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Atmospheric Chemistry and Physics Discussions

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

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General comments:

- The first objection I have is, that, after reading the paper, remain Unconvinced with the general form of the message/conclusions of the paper. This can be remedied with either: additional data on measurement quality assurance, or by reducing the general conclusions to being slightly more specific (for instance: abstract line 22-26 and 4. Conclusions p 22643 line 14-19).

- If you would choose to be more precise on the experimental conditions, that would result in a high-quality paper and it would enhance the citation possibilities of this paper,

I think.

We would like to thank the reviewer for their comments, they are much appreciated. We hope that we are able to answer these general points in response to the specific comments below.

- In detail:

- 1) The authors write in Abstract line 24:

By proven what is meant is that the instrument performance has been validated against a nebulised aerosol of known composition, which is described in section 2.2. Additionally these are instruments which have been used extensively to measure hygroscopic growth.

To clarify this point the word "proven" is replaced with "validated and extensively used".

- 2) "quality assurance procedures"

Yes this refers largely to Gysel et al. 2009. Extra details are added after this statement to quantify the quality assurance steps taken:

"The raw data in checked on a scan by scan basis to ensure there are no anomalies in the in the recorded counts, temperatures, pressures, flows and RHs. Scans with less than 50 total counts are disregarded. When running at a constant RH any scans where the average RH is outside 1.5% of the set point are disregarded. "

- 3)

Good agreement is defined by agreement with the theoretical data within the measurement uncertainty. In the case of each instrument the agreement is within experimental error. This is clarified in Sect. 2.4 by replacing the word "good" and stating that the agreement is consistent with the estimated uncertainties

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For the measurements shown an average GF(RH) would not really make sense as humidograms are being run.

Taking the growth factor of 1.72 (the growth factor closest to the theoretical value measured by all 3 HTDMAs) we see that the RH set by the HTMDA is within the stated accuracy of the dew point mirrors. For HMAN: RHHTMDA=90.6 RHADDEM=90.2, for HPSI: RHHTMDA=89.9 RHADDEM=90.6 and f or HQUT: RHHTMDA=90.8 RHADDEM=90.6. The difference in the theoretical (ADDEM) values is due to the Kelvin effect. We note that the data points specifically noted: Hpsi 1.75 (a touch lower than estimated by the reviewer) is at 90.25% RH the theoretical value at this RH is 1.704 and therefore falls within the uncertainty. Similarly the difference between the HMAN value at 1.675 and theory is 0.052. We therefore compare the modelled and measured RHs close to 90% RH in the revised text.

- 3b) In figure 1 right hand panel only simultaneous measurements are presented so all environmental conditions were identical. More measurement points are available though not always with each instrument measuring simultaneously and over the same RH range. The number of HTDMA data points in the original figure was 34. This has now been extended to 71, to show the measurements down to 60% RH as requested by reviewer#1. Many more data points could be should from different humidograms, however no more information can be gained. The measurement uncertainty (indicated by the error bars) is due to 2 main factors

i. there is an intrinsic measurement uncertainty in the HTDMA technique due to precision of instrument this is assumed to be $\sim\pm 0.02$ in growth factor space.

ii. there is the accuracy and precision of the RH measurement. All 3 HTDMAs utilise dew point mirrors to measure the RH, with an accuracy of $\sim$1.2% at 90% RH. The change in GF(RH=90%)=1.7 for ΔRH=1-1.5% is $\sim$0.04-0.06.

Thus the for a well calibrated HTDMA instrument measuring pure ammonium sulphate at 90% RH the measured GF is expected to be within about 0.05 of the theoretical

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value. The error is then calculated in the same way for all points on the humidogram in Fig 1. Therefore as the large majority of the data points agree with the theory within error we ascertain that the criteria within Duplissy et al 2009 which states that the GF bias should be less no more than ∼3% is met.

The uncertainty for the pure organic humidograms is calculated in the same way and shows when there is and isn't a difference in the measured GFs beyond the calculated uncertainty consistent with the definition within Duplissy et al 2009.

An explanation of the error bars in added in section 2.4.

- 3c)

The QUT H-TDMA (Hqut) has been previously tested at high humidities (above 90%RH). An example of the HGF measurements with the Hqut. of Ammonium sulphate. at humidities up to 97%RH has been previously published in Johnson et al 2008 (see figure 7.). Good agreement with the theoretical prediction has been observed confirming the quality of the Hqut data measured at high RH.

Johnson et al 2008 is referenced in the manuscript.

- 3d) Reviewer #1 makes a similar observation. Each HTMDA measured at a different dry diameter, this is clarified in the text. The theoretical values are so close they are covered by the original line.

- 3e) What are the error bars in right panel of Fig 1?

This should have been made clearer. Our response to 3b. gives details.

- 4)

Each main point is addressed one by one:

i. Yes, we meant Figure 2. This is corrected.

ii. The range of kappa values was chosen to represent the full range measured by each

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HTDMA and covers full RH uncertainty range at 90% RH for each HTDMA. In growth factor space this is slightly larger for HPSI because the magnitude of the growth factors measured is larger. The choice for the kappa range is clarified in the text.

iii. We believe the conclusions are valid given that the HTDMAs have been shown to operate within the limiting accuracy of their respective RH measurements (1-1,5%). The important thing to realise is that it is uncertainty in the GF as a result of the RH uncertainty which causes it to be larger than $\sim$0.02 when the HTDMA is operated humid. The magnitude of the uncertainty in GF will therefore be larger when the change in GF as a function of RH is larger such as for ammonium sulphate at 90% RH compared to SOA with a GF of 1.15 for example. As stated previously the criteria in Duplissy 2009 are met for ammonium sulphate.

iv. We have changed proven to “validated and extensively used” to clarify what we mean in the abstract.

v. And extensively quality assured is added to the sentence.

vi. It is possible if you believe your HTDMA measurement. Our point is (in this paper), that there is no definitive reason to disbelieve one of the HTDMAs. Clearly further work is required to determine and resolve the measurement discrepancies

vii. "Conclusions" p 22643 line 14, for instance, "SOA measurements" Have changed to : "the measurements of alpha-pinene SOA".

viii. p 22643 line 17 might be better "properties of SOA from smog chambers"? changed to "properties of the SOA particles" such that we are referring to the SOA produced during these experiments.

ix. Reference to the Ziese study is added in the context of the ability to obtain hygroscopic data at high RHs.

- Second main point:

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- P 22634 line 6-9: The HPSI was always on a different inlet to the CCNc , HMAN and HQUT. The inlets to the chamber are all identical stainless steel tubes. One would hope using a different inlet would make no difference.

The possibility that the temperature difference caused the difference was investigated during the experiments. HPSI was opened up to the laboratory and operated at ambient temperature. Operating at ambient temperature the growth factor remained the same as when it was operated in its temperature controlled box and whilst HMAN and HQUT measured a lower GF operating at the same laboratory temperature. So from this simple test we saw no difference, indicating that HPSI's results are not affected by the 4 degree temperature drop. Investigating the volatilisation for these relatively small temperature changes might be worth considering for future experiments. Another point to consider is the CCN counter which controls the temperature of the sample altering it by $\sim$2 to 15 degrees depending on the supersaturation set.

If we only considered the instruments on the same inlet this would be the case. We make it clear in the text that the CCNc is on the same inlet as HMAN and HQUT more studies will to required to see if there is an effect due to this.

- Minor points

- P 22625-26 line 1-3:

Have started a new sentence after Meyer et al. (2009).

- P 22628 line 15-22:

This section has been restructured somewhat based on the previous comments. Have changed "these" to "the" to be more logical.

- P 22631 line 20 "detection"

Corrected.

- P 22632 line 5 and line 18:

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Humidogram data was taken once the growth factor and the number size distribution had stabilised. This was after ∼6 hours. Data was then taken by the HTMDAs over the next 10 or so hours.

This detail is added to the text.

- line 16-17: "from about 40% up to about 95

Have specified for each HTDMA.

- Fig 4 x-axis:

Corrected both o these mistakes.

- Fig 5, 6, and 7: the right hand scales are not the same...

Changed all axes to be the same

- Concerning the temperature in the smog chamber hall laboratory...

The temperature data was taken from the HMAN T sensors . Unfortunately there is not a sensor outside of the air-conditioned smog chamber in the laboratory. The range of temperatures experienced according to the HTMDA's sensors during the experiments is given in the new table requested by reviewer #1. Even during the night the indoor temperature did not drop significantly.

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

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