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Surface tension of solutions containing dicarboxylic acids with ammonium sulfate, D-glucose, or humic acid

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ABSTRACT

To examine the effect of organic/inorganic solutes on the properties of cloud condensation nuclei and other atmospheric aerosols, the surface tension of various mixtures including dicarboxylic acids, ammonium sulfate, D-glucose, and humic acid sodium salt (NaHA) was measured at 20 °C using the Wilhelmy plate method. An equimolar mixture of oxalic/malonic acids (0–0.5 mol/kg water) was added to ammonium sulfate (0.1–3 mol/kg water), D-glucose (0.01–2 mol/kg water), and NaHA (0.1–10 g/kg water) solutions. The surface tensions of ammonium sulfate, D-glucose, and NaHA solutions decreased as the concentration of the added oxalic/malonic acid mixture increased. The trend in surface tension for the ammonium sulfate solution is roughly similar with and without the addition of the oxalic/malonic acid mixture. However, the surface tension trend for the D-glucose and NaHA solutions with the oxalic/malonic acid mixtures does not follow that of pure D-glucose and NaHA solutions. With the presence of oxalic/malonic acids in the solution, the surface tension increase by D-glucose becomes larger and the surface tension reduction by NaHA becomes smaller.

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1. Introduction

Atmospheric aerosols are well known to influence global climate change via two mechanisms, the direct and indirect effects. Aerosols can absorb and scatter solar radiation (direct effect), or they can modify cloud properties by acting as cloud condensation nuclei (indirect effect). These effects are highly dependent on the properties of the aerosol, and thus its composition. For example, decreases in the surface tension will lower the critical super-saturation required for an aerosol to be activated into a cloud droplet (Köhler, 1936). A lower critical super-saturation increases the number of cloud droplets, contributing to an increase in cloud albedo and a change in cloud life time (*e.g.*, Twomey, 1974, 1977; IPCC (Intergovernmental Panel on Climate Change), 2007; Kanakidou et al., 2005; Novakov & Penner, 1993). Therefore, understanding the physicochemical properties of aerosols is important.

Previous studies have examined the surface tension of aqueous solutions containing an organic component and an inorganic salt (*e.g.*, Tuckermann, 2007; Shulman et al., 1996; Li et al., 1998; Kiss et al., 2005; Prisle et al., 2011; Topping et al., 2007; Raatikainen et al., 2008). According to some of these studies, the effect of inorganic salt on the overall surface tension can be altered by adding organic acid to the solution.

The surface tension of aqueous solutions containing mixtures of water soluble organic compounds also has been measured in previous studies. Tuckermann and Cammenga (2004) measured the surface tension of aqueous solutions

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containing various water-soluble organic compounds and reported that the surface tension of a mixture can be calculated as the sum of contributions from each single component. Henning et al. (2005) proposed that the surface tension of a mixture can be calculated as a weighted sum based on the carbon content of each component. In contrast, Lee (2013) measured the surface tension of various dicarboxylic acid mixtures and found that the overall surface tension closely follows that of the most surface-active dicarboxylic acid alone. These different and somewhat conflicting results may stem from the different mixtures studied. More experimental studies involving the surface tension of multi-component mixtures are needed to reduce these conflicts.

This study examined how the presence of dicarboxylic acids influences the surface tension of ammonium sulfate, D-glucose and humic acid sodium salt (NaHA). These three compounds were chosen, based on their abundance and solubility (*e.g.*, Jia et al., 2010; Caseiro et al., 2007; Pope et al., 2010; Brooks et al., 2004; Hatch et al., 2009; Lide, 2009; Stone et al., 2009), to represent three categories of water-soluble compounds: inorganic salts, low molecular mass water-soluble organic carbon (WSOC), and high molecular mass WSOC (known as humic-like substances or HULIS). Oxalic and malonic acids were chosen to represent the dicarboxylic acid group due to their abundance in the atmosphere (*e.g.*, Braban et al., 2003; Sun & Ariya, 2006).

Surface tensions for these mixtures, spanning a range of concentrations, were measured at 20 °C using the Wilhelmy plate method. The results were fitted either to a linear equation (for ammonium sulfate and D-glucose mixtures), or the Szyskowski equation (for NaHA mixtures). With these parameters, the effect of dicarboxylic acids on the physicochemical characteristics of the solutions could be examined.

2. Experimental procedures

2.1. Materials

The properties of all compounds used in this study are listed in Table 1, including the chemical formula, molecular mass (g/mol), density (g/cm^3), solubility (g/100 g of water), and manufacturer information.

2.2. Surface tension measurements

A thermostated tensiometer (Krüss K11, Germany) based on the Wilhelmy plate method was used to measure the surface tensions of mixed solutions. The Wilhelmy plate method (Hyvärinen et al., 2006; Aumann et al., 2010) is frequently used to measure the surface tension of a solution, along with the Du Nouy ring (Henning et al., 2005; Tuckermann, 2007) and pendant drop method (Svenningsson et al., 2006; Asa-Awuku et al., 2008).

Details regarding the equipment and the procedure for surface tension measurement can be found elsewhere (Lee, 2013). Briefly, a thin and small platinum plate, which hangs on the balance, measures the equilibrium surface tension of the aqueous solution at the air–liquid interface. The solution container (a small beaker) was surrounded by water in a larger beaker to create a double boiler system, as shown (5) in Fig. 1. Temperature control of the water bath was achieved via a thermostat regulator. This configuration transfers heat uniformly to the solution and suppresses variations due to the ambient air temperature; it also allowed the use of a smaller volume of solution for measurement. Figure 1 also shows a schematic of the tensiometer – the surface tension of the aqueous solution was measured by submerging a small platinum plate several times. The same plate was used for all measurements. Prior to each determination, the plate was rinsed multiple times with Milli-Q water (18.2 M) and acetone, and then flame dried to remove residual chemicals.

Table 1
Chemical properties of organic compounds.

Compound name	Chemical formula	Molar mass (g/mol)	Density (g/cm^3)	Solubility (g/100 g)	Manufacturer
Oxalic acid	$C_2H_2O_4$	90.03	1.900[a]	12[b,e]	Sigma-Aldrich
Malonic acid	$C_3H_4O_4$	104.06	1.619[a]	161[b,e]	Alfa Aesar
Ammonium sulfate	$(NH_4)_2SO_4$	132.14	1.77[a]	76.4[a,e]	Sigma-Aldrich
D-glucose	$C_6H_{12}O_6$	180.17	1.562[a]	81.8[a,f]	Sigma-Aldrich
Humic acid sodium salt (NaHA)	N/A[c]	> 2000[d]	N/A[c]	N/A[c]	Sigma-Aldrich

[a] Lide (2009).
[b] Summarized in Saxena and Hildemann (1996).
[c] No data available in MSDS.
[d] Pope et al. (2010).
[e] Solubility at 25 °C.
[f] Solubility at 15 °C.

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