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Water uptake of multicomponent organic mixtures and their influence on hygroscopicity of inorganic salts

Yuanyuan Wang[1,2,3], Bo Jing[1,2,3], Yucong Guo[2], Junling Li[2], Shengrui Tong[2], Yunhong Zhang[1,*], Maofa Ge[2,*]

1. The Institute of Chemical Physics, School of Chemistry, Beijing Institute of Technology, Beijing 100081, China
2. Beijing National Laboratory for Molecular Sciences (BNLMS), State Key Laboratory for Structural Chemistry of Unstable and Stable Species, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China

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ABSTRACT

The hygroscopic behaviors of atmospherically relevant multicomponent water soluble organic compounds (WSOCs) and their effects on ammonium sulfate (AS) and sodium chloride were investigated using a hygroscopicity tandem differential mobility analyzer (HTDMA) in the relative humidity (RH) range of 5%–90%. The measured hygroscopic growth was compared with predictions from the Extended-Aerosol Inorganics Model (E-AIM) and Zdanovskii–Stokes–Robinson (ZSR) method. The equal mass multicomponent WSOCs mixture containing levoglucosan, succinic acid, phthalic acid and humic acid showed gradual water uptake without obvious phase change over the whole RH range. It was found that the organic content played an important role in the water uptake of mixed particles. When organic content was dominant in the mixture (75%), the measured hygroscopic growth was higher than predictions from the E-AIM or ZSR relation, especially under high RH conditions. For mass fractions of organics not larger than 50%, the hygroscopic growth of mixtures was in good agreement with model predictions. The influence of interactions between inorganic and organic components on the hygroscopicity of mixed particles was related to the salt type and organic content. These results could contribute to understanding of the hygroscopic behaviors of multicomponent aerosol particles.
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Introduction

In the atmosphere, the interaction between water vapor and aerosol particles has a significant influence on many atmospheric processes. The water uptake of aerosol particles could affect their dry and wet deposition, atmospheric visibility, human health, solar radiation and heterogeneous chemical reactions (Pandis et al., 1995; Poschl, 2005). Atmospheric aerosol particles are generally complex mixtures containing numerous inorganic salts and organic materials, which significantly contribute to the hygroscopic growth of particles. The hygroscopic properties of inorganic salts present in the atmosphere have been well known (Ansari and Pandis, 1999; Colberg et al., 2003). However, the hygroscopic behaviors of organics have not been well characterized due to the kinds of species and uncertainties in hygroscopicity and phase transitions (Decesari et al., 2000; Mader et al., 2004). Field measurements have shown that a large mass fraction of total

* Corresponding authors.
E-mail addresses: yhz@bit.edu.cn (Y. Zhang), gemaofa@iccas.ac.cn (M. Ge).
[3] These authors contributed equally to this work.

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atmospheric fine particles is organic matter, in which water soluble organic compounds (WSOCs) are the major hygroscopic constituents (Decesari et al., 2005; Saxena and Hildemann, 1996). The water soluble organic fraction can affect the water uptake behaviors of mixed particles (Gysel et al., 2004).

According to the method proposed by Fuzzi et al. (2001), complex WSOCs can be divided into three different classes: (1) neutral compounds; (2) mono-/di-carboxylic acids; and (3) polycarboxylic acids (PA). Decesari et al. (2001) found that PA constituted up to 40% of total WSOCs by mass. Due to their structural similarity to humic acid, PA are also called humic-like substances (HULIS). Based on the classification method, a corresponding model substance can be selected to investigate the properties of WSOCs. As a biomass burning product tracer, levoglucosan is usually chosen as the model species of neutral compounds. Succinic acid and phthalic acid can be selected as surrogates of dicarboxylic acids because of their abundant contents in atmospheric particles. Humic acid is used to represent the HULIS (Svenningsson et al., 2006; Zamora and Jacobson, 2013).

Previous studies have mainly focused on the hygroscopic properties of individual WSOCs, especially straight chain dicarboxylic acids and their mixtures with inorganic salts such as ammonium sulfate (AS) or sodium chloride (Braban et al., 2003; Brooks et al., 2003; Cruz and Pandis, 2000; Peng et al., 2001; Pope et al., 2010; Prenni et al., 2001). Only a few studies have involved the water uptake behaviors of multi-component WSOC aerosols (Mochida and Kawamura, 2004; Svenningsson et al., 2006; Zamora and Jacobson, 2013). Marcolli et al. (2004) considered that the interactions between organic components may increase the solubility of single species, and mixed aerosol particles may remain liquid irrespective of ambient humidity. Since the interaction between species has potential impact on the phase and water uptake of aerosols, it is necessary to investigate the hygroscopicity of multicomponent organic aerosols and their influence on the water uptake of inorganic salts. Thus, further study on the hygroscopicity of internally mixed particles containing multicomponent WSOCs with inorganic salts could help to understand the hygroscopic properties and phase state of atmospheric aerosols under different humidity conditions. Furthermore, the measurement data can be used to evaluate the predictive ability of aerosol thermodynamic models (Peng et al., 2001).

In this study, hygroscopic growth was measured using a hygroscopicity tandem differential mobility analyzer (HTDMA) for atmospheric multicomponent WSOC mixtures and their internal mixtures with inorganic salts, including AS or sodium chloride (NaCl). The multicomponent WSOC mixture was composed of a mix of equal masses of four organics: levoglucosan, succinic acid, phthalic acid and humic acid, selected as the model species of the WSOCs. The mixing ratios of WSOCs and inorganic salts were related to the water uptake behavior of the mixed particles. Furthermore, the measured hygroscopic growth of the mixtures was compared with predicted curves from the Extended-Aerosol Inorganics Model (E-AIM) or Zdanovskii–Stokes–Robinson (ZSR) method.

1. Experiment and model section

1.1. Sample preparation

The properties of substances studied in this work are given in Table 1, and the chemical structural formulas of organic compounds are shown in Fig. 1. 100 mg humic acid was dissolved in 100 ml ultrapure water (EASY Pure®II UF ultrapure water system, 18.2 MΩ cm). The humic acid could not be dissolved completely, and the aqueous suspension was filtered with filter paper (Whatman, medium speed). The elemental analysis showed that the mass fractions of carbon, hydrogen and nitrogen in the sample were 41.32%, 3.29%, 1.21%, respectively. In order to calibrate the concentration of humic acid filtrate, a total organic carbon analyzer (TOC, Analytik jena multi N/C 2100) was used to measure the carbon concentration. The concentration of humic acid in the filtrate (g/mL) was calculated from the concentration of carbon in the filtrate measured by TOC (g/mL) divided by the mass fraction of carbon in the solid humic acid sample.

The filtrate with calibrated concentration was used for the following mixed solution preparation. The other three organics (levoglucosan, succinic acid, phthalic acid) in equal mass were added into the filtrate to prepare the MIXORG (levoglucosan, succinic acid, phthalic acid, humic acid, by equal mass) solution, and the amount of each species was equal to that of humic acid. The obtained solution was diluted to around 0.1% total mass fraction. The bulk solutions containing inorganic salt (AS or NaCl) and MIXORG at different mass ratios were also prepared in a similar way.

1.2. Hygroscopic growth measurements

The HTDMA has been fully described in our previous study (Jing et al., 2015) and will be briefly introduced here. The polydispersed aerosols were generated from a constant output atomizer (MSP, 1500) containing prepared bulk solution. After leaving the atomizer, the particles passed through silica gel diffusion dryers (SDD) and a Perma Pure drying tube (PD-100T-24MSS), where the aerosol flow was dried to below 5% relative humidity (RH).

Table 1 – Properties of species used in this study.

Substance	Chemical formula	MW (g/mol)	ρ (g/cm^3)	Supplier/purity
Ammonium sulfate	$(NH_4)_2SO_4$	132.1	1.769	Alfa Aesar, 99.95%
Sodium chloride	NaCl	58.44	2.165	Alfa Aesar, 99.99%
Levoglucosan	$C_6H_{10}O_5$	162.1	1.62	Aldrich, 99%
Succinic acid	$C_4H_6O_4$	118.1	1.57	Sigma-Aldrich, ≥99.5%
Phthalic acid	$C_8H_6O_4$	166.1	1.59	Sigma-Aldrich, ≥99.5%
Humic acid	–	–	1.5	Aldrich

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