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Size-resolved effective density of submicron particles during summertime in the rural atmosphere of Beijing, China

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ABSTRACT

Particle density is an important physical property of atmospheric particles. The information on high time-resolution size-resolved particle density is essential for understanding the atmospheric physical and chemical aging processes of aerosols particles. In the present study, a centrifugal particle mass analyzer (CPMA) combined with a differential mobility analyzer (DMA) was deployed to determine the size-resolved effective density of 50 to 350 nm particles at a rural site of Beijing during summer 2016. The measured particle effective densities decreased with increasing particle sizes and ranged from 1.43 to 1.55 g/cm^3, on average. The effective particle density distributions were dominated by a mode peaked at around 1.5 g/cm^3 for 50 to 350 nm particles. Extra modes with peaks at 1.0, 0.8, and 0.6 g/cm^3 for 150, 240, and 350 nm particles, which might be freshly emitted soot particles, were observed during intensive primary emissions episodes. The particle effective densities showed a diurnal variation pattern, with higher values during daytime. A case study showed that the effective density of Aitken mode particles during the new particle formation (NPF) event decreased considerably, indicating the significant contribution of organics to new particle growth.

Introduction

Atmospheric particles play a profound role in air quality, human health, regional visibility, and global climate change (Dockery and Rd, 1994; Edenhofer and Seyboth, 2013; Laden et al., 2000). These effects strongly depend on their size distribution, chemical compositions, and morphology. The particle properties may vary with their sources and change *via* physical and chemical aging processes in the atmosphere. Density is one of the most important physical properties for atmospheric aerosols, which may link to particle emission sources and atmospheric aging processes. For these complex properties of aerosols, the particle density can act as a good predictor (Pitz et al., 2008). The effective density function is defined as the mass of a particle divided by the volume of its mobility equivalent sphere. In previous studies, effective density was used as a tracer for new particle formation and for aging processes, since density changes in response to chemical reactions, restructuring agglomerates and so on (Katrib

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et al., 2005; Pagels et al., 2009; Zhang et al., 2008). In addition, the size-resolved particle density can be used to determine the relationship between mobility and aerodynamic diameter and to convert high time resolution ambient particle number distributions to mass concentration related to air quality and visibility (McMurry et al., 2002).

The particle effective density can be determined if one of the following combinations is known: mobility size-aerodynamic size, mobility size-particle mass, or aerodynamic size-particle mass. In principle, each individual particle may have different density, so that for a given mobility size, one could then define a density distribution. An obvious way to determine the particle density distribution is to measure the density of a large number of individual particles. Several online techniques that can provide real-time information on particle density have been developed. A novel technique was developed to detect the size-resolved particle effective density in real-time by coupling a tandem differential mobility analyzer with an aerosol particle mass analyzer (APM) (McMurry et al., 2002), and the differential mobility analyzer (DMA) combined with APM system has been applied in extensive laboratory experiments as well as field measurements (Charvet et al., 2014; Malloy et al., 2009; Nakao et al., 2013; Zhang et al., 2008). By using similar technology, the Couette centrifugal particle mass analyzer (CPMA) was first introduced in 2005 as a new particle mass classifier comparable with APM (Olfert and Collings, 2005). The advantage of APM and CPMA is the ability to record the particle mass without the need to collect particles for weighing. Unlike the APM analyzer, CPMA uses a stable system of forces to improve the transfer function of the classifier (Olfert and Collings, 2005). A CPMA along with a DMA was employed to measure the effective density of the particles emitted from a light-duty diesel vehicle (Olfert et al., 2007) and gasoline direct-injection passenger vehicles (Momenimovahed and Olfert, 2015).

Because air pollution in China is different in comparison to Europe and the USA, in terms of the specific pollutant mixtures and the processes involved in pollutant transformation (Guo et al., 2012, 2014; Hallquist et al., 2016), several studies have been deployed in China to investigate particle sources (Guo et al., 2013), chemical compositions (Guo et al., 2010), number size distribution (Wu et al., 2007; Yue et al., 2013), and atmospheric processing (Hu et al., 2017) of atmospheric aerosols in the last decade. However, only a few studies have been carried out to measure the particle effective density in Beijing (Gao et al., 2007; Hu et al., 2012; Yue et al., 2010a) and Shanghai (Gao et al., 2007; Xie et al., 2017; Yin et al., 2015). Gao et al. (2007) reported that the densities of $PM_{2.5}$ (particles with aerodynamic diameter smaller than 2.5 μm) were 1.5 and 1.7 g/cm^3 in Beijing and Shanghai, respectively. Hu et al. (2012) estimated the size-resolved ambient particle density in the winter season in Beijing and found that the effective densities of ambient particles were 1.62 ± 0.38 g/cm^3 for $PM_{1.8}$ and 1.67 ± 0.37 g/cm^3 for PM_{10} by combining the particle number, mass and chemical size distributions.

In this study, we conducted a measurement of the size-resolved effective density of submicron particles in a rural site of Beijing, China using a DMA-CPMA-CPC (condensed particle counter) system during an intensive field campaign. Here, we represent the size dependency of particle density and its link to chemical compositions.

1. Materials and methods

1.1. Sampling site

The intensive field campaign (Photochemical Smog in China) was conducted at the Changping site from May to June 2016. The Changping site (40.14°N, 116.11°E) is located in the northwest of Beijing, about 40 km to Beijing downtown. There were no significant emission sources nearby the sampling site. The instruments were installed in the laboratory sitting on the top floor of a four-floor building. The temperature of the laboratory (around 25°C) was well-controlled by an air conditioner. The relative humidity (RH) of the sampled air was kept to below 30% using a silica gel dryer and a Nafion dryer in series.

1.2. Measurement of the particle effective density

The available data for CPMA was from 14 June to 23 June, 2016. The size-resolved particle density was detected by a DMA-CPMA-CPC system (Olfert et al., 2007). The CPMA (version 1.53, Cambustion Ltd., UK) filtered particles by the ratio of their mass to charge. The CPMA was placed downstream of a DMA (model 3081, TSI, USA), and the combined DMA-CPMA was used to select for the mobility diameter, prior to a condensation particle counter (CPC, model 3772, TSI, USA). The CPMA voltage was scanned across a range of mass-to-charge ratios while the number concentration of particles corresponding to each mass-to-charge ratio was recorded by a CPC. By stepping the voltage and/or rotational speed of the CPMA, the particle number concentration was recorded as a function of the CPMA operating condition. The control software for continuous measurement system of CPMA based on LabVIEW (Computer Software Copyright Registration Certificate No. 2044928, China) was developed to control the instrument.

The particle densities of 50, 80, 100, 150, 240, and 350 nm were measured in each DMA-CPMA-CPC cycle. A DMA-CPMA-CPC cycle could create a density distribution by recording numbers of particles for 30 different densities between 0.30 and 2.60 g/cm^3 for the six distinct particle mobility diameters. Each measurement cycle took about 1 hr to complete. Before the field observation, the DMA-CPMA-CPC system was calibrated using Polystyrene Latex (PSL) particles (47, 64, 102, 202, and 304 nm). The measured average effective density of PSL particles was ρ_{eff} = 1.04 ± 0.01 g/cm^3, which agreed well with the PSL material density (1.05 g/cm^3).

The particle effective density can be calculated by combining mobility and mass measurements assuming a spherical particle. Thus, the particle physical diameter equals the electrical mobility diameter (d_m) selected by a DMA. The effective density (ρ_{eff}) can be calculated by Eq. (1):

$$\rho_{eff} = \frac{m_p}{\frac{\pi}{6} \times {d_m}^3} \quad (1)$$

where m_p stands for the particle mass obtained by a CPMA (Olfert and Collings, 2005).

The CPMA classifies an aerosol stream by the particle mass to electrical charge ratio, m/q. It uses opposing centrifugal and