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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ternary solution of sodium chloride, succinic acid and water – surface tension and its influence on cloud droplet activation" *by* J. Vanhanen et al.

J. Vanhanen et al.

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The referee has reviewed the paper and given suggestions related to the language of the paper, and has raised also some questions concerning the surface tension parameterization and cloud model simulations. The authors therefore propose to submit a revised manuscript that takes the aspects noted by the referee into account. The changes are listed below. The comments of the referee are italicized.

The language of the abstract should be refined in order to emphasize the novel contributions of this work. Also, it should be made clear in the abstract that the parameterization was extrapolated from concentration range of the experimental data; "An equation based on thermodynamical relations was fitted to the data" is understated to

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the point of being misleading. This is described quite clearly and satisfactorily in the first paragraph of the conclusion, perhaps try to emulate that in the abstract.

-The description of the fitting procedure and the extrapolation of the data will be elaborated in the abstract.

Introduction, paragraph 2: since the authors are focusing on NaCl as a sea-salt proxy they should mention the incorporation of organics at the sea-surface when sea salt aerosols form via bubble bursting (Gershey, 1983). The authors should also provide citations of field observations of succinic acid in marine aerosol.

-Direct observations of succinic acid in marine aerosols can be found in Legrand et al. 2007. The connection of organics and sea salt will be deliberated more in the text. Also the fact that the organics at the sea surface have a contribution on the organics mass in marine aerosol particles will be mentioned (Gershey, 1983; Ellison er al., 1999; Tervahattu et al., 2002a, b).

Section 2.3: While the density estimation in equations 2-3 is an elegant approach, measuring the density of a liquid is a simple matter, so it is not clear why the theory is necessary. The authors should at least "spot check" the theory results with a few direct measurements of the density of their ternary solutions at different concentrations to show consistency.

-The density was point checked by performing additional measurements. The deviation between the estimation and the measurements was less than 0.2%. These results will be added to the revised version of the paper.

Section 3.1.2: "Surface tension of the solution decreased with increasing temperature as expected." This statement should be supported with a reference.

-Reference for surface tension lowering due to increase in temperature will be added (Bikerman 1970)

Section 3.1.2: Please provide more discussion regarding the differences in these re-

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sults vs. the observations of Tuckerman (2007) and Kiss (2005.)

-More discussion concerning the differences in the results of our work and the work of Tuckermann (2007) and Kiss et al. (2005) will be added.

The authors should provide an estimate of the uncertainty of the data in Table 2 based on propagation of the measurement uncertainty, and its impact on the parameterization.

-The estimation of the uncertainty of the parameterization is discussed in the section 2.1. The corresponding sentence is elaborated.

Extrapolating surface tension measurements in this system out of the range of concentrations measured is problematic. For one, the authors assume that the aqueous phase of the aerosol will become supersaturated in both succinic acid and sodium chloride. However, as the aerosol becomes supersaturated in NaCl, the succinic acid may be "salted-out" of solution and crystallize (Setschenow, 1889). This should be commented upon directly in the text.

-The salting out will be considered more in the text while comparing the results to the results of Tuckermann (2007) and Kiss et al. (2005).

An additional important point for discussion is that the authors neglect the formation of micelles or other phases as particle composition temperature changes, while these phenomena can change surface tension and water uptake (Cistola et al., 1988;Tabazadeh, 2005). The authors make this omission clear in the text, but given the potential significance of these phenomena to CCN activation, they should provide a justification based on published values of the critical micelle concentration, etc. for why this is an acceptable assumption. The authors characterization of these processes as "macroscopic phenomena" and continuing on to say "despite these uncertainties, the extended parameterization is applicable in models simulating microscopic phenomena such as nucleation" misses the point.

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-The referee pointed out the importance of micelle formation when considering CCN activation. We made calculations on the concentrations of succinic acid in critical supersaturation. This way we could assess the possibility of the micelle formation in the liquid. The maximum mole fraction of succinic acid used in the surface tension measurements is 0.0075. Using the geometric mean value of the Aitken mode (45nm) the mole fraction of succinic acid at the critical supersaturation is 0.003. Because we did not find micelle formation in our surface tension measurements, this means that micelle formation will not affect the CCN activation. Although there is no micelle formation taking place at the concentration range relevant to the CCN activation, it might take place at higher acid concentrations. Unfortunately this is not accounted for by our parameterization. This fact will be clarified in the text.

- CCN activation is characterized as macroscopic phenomena due to the fact that we can be sure about the phase of the droplets and therefore bulk properties of the mixtures can be used. In nucleation the situation is different. Nucleation is the first step of a phase transition, and will always take place before any other phase related phenomena, such as micelle formation can occur. The parameterization obtained from the measurements and estimations are done in order to obtain the best estimate of surface tensions of the mixture beyond the solubility limits, which is needed in nucleation models. These types of parameterizations have been used earlier by Gaman et al. 2004, Hyvärinen et al. 2006 and Riipinen et al. 2007.

The authors should also provide an explanation, if there is any, for why it makes physical sense to extrapolate from the surface tension of an ordered surface film of succinic acid on an aqueous substrate to that of a supercooled succinic acid liquid, and likewise from an aqueous NaCl solution to a molten NaCl salt.

- This is done in order to get additional information on the surface tension of the mixture beyond the solubility limits (see e.g. Gaman et al. 2004 and Hyvärinen et al. 2006. Sensitivity analysis by Gaman et al. (2004) shows that this surface tension estimation method has reasonable accuracy for atmospheric nucleation studies.

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Conclusion, final paragraph, first sentence: "relevance" should be replaced by "impact" or similar.

-In the conclusions in the final paragraph: relevance will be changed to impact or similar.

Overall, the paper needs to be edited carefully for English language. Many misunderstandings may arise from language errors in this manuscript: for example, the first complete sentence after equation 7 reads "Few assumptions had to be made in order to obtain a fit that represents surface tension of the ternary solution beyond the solubility limits (Table 1)" Whereas surely the authors meant "A few assumptions" and in fact it would be more accurate to say "Several assumptions".

-The paper will be re-edited for English language.

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