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Comment

## ***Interactive comment on “Increased cloud activation potential of secondary organic aerosol for atmospheric mass loadings” by S. M. King et al.***

**S. M. King et al.**

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The authors thank the reviewer for the helpful comments, which are shown below in bold. The author responses are in normal font, and modifications to the manuscript are shown in italics.

**This is an interesting manuscript, clearly worthy of publication in ACP. However, I believe that the optimization with respect to surface tension has not been carried out in a thermodynamically consistent way, and the authors should redo the exercise for the final paper. A 10% reduction of surface tension at the critical size means that the organics in the droplet are quite surface active, and will be highly concentrated in the surface layer. At the same time, they are depleted from the**

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bulk of the droplet, which causes the Raoult term to become smaller, counter-acting the surface tension influence on critical supersaturation. This "surface partitioning" effect, which follows from Gibbs surface thermodynamics and finite size of the droplet, has been discussed in a number of recent papers (see list below). Lab experiments (Sorjamaa et al., Prisle et al.) have provided strong evidence that the effect is real.

Sorjamaa R. et al., ACP 4, 2107 (2004) Sorjamaa R. and Laaksonen A., J. Aerosol Sci. 37, 1730 (2006) Kokkola H. et al., GRL 33, L10816 (2006) Topping D.O. et al., ACP 7, 2371 (2007) Prisle N. et al., Tellus 60B, 416 (2008) Duplissy J. Et al, GRL 35, L03818 (2008) Wex H. et al., J Atmos. Sci. 65, 4004 (2008)

We agree with Professor Laaksonen that the suggested reduction in surface tension may be influenced by the possibility of partitioning from the bulk phase to the surface layer. However, the analysis suggested requires more specific knowledge of the properties of the condensed phase than is available with the current data set. The condensed phase in the system described is composed of an unknown large quantity of organic species, and many assumptions have already been presented for the existing analysis. As suggested by the reviewer, we consulted the Supplementary section of the Duplissy et al., who "employed a model substance whose properties were altered until the  $GF_D$  and  $S_{crit}$  were reproduced." In order to include the surface partitioning effect, they had "altogether five parameters that can be adjusted within some limits to model the hygroscopicity and critical supersaturation." These parameters include compound-specific constants used in the van Laar equation and Szyskowski equation, which are currently not included in this manuscript's analysis. Therefore, the proposed analysis, given the large number of free parameters necessary, is beyond the scope of this manuscript.

Moreover, we emphasize that the reduction of 10% in surface tension is inferred from sensitivity analyses described in the manuscript, under the condition that all other pa-

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rameters are held equal to the initial values, and is presented in order to highlight the greater sensitivity of CCN activity to surface tension in contrast to other physicochemical parameters. Part of the intent and scope of this manuscript is the presentation of a simple Köhler framework that can explain the CCN activity of particles with complex chemical composition.

Nevertheless, Professor Laaksonen makes a good point regarding the potential effect of surface partitioning. In indirect partial response, we note that some of our modifications to the text in response to reviewers #1 and #2 broaden the scope of explanations to possible factors other than surface tension. In a more direct response, we modified the text accordingly, as follows:

[pg. 1685, lines 23+] *"The sensitivity to another possibility, namely a 10% increase in the van't Hoff factor  $i_{\text{ORG}}$  at lower loadings (e.g., resulting from an increase in the relative fraction of carboxylic acids, which dissociate, or from nonidealities introduced by interactions between the organic and the inorganic components), is approximately equivalent to that of a 25% decrease in  $V_{m,\text{ORG}}$  (King et al., 2007). Combined changes within the particle can also be more complex than represented in our model. For example, the possible partitioning of more surface-active molecules from the droplet interior to the surface layer can change the droplet composition sufficiently to increase the water activity, thereby counteracting the effect of surface tension reduction (Sorjamaa et al., 2004; Prisle et al., 2008). This kind of possibility is not explicitly considered in our analysis."*

**I would also like to point out that Duplissy et al. (see supplement) carried out somewhat similar sensitivity analysis as has been done in the present ms, by altering surface tension (taking account of surface partitioning), molar volume of the organic, and water activity coefficient. A comparison might be worthwhile.**

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We have cited the related paper by Duplissy et al. in the text, but further comparisons are not explicitly made by us because the observed decrease in CCN activity in this study occurs at organic loadings that are lower than those reached in Duplissy et al. In the revised text, we have emphasized this point.

**As a minor point: there are no page limitations in ACP, why is a separate supplement needed?**

Our thinking is that we wanted to present a focused document to the reader to get across our points. Therefore, we made decisions about what text, tables, and figures to include in the main text that we think will be of interest to most readers. We also felt that some readers will also be interested in the more detailed accounts, so we included that information in the supplementary material.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1669, 2009.

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