



Effect of mixing structure on the hygroscopic behavior of ultrafine ammonium sulfate particles mixed with succinic acid and levoglucosan



Shila Maskey, Ka Yan Chong, Gibaek Kim, Jae-Seok Kim, Arshad Ali, Kihong Park*

National Leading Research Laboratory, School of Environmental Science and Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju, South Korea

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ABSTRACT

Understanding the interactions between water and atmospheric aerosols is critical for estimating their impact on the radiation budget and cloud formation. The hygroscopic behavior of ultrafine (<100 nm) ammonium sulfate particles internally mixed with either succinic acid (slightly soluble) or levoglucosan (soluble) in different mixing structures (core-shell vs. well-mixed) were measured using a hygroscopicity tandem differential mobility analyzer (HTDMA). During the hydration process (6–92% relative humidity (RH)), the size of core-shell particles (ammonium sulfate and succinic acid) remained unchanged until a slow increase in particle size occurred at 79% RH; however, an abrupt increase in size (i.e., a clear deliquescence) was observed at ~72% RH for well-mixed particles with a similar volume fraction to the core-shell particles (80:20 by volume). This increase might occur because the shell hindered the complete dissolution of the core-shell particles below 92% RH. The onset RH value was lower for the ammonium sulfate/levoglucosan core-shell particles than the ammonium sulfate/succinic acid core-shell particles due to levoglucosan's higher solubility relative to succinic acid. The growth factor (GF) of the core-shell particles was lower than that of the well-mixed particles, while the GF of the ammonium sulfate/levoglucosan particles was higher than that of ammonium sulfate/succinic acid particles with the same volume fractions. As the volume fraction of the organic species increased, the GF decreased. The data suggest that the mixing structure is also important when determining hygroscopic behavior of the mixed particles.

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1. Introduction

The hygroscopicity of atmospheric particles (i.e., their ability to attract and retain water molecules from the surrounding environment) is important for the radiation balance, visibility impairment, cloud droplet formation and growth, and heterogeneous chemistry (Liu, Zhang, Cheng, Hu, & Han, 2012; Lohmann & Feichter, 2005; Seinfeld & Pandis, 1998). When aerosol particles are exposed to high relative humidity (RH), the physical, chemical, and optical properties of the particles change due to their increased water content. These changes account for the

largest portion of the uncertainty in estimates of aerosols' effects on climate change (Intergovernmental Panel on Climate Change, 2007). The magnitude of the aerosol particles' effect on cloud formation and radiation balance depends on their size, phase, chemical composition, and hygroscopicity (Finlayson-Pitts & Pitts, 1999; Seinfeld & Pandis, 1998). Therefore, understanding the fundamental interactions between water and atmospheric aerosols is critical for estimating their impact on global climate change.

Atmospheric aerosols encompass a complex mixture of various inorganic (e.g., sea salt, sulfate, and nitrate) and organic species with diverse physical and thermodynamic properties (Clegg & Seinfeld, 2006a; Henning et al., 2005; Marcolli, Luo, & Peter, 2004; Raymond & Pandis, 2003). The organic fraction accounts for 10–70% of the fine aerosols' total mass, depending on the sample location and season (Saxena & Hildemann, 1996). Studies have revealed the importance of organic aerosols on the light scattering, hygroscopicity, and phase transition properties of multi-component atmospheric aerosols (Lightstone, Onasch, Imre, & Oatis, 2000; Saxena, Hildemann, McMurry, & Seinfeld, 1995).

Abbreviations: AIM, aerosol inorganic model; CPC, condensation particle counter; DMA, differential mobility analyzer; DRH, deliquescence relative humidity; GF, growth factor; HTDMA, hygroscopicity tandem differential mobility analyzer; RH, relative humidity; SMPS, scanning mobility particle sizer.

* Corresponding author.

E-mail address: kpark@gist.ac.kr (K. Park).

Therefore, hygroscopic measurements of inorganic species as a single component cannot explain the observed hygroscopicity of atmospheric aerosols during laboratory or field measurements. Many studies have explored the effects of organic compounds on the hygroscopicity of inorganic aerosols utilizing various analytical techniques, such as optical microscopy, hygroscopicity tandem differential mobility analyzer (HTDMA), infrared (IR) spectroscopy, and electrodynamic balance (EDB) methods (Chan & Chan, 2003; Choi & Chan, 2002; Clegg & Seinfeld, 2006b; Cruz & Pandis, 2000; Marcolli et al., 2004; Meyer et al., 2009; Miñambres, Sánchez, Castaño, & Basterretxea, 2010; Park, Kim, & Miller, 2009a; Park, Kim, & Seung, 2009b; Parsons, Mak, Lipetz, & Bertram, 2004; Shi, Ge, & Wang, 2012; Sjogren et al., 2007; Svenningsson et al., 2006; Zardini et al., 2008). Most of the earlier studies investigating the hygroscopic behavior of multi-component aerosols of various organic and inorganic compounds have been performed with internally mixed particles generated from homogeneously mixed solutions. Because atmospheric particles are emitted from various sources, such as vehicles, industrial plants, biomass burning, oceans, and deserts, and undergo different aging processes, such as heterogeneous reactions, condensation, coagulation, and oxidation, they may exist in various mixing structures that include multiple chemical species. Many researchers have reported the existence of aerosol particles in different mixing structures in the atmosphere, such as externally mixed particles (each particle consists of a single chemical species) (Dey, Tripathi, & Mishra, 2008; Ganguly, Jayaraman, Rajesh, & Gadhavi, 2006) and internally mixed particles (each particle contains a mixture of various chemical species) (Bond & Bergstrom, 2006; Dey et al., 2008; Ganguly et al., 2006; Naoe & Okada, 2001; Péré, Mallet, Bessagnet, & Pont, 2009; Zhang, McMurry, Hering, & Casuccio, 1993). The internally mixed particles may exist in different mixing structures, such as well-mixed (multiple components in a homogeneous mixture) and core-shell (a solid core coated with different chemical species) (Adachi & Buseck, 2008; Jacobson, 2001; Shamjad et al., 2012). The organic coating on atmospheric aerosol particles has a significant effect on their hygroscopicity (Chan, Lee, & Chan, 2006). Therefore, the mixing structure of atmospheric aerosols with respect to hygroscopicity is currently one of the largest contributors to uncertainty in the estimation of climate change. However, to date, few laboratory studies have investigated the effect of organic coatings on the hygroscopicity of inorganic solid core particles. Andrews and Larson (1993) concluded that an organic coating film lowered the deliquescence relative humidity (DRH) of pure inorganic aerosol particles. Shamjad et al. (2012) reported that the water absorption enhancement of mixed black carbon particles varies according to the thickness of the shell and core black carbon mass, suggesting the importance of considering aerosol-mixing states while calculating their radiative forcing. Chan et al. (2006) measured the hygroscopicity of solid ammonium sulfate particles coated with water-soluble glutaric acid in two consecutive cycles of hydration and dehydration utilizing the EDB. These researchers concluded that the different deliquescence behavior of the particles between the two cycles of hydration and dehydration were caused by the different mixing structures of the particles (most likely, core-shell and well-mixed particles in first and second cycles, respectively). Although this work explained the cause of the difference in the deliquescence behavior of the mixed particles in two cycles, the hygroscopic properties of the mixed particles with different mixing structures were not fully investigated.

In this study, the DRH and growth factor (GF) of ultrafine (<100 nm) ammonium sulfate particles coated with a certain thickness of water-soluble organic species, such as succinic acid and levoglucosan (i.e., a core-shell mixing structure) were measured

using HTDMA. Additionally, the DRH and GF of internally mixed ammonium sulfate particles produced from a well-mixed aqueous solution of ammonium sulfate and the organic species (i.e., well-mixed mixture) with a similar volume fraction to that of the core-shell mixture were investigated. Ammonium sulfate was selected as the inorganic species because it is ubiquitous in ultrafine- and fine-sized atmospheric aerosols. For the water-soluble organic species, succinic acid (slightly soluble) and levoglucosan (soluble) were selected. Low-molecular-weight dicarboxylic acids, such as succinic acid, commonly occur in the atmosphere due to the direct emission or oxidation of volatile compounds; these compounds tend to condense on preexisting atmospheric particles (Chebbi & Carlier, 1996; Kerminen et al., 2000; Lightstone et al., 2000; Pavuluri, Kawamura, Kikuta, Tachibana, & Aggarwal, 2012). Levoglucosan represents an important fraction of water-soluble organic carbon in atmospheric particles influenced by residential wood burning and wildfires and is recognized as an indicator of biomass-burning aerosols (Hoffmann, Tilgner, Iinuma, & Herrmann, 2010; Mochida & Kawamura, 2004).

2. Experimental

To generate ultrafine particles in a core-shell mixing structure (referred to as core-shell particles hereafter), an atomizer, a differential mobility analyzer (DMA), and a silicone oil bath vaporizer were used, as depicted in Fig. 1. The core particles were generated from an aqueous 0.1 wt% solution of ammonium sulfate (SIGMA ALDRICH, St. Louis, MO 63103, USA) by an atomizer (TSI 3076, USA). The particles were first dried to ~6% RH using diffusion driers with silica gel in outer tubes 53.5 cm long, and particles within a specific size range (70–80 nm) were selected by the first DMA (i.e., monodisperse). Stainless steel and Tygon tubes were used for the system. These selected particles were introduced to a silicone oil bath vaporizer, where a hard glass test tube was partially filled with an aqueous 0.1 wt% solution of organic species (succinic acid (99% purity, Sigma Aldrich, USA) or levoglucosan (99% purity, Sigma Aldrich, USA)) to be used as a coating material using a diffuser (Moteki & Kondo, 2007). The organic liquid was evaporated by heating, and the vapor was entrained into the aerosol flow and subsequently condensed onto the particles after cooling to room temperature, leading to the production of coated particles (i.e., core-shell particles). A diffusion drier with silica gel in its 53.5 cm long outer tubes was used to remove water vapor after the vaporization stage. TEM images of “core-shell” particle support that the coating material is organic. The oil bath temperatures were maintained at ~45 °C and ~50 °C for succinic acid and levoglucosan, respectively. At these temperatures, the amounts of organic vapors evaporated from the solutions were insufficient to trigger homogenous nucleation after cooling to room temperature. Small nanoparticles were not observed in size distribution measurements after the coating step, suggesting that homogenous nucleation did not occur. The resulting particle size was measured using scanning mobility particle sizer (SMPS) containing a DMA (TSI 3080, USA) and a condensation particle counter (CPC) (TSI 3022A, USA). The thickness and volume of the coated material were calculated by assuming the particles were spherical. As examined by microscopy, the produced particles are imperfect spheres. Ultrafine particles in a well-mixed mixing structure (referred to as well-mixed particles hereafter) were produced using an aqueous solution prepared by mixing the species in approximately the same volume fractions as those in the core-shell particles and aerosolized by the atomizer (Sjogren et al., 2007). The generated aerosols were subsequently dried using diffusion driers.

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