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Discussion Paper



Interactive comment on "The vapor pressures and activities of dicarboxylic acids reconsidered: the impact of the physical state of the aerosol" by V. Soonsin et al.

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Received and published: 1 December 2010

Comments to Referee # 1

The authors would like to sincerely thank anonymous referee # 1 for his/her constructive comments. We have addressed the referee's concerns point-by-point as shown in the following. Our comments are given in italic.

(1) Even though the estimation of vapor pressures of solid particles has been described in detail previously, it would be helpful if more specific details were included in this manuscript. From the authors' previous Optics Express article, it is clear that the simulating the particle morphology is key to retrieving vapor pressures. However, there is very little information given in this manuscript, for example, as to what the radius actually means for a particle that is presumably non-spherical.

We changed and extended the corresponding description to state explicitly that we apply the equivalent sphere radius approximation for retrieving radius change rates. However we do not want to discuss all the details which support the validity of this assumption since these details are already published. New description reads (page 20519, lines 24-26): "For solid, in general non-spherical particles, we need to consider the actual particle shape. The only information we gain from the TAOS pattern is the non-sphericity of the solid particle but not its actual shape. Using an equivalent sphere radius approximation we can still deduce evaporation rates from optical resonance spectroscopy by assigning a size parameter to a specific resonance in the spectra and following its temporal evolution. This is explained in detail in Zardini et al. (2009, 2010). The results depend only slightly on whether the equivalent sphere radius is assigned to the minimum enclosing ball radius of the non-spherical particle or its mean radius. We estimated the relative error in vapor pressure using this approximation to evaluate the optical resonance spectra together with uncertainties in gas phase diffusivities to be 35%."

(2) Given the uncertainties in estimating vapor pressures from the resonance spectrum for solid particles, the authors should say why they used this method to retrieve the evolving radius of the particle, rather than measuring the mass directly from the DC balancing voltage in the EDB measurement.

It is possible to retrieve the evaporation rate directly from the mass change data, which appears to be especially attractive for solid particles because the ambiguity of assign-

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ing an equivalent sphere radius does not arise. However, for low vapor pressures with corresponding small evaporation rates, the precision in the mass change rates is limited by the overall drift of the DC balancing voltage feedback loop, which relies on a video image of the particle. This becomes especially important for small particles. It turned out that only for high vapor pressures (corresponding to temperatures above ca. 25°C) the pressures inferred from mass change data are of the same precision as those inferred from the optical resonance spectra of the solid particles. We added this explanation to the text.

(3) The argument made that the relative evaporation rates recorded are those for amorphous and crystalline glutaric acid in Figure 4 is convincing. Figure 9 shows a similar diagram for succinic acid that includes the efflorescence point. It would be helpful if the glutaric acid data set were presented in the same way. In Figure 9, it is unclear from the caption what time periods the two lower evaporation rates were determined over.

We do not have similar data for glutaric acid, because the relative humidity has to be constant during a measurement to be able to evaluate the evaporation rates. Thus it was an exceptional occurrence in the case of succinic acid (Fig. 9) that we observed efflorescence by chance while we intended to study the evaporation of the liquid at constant RH. To perform an experiment on purpose would mean to run it for extremely long times with extremely slow rates of drying to meet the constant RH requirement and allowing efflorescence to happen. We did not attempt to perform such an experiment for glutaric acid. We added the time period (150 ks to 213 ks) for the lower evaporation rate to the caption of Fig. 9.

(4) An article has recently appeared in the Journal of Physical Chemistry A (ASAP) which appears to present vapor pressures of dicarboyxlic acid components with and without an inorganic component by Clegg and Cox (DOI: 10.1021/jp1052979). It might

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be appropriate to consider the consistency of this work with the current study.

The mentioned article is now published (Pope, F. D., Tong, H.-J., Dennis-Smither, B. J., Griffiths, P. T., Clegg, S. L., Reid, J. P., and Cox, R. A.: Studies of Single Aerosol Particles Containing Malonic Acid, Glutaric Acid, and Their Mixtures with Sodium Chloride. II. Liquid-State Vapor Pressures of the Acids, J. Phys. Chem. A, 114, 10156-10165, 2010.) and we include their data for malonic and glutaric acid in Table 3 as well as in Fig. 13 in the revised version and compare it with our measurements. The agreement for the supercooled melt vapor pressures is excellent for both acids.

Minor typographical/grammatical errors:

(1) Line 15 page 20517 and lines 3 and 7 page 20518: These sentences using the words 'allow(s) to' are grammatically incorrect and should be reworded as 'over binary solutions allow the organic activity to be directly obtained...', 'only techniques which allow the vapor pressures of semivolatile substances to be determined...', and '...that it allows the unambiguous identification of the physical state...'

We have changed the above sentences as suggested by the referee.

(2) Line 4 page 20527: sentence starting 'Because' does not make sense.

We have deleted 'Because'.

(3) Figure 3 caption: the filled square symbol should be a circle.

We have changed the filled square symbol to a filled circle symbol.

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