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Effects of meteorology and secondary particle formation on visibility during heavy haze events in Beijing, China

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HIGHLIGHTS

- The cases of haze formation in Beijing, China were analyzed.
- The effects of RH on PM2.5 concentration and visibility were studied.
- Gas-phase to particle-phase conversion under different visibility was analyzed.
- With high RH, the conversion SO2 to SO4= accounted for 20%.

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ABSTRACT

The causes of haze formation in Beijing, China were analyzed based on a comprehensive measurement, including PBL (planetary boundary layer), aerosol composition and concentrations, and several important meteorological parameters such as visibility, RH (relative humidity), and wind speed/direction. The measurement was conducted in an urban location from Nov. 16, 2012 to Jan. 15, 2013. During the period, the visibility varied from >20 km to less than a kilometer, with a minimum visibility of 667 m, causing 16 haze occurrences. During the haze occurrences, the wind speeds were less than 1 m/s, and the concentrations of $PM_{2.5}$ (particle matter with radius less than 2.5 μm) were often exceeded 200 μg/m^3. The correlation between $PM_{2.5}$ concentration and visibility under different RH values shows that visibility was exponentially decreased with the increase of $PM_{2.5}$ concentrations when RH was less than 80%. However, when RH was higher than 80%, the relationship was no longer to follow the exponentially decreasing trend, and the visibility maintained in very low values, even with low $PM_{2.5}$ concentrations. Under this condition, the hygroscopic growth of particles played important roles, and a large amount of water vapor acted as particle matter (PM) for the reduction of visibility. The variations of meteorological parameters (RH, PBL heights, and WS (wind speed)), chemical species in gas-phase (CO, O_3, SO_2, and NOx), and gas-phase to particle-phase conversions under different visibility ranges were analyzed. The results show that from high visibility (>20 km) to low visibility (<2 km), the averaged PBL decreased from 1.24 km to 0.53 km; wind speeds reduced from 1 m/s to 0.5 m/s; and CO increased from 0.5 ppmv to 4.0 ppmv, suggesting that weaker transport/diffusion caused the haze occurrences. This study also found that the formation of SPM (secondary particle matter) was accelerated in the haze events. The conversions between SO_2 and $SO_4^{=}$ as well as NOx to NO_3^- increased, especially under high humidity conditions. When the averaged RH was 70%, the conversions between SO_2 and $SO_4^{=}$ accounted for about 20% concentration of $PM_{2.5}$, indicating that formation of secondary particle matter had important contribution for the haze occurrences in Beijing.

1. Introduction

High occurrence of haze events (visibility lower than 10 km) in Beijing, the capital city of China, causes deeply concern in the scientific community in recent years. This severe environmental problem has widely impacts on the people's life, traffic, climate, and other important aspects (Charlson et al., 1987; Ramanathan and Vogelmann, 1997; Tegen et al., 2000; Yu et al., 2002; Tie et al., 2009a,b). In haze events, the concentrations of $PM_{2.5}$ (the particle matters with the radius less or equal to 2.5 μm) rapidly increased, with a maximum of 600 μg/m^3 (Quan et al., 2013). The extremely high aerosol concentrations caused a very low visibility, and the hygroscopic growth of aerosol particles due to increased RH (relative humidity) in haze events led to further

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increase their effects on atmospheric visibility (Liu et al., 2011; Quan et al., 2011).

The aerosol concentrations in atmosphere are affected by several factors, including pollutant emissions, atmospheric advection/diffusion, and second aerosol formation etc. (He et al., 2001; Yang et al., 2011; Sun et al., 2013). Large pollutant emission in north China plain (NCP) is a dominant reason for causing the high aerosol concentrations in this region (Tie et al., 2006; Guinot et al., 2007; Chan and Yao, 2008), and unfavorable meteorological conditions can further increase aerosol concentrations. First, there is generally a barrier (very low mixing rate) at the top of the PBL to prevent particles being across from the PBL to the free troposphere (Han et al., 2009; Zhang et al., 2009). As a result, aerosol particles are constrained in the PBL, and aerosol concentrations are anti-correlated with the PBL heights (Zhang et al., 2009; Quan et al., 2013). Second, the aerosol particles are horizontally transported from source regions to downwind regions, and the rate of the transport depends strongly upon wind speeds. In addition to the factors of emission/transport/diffusion, the formation of aerosols might be enhanced due to the participation of liquid phase reactions (Zhang et al., 2013), especially for inorganic components (sulfate, nitrate, ammonia, etc.).

In this work, a comprehensive measurement was carried out during a period (with 16 haze occurrences) from Nov. 16, 2012 to Jan. 15, 2013 to investigate the causes of the occurrences of the haze events. The framework of this study as the following orders; (1) The instruments and measurements were described; (2) The analysis of the results was given. The analysis focuses on the following issues: (a) the characteristics of the haze events, (b) the effects RH on visibility, (c) the relationship between visibility and meteorological and particle conditions, (d) the relative contributions of meteorological and particle conditions to visibility, and (e) the relative contributions of particle composition to visibility in the haze events.

2. Instruments and measurements

A comprehensive measurement was conducted from Nov. 16, 2012 to Jan. 15, 2013 in Beijing located at Baolian (BL) meteorological station, China Meteorological Administration (CMA) (39°56′N, 116°17′E). In the measurement, atmospheric visibility, mass concentration of $PM_{2.5}$, chemical composition of non-refractory submicron particles (NR-PM_1), gaseous pollutants (SO_2, NOx, carbon monoxide (CO), ozone (O_3)), and meteorological variables (such as temperature, PBL heights, RH, pressure, wind speed, and wind direction) were observed simultaneously.

The mass concentration of $PM_{2.5}$ was observed by a R&P model 1400a Tapered Element Oscillating Microbalance (TEOM, Thermo Scientific Co., USA) instrument with a 2.5 μm cyclone inlet. The collocated gaseous species including CO, SO_2, NOx and O_3 were observed by various gas analyzer (Thermo Scientific Co., USA).

Chemical composition of NR-PM_1 was measured by an Aerodyne high-resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS). The sampling time resolution was 2 min. The measured composition of particles included sulfate ($SO_4^{=}$), nitrate (NO_3^-), ammonium (NH_4), chloride (Chl), three primary organic (OA) aerosols, including hydrocarbon-like OA (HOA), cooking OA (COA), and coal combustion OA (CCOA), and one secondary OA aerosol, i.e., oxygenated OA (OOA). According to their origin, PM can be defined into two classes: primary PM (PPM), including, Chl, HOA, COA, CCOA, and secondary PM (SPM), including $SO_4^{=}$, NO_3^-, NH_4, and OOA.

The evolution of PBL was observed by a micro-pulse lidar (MPL-4B, Sigmaspace Co., USA). Atmospheric visibility was observed by a PWD20 (Vaisala Co., Finland), and meteorology variables were observed by WXT-510 (Vaisala Co., Finland). Detailed instructions of above instruments were given by Quan et al. (2013).

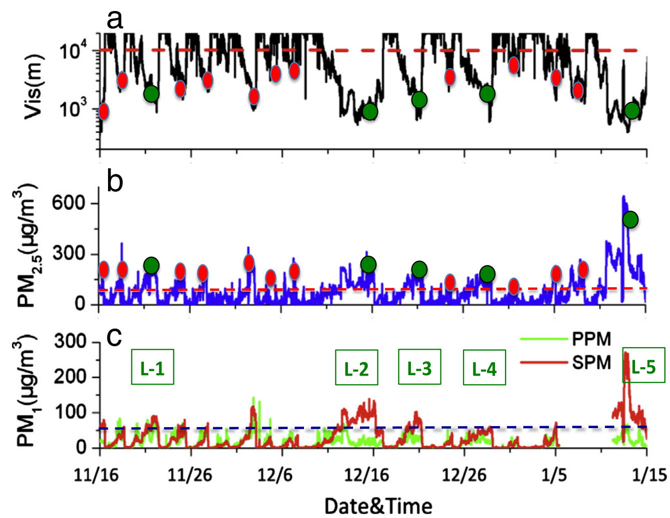

Fig. 1. The measured visibility (a), $PM_{2.5}$ concentrations (b), and PM_1 (c) from Nov. 16, 2012 to Jan. 15, 2013 at Baolian station in Beijing. There were 16 haze occurrences during this period. The red dots show short-haze events (1–2 days). The blue dots show long-haze events (longer than 3 days), which were also indicated by L-1 to L-5 in panel c. In panel c, the green line presents for PPM and red line for SPM. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3. Result and analysis

3.1. Characteristics of haze events

According to the definition by CMA (Chinese Meteorological Administration), a haze event is defined by the following conditions, i.e., visibility < 10 km and RH < 90%. During the 2 month experiment period (from Nov. 16, 2012 to Jan. 15, 2013), there were totally 16 haze events, indicating by the red and green dots (see Fig. 1). The time of haze events comprised approximately 60% of the total time in the 2 months, indicating that during wintertime in Beijing, the haze events were very active. During the 16 haze events, the $PM_{2.5}$ concentrations were higher than 100 μg/m^3 (see panel b in Fig. 1), suggesting that the high particle concentration was a main reason for causing the haze events. Considering the CMA definition and the measured results, the haze and heavy haze events were clarified in this study as average concentration of $PM_{2.5}$ is above 75 μg/m^3 and visibility is less than 5 km for more than 6 h, and average concentration of $PM_{2.5}$ is above 150 μg/m^3 and visibility is less than 2 km for more than 12 h, respectively. In addition to this general conclusion, more detailed feathers of the haze events were found, including: (1) The duration of haze events ranged from 1 to 2 days ("short haze" marked by red dots) to 3–6 days ("long haze" marked by green dots). (2) During the "long haze" events, the $PM_{2.5}$ concentrations were extremely high, causing the occurrences of heavy haze (visibility < 1 km). For example, on Jan. 13, 2013, the $PM_{2.5}$ concentrations reached to a maximum value of 600 μg/m^3, resulting in a very poor visibility condition (a few hundred meters). (3) The formation of the haze events was in an accumulation mode, especially for the "long haze" events. This feather was clearly indicated during the "long haze" period from Dec. 11 to 16, 2012. During the period, the $PM_{2.5}$ concentration gradually increased from 50 μg/m^3 to 200 μg/m^3 in 5 days. (4) In contrast to the accumulation mode of the haze formation, the disappearance of the haze events was generally in a quick mode. For example, on Dec. 15, 2012, the $PM_{2.5}$ concentration was about 300 μg/m^3 and the visibility was about 1 km. On the next 2 days (Dec. 16–17, 2012), the $PM_{2.5}$ concentration decreased to a small value, and the visibility increased to about 20 km. (5) The secondary particles (SPM) had important contribution to the haze events, especially during the "long haze" events. Fig. 1 (panel c) shows that the measured primary particles (PPM) had a similar magnitude in concentrations compared to the