

Interactive comment on “Cloud droplet activation and surface tension of mixtures of slightly soluble organics and inorganic salt” by S. Henning et al.

S. Henning et al.

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Referee 2

We would like to thank the referee for the useful and constructive comments.

General comments

The referee asks if it is possible that kinetic effects in the CCN chamber are responsible for the difference between dry and wet particle behavior – could it be that the dry particles do not activate because they do not get enough time in the chamber?

The intensity of scattered light (at an angle of 45 degrees) is measured as a function of time and the particles get 20 seconds to activate. We re-checked the scattered light signal as a function of time for all mixtures presented (Figure 6). The time until the peak

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of this signal is less than 10 second for all particle sizes investigated and for both wet and dry conditions. Since the peak of the signal corresponds to an activated fraction close to one, and since this occurs at a time significantly shorter than 20 seconds it seems reasonable to believe that wet as well as dry particles have sufficient time to activate in the chamber under all conditions studied. This may not be the case for all organic aerosols and it is something that we will investigate in the future.

We agree with the reviewer that in the atmosphere particles in a below-cloud environment might have a long time to equilibrate to ambient relative humidity. This will however not affect the critical supersaturation of the dry particles studied herein since they have to cross a saturation ratio barrier above one to activate.

Specific comments

P. 7465: text is changed to: It has recently been suggested (Li et al. 1998, Sorjamaa et al. 2004) that partitioning of surface active organic compounds between bulk and surface in activating droplets affects the critical supersaturation. This was shown experimentally and theoretically for sodium dodecyl sulfate (SDS) particles and theoretically for particles consisting of ammonium sulfate and cis-pinonic acid. Since the surface activity of the compounds studied herein is small the effect of partitioning is not taken into account in this work.

p. 7566: “as follows” is added to the text.

p. 7469 We ascribe the non-zero values at the lowest Sc values to double charged particles as explained Bilde and Svenningsson 2004. We estimate that the accuracy of a single measurement of the number of activated particles at one given supersaturation is 20 %. This explains that values can be below and above one. To improve data-quality we do however not use single measurements, but take averages of several scans.

p.7470 The data in Figure 3 do not have the DMA transfer function applied and the lines are just connecting the individual data points. The text on page 7470 line 6 has

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been changed to: The critical supersaturation is the point of 50 % activation and the DMA transfer function was taken into account when finding this point.

P. 7471 Line 1-2 has been appended to the prior paragraph to make it clear that they are connected to Figure 4. We did not measure surface tensions of solutions of the pure compounds since they already exist in the literature. Figure 4 is quite busy as it is and we therefore prefer not to include the pure compounds.

p. 7471: The figure text has been clarified:

..with diameters of 50 and 100 nm for pane (a) and (b) and 40, 50 and 100 nm for pane (c).

Figure 6 Great care was taken to ensure proper presentation of all data points and readability of the legend text. Moving the legend to pane (a) would affect the readability. Therefore we prefer to keep the legend in pane (c) and add the following text to the figure caption: Please note the legend in pane (c).

Text: The text has been modified to give a more detailed introduction to Figure 6:

Experimentally obtained critical supersaturations for initially dry particles (solid squares) and solution droplets (open circles) are given in Figure 6. Theoretical critical supersaturations ($S_{c,theo}$) calculated using the surface tension of water (σ_w) are shown as grey lines and $S_{c,theo}$ calculated using the surface tension at activation given in Equation 7 (σ) are shown as black lines. In both cases unbroken lines represent particles that were initially dry and dashed lines represent particles that were initially solution droplets. It is clear from the measurements that there is a pronounced difference between the critical supersaturation of initially dry particles and solution droplets (containing the same amount of organic acids and NaCl) when adipic acid is the dominating species. For a 90 nm particle containing 93% adipic acid the critical supersaturation is 0.32% for an initially dry particle. In comparison the critical supersaturation is 0.48% if the particle initially existed as an aqueous solution droplet. This dependence of particle

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phase is related to the solubility of the organic compounds as explained previously and as can also be seen from Figure 1. When the particle is initially dry it has a solid core at activation and the Köhler curve has two or three maxima. The first two maxima can be seen as a barrier to activation caused by the solid core. When the particle exists initially as a solution droplet there is no such barrier and activation follows the "traditional" Köhler curve corresponding to infinite solubility of the organic compounds. The difference in critical supersaturation between particles with different phases is dependent on the initial dry particle size and the effect is largest for the smallest particles. It can be seen from Figure 6 that as adipic acid is replaced by the more soluble succinic acid the importance of particle phase decreases because succinic acid is more soluble than adipic acid. Solution droplets of the same size but different chemical compositions activate at quite similar supersaturations while critical supersaturations for initially dry particles (of the same initial size) can span a wide range. For solution droplets experimental values and theory taking the parameterized surface tension of the solution into account agree very well for all mixture types. In case of dry particles another effect seems to dominate the lowering of surface tension and the small particles activate at a higher critical supersaturation than predicted.

Page 7471 eq. 6 and 7 and Figure 6: To make the connection between the surface tension terms described in eq.6 and 7 and the surface tension terms used to obtain the theoretical dry diameter / critical supersaturation lines in Figure 6 clearer we applied uniform variable names in the equations and the figure legend.

Minor technical comments: We agree with the reviewer and made changes according to the suggestions of the reviewer.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 7463, 2004.

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