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Chemical composition and sources of PM_1 and $PM_{2.5}$ in Beijing in autumn

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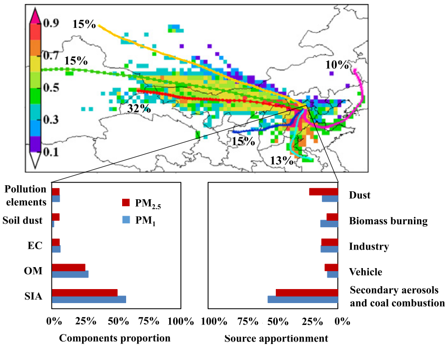
HIGHLIGHTS

- Simultaneous observation of PM1 and PM2.5 were conducted.
- Chemical composition of PM1 and PM2.5 were investigated.
- Three typical weather patterns caused heavy pollutions in autumn.
- Five contribution sources were identified for PM1 and PM2.5.
- Source areas of PM1 and PM1-2.5 were generally similar, but different during heavy pollution periods.

GRAPHICAL ABSTRACT



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ABSTRACT

Beijing, the capital of China, suffers from severe atmospheric aerosol pollution; nevertheless, a comprehensive study of the constituents and sources of PM_1 is still lacking, and the differences between PM_1 and $PM_{2.5}$ are still unclear. In this study, an intensive observation was conducted to reveal the pollution characteristics of PM_1 and $PM_{2.5}$ in Beijing in autumn. Positive matrix factorization (PMF), backward trajectories and a potential source contribution function (PSCF) model were used to identify the source categories and source areas of PM_1 and $PM_{2.5}$. The results showed that the average concentrations of PM_1 and $PM_{2.5}$ reached 78.20 μg/m^3 and 95.47 μg/m^3 during the study period, respectively. PM_1 contributed greatly to $PM_{2.5}$. The $PM_1/PM_{2.5}$ value increased from 73.6% to 90.1% with PM_1 concentration growing from <50 μg/m^3 to >150 μg/m^3. Higher secondary inorganic aerosol (SIA) proportions (31.3%–70.8%) were found in PM_1. The higher fraction of SIA, OC, EC and typical elements in PM_1 illustrated that anthropogenic components accumulated more in smaller size particles. Three typical weather patterns causing the heavy pollution in autumn were found as follows: (1) Siberian high and uniform high pressure field, (2) cold front and low-voltage system, and (3) uniform low pressure field. A PMF analysis indicated that secondary aerosols and coal combustion, vehicle, industry, biomass burning, and dust were the important sources of PM, accounting for 53.8%, 8.0%, 13.0%, 13.2% and 12.0% of PM_1, respectively, and for 47.5%, 9.9%, 12.4%, 8.4% and 21.8% of $PM_{2.5}$, respectively. The HYSPLIT and chemical components analysis indicated the potential contribution from biomass burning and fertilization ammonia emissions to PM_1 in autumn. The source areas were similar for PM_1 and $PM_{1–2.5}$ under general polluted conditions, but during the heavily polluted periods, the source areas were distributed in farther regions from Beijing for PM_1 than for $PM_{1–2.5}$.

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1. Introduction

Particulate matter (PM) is an important atmospheric pollutant. It has substantial impacts on air quality (Li et al., 2017), human health (Huang et al., 2017), atmospheric visibility (Zhou et al., 2015) and climate change (Ding et al., 2017). China is suffering from severe $PM_{2.5}$ (fine particulate matter with aerodynamic diameters <2.5 μm) pollution, especially in its capital – Beijing. The annual average concentration of $PM_{2.5}$ in Beijing was 69.70–122.40 μg/m^3 during the past decade (Lang et al., 2017), 2.0–3.5 times the national standard (35 μg/m^3). As a result, $PM_{2.5}$ has received a great deal of attention in recent studies (Yao et al., 2016; Zikova et al., 2016). As a smaller diameter particulate matter, submicron aerosols (PM_1, fine particulate matter with aerodynamic diameters <1 μm) played more significant roles on visibility degradation, climate change and human health hazards (Rattanavaraha et al., 2017). However, PM_1 has received much less attention than $PM_{2.5}$ (Hu et al., 2016; Wang et al., 2016). Investigating the chemical composition and sources of PM_1 is necessary and important for further studies on its environmental, health and climate implications, as well as for the effective mitigation of both PM_1 and $PM_{2.5}$ pollutions.

Some studies have been conducted on the PM_1 pollution characteristics of Beijing based on online monitoring. For example, Sun et al. (2015) measured the non-refractory submicron aerosol (NR-PM_1) in Beijing and analyzed the diurnal variations of NR-PM_1; Q. Q. Wang et al. (2015) carried out a real-time measurement of NR-PM_1 composition by Aerosol Chemical Speciation Monitor (ACSM) from October to December, reporting that ammonium salts made a large contribution to particle extinction. The above online monitoring could provide time frequency data, but the species only included organics, sulfate, nitrate, ammonium and chloride, while the concentrations of other important components (e.g., other water-soluble ions, elements and elemental carbon (EC)) can't be obtained. The lack of comprehensive species data would limit the further research of PM_1 to some extent. For example, the source apportionment of PM_1 could not be conducted, and as a result, the sources and their contributions to the total PM_1 in Beijing are still unclear. In addition, the comparisons of PM_1 and $PM_{2.5}$ need to be further studied.

The transition season between summer and winter had opposite climatic characteristics (Text S1), and autumn had complex and variable meteorological conditions, which could influence the air quality, especially heavy pollution processes, to a great extent (Chen et al., 2015; J. Zhao et al. 2017). In addition, autumn is a typical farming season, with agriculture-related activities and emissions (e.g., fertilization and biomass burning), this would also have an impact on the PM_1 and $PM_{2.5}$ pollution. According to China's Ministry of Environmental Protection (http://www.zhb.gov.cn/), the average concentration of $PM_{2.5}$ (79.9 μg/m^3) of Beijing in autumn is comparable to that in winter (81.0 μg/m^3) in 2016. This finding indicated that the PM pollution of Beijing in autumn is also severe. A few studies have focused on PM_1 in winter (Jiang et al., 2015; Zhang et al., 2014); however, less attention has been devoted to PM_1 in autumn.

Consequently, in this study, we conducted simultaneous observations of PM_1 and $PM_{2.5}$ in Beijing in autumn. The chemical components, including elements, ions, organic carbon (OC) and EC were determined. The chemical composition characteristics of PM_1 and $PM_{2.5}$ were analyzed (3.1). Heavy pollution periods and their driving meteorological field patterns in autumn were investigated (3.2). The contributions from emission sources of PM_1 and $PM_{2.5}$ were then obtained by PMF analysis (3.3). The source areas of PM_1, $PM_{1-2.5}$ and typical chemical components were also identified based on a backward trajectory and potential source contribution function (PSCF) analysis (3.4).

2. Methods

2.1. PM_1 and $PM_{2.5}$ sampling

The PM_1 and $PM_{2.5}$ sampling was conducted on the roof of the Science Building of Beijing Normal University (BNU) (39.96°N, 116.36°E) at a height of approximately 35 m from the ground. The sampling site was located in residential and commercial areas surrounded by dense traffic. Samples were collected for 24 h (from 09:00 a.m. to 09:00 a.m., BST) from 10th October to 4th November 2016. A total of 52 samples were collected.

A four-channel Spiral Ambient Speciation Sampler (URG-3000ABC, USA) with a flow rate of 16.67 L min^{-1} for each channel was used to collect the PM samples. The first and second channels were fitted with 47 mm Whatman 41 filters (Whatman Inc., Maidstone, UK) for PM_1 and $PM_{2.5}$ mass collection and subsequent ion and elemental analyses. The third and fourth channels were used to collect PM_1 and $PM_{2.5}$ on 47 mm quartz fiber filters (Whatman Inc., Maidstone, UK) for OC and EC analyses. After collection, the samples were placed into polyethylene plastic bags and saved in a refrigerator. All the procedures underwent strict quality control to avoid possible sample contamination. The weight of the filters was determined with an electronic balance (Sartorius TB-215D) before and after sampling, with a precision of 0.01 mg. The filters were equilibrated for 48 h after sampling and weighed under conditions with a temperature of 20 ± 5 °C and relative humidity of 40 ± 2%.

2.2. Chemical analysis

OC and EC in PM_1 and $PM_{2.5}$ were determined with an Optical Carbon Analyzer Model (DRI, Desert research institute, Model 2001A). The detection limits for TOC and TEC were 0.82 μg C cm^{-3} and 0.20 μg C cm^{-3}, respectively. The determination precision of the carbon components was below 5%. Four OC fractions, including OC1, OC2, OC3 and OC4, were produced in a helium atmosphere at 140 °C, 280 °C, 480 °C and 580 °C, respectively. Three EC fractions were produced in a 2% oxygen/98% helium atmosphere at 580 °C, 740 °C, and 840 °C for EC1, EC2, and EC3, respectively. Half of each 47 mm Whatman 41 filter sample was used for ion analysis. The filters were soaked in 10 mL of deionized water and extracted for 40 min in an ultrasonic extractor. For the high concentration samples, samples were analyzed after being diluted. A blank filter was analyzed as the background value of the ions to ensure the accuracy of analysis. The concentrations of 10 ions (NO_3^-, SO_4^{2-}, NH_4^+, NO_2^-, K^+, Na^+, Ca^{2+}, Mg^{2+}, F^- and Cl^-) were determined by ion chromatography (Metrohm 861 Advanced Compact IC, SUI). The detection limits were <0.04 mg L^{-1} for anions and 0.006 mg L^{-1} for cations. The relative standard deviation of each ion concentration was <5%. The other half of each Whatman 41 filter sample was used for elemental analysis. Total metals were extracted with 3 mL of concentrated HNO_3, 1 mL of concentrated $HClO_4$ and 1 mL of concentrated HF in the polytetrafluoroethylene high-pressure tank under 170 °C held for 4 h. After cooling, the tank was placed on an electric hot plate (150 °C) to drive off the acid. The elements that were determined by inductively coupled plasma mass spectrometry (ICP-MS, 7500a, Thermo) included Na, Mg, Al, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Cd, Sb, Ce, Eu and Pb. S was determined by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). The relative standard deviations of elements were also generally below 5%. In addition, all the above measurement procedures underwent strict quality-control to avoid possible contamination of the samples. More details on the chemical analysis can be found in X. W. Yang et al. (2017).

2.3. PMF model

PMF is an effective source apportionment receptor model that decomposes a matrix of sample data into the following two matrices: factor profiles and factor contributions. EPA-PMF version 5.0 was applied in this study to determine the source types of PM_1 and $PM_{2.5}$. The species input model included OC, EC, NO_3^-, SO_4^{2-}, NH_4^+, K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, Na, Mg, Al, S, Ca, Ti, Mn, Fe, Ni, Cu, Zn, Sr, and Pb. Some species with unusually low or high concentrations were excluded to avoid error. If the concentration was less than or equal to the method