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Investigation of ambient aerosol effective density with and without using a catalytic stripper

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ABSTRACT

Size-resolved effective densities of ambient aerosols in Riverside, CA were determined over 4 periods during 2015–2016. A DMA-CPMA-CPC technique was used to measure effective density for particles with selected diameters of 50, 70, 101 and 152 nm. A catalytic stripper (CS) was used alternately to remove the volatile fraction of aerosol before density measurements. Aerosol non-refractory composition measurement was conducted in June 2016 campaign to understand the effect of chemical composition on particle density. The average densities for particles over all the measurement campaigns over BP mode (i.e. bypassing the CS) were 1.17 g/cm^3 at 50 nm and 1.25–1.28 g/cm^3 at 70, 101 and 152 nm. The average density after CS conditioning (CS mode) showed a decreasing trend from 1.22 g/cm^3 to 1.04 g/cm^3, with increase in the selected size, and a mass fractal dimension (D_f) of 2.85. Both the BP and CS mode particles showed the lowest effective density at 6–9 am and highest density at 11 a.m.-3 pm. The diurnal variation of density for both modes became more intensive as particle size increased. The variation was also more intense for the CS mode compared to the BP mode. Organic aerosol and ammonium nitrate mass in the size range of density measurements correlated well positively ($R^2 = 0.78$) and negatively ($R^2 = 0.62$), respectively with BP mode effective density. The study provides an update to the aerosol density profiles of a well-known receptor site (Riverside, CA) and investigates the transformation of particles in different seasons. The effective density profiles will be used in a follow-up study to better estimate the respiratory-deposited ambient aerosol mass.

1. Introduction

Atmospheric aerosol plays an important role in human health, air quality, and climate change. Atmospheric aerosol is complex and dynamic in its chemical composition and physical mixing state. Effective density is an important property to understand the mixing state, transport, and depositional characteristics of particles in the ambient atmosphere and human respiratory system. Conventionally, average bulk density is used to convert lung-deposited particle number to mass assuming spherical particle shape, which leads to some degree of uncertainty in the estimate of the deposited mass.

Since McMurry et al. (2002) have developed an online size-resolved effective density measurement method using Differential Mobility Analyzer (DMA)-Aerosol Particle Mass analyzer (APM) technique, this technique has been applied to fresh soot and ambient aerosol to examine morphology and mass-mobility relationships (Geller et al., 2006; Levy et al., 2014; Rissler et al., 2014).

Effective density, ρ_{eff}, is defined as the ratio of particle mass (m) to volume of a sphere with the mobility equivalent diameter (d_m):

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

The mass-mobility exponent (D_m) is an indirect measure of the morphology of irregularly shaped fractal-like particles. D_m can be determined from known effective densities of particles at different mobility sizes (Park et al., 2003):

$$\rho_{eff} = C d_m^{D_m - 3} \quad (2)$$

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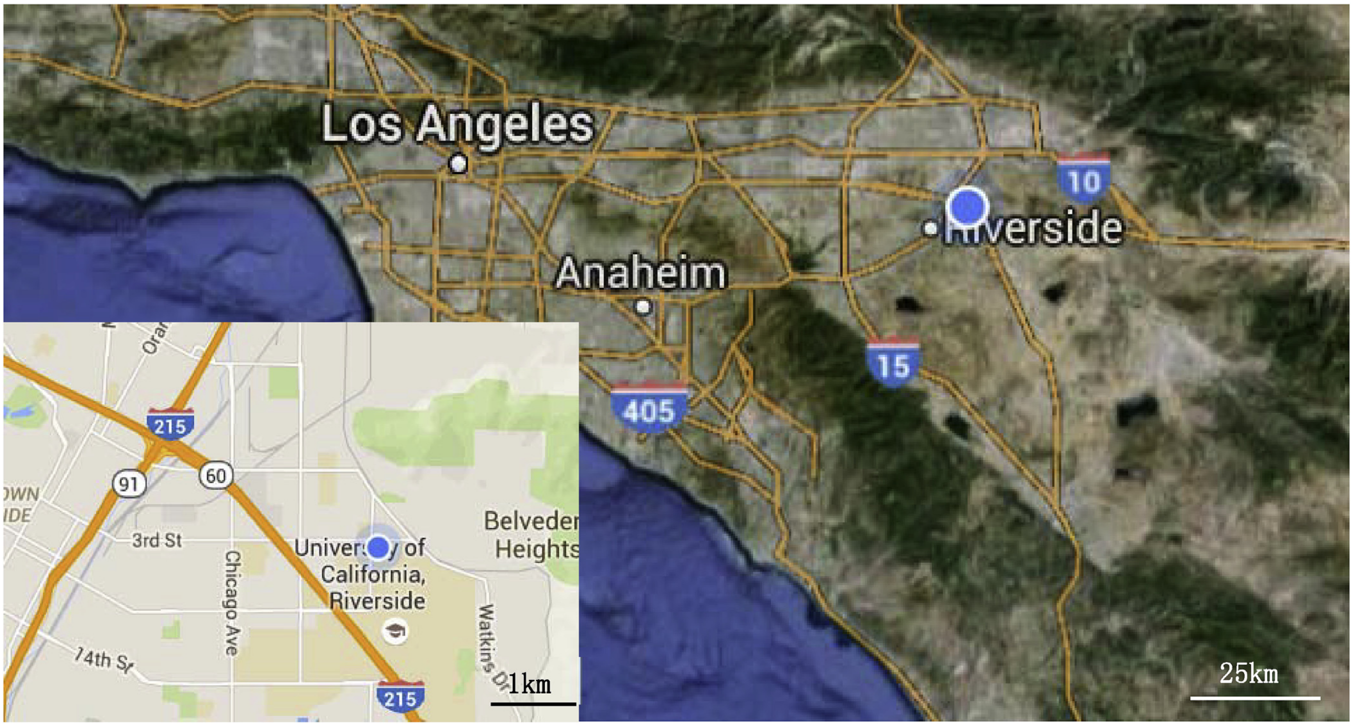
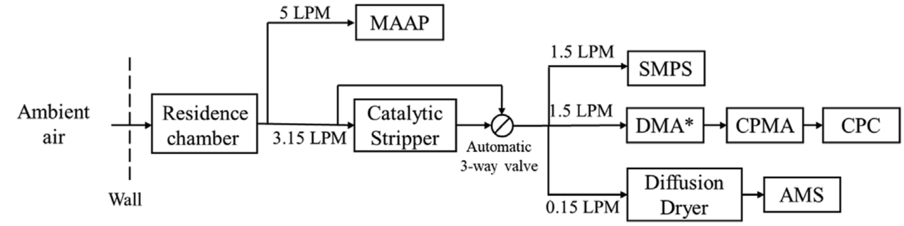


Fig. 1. Map of the sampling site and its distance from the closest highway.



* DMA is operated with sheath flow rate 7.5LPM.

Fig. 2. Schematic of the experimental setup.

The mass-mobility exponent is the same as the fractal dimension, D_f, when the ratio between radius of gyration (R_g) and mobility diameter (d_m) is constant (Maricq and Xu, 2004; Park et al., 2003; Sorensen, 2011).

ρ_{eff} and D_f (or D_m) can be determined by measuring particle mass at different mobility diameters using a DMA- APM system (McMurry et al., 2002) or DMA-centrifugal particle mass analyzer (CPMA) (Olfert and Collings, 2005). The CPMA technique works similarly to APM and has a better instrument inversion transfer function than the APM (Olfert et al., 2007).

The number of studies on particle effective density of ambient aerosols has gradually increased thanks to the establishment of measurement methods and advancement of instrumentation. McMurry et al. (2002) demonstrated capability of DMA-APM system and applied it first for the measurement of effective density of ambient aerosol in Atlanta, GA. They showed the measured density of 1.5–1.7 g/cm^3 agrees with the density calculated based on chemical composition for two selected sizes of 107 and 309 nm particles. Using the same technique, Geller et al. (2006) conducted measurement at various locations in Los Angeles Basin. They observed bimodal density distributions at a location where both traffic and background aerosols were present. They also showed that effective density is driven by photochemistry and meteorology at a receptor site in Riverside, California. The particle effective density of 50 nm particles rapidly dropped from 1.4 g/cm^3 in the mid afternoon to a value of 1.2 g/cm^3 by sunset. The value of 1.2 g/cm^3 is an assumed density for organic aerosol by Turpin and Lim (2001). Spencer et al. (2007) confirmed that effective density varies dynamically, as much as 40%, within 16 h in Riverside, CA during photochemical seasons. They found a correlation between effective density and ambient water content. However, they assumed the correlation might be an artifact due to evaporation in the aerodynamic lens of their aerosol mass spectrometer. Levy et al. (2014) conducted continuous density measurements at US-Mexico border near Tijuana, Mexico for one-week in June, 2010. They reported mixing state and effective density of ambient particles in the size range of 46–240 nm. They showed that the 46 nm particles have the most distinctive diurnal cycle, with the lowest density in the afternoon (at 1–4 pm) likely associated with fresh black carbon emission from vehicles, and the highest in the early morning (at 1–4 a.m.), suggesting the presence of primary organic aerosol. Levy et al. (2013) also conducted density measurement in Houston, Texas. They showed that the effective density has a minimum during morning rush hour and it increases from morning to the afternoon (i.e. 7 a.m. to 5 p.m.) likely due to particle-phase sulfate and oxidized organic components. Rissler et al. (2014) conducted semi-continuous density measurement at an open street canyon in central Copenhagen, Denmark during winter season. They reported both soot and more dense particles were present in 50–400 nm size range. Additionally, they used a thermal denuder in-between DMA and APM and measured the volatile mass fraction of soot as ~10% and those of dense particles as ~80–100%. Yin et al. (2015) conducted a five-week long