

Interactive comment on “Widening the gap between measurement and modelling of secondary organic aerosol properties?” by N. Good et al.

Anonymous Referee #1

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This manuscript describes the hygroscopicity of freshly generated SOA and the difficulties connected to both the measurements and the modeling trying to link the sub-saturated data to supersaturation and cloud droplet activation. It is clear that in this study, instrumental differences were rather large, which of course affects the modeling. The authors discuss the possible causes for these differences in a clear way and have produced a well written manuscript. I recommend this paper to be published, after modifications and clarifications described below.

General comments:

1) It would be preferable to move some of the system description included in chapters

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3.1 – 3.3 to chapter 2.1. There are also references to specific system setups described in Duplissy et al. (2009) which would be very helpful to have also in this paper. I suggest that you include a table describing the different H-TDMA systems with respect to residence times between Teflon bag and system drier, times between driers and DMA1, times between the two DMA:s, closed loop or open end, operating temperatures (which may be important for the gas to particle partitioning of the organic vapors) etc. and the possible artifacts that can come from the different setups. Everything that you later discuss in chapter 3 and which would be much easier to follow with a table describing the systems.

2) Reading the paper it was not really clear what different experiments were done and what measurements were carried out in the seeded and unseeded experiments. In chapter 2.1 you for instance state that subsaturated water uptake was measured using 3 different HTDMAs, but it is not clear that you are talking about only the unseeded experiments. I would suggest dividing this chapter into two parts, describing the seeded and unseeded experiments separately, what measurements were done and at which times etc., so that you cannot misunderstand which measurements techniques were used on which experiment.

Specific comments:

1) Page 22621 row 27: You say that you cannot say if any of the processes are important, but in fact at least one of them is important, since it was not possible to ideally predict the activation super saturation ratios. Do you mean that you cannot distinguish between them or that your differing H-TDMA results make you not trust the subsaturated data? You also say that you cannot say if the water activity coefficient changes significantly as a function of solute concentration, but according to Figure 2, the kappa values from the Manchester H-TDMA seem to be increasing with RH. Is this not a significant trend or do you dismiss it because of the differing results from the two other H-TDMAS?

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- 2) Page 22626, line 5: HTDMA is spelled wrong.
- 3) You describe a number of possible explanations for the difference in H-TDMA results. The H-TDMA systems are all referenced, but since systems are continuously rebuilt, I would like to know if closed loop were used for all 6 DMAs. Considering that you have a lot of newly condensed semivolatiles which can be quite sensitive regarding particle to gas partitioning, this can be of importance. An open end DMA will introduce new “clean” gas phase which will lead to an evaporation from the particles and hence a decreased growth factor.
- 4) For the seeded experiments, the kappa values of the SOA changed with time, is this time dependency included in the seeded experiments?
- 5) Page 22626 line 7: After RH, there is an extra “)” included.
- 6) Page 22627, line 3: “Beacause the CCN activation. . .”. Why is this the case? Can you explain this a bit more thorough?
- 7) Page 22627, line 5: “In each sampling mode. . .”. What does sampling mode mean, is it one dry size or a span of saturation ratios or something else?
- 8) Figure 1, right panel: Are these data points a mix of 100, 105 and 150 nm dry size particles? In that case you should include “theory” including the Kelvin effect and specify which points are which. Otherwise just use 100 nm and specify that in the paper.
- 9) Page 22628, line 8: “relatively unimportant”. Specify for the reader who does not know the Kelvin effect by heart. It is about 1%.
- 10) Page 22628, line 16: “only the deliquesced. . .” Why? I would like you to include the rest of the points. Was the Manchester curve “smeared” or sharp? What about the other systems? A smeared deliquescence point may like you say indicate temperature gradients in the second DMA, or if it the same for all systems – maybe a slightly contaminated salt solution. Hard to follow your discussion without the points included in

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the plot.

11) Why are you using the parameterization of Suwanee river Fulvic acid? I would expect Suwanee river Fulvic acid is more surface active than the fresh SOA that you are measuring. Can you somehow motivate this choice?

12) Page 22631, line 20: “nominal detenction efficieny”. Detection is spelled wrong, and what do you mean by “nominal”? I guess you mean the 50%-value, but it not clear.

13) Page 22631, line 20: “Several hours after..” What is it that quickly decreases to zero? That nucleation completely stops is not connected to the wall losses, so the sentence is a bit hard to follow.

14) Page 22631, line 22: “Using an estimate..” Please divide this sentence into two parts. It is too long.

15) Page 22632 line 4 and below: It is a bit hard to follow this without the figure in Duplissy et al. It would be helpful to include it in this paper also, since it is essential to this study.

16) After how long time are the SOA humidograms in Fig 2 taken, how long did they take, which of the two pure SOA experiments do they come from and what dry size are they performed at? In figure 2 it says “for the unseeded SOA experiments”. Does this mean that both experiments are in this figure? This is not clear.

17) Page 22635, line 3: Why did you use 500 g/mol? Did this give the best ADDEM fit to CCNC data? Alphapinene has a mass of 136 so I guess you presume some oligomerization?

18) Page 22638, line 17: “each HTMDA appears to converge towards a model scenario”. 1) HTDMA is spelled wrong. 2) I do not understand what you mean; does it converge with increasing or decreasing diameter? I cannot see this. Please clarify.

19) Why does including surface to bulk partitioning increase the predicted critical su-

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persaturation values? Should not soluble entities that were “hidden in the surface” in the first approximation (only using the surface tension parameterization) migrate from the surface to the bulk as the water activity increases, hence increasing the number of free entities in the solution and decreasing the critical supersaturation. At least if the partitioning is included both for sub- and supersaturation, this should be the case.

20) Fig.4 “for each molecular weight and density pair”. Why is the number of green circles different for different Dry sizes? Which pairs were used?

21) If I understand correctly, no surface to bulk partitioning was used in the seeded experiments, only a parameterization of surface tension. Why is this?

22) It is unclear why you use 90% RH values for the kappa parameter for the seeded experiments, when the solubility seems to increase above 90%, at least for the PSI system.

23) Figure 4: This plot is hard to read. I suggest making lin-lin plots, with saturation ratios on both axis. Use x-coordinate from measured supersaturations and the y-coordinate for the modeled ones. This way it will be much easier to see the trends and absolute numbers.

24) Figure 4: Why do the ADDEM model including surface tension and surface to bulk partitioning always give the “correct” Sc independent of the HTDMA data? In chapter 2.3.1. you state that the surface tension parameterization is based on Suwanee river fulvic acid, but in the abstract you say that reconciliation required widely different assumed surface tension behavior. What surface tension is really included in the black crosses in figure 4?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 22619, 2009.

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