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Water soluble fraction of Asian dust particles

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ABSTRACT

The volume fraction (ε) of water soluble material in atmospheric aerosol particles is an important parameter related to their hygroscopicity and activation processes to form cloud and ice particles. To estimate ε of coarse dust particles, confocal scanning laser microscope was applied to measure the volume difference of individual particles before and after water dialysis directly. Individual particles (sphere equivalent diameter approx. 1–8 μm) of Asian reference dusts (CJ1 and CJ2) and atmospheric coarse particles during four Asian dust events were analyzed to ascertain ε. Median values of ε for CJ1 and CJ2 were, respectively, 29% and 13% with no size trend. Median values of ε for coarse aerosol particles during four dust events were 18–42%, which show nearly pure (low ε) to aged (higher ε possibly attributable to addition of sea salts and other water soluble salts) Asian dust. Dust particles with high ε are potentially important for acting as giant CCN. Therefore the aging of dust particles during transport might enhance the number of giant CCN over the North Pacific.

1. Introduction

Atmospheric aerosol particles consist of various components such as water soluble (e.g., $(NH_4)_2SO_4$ and NaCl) and insoluble (e.g. SiO_2) materials. The mixing state of aerosol particles with water soluble constituents, which is an important information for atmospheric sciences (e.g., Junge and McLaren, 1971), is related to direct and indirect effects of aerosol particles on climate (Denman et al., 2007). Mineral dust particles derived from arid and semiarid regions are known to act as efficient ice nuclei (Pruppacher and Klett, 1997). However, the mixing state of mineral dust particles with water soluble salts changes the conditions (temperature and supersaturation) of ice nucleation (Hoose and Möhler, 2012). Mineral dust particles are often mixed with soluble salts by soil salinization at their source area (Abuduwailia et al., 2008; Liu et al., 2011; Wang et al., 2012) and aging processes during atmospheric transport (Andreae et al., 1986; Levin et al., 1996; Niimura et al., 1998; Okada et al., 1990; Sullivan et al., 2007; Wurzler et al., 2000). Although Kumar et al. (2011) reported that freshly emitted "pure" dusts might act as a cloud condensation nuclei (CCN), dust particles with water soluble salts are regarded acting as a giant CCN or as a collision–coalescence initiator during the early stage of precipitation formation (Levin et al., 1996, 2005; Rudich et al., 2002; Wurzler et al., 2000). Recent model simulation has shown that aged dusts with soluble salts can deplete in-cloud supersaturation of water vapor substantially during the initial stages of cloud formation (Karydis et al., 2011). Consequently, the mixing state of mineral dusts with water soluble salts is an important parameter that is useful to characterize their role in cloud-precipitation formation.

Several methods have been used to measure the water soluble fraction of aerosol particles (Kandler and Schütz, 2007). To analyze the soluble volume fraction of individual particles smaller than 1 μm, Okada (1983) used volume differences on single particles before and after water dialysis using an electron microscope. He applied a double-shadowing technique to ascertain the height of particles on the semipermeable membrane (collodion film), and estimated the soluble volume fraction (ε) of individual sub-micrometer particles assuming two model shapes of particles. The ε of individual particles is defined as

$$\varepsilon = \frac{(V_{before} - V_{after})}{V_{before}} \times 100[\%], \qquad (1)$$

where V_{before} and V_{after} respectively denote the volumes of individual particles before and after water dialysis.

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Recently, confocal scanning laser microscopy (CSLM) has been applied to measure the height distribution directly: the volume of particles for the super micrometer size range (Osada et al., 2011). Because the volume measurement by CSLM need not assume a model shape for particles, the results are expected to be more precise than those obtained using the conventional method. Although hygroscopic parameters such as conventionally used ε (Junge and McLaren, 1971) and recently developed κ (Petters and Kreidenweis, 2007) have been frequently estimated for sub-micrometer particles, few studies of super-micrometer dust particles with water soluble salts have been reported (Kandler and Schütz, 2007).

Despite its well-recognized importance, assessments of dust impacts on cloud formation processes and consequent climate remain highly uncertain. Asian dust particles that originate from arid and semiarid regions of the continental Asia are often transported and spread over northeastern Asia and the North Pacific. In this study, to provide basic data related to the mixing state of dust particles at the source and receptor areas, size-segregated ε data were reported for 1) frequently used Asian reference dusts (CJ1 and CJ2) and 2) coarse particles during Asian dust events sampled in Nagoya, central Japan.

2. Method and samples

2.1. Volume measurement of individual dust particles

A confocal scanning laser microscope (CSLM, VK8700; Keyence Co., Japan) was used to measure the individual particle volume. A red laser (658 nm) was used to detect the height of a target with nominal vertical resolution of 10 nm. Magnification of an object lens was used as $\times 100$ with an optical zoom of $\times 6$ or $\times 1$ depending on the dust size. The reproducibility of volume measurements was $\pm 2.5\%$ for particles larger than approximately 2 μm diameter, as evaluated using measurements with standard polystyrene latex particles (1.58 ± 0.02 μm, 1.99 ± 0.02 μm, 3.01 ± 0.02 μm, and 10.00 ± 0.08 μm in diameter; Duke Scientific Corp.). Because the particle volume was obtained as the height distribution based on downward laser measurements, overestimation might occur for a particle having blind or void volumes under the peripheral line. For example, a sphere is observed as a cylinder with a half-size spherical cap at the top, resulting in an overestimation of volume as 25% larger than a sphere. This might be a maximum figure because most dust particles can approximate oblong, rectangular, or disk, and they normally lie flat. Therefore, the overestimation of the blind volume is expected to be much less than 25% for the case of a sphere.

Atmospheric and reference dust particles were collected using an impactor (aerodynamic 50% cutoff diameter is approximately 2 μm at a flow rate of 1 L min^{-1}) with a Nuclepore filter (pore size is 0.05 μm with porosity of 4.7%) as a collection sheet. Reference dusts were suspended by vibration and diluted using clean air to adjust the appropriate superficial density on the collection sheet. Because the Nuclepore filter membrane has a flat and smooth surface, which is an important feature as a datum plane of height measurements by CSLM, it is a superior material for water dialysis. To remove water soluble salts on dust particles, the filter membrane was floated on ultrapure water at room temperature for 3 h with the collection surface side upward (Okada, 1983; Osada et al., 2011). After dialysis, the water-insoluble residue was dried in a desiccator at RH $<$ 10% for more than 6 h. The dust particle volume was measured using CSLM, assisted by macro-size markings and striations on the filter membrane to identify the same particle before and after dialysis. Changes in reflectivity of the dust surface after dialysis were adjustable for measurements. The measurements were conducted under ambient laboratory conditions with RH from approximately 40% to 65%. Because the atmospheric dust samples were also stored in a desiccator, the equilibration history of dust particles with water molecule was almost equal to that before and after dialysis. Efficiency and validity of water dialysis using the Nuclepore filter were confirmed by silt-size test dust particles and were reported elsewhere (Osada et al., 2011). Based on preliminary experiments, ε values of less than 10% are not significant for the existence of water soluble salts because of larger errors in measurements of dust particles having irregular shape, unlike reproducibility (2.5%) for standard latex particles having a smooth and regular shape.

Fig. 1 presents an example of water dialysis for the coarse particles sampled during an Asian dust event at Nagoya, Japan. Upper and lower rows present results obtained before and after dialysis. Left and right columns respectively show maps of reflected laser intensity and height distribution as scaled by the color bar shown at the right. Horizontal red bars represent the scale of 10 μm. In Fig. 1, the volume of larger particles before and after dialysis was, respectively, 22.8 μm^3 and 7.1 μm^3, calculating ε as 69%. Similarly, ε for smaller particle in Fig. 1 was 32%. Although the volume change was large for the larger particle, the difference in sphere equivalent diameter was from 3.52 to 2.38 μm, showing 32% reduction from the original diameter. Fig. 2 shows the relation of equivalent diameters between volume as a sphere and area as a circle based on the projection area for aerosol particles discussed later in this study. Circle equivalent diameters were approximately 1.6 times larger than sphere equivalent diameters, suggesting that the dust particle

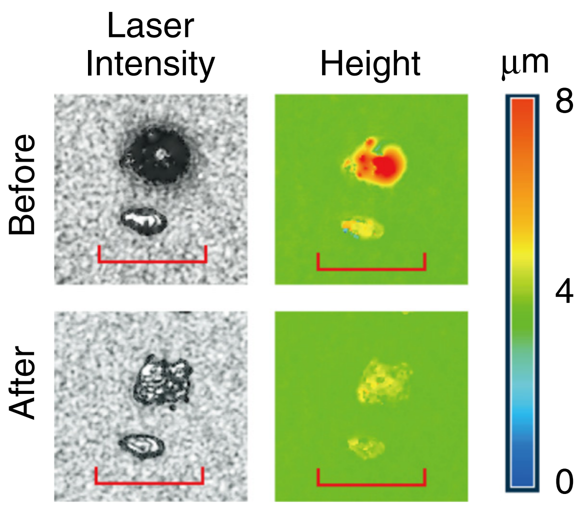

Fig. 1. Examples of the water dialysis of coarse particles sampled on 21 May 2010 at Nagoya, Japan. Upper and lower rows present results obtained before and after water dialysis. Left and right columns respectively show maps of reflected laser intensity and height distribution as scaled by the color bar at the right. Horizontal red bars represent the scale of 10 μm.