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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 2: Theoretical approaches" *by* M. D. Petters et al.

M. D. Petters et al.

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We thank the two reviewers for their comments on the manuscript. The original reviewers comments are in regular type. Our responses to the comments are in *italic type*.

Reviewer #1

General comments

In this manuscript, the authors have investigated when information on the hygroscopic growth of secondary organic aerosol (SOA) at RH<100% can be used to predict the cloud condensation nucleation properties of the particles accurately. The motivation for the work lies in previous studies where contrasting results were obtained in this respect. The authors present two theoretical models to explore the phenomena and through performing model calculations, are able to give a sound explanation for the observations. The manuscript clearly fulfills the criteria for publication in Atmospheric Chemistry and Physics, and therefore I recommend the manuscript for publication after the authors have considered the following minor remarks.

Specific comments

Regarding atmospheric implications of the work. The results suggest that the cloud condensation nucleation properties of atmospheric SOA can be parameterized using a single parameter, the so-called kappa parameter, of which value varies over a relatively narrow range (the last paragraph of Section 4). However, the conclusion is based on laboratory studies employing only a few aerosol precursors and oxidation mechanisms that represent only a small fraction of conditions met in the atmosphere. Therefore, I'd be more careful about making such a conclusion. The authors could elaborate this point a little bit further.

In the last paragraph we primarily discuss the similarity of CCN activity for α-pinene and other monoterpene precursors and it was not our intention to extrapolate this result to all SOA in the atmosphere. We changed the wording to reflect this: "Although this view disregards the complexity of the underlying water relationship it suggests a concise parameterization of the activation behavior of monoterpene-derived SOA particles can be formulated."

Technical comments

1. Equation 11. The order of square brackets and brackets should be reversed in the right-hand side of the equation.

Corrected.

2. Page 20853, line 6. Should be "hygroscopicity", not "hygroscopicy".

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Corrected.

3. Page 20854, line 16. Replace "in" with "at".

Corrected.

4. Acknowledgements. Should be "ACCENT", not "ACCNET".

Corrected.

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Reviewer #2

This paper is a second part of a study where hygroscopic growth and cloud droplet forming potential of secondary organic aerosol is explored. Compared to previous studies hygroscopic growth is successfully studied at RH 's between 95% and 99.6%, and it has been found that information from that range can be used to estimate supersaturation needed to activate those particles. The authors have chosen two models to explain observations, and especially usage of a non-ideal model and relating discussion merits publication. I found this manuscript very interesting, and suitable to be published in Atmospheric Chemistry and Physics after minor revision. However, as the manuscript is the second part and uses data presented in the first part, I cannot recommend publication before the first part is also published as a separate paper.

Overall comments:

The Part I manuscript (Wex et al., 2009) is now posted on ACPD and the discussion phase is completed. We will coordinate submission of the revised versions of both manuscripts.

1. Why did you choose to use two-component system with ideal model. I did not find any proof that it would behave clearly better than the single-component system.

We believe that there is a slight misunderstanding about the two-component model. It assumes ideal ZSR based mixing (ideal mixing of nonideal components), not ideal solution behavior overall. The reason for introducing a two-component model is that it is frequently invoked to conceptually explain the behavior of organic aerosols (e.g. Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics of β-caryophyllene secondary organic aerosol, Atmos. Chem. Phys., 9, 795-812). Our main point is that the observations can be fit to either model and that agreement with any particular model in itself does not constitute proof for either model to be correct.

Related to that, why have you chosen to use two components also in case of non-ideal model for type 2 aerosol but only one component for types 1 and 3. At the moment it seems that the choice of the number of components is quite arbitrary.

Both models, the pseudo-binary hygroscopicity model and the non-ideal model are applied for all types studied as shown in Fig. 2 and Fig. 4. We therefore fully analyze all of our data assuming either one or two components. The purpose of this analysis is to demonstrate that even detailed data are insufficient to constrain the number of effective components.

2. Could you please use more clear notation in the equations. After careful reading it is clear that gf^3 means cube of growth factor, but as g is also Gibbs free energy and f is defined by equation 7, the notation gf^3 is not so clear.

We clarified this issue by referring to the hygroscopic growth factor directly by D/Dd instead of gf.

Minor comments:

What does xi stand for in Eq 5.? If it is a mole fraction, how could it even get values larger than 1.

In Eq. 5 x_i *denotes the dissolved volume fraction of the solute, as defined in Raymond and Pandis (2003) and Petters and Kreidenweis (2008). The dissolved volume fraction cannot be greater than unity and hence the constraining function* $H(x_i)$ *is needed. We did not realize that this definition conflicted with the later definition of mole fraction. We therefore have changed the notation in Eq. 5 and added additional explanation.*

page 20840, lines 6-7: Why is the system non-ideal at RH less than 98%. It is quite contradicting if you state in the next sentence that both models, ideal and non-ideal, give equally good agreement with the measurements.

We believe that there is some misunderstanding about the pseudo binary mixture of infinitely water soluble compounds and sparingly soluble compounds. This model does

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not assume ideal solution behavior. It assumes that two distinct classes of compounds are presents, that each can be modeled using an effective hygroscopicity parameter (which subsumes all non-ideal behavior into one parameter), that one of the classes compounds is only sparingly soluble, and that the two classes of compounds do not interact which each other. We therefore do not see that there is a contradiction in those statements.

page 20846, line 4: What effect?

We reworded the sentence to be more specific: "In the presence of water vapor during the reaction (Type III) the aerosol is more hygroscopic than both Types I and II."

page 20848, line 19: Would it be better to say that "SOA can be comprised of hundreds of components" as you do not know it for sure based on your experiments.

We changed to wording as suggested.

page 20849, line 25: Water activity coefficient is always dependent on mole fractions for non-ideal solutions.

The reviewer is correct. However, we meant to convey a different point. The water mole fractions that are inferred from the data at a specified water activity depend on the parameter f*. The uncertainty in* x_w *is estimated based on a range in* f *and this range is indicated in the leftmost panels of Figure 4. Unfortunately the error in inferred mole fractions cannot be plotted the same way since the derived activity coefficients will also depend on the choice of* f*. This was not clearly communicated and we revised the sentence as follows: "We note that uncertainties in the mole fraction shown in the left panel cannot be plotted in the same way in the middle panel because the error in derived water activity propagates into the derived activity coefficients, i.e. a larger* x_w *(calculated from a smaller assumed* f*) implies a smaller* γ_w*."*

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 20839, 2008.

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