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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud condensation nuclei closure study on summer arctic aerosol" *by* M. Martin et al.

Anonymous Referee #2

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This manuscript presents results of a closure study on CCN measurements conducted during the ASCOS campaign. In their analysis, the authors fitted the experimental measurements using the k-Khöler theory to constrain values for korg. Subsequently, the authors used the best fit permutation parameters to derive ktot during the campaign. The most interesting aspect in this study is that model and measurements could not be brought into agreement for the highest supersaturations, suggesting an increase in the organic fraction composition with decreasing particle size with respect to the average composition measured by an AMS instrument. The paper is well written, with plenty of details on the measurements and procedures applied. To my view, this is a high quality study which provides an interesting contribution and deserves being published in ACP. I have however, some concerns, regarding the analysis conducted by the authors to constrain korg and ktot and conclusions therein derived, that should

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Discussion Paper

be addressed before publication in ACP.

Major comments (text between " " are manuscript quotations)

-Assuming that the dissolution behaviour of marine biogenic organics deviates from non-ideality (i.e. korg=constant) variations of korg between different supersaturations (i.e. particle sizes) in this study could be attributed, as the authors postulate, to an underprediction of the organic composition with respect to the average mass fraction measured by the AMS instrument. Under this hypothesis, it is pertinent to think that the underestimation of the organic fraction in turn leads to underestimations of korg, thus explaining the decreasing value of korg required to fit model and measurements for increasing supersaturations. Because of this underestimation, the values of korg in this study would not be valid to provide a lower estimate for the hygroscopic parameter. It seems difficult to constrain a lower range for korg from these measurements without information on size-resolved particle composition. For this reason I strongly recommend the authors to state that they found an upper estimate for korg of 0.2 and that further study is needed to provide a lower estimate for this parameter. Statements in abstract and text that korg as low as 0.02 is necessary to fit the results and model would not be valid, as this is very likely to be an underestimated value. It is interesting to note that the upper range found for korg in this study is consistent with the korg range between 0.073-0.164 found for marine biogenic organics in the CCN closure by Fuentes et al. (2011). A reference to this work would certainly support the findings in this study.

-Derivation of ktot using the parameters from a particular permutation (permutation 13) leads to an overprediction and underprediction of the CCN number on different periods of the cruise (page 8819 and Figure 6). This is due to the fact that ktot needs to be constrained using the extreme values defining the uncertainty range for the fitting parameters (i.e. korg=0-0.2 and density=1-1.6), rather than using a particular permutation case. I strongly recommend the authors to re-analyse this part of their study in this manner, as it will provide adequate upper and lower estimates of ktot that will be useful for comparison in future field studies.

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-The abstract needs to be more concise and less ambiguous. For instance, the abstract should clearly state that findings in this study are 1) an upper range for korg of 0.2 and 2) a lack of agreement between measurements and model at high supersaturations which suggests increasing organic mass fractions for decreasing particle size. It would be very helpful to include information on the estimates for ktot and on the average mass organic fraction found with the AMS (i.e. 36%) in the abstract. General statements should be more consistent with the findings. For instance, the abstract should state that the upper range of korg in this study points at the organic material being from sparingly soluble to effectively insoluble. The uncertainty range for korg is too large to state that the marine organics are purely insoluble.

-Can the authors provide information of the sensitivity of the CCN number calculation to variations in density and korg? are the calculations more sensitive to korg than to the density value?

-Page 8814, lines 28-29: "Surface tension of water (0.072Nm$-$1 at the given temperature in the laboratory) was assumed for all calculations." This assumption needs to be better supported. Because the organic matter could suppress the surface tension of aerosol particles, the authors should discuss why they used the surface tension of water for their calculations and how this assumption affected their conclusions. I do agree, however, with the authors that this assumption is valid, since marine organics have been shown to present low surfactancy properties, with reductions of the surface tension from 0.5-5% at the point of activation for compositions similar to those in this study (Fuentes et al. , 2011). Indeed, a reduction in the surface tension would also lead to larger deviations between estimations and observations, which also support the authors' assumption.

-Conclusions (8820, line 13). "Assuming an internally mixed aerosol and an insoluble or only slightly soluble organic volume fraction." I recommend replacing "slightly" soluble by the term "sparingly" soluble organic fraction, which is a more common term in the literature to define compounds of limited solubility (Petters and Kreidenweis, 2008).

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The “slightly soluble” definition may induce to think that it is nearly insoluble matter, while korg could be as high as 0.2.

-Conclusions (8820, line 18) “One way to explain this is by assuming that the smaller particles have a different composition than the larger ones, presumably a non-hygroscopic organic fraction.” As stated above, the korg uncertainty range (korg <0.2) is too large to state that the marine organics are purely non-hygroscopic or purely insoluble. The fact that korg needs to approach zero to bring measurements and model into agreement in some cases is due to the underestimation of the organic fraction. Rather than stating that the organic matter is non-hygroscopic it is more rigorous to state that it presents low hygroscopicity.

-(8820, line 23) “This means, that the organic fraction of the aerosols was nearly non-hygroscopic and does thus not contribute to droplet growth.” As discussed above, an upper value for korg of 0.2 is certainly low but not non-hygroscopic, so there should be some contribution to the particle growth. Please, modify text accordingly.

-(8820, line 27) ktotal needs to be better constrained using the korg uncertainty range between 0-0.2 and density between 1-1.6, as described above, rather than using the parameters of a best fit permutation. This part of the conclusions needs to be updated with new upper and lower estimations of ktotal.

-The fact that korg presents a value <0.2, and that the droplet surface tension at activation can be assumed equal to pure water, implies that for a given particle size, an increase in the particle organic enrichment would lead to a depression of the CCN activity and hygroscopicity of the particles. I believe this would be a relevant finding, worthy to be included in the abstract and supported with similar conclusions in Leck et al. 2002 and Fuentes et al., 2011 in the discussion section.

Minor comments

Abstract

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-“For the two highest measured supersaturations, 0.73 and 0.41%, closure could not be achieved with the investigated settings concerning hygroscopicity and density. The calculated CCN number concentration was always higher than the measured one.” The last sentence may lead to confusion because it seems to refer to the whole dataset, i.e., all supersaturations.

-“At 0.20, 0.15 and 0.10% supersaturation, the measured CCN number can be represented with different parameters for the hygroscopicity and density of the particles.” This sentence is confusing. I guess the authors mean that different combinations of the hygroscopicity parameter and density allowed fitting the model to the experimental measurements?

Introduction, results and discussion

-Page 8805, lines 4-6 and page 8806, lines 3-4. “However, the hygroscopic properties, the cloud nucleating ability of these biogenic particles, and their source and sink strengths are still not well understood.” The authors should also mention that some work has been done to explore the CCN properties and hygroscopic properties of particles enriched with marine biogenic organic matter (e.g. Moore et al. 2008, Fuentes et al., 2011), that shows that marine organics depress the hygroscopicity and CCN nuclei activity of particles.

-Page 8815, lines 5-11. I do not think that Figure 2 is necessary since it does not provide any additional information with respect to results in Figure 3. In fact, Figure 2 is difficult to interpret, without first knowing about the calculations set presented in Figure 3.

8814, lines 26-27: How are the cases of insolubility for the organic fraction implemented in the model?

-8818, line25, “Permuation”- change to permutation 8819, line6, “The value is varying between 0.1 and 0.4 over this time period”.-change to” The value varied between 0.1

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and 0.4 over this time period"

-page 8820, line 16, "Hygrosopicity" change to hygroscopicity

-8820, line 21) "Results from counter 1 at 0.10% SS give an upper limit of _org = 0.2, since the assumption of a more hygroscopic organic fraction results in overpredicted CCN concentrations." What do the authors mean here by "more hygroscopic"? hygroscopicity above 0.2? please, clarify this in the text.

-I recommend checking the text for typos.

References

Fuentes, E., Coe, H., Green, D., and McFiggans, G.: On the impacts of phytoplankton-derived organic matter on the properties of the primary marine aerosol – Part 2: Composition, hygroscopicity and cloud condensation activity, Atmos. Chem. Phys., 11, 2585-2602, 2011.

Leck, C., Norman, M., Bigg, E. K., and Hillamo, R.: Chemical composition and sources of the high Arctic aerosol relevant for fog and cloud formation, J. Geophys. Res., 107(D12), doi:10.1029/2001JD001463, 4135, 2002.

Moore, R. H., Ingall, E. D., Sorooshian, A., and Nenes, A.: Molar mass, surface tension, and droplet growth kinetics of marine organics from measurements of CCN activity, Geophys. Res. Lett., 35, L07801, doi:10.1029/2008GL033350, 2008.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 8801, 2011.