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Source apportionment of urban fine particle number concentration during summertime in Beijing

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HIGHLIGHTS

- Eight fine particle sources were identified in Beijing using PMF method.
- This was achieved with high-time resolved PSD and PCC data.
- Traffic and combustion aerosol were the dominant sources for particle numbers.
- Regionally transported aerosol and combustions contributed most to particle volumes.
- Local and remote traffic showed distinct size distribution characteristic.

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ABSTRACT

Continuous particle number size distributions (15 nm–2.5 μm), particle chemical compositions, gaseous species and meteorological variables were collected at an urban site in Beijing to investigate the source apportionment of ambient fine particle number concentrations. Hourly data sets were analyzed using the positive matrix factorisation (PMF) which identified a total of eight factors: two traffic factors, two combustion factors, secondary nitrate factors, secondary sulfate + secondary organic aerosol (SOA), fugitive dust and regionally transported aerosol. Traffic (47.9%) and combustion (29.7%) aerosol were found to dominate the particle number concentrations, whereas the most important sources for particle volume concentrations were found to be regionally transported aerosol (30.9%) and combustions (30.1%). Although the diurnal pattern of each of the two traffic factors closely followed traffic rush hour for Beijing, their size modes were different suggesting that these factors might represent local and remote emissions. Biomass burning and coal-fired power plant aerosol were distinguished according to their size modes and chemical species associated with them. Secondary compounds showed similar bimodal particle number size distribution, the distinct diurnal pattern distinguished these factors as secondary nitrate and mixed source of secondary sulfate and SOA. Regionally transported material was characterized by accumulation mode particles. Overall, the introduction of combinations of particle number size distributions and chemical composition data in PMF model is successful at separating the components and quantifying relative contributions to the particle number and volume size distributions in the complex urban atmosphere.

1. Introduction

Numerous epidemiological studies have shown a consistent association of cardiovascular hospital admissions with particulate mass concentrations (Peters et al., 2000; Dominici et al., 2005; Brook et al., 2010). In addition to mass, evidence from many studies indicated that the particle number concentration (mainly ultrafine particles, UFP, diameter <100 nm) may be a cause of adverse health effects, especially cardiovascular diseases (Wichmann et al., 2000; Delfino et al., 2005). Recently, a series of studies conducted in Beijing clearly confirmed the relationship between particle number concentration and cardiovascular mortality (Leitte et al., 2011; Breitner et al., 2011; Liu et al., 2013). Since particles in ambient air are generally mixtures of materials directly

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released from different sources or formed as a product of gas to particle conversion, a better understanding of the source attribution of particles in the urban atmosphere is not only important for investigating the associations between specific particle sources and health, but also for policy makers to introduce effective abatement strategies.

As the capital of China and a major megacity, Beijing has been experiencing great changes in the last few years. The rapid increase of the economy, population and motor vehicles makes Beijing become one of the most polluted megacities in the world. A total of 15 stages of control measures for air pollution have been implemented since 1998 and were enhanced strongly for the 2008 Olympic Games. Primary pollutants such as SO_2 and PM_{10} have decreased steadily in the last 10 years (Chan and Yao, 2008); however, fine particles remain at high levels, suggesting that past control measures have not been effective in reducing emission sources of fine particles. A clear understanding of the sources of fine particles in Beijing will effectively promote the implementation of follow-up abatement policies.

In the past decade, Beijing's atmospheric particulate pollution problem has seen a growing volume of scientific publications on source apportionment attempts. Receptor modelling methods such as positive matrix factorisation (PMF) and chemical mass balance (CMB) were mostly used. These conventional source apportionment studies typically used chemical composition data from filter sampling to provide information on particle matter sources (Polissar et al., 2001; Song et al., 2006, 2007; Xie et al., 2008). Once particles are emitted from a given source, their size, number, and chemical composition change by several mechanisms until they are ultimately removed by natural processes (Seinfeld and Pandis, 2006). Particle number size distributions can help characterise source contributions and even estimate emission factors (Charron and Harrison, 2003; Janhäll et al., 2004; Costabile et al., 2009). However, conventional source apportionment studies cannot provide such information, as only particulate chemical composition is considered in these receptor modelling methods. Furthermore, the filter-sampling period usually lasts 24 h or even longer, obscuring the dynamic changes of particle size, number, and chemical composition. The result is a lack of ready source emission information. More recently, several studies have been made to use particle number size distribution data to elicit source information (Kim et al., 2004; Zhou et al., 2004, 2005b; Ogulei et al., 2006; Ogulei et al., 2007; Krecl et al., 2008; Gu et al., 2011; Harrison et al., 2011). Some of these efforts have involved the simultaneous multivariate analysis of particle number size distributions and chemical composition data (Zhou et al., 2005a; Ogulei et al., 2006; Pey et al., 2009). A new result reported by Wang et al. (2013) using particle number size distributions identified four sources (local and remote traffic emissions, combustion sources as well as secondary transformation) contributed to particle number concentrations during summertime in Beijing. However, more complete source apportionments were impossible unless the particle number size distributions could be combined with highly time resolved particle composition measurements (Viana et al., 2008). Still, high-temporal-resolution of chemical composition data is limited in these studies and obscuring a deeper understanding of the source apportionment.

The objective of this study is to identify possible sources of fine particle number concentrations in Beijing, China. The data set is obtained from an intensive summer observation and consists of one-month samples averaged to an hour resolution. Particle number size distributions (PSD) data set (diameter 15–2500 nm) combined with measured particle chemical composition (PCC) and gaseous composition data was analysed using positive matrix factorisation (PMF). By investigating high-resolution particle number size distributions and particle chemical composition data, it is possible to more clearly identify and apportion contribution from those sources that contribute more to the particle number concentrations than to the particle mass concentrations.

2. Experimental methods

2.1. Sampling site

The sampling site was on the rooftop of a two-story building in the courtyard of the Institute of Atmospheric Physics (IAP, 39°58′N, 116°22′E), which is located 10 km to northwest of the centre of Beijing (Tiananmen Square). The site is approximately 1 km from the 3rd Ring Road, 250 m from the Jingzang Expressway G6, running north-south to its east, and 125 m from the Beitucheng West Road running east-west to its north. A few field experimental campaigns have been conducted at this urban site (Chan et al., 2005; Liu et al., 2012; Sun et al., 2013). This site is surrounded by heavy traffic, restaurants, residential areas, and research institutions. Thus, the observations could be typical of the general urban pollution in Beijing. Sampling instruments were installed in an air-conditioned room approximately 10 m above ground level.

2.2. Data description

An intensive observation period ran from 30th July to 30th August 2011. Particle number size distributions from 14.5 to 710.5 nm (mobility diameter, *D*m) were determined using a scanning mobility particle sizer (SMPS), comprising a model TSI 3080 electrostatic classifier and a model TSI 3775 condensation particle counter (CPC), complemented by a TSI aerosol particle sizer (APS) 3321, which measures particle aerodynamic diameters (*D*a) within a range of 0.5–20 μm. Ambient air was sampled into the SMPS and APS from a 0.5 inch (outer diameter, 2.0-m) stainless steel tube with a low flow PM_{10} inlet. The total flow through the stainless steel tube was 5 L min^{-1}, out of which about 0.3 L min^{-1} and 1.0 L min^{-1} were sampled by the SMPS and APS from two 0.25 inch (outer diameter, 0.5-m) stainless steel tubes, respectively. The relative humidity within the systems was kept below 30% by adding a silica-gel dryer in the inlet line and also in the sheath air cycle to avoid condensation of water in the inlet systems during summertime. The sheath flow of SMPS and APS were 3.0 L min^{-1} and 4.0 L min^{-1}, respectively. Operational parameters (flows and high voltage values) of the SMPS and APS were checked and calibrated before and after the intensive observation period. Size distributions were scanned every 10 min. Size-dependent diffusional and gravitational losses for the inlet line have been corrected by using the empirical functions given by Willeke and Baron (1993). The diffusion loss was estimated to be ~20% for the smallest measureable particles of 14.5 nm, diffusion loss could be negligible for particles between 100 nm and 1000 nm, and estimated to be ~3% for particles of 2.5 μm. The gravitational loss could be negligible for particles below 1000 nm, and estimated to be ~3% for particles of 2.5 μm. The corresponding correction was made for SMPS and APS data. The data collected from these two instruments were averaged into hourly spectra and merged into one particle size spectrum matrix (*D*m: 14.5–2500 nm) following the method of Beddows et al. (2010). The mobility diameter was used throughout the paper. A detailed description about the merging procedure can be found in supplemental materials.

Hourly means of particle chemical composition size distributions (organic matter, sulfate, nitrate, ammonium and chlorine) from 30 to 1500 nm (vacuum dynamic diameter, corresponding to approximately 20–1000 nm in mobility diameter assuming an average particle density of 1.5 g cm^{-3} and a shape factor of 1) during the study period were measured using an Aerodyne high-