

Water uptake characteristics of individual atmospheric particles having coatings

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Abstract

We used an environmental transmission electron microscope to observe deliquescence and hygroscopic growth of atmospheric particles with hygroscopic coatings over the range 0–100% relative humidity (RH). The particles were collected from polluted and clean environments. Types included a sulfate-coated NaCl/silicate aggregate particle, a sulfate-coated sea-salt particle, and a Mg-rich, chloride-coated sea-salt particle. They all exhibited initial water uptake between 50% and 60% RH, although the first major morphological changes occurred at 70% RH. A deliquescence sphere, adjacent to the core particle, formed between 70% and 76% RH when deliquescence occurred or when the liquid phase was able to break out of the solid exterior coating. The deliquescence sphere grew to engulf the particle with increasing RH. Some particles developed a splatter zone associated with a particle coating. Efflorescence occurred over the range 49–44% RH. Our results indicate that some coated particles undergo a multi-step deliquescence process and that composition of the different phases within the coating affects deliquescence and hygroscopic growth below 76% RH. Above 76% RH, the dominant hygroscopic growth was due to water uptake by NaCl. Efflorescence of these particles also was strongly linked to NaCl, although the presence of other phases inhibited formation of a single NaCl crystal. Our results show that the observed coatings can both enhance particle solubility and lower the effective deliquescence RH of the particle. Thus, these coatings cause important phase and size changes for aerosol particles that could feed back into many other chemical and physical processes that contribute to radiative forcing within the atmosphere.

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1. Introduction

Hygroscopic coatings on aerosol particles can alter the ways in which they influence atmospheric chemistry and Earth's radiative budget. In the dry state, they can affect particle surface interactions with light and other atmospheric constituents, e.g.,

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coatings will enhance or retard the reactivity of aerosol particles in the atmosphere (Folkers et al., 2003; Usher et al., 2003). In addition, they can affect deliquescence and hygroscopic growth of the core particle at elevated relative humidity (RH). Change of phase from a dry particle to a solution droplet is associated with changes in size, shape (from non-spherical to spherical), and chemistry, e.g., a heterogeneously mixed solid particle with separate phases in the core and coating, may become a homogeneously mixed solution droplet in which all constituents are water soluble. The solid–liquid phase change of atmospheric particles can also affect their ability to scatter light and their impact on the catalysis of liquid-phase reactions (Martin, 2000).

Coatings on individual particles can be observed using electron-beam techniques in samples from a range of marine and continental environments (e.g., Buseck and Pósfai, 1999; Martin and Han, 2000). Coatings commonly result during mixing of different air masses, e.g., Asian pollution outflows into the Pacific marine air or oil-pollution plumes in desert environments (Levin et al., 1996; Parungo et al., 1992). Coated particles in these polluted environments range from ammonium sulfate on soot to sulfates on mineral dust. Although coatings commonly result from anthropogenic components, natural processes also form coatings. For example, organic molecules on the surface of the ocean can coat the surface of sea-salt particles during wave-breaking action. In addition, aging, oxidation, and reactions with other soluble atmospheric components all contribute to the formation of coatings. Thus, the proportion of coated aerosol particles within a population is likely to increase with time, typically resulting in a greater fraction of hydrophilic particles in the atmosphere.

In multi-component coated particles, both the coating and core compounds can impact the hygroscopic behavior of the aerosol particle. Compounds in coatings can impact the hygroscopic properties of the core phases in two ways: by lowering the deliquescence RH (DRH) (Tang and Munkelwitz, 1993) and by reducing or enhancing overall water uptake of the core phases. For example, some organic compounds are hypothesized to make the surface of an aerosol particle hydrophobic, thus potentially impacting water uptake (Ellison et al., 1999). The insoluble or weakly soluble core phases can also affect the hygroscopic behavior of the aerosol particle, e.g., mineral dust particles can

induce heterogeneous nucleation and efflorescence of aqueous coatings (Han et al., 2002; Han and Martin, 1999; Martin et al., 2001).

Sulfate coatings, in particular, are common on mineral dust and sea-salt aerosol particles. Sulfate coatings can arise from H_2SO_4 deposition after gas-phase oxidation of SO_2 or dimethylsulfide (DMS). Furthermore, they can be created by cloud processing, in which particles scavenge anthropogenic sulfates (Parungo et al., 1992; Wurzler et al., 2000). Coatings on mineral dust have been reported from the Mediterranean and Asian regions, where air masses rich in anthropogenic SO_2 or sulfates interact with desert dust (Formenti et al., 2003; Levin et al., 2005). The heterogeneous reaction of SO_2 with mineral dust is hypothesized to reduce the load of anthropogenic sulfate in the atmosphere because of the increased deposition rate of coarse-mode particles (Bauer and Koch, 2005; Levin et al., 1996; Parungo et al., 1992). Sulfate coatings have also been reported on sea-salt particles in most marine air masses (Li et al., 2003; Pósfai et al., 1995; Sievering et al., 1999). The coatings develop during initial formation of sea-salt particles through interaction of emissions of DMS from phytoplankton or through reaction of anthropogenic sulfates with sea salt (Buseck and Pósfai, 1999; Li et al., 2003; Pósfai et al., 1995; Zhao et al., 2006).

We investigated the physical and chemical behavior of coated mineral and sea-salt aerosol particles over a range of atmospheric RH values using an environmental transmission electron microscope (ETEM). We present results for a sulfate-coated NaCl/silicate particle, a sulfate-coated sea-salt particle, and a Mg-rich chloride-coated sea-salt particle as representative of certain reactions in the atmosphere. Changes in size, shape, and phase of these individual particles from 0% to 100% RH are documented, providing information on the hygroscopic behavior of coated particles.

2. Methods

2.1. Sampling

We studied aerosol particles from the pollution plume of an oil-refinery in the desert near Habshan, United Arab Emirates (UAE, 23/6/2002); marine air at Cape Grim, Australia (ACE-1, 11/12/1995); and polluted marine air at the Scripps Institution of Oceanography in San Diego, USA (SIO, 21/1/2006). Aerosol particles were directly impacted onto

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