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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1 – Evidence from measurements" *by* H. Wex et al.

H. Wex et al.

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Answer to Referee 2:

The authors thank the Referee for his kind comments.

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Your comment: The work presented is solid, well-documented, and merits publication in ACP. I note no substantive issues that would preclude acceptance in its current form. I do have two suggestions that might guide the authors' future efforts (and might be commented on, at least, in the current work). First, as was noted by the other reviewer, the formation conditions for the SOA in these experiments was very different

from those found in the ambient atmosphere. Since recent studies have suggested that these differences can affect some aerosol properties, it would be useful to repeat the study with more realistic conditions.

Our response: We agree with the reviewer that mass loading effects may be important and deserve further study. We now discuss the effect of different aerosol mass concentration we used in our study and included how this compares to literature results (at the end of the Discussion section, p. 17, line 7 ff): " ... . However, organic aerosol mass concentrations in different experiments we conducted varied from 3 to 1000 micro g/m3, but we did not observe significant changes in LACIS- or CCNc-derived hygroscopicities. This is consistent with the observation of King et al. [2009], who examined a-pinene/O3 (dark ozonolysis) particles that were internally mixed with ammonium sulfate, and who only observed an effect on hygroscopicity at mass concentrations that were lower than those achieved in our study. The extent to which different compounds (differing in their functional groups) were present in SOA in our experiment, as compared to SOA examined in Duplissy et al. [2008] or present in the atmosphere, can depend on the actual precursors and on the conditions under which SOA formation and subsequent aging takes place. Nevertheless, our work clarified the origin of the previously often reported gap between low hygroscopic growth and good CCN activity of laboratory produced SOA particles. It raises a note of caution for future atmospheric studies by pointing out that assuming a constant hygroscopicity over the whole range from water sub- to super-saturation might be erroneous."

To achieve more realistic conditions we make the following note: the SOA generation system as used in our study cannot generate aerosol mass concentrations less than 3 micro g m-3. To achieve such low organic aerosol concentrations the use of seed particles becomes necessary as was demonstrated by King et al. (2009) in ACPD. Although it is possible to retrieve organic aerosol hygroscopicity from such data through mixing rules, it is necessary to then separate mass loading effects from non-ideal, i.e. non-ZSR, mixing effects. Thus there are considerable experimental challenges to

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accomplish atmospherically relevant mass loadings. This is further complicated by the challenge to more accurately simulate the SOA chemistry under laboratory conditions. Such simulations will require more accurate control over NOx, HOx, and other VOCs that are present in the atmosphere but not, or in the wrong proportions, in the reactor. Also it will be necessary to include multi-generational evolution of the SOA composition. These effects are clearly beyond the scope of this study and will hopefully be addressed in future studies on the subject.

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Your comment: Second, there were at most two repetitions for any of the experiments presented here. The formation of SOA is complex enough that conclusions on so few experiments must be viewed with some skepticism. Without ignoring the significant effort required to make these measurements, the work would be considerably improved by repeating the experiments and presenting the additional data.

Our response: We conducted more experiments than we showed in this manuscript. When we repeated the same experimental conditions (see e.g. exp-4/exp-7 and exp-1/exp-2, as shown in the manuscript) we obtained similar results, although these experiments were done on different days (which included turning on and off of the instrumentation and particle generation in between). As we wanted to test the dependency of the SOA on different experimental variables (ozone and water-vapor concentration), we did not have a chance to repeat the same experiment more often. However, in general, the three different SOA-types showed similar behavior, with only some small dependencies on the ozone and water vapor concentration, so we are confident in the repeatability of our data, especially since those experiments that we repeated agreed so well. We stress this stronger now in the manuscript now (p. 11, last sentence (continued on p. 12)): ”These repetitions of measurements always took place on different days, and the similarity in the measured values shows the good reproducibility of the experiments.”

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Interactive Comment

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Your comment: The figures are overall clear and helpful for understand. My lone critique is that the contour lines on Figures 7 and 8 are rather closely spaced for such small figures, to the point that it is difficult to see the key result that the data systematically cross the contours.

Our response: Although we agree with the reviewer that the isolines are tightly spaced, we prefer to keep the figures as they are.

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Your comment: In terms of presentation, the most outstanding feature is the need for additional proofreading prior to final publication. There are too many glaring mistakes to ignore- most should have been found and removed prior to submission. To note two examples, there is a grammatical mistake in the very first sentence of the abstract, and the titles of sections 4.1 and 4.2 are identical.

Our response: We carefully proof-read the manuscript and eliminated typographical and grammatical mistakes.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 955, 2009.

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Interactive Comment