in a rapid increase of $PM_{2.5}$ concentration (L.L. Wang et al., 2015; Zheng et al., 2015). Earlier studies found that the high mass concentration of particulate matter (PM) in Beijing was always associated with southerly winds and frequently caused by the transport of pollutants from the south areas of Beijing (Sun et al., 2015; Yang et al., 2015). Regional transport contributed 28–36% of the $PM_{2.5}$ mass concentration in Beijing in 2012 and 2013 according to the results of source apportionment published by Beijing Environmental Protection Bureau.

It was also found that the secondary formation, i.e., the gas-to-particle conversions in the atmosphere, substantially contributed to the $PM_{2.5}$ pollution in Beijing (Cheng et al., 2016; Huang et al., 2014; Wang et al., 2016). During the haze pollution period, secondary species, especially inorganic ions such as sulfate and nitrate, dominated the $PM_{2.5}$ at levels of 40–50% (Pan et al., 2016; Wang et al., 2012; Zhang et al., 2016). In haze pollution periods, the relative humidity is usually high and ozone concentration is usually low. The increases of secondary inorganic species are most likely from heterogeneous reactions rather than photochemical reactions (Han et al., 2015; Huang et al., 2014; Y. Sun et al., 2013; Quan et al., 2015; Wang et al., 2012; Zhang et al., 2016). Wang et al. (2016) and Cheng et al. (2016) confirmed that the production of sulfate and nitrate via heterogeneous reactions on aqueous particles had led to severe haze pollution in polluted environments with high relative humidity. Thus, the winter haze pollution in Beijing during winter was regarded as a consequence of the combination of meteorological conditions, primary emissions (coal combustion), heterogeneous reactions and regional transport (Y.L. Sun et al., 2013; Sun et al., 2015).

It is generally known that the $PM_{2.5}$ concentration will increase with the development of haze pollution. However, the increase of $PM_{2.5}$ and the reasons leading to the increase have not been well examined at the different levels of haze pollution. The emissions remain nearly constant over a given season. A recent study also showed that primary emissions are nearly constant in both clean and pollution periods (Zheng et al., 2015). Our study aimed to reveal the contribution of heterogeneous reactions and regional transport combined with weather conditions to the increase of $PM_{2.5}$ in Beijing in haze episodes. As a result, in this study, we show the observational results of particle size distribution and $PM_{2.5}$ composition at an urban site in Beijing, describe the $PM_{2.5}$ increase combining with temporal and spatial meteorological conditions, and discuss the roles of heterogeneous reactions and regional transport on $PM_{2.5}$ increase during different stages of haze occurrence.

2. Instruments and measurements

2.1. Experiment site

Observations were carried out on the roof of a two-story experiment building in the tower division of the Institute of Atmospheric Physics (IAP, Fig. 1), Chinese Academy of Sciences, during a late winter period from February 19 to March 12, 2014. The observation site is located in the north part of Beijing (39°58′ N, 116°22′ E). It is close to several major roads including a highway and is surrounded by residences and restaurants. It is a typical urban site in Beijing, and several studies on air pollutants in the urban atmosphere have been conducted at this site (Jing et al., 2015; Wu et al., 2016).

2.2. Online monitoring of $PM_{2.5}$ mass concentration and meteorological parameters

Real-time $PM_{2.5}$ mass concentrations were measured using a hybrid beta attenuation particulate monitor (Model 5030 SHARP, Thermo Scientific, USA). The monitor, which is a USEPA Class III Federal Equivalent Method (FEM) analyzer (U.S. EPA, 2011), synchronized the light scattering on fundamental beta attenuation to provide high resolution measurement of airborne particulates. Ambient air was drawn into a single inlet with $PM_{2.5}$ cutoff, passed through a silicon diffusion dryer and then entered $PM_{2.5}$ online measurements at a nominal flow rate of 16.7 L min^{-1}. Concentrations of other air pollutants, including SO_2, NO_2 and O_3, were obtained from a nearby site (approximately 2 km northeast of our site) at the Olympic Sports Center, which is operated by the Beijing Municipal Environmental Monitoring Center (http://www.bjmemc.com.cn/g68.aspx). The hourly $PM_{2.5}$ concentrations observed at our site were highly consistent with those at the Olympic Sports Center site with a linear slope of 0.94 and a square of correlation coefficient equal to 0.92. The meteorological conditions (including wind direction, wind speed, temperature and relative humidity) at 15 altitude levels (e.g., 8 m, 15 m, 32 m, 47 m, 65 m, 80 m, 100 m, 120 m, 140 m, 160 m, 180 m, 200 m, 240 m, 280 m, and 320 m) were obtained from the IAP 325 m meteorological tower, which is approximately 30 m from our observation site.

2.3. $PM_{2.5}$ sample collection and chemical analysis

A R&P Partisol® Model 2025 dichotomous sequential PM air sampler (Thermo, USA) and a MiniVol TAS PM sampler (Airmetrics, USA) were used in parallel to collect $PM_{2.5}$ samples between February 24 and March 12, 2014. The R&P Partisol® sampler was equipped with a 47 mm Teflon filter (Whatman PTFE) and operated at a nominal flow rate of 16.7 L min^{-1}. The MiniVol sampler was equipped with a 47 mm quartz filter (Whatman QM-A) and operated at 5 L min^{-1}. Samples were collected twice per day with one during the day (from 7:00 to 19:00 at local time) and the other at night (from 19:00 to 7:00 of the next day). After the collection of a sample, the sampled filter was stored in a freezer at −18 °C until the subsequent analysis, to minimize the possible evaporation of volatile components. Samples were analyzed gravimetrically for particle mass concentration using an Electronic balance with a detection limit of 1 μg (Sartorius, Göttingen, Germany) after 24-h equilibration at a temperature between 20 °C and 23 °C and

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