



Size-resolved aerosol chemical analysis of extreme haze pollution events during early 2013 in urban Beijing, China



Shili Tian, Yuepeng Pan*, Zirui Liu, Tianxue Wen, Yuesi Wang*

State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

HIGHLIGHTS

- Anthropogenic species substantially accumulated in both fine and coarse particles.
- Secondary organic carbon in PM_{1.1} decreased from clear to haze days.
- The mass peak shifted to larger particles from clear to haze days.
- The NO₃⁻/SO₄²⁻ ratio decreased with enhanced haze pollution.
- Both mobile local and stationary regional sources were vital for haze formation.

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ABSTRACT

Using size-resolved filter sampling and chemical characterization, high concentrations of water-soluble ions, carbonaceous species and heavy metals were found in both fine (PM_{2.1}) and coarse (PM_{2.1-9}) particles in Beijing during haze events in early 2013. Even on clear days, average mass concentration of submicron particles (PM_{1.1}) was several times higher than that previously measured in most of abroad urban areas. A high concentration of particulate matter on haze days weakens the incident solar radiation, which reduces the generation rate of secondary organic carbon in PM_{1.1}. We show that the peak mass concentration of particles shifted from 0.43–0.65 μm on clear days to 0.65–1.1 μm on lightly polluted days and to 1.1–2.1 μm on heavily polluted days. The peak shifts were also found for the following species: organic carbon, elemental carbon, NH₄⁺, SO₄²⁻, NO₃⁻, K, Cu, Zn, Cd and Pb. Our findings demonstrate that secondary inorganic aerosols (36%) and organic matter (26%) dominated the fine particle mass on heavily polluted days, while their contribution reduced to 29% and 18%, respectively, on clear days. Besides fine particles, anthropogenic chemical species also substantially accumulated in the coarse mode, which suggests that particles with aerodynamic diameter larger than 2.1 μm cannot be neglected during severe haze events.

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1. Introduction

Haze is defined as a weather phenomenon featuring a high concentration of fine particles that leads to a visibility of less than 10 km at a relative humidity (RH) lower than 90% [1–3]. This severe air pollution can not only decrease air quality and endanger public health but can also alter the radiation budget and the hydrological cycle [4,5]. As a consequence, the persistence and severity of haze events have resulted in widespread public concern. In recent years, China has faced serious haze pollution due to vast energy consumption and a rapid increase in the number of vehicles. However, further research is necessary to understand the characteristics, formation and evolution of large-area and long-lasting haze pollution events across China.

* Corresponding authors. Tel.: +86 01082020530; fax: +86 01062362389.
E-mail addresses: panyuepeng@mail.iap.ac.cn (Y. Pan), wys@mail.iap.ac.cn (Y. Wang).

In January 2013, several of the most severe haze events on record swept across most of east-central cities and covered a quarter of the total land area in China. These severe wintertime haze events included substantial regional particulate matter (PM) pollution according to both satellite- and surface-based aerosol optical depth (AOD) observations and model simulations [6–8]. In addition, high concentrations of important trace gases, such as carbon monoxide (CO), sulfur dioxide (SO₂) and ammonia (NH₃), and ammonium sulfate aerosols ((NH₄)₂SO₄) were found using IASI (Infrared Atmospheric Sounding Interferometer) satellite measurements [9]. Based on surface measurements, we previously found that extremely high nitric oxide (NO_x) concentrations played either a direct or indirect role in the rapid secondary transformation of coal-burning SO₂ into sulfate aerosols during haze episodes [10]. In addition to sulfate and nitrate, high concentrations of ammonium and organic carbon were also observed in Beijing and Shanghai during these severe regional haze events (e.g., Jan. 12–13 and Jan. 29–30, 2013) [11–13]. However, most of these studies focused only on inorganic and organic chemical compounds in single particle fractions (e.g., PM₁ or PM_{2.5}). To date, the size-resolved organic carbon, elemental carbon and water-soluble ion chemistry for these haze episodes remains unclear, especially regarding water-soluble organic carbon and toxic

metals. Knowledge of the size distribution of various chemical species is important for understanding the physical and chemical atmospheric processes that affect aerosol properties during haze episodes [14]. Haze pollution is characterized by elevated levels of fine particles, which have severe adverse effects on human health, visibility and climate forcing due to their chemical compositions and longer lifetimes in the atmosphere [15]. Coarse particle superposition is also an important factor for the reduced visibility during haze pollution [10]. Besides, the deposition of acids and toxic species through coarse particles causes adverse effects in ecosystems [16]. Such size-resolved chemical investigations are especially needed in megacities, such as Beijing, that have been subjected to periodic haze pollution in recent years.

In this study, two cascade impactors simultaneously collected size-segregated airborne particles in urban Beijing, where prolonged haze episodes occurred in January and February of 2013. To our knowledge, these are the only measurements obtained in China during these haze periods to characterize the size distributions of chemical species (i.e., water-soluble ions, carbonaceous species and elements). The unique datasets may improve our knowledge of aerosol properties during haze pollution episodes in Beijing and thereby provide baseline information for future measurement and modeling studies.

2. Materials and methods

2.1. Sampling site

The sampling site was on the roof of a building at the Institute of Atmospheric Physics (IAP). The building is approximately 15 m above the ground, located near a residential area in North Beijing (39°58'N, 116°22'E) and situated between the 3rd and 4th ring roads (Fig. S1). The site is approximately 1 km from the 3rd ring road, 200 m west of G6 Highway (which runs north-south) and 50 m south of Beitucheng West Road (which runs east-west). Daily average traffic volumes for 3rd and 4th ring roads were around 10 million/day [17]. There were no industrial sources of atmospheric pollutants during the study period. The experimental campaign was conducted between Jan. 1 and Feb. 28, 2013.

2.2. Sample collection

To obtain comprehensive chemical characteristics of the size-segregated airborne particles, two nine-stage samplers (Andersen Series 20-800, USA) were used to simultaneously collect particles at a flow rate of 28.31 min^{-1} with cutoff points of 0.43, 0.65, 1.1, 2.1, 3.3, 4.7, 5.8 and $9.0 \mu\text{m}$. Quartz fiber filters (for water-soluble ions and carbonaceous species analysis) and cellulose membranes (for element analysis) were used as substrates in the Andersen samplers; both substrates had a diameter of 81 mm. Each set of the size-segregated samples was continuously collected for 24 or 48 h. Regular aerosol sampling was conducted for 48 h from 10:00 LT on Monday to 10:00 LT on Wednesday every week. When an air pollution episode occurred, we attempted to densify the sampling to capture the formation and dissipation processes of the haze pollution. Therefore, a relative short sampling time last for 24 h was performed. The quartz fiber filters were wrapped with aluminum foil and pre-heated at 800°C for 2 h to remove all organic material. The filters were subsequently conditioned in a dryer (temperature: 25°C ; humidity: 10%) for 72 h before weighing. The filters were weighed before and after sampling on a microbalance with a balance sensitivity of $\pm 0.01 \text{ mg}$. After re-weighing the samples, the exposed filters were divided into different portions (see below) with clean tools and prepared for the chemical analysis. Each filter was weighed three times before and after sampling, and the average value was used. Blanks and duplicate sample analyses were performed for approximately 10% of the samples. Blank filters were processed simultaneously with the field samples. In addition, the PM_{10} , $\text{PM}_{2.5}$ and PM_1 concentrations were measured with RP1400 instruments, which provide continuous direct mass measurements of particles through a tapered element oscillating microbalance (TEOM) [18]. Meanwhile, the meteorological parameters, including temperature, humidity, wind speed and

wind direction, were observed using an automatic meteorological observation instrument (Milos520, Vaisala, Finland).

2.3. Chemical analyses

A quarter of each quartz filter was cut and extracted using 25 ml of deionized water (Millipore, $18.2 \text{ M}\Omega$) and an ultrasonic bath at room temperature for 30 min. The extraction liquid was filtered using $0.22 \mu\text{m}$ filters and subsequently analyzed to determine the Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , NO_3^- and SO_4^{2-} concentrations with an ion chromatograph (DIONEX, ICS-90, USA). Water-soluble organic carbon (WSOC) concentrations were analyzed using a Multi N/C 3000 Analyzer (Analytik Jena AG, Germany). After another quarter of each quartz filter was cut, a thermal/optical carbon aerosol analyzer (DRI Model 2001A, Desert Research Institute, USA) was used to examine the organic carbon (OC) and elemental carbon (EC) loaded on the samples.

A quarter of the cellulose membrane was digested in a mixture of concentrated HNO_3 (6 ml), HCl (2 ml) and HF (0.2 ml) using a closed vessel microwave digestion system (MARS5, CEM Corporation, Matthews, NC, USA). The concentrations of 14 trace elements (TEs) (Na, Mg, Al, K, Ca, V, Cr, Mn, Fe, Cu, Zn, As, Cd and Pb) in the digests were determined using Agilent 7500 ce inductively coupled plasma mass spectrometry (ICP-MS, Agilent Technologies, Tokyo, Japan). The extraction and analysis methods of all of the chemical species have been reported in previous studies [19–22]. Most of the information about the instruments (e.g., detection limit, precision and calibration) and the results of data quality control work can also be found in these cited papers.

3. Results and discussion

3.1. Mass concentrations of particles

3.1.1. Online measurement of particulate matter

The hourly concentrations of PM_{10} , $\text{PM}_{2.5}$ and PM_1 in Beijing, along with the meteorological parameters (i.e., temperature, RH, wind speed and direction), from Jan. 1 to Feb. 28, 2013, are depicted in Fig. 1. Surprisingly, there were only 4 days in which the daily average PM_{10} concentration was less than the Chinese National Ambient Air Quality Standard (GB3095-2012) of $50 \mu\text{g m}^{-3}$ (NAAQS, Grade I) (Table 1). This finding suggests that 27 days in January exhibited serious particulate pollution. In addition, 17 days had daily average PM_{10} values that exceeded the NAAQS Grade II level of $150 \mu\text{g m}^{-3}$, which is harmful to human health [23]. Besides PM_{10} , the pollution from $\text{PM}_{2.5}$ and PM_1 was also severe in January. For example, the highest instantaneous 5-min

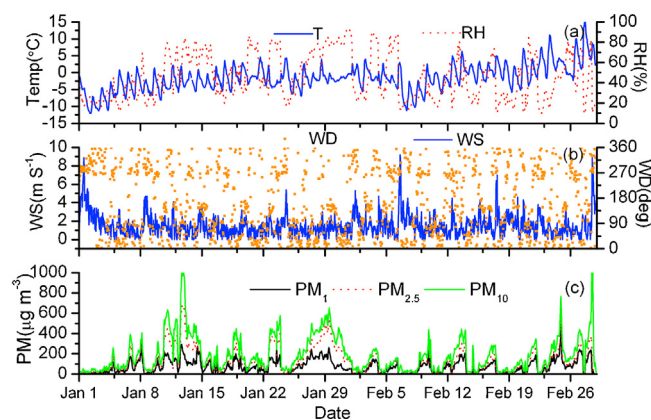


Fig. 1. Temporal variations of (a) ambient temperature and relative humidity; (b) wind direction and wind speed; (c) PM_1 , $\text{PM}_{2.5}$ and PM_{10} concentrations from Jan. 1 to Feb. 28, 2013.

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