

Interactive comment on “Consistency between parameterisations of aerosol hygroscopicity and CCN activity during the RHaMBLe Discovery cruise” by N. Good et al.

N. Good et al.

n.good@opgc.univ-bpclermont.fr

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Anonymous Referee #1

Firstly we would like to thank the referee for their comments they are much appreciated.

Specific:

- page 22660, abstract: The abstract is a little vague and needs to be more specific in summarizing the main results of the paper.

Parts of the abstract have been rewritten to clarify the main points in the paper

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- page 22663, eq. 1 and 2: I suggest to move the 'description' of the Koehler theory to section 2. The Koehler equation could be given and the different approaches and assumptions used/made could be introduced.

A sub section is added to section 2 entitled "Describing the aerosol water uptake - Köhler theory". This now contains the Köhler theory descriptions.

- page 22663, line 16 ff: I think I have an idea what the authors are trying to say here. However some rewording could be useful.

An additional sentence putting the statement into context is added.

- page 22664, line 4: The expression 'large aerosol populations' seems a little vague.

Changed sentence to:

"... for example it may be possible to describe the hygroscopicity in broad aerosol classes e.g. marine or continental in terms of just a single kappa for modelling purposes."

- page 22664, line 19ff: Giving this reasoning at the end of the introduction is somewhat unusual. Authors might think of moving it upfront.

This reasoning has been moved to the start of the second paragraph of the introduction.

- page 22666, line 9: performed -> measured

Changed performed to recorded.

- page 22666, section 2.3: I suggest to restructure this section in the following way:
- introduce CCNC - describe the CCNC's calibration - explain how it was used, i.e. subsequent to the DMPS ...

Section 2.3 has been restructured accordingly.

- page 22667, line 14: I don't see how a temperature gradient could deem a supersaturation. Rewording necessary.

Under the assumption that the other operating conditions of the instrument are held constant the variable determining the SS in the CCNc's column is the temperature. However, for clarity this is reworded simply as supersaturation.

- page 22667, line 25ff: Shouldn't it be the other way round (sulphate rich vs. sulphate poor regions)?

Corrected

- page 22668, lines 8/9 and 9/10: “: : component compounds” -> sounds wrong.

Have changed to "individual components"

- page 22668, lines 12 - 16: I guess this means that the same growth factor was assumed for different dry particle sizes and the corresponding moles of organic solute were then determined assuming the surface tension of water ? More explanation is required here. Part of the explanation could already been given when introducing the Koehler equation and referenced here.

A clear explanation of the method is added. An kappa value of 0.085 is assumed for the organics (equivalent to a growth factor of ~ 1.2 at 90% for $D_0 \rightarrow \infty$). The size dependence is then calculate according to "kappa-Kohler equation" (3) i.e. assuming the surface tension of water in the Kelvin term. Also see our response to reviewer #1 question on the sensitivity to the organic choice of the organic growth factor.

- page 22668, lines 22: the composition -> the composition measured by AMS

Correct (this phrase has now been removed as a consequence of the re-ordering/wording requested elsewhere)

- page 22669, lines 6 and 7: The statement is a little vague. And to my understanding this statement is not necessarily true. It should be explained briefly under which assumptions kappa varies with size and RH and why (non-ideality, slightly soluble substances (change in surface tension?))

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True, the point is clarified.

- page 22670, lines 1 and 2: This is confusing. Why not integrate the number size distribution from D_{thres} to infinity ?

The CN number distribution is integrated from the threshold diameter for CCN activation to the maximum dry size measured by the DMPS in order to calculate the CCN number. This step is now described in the passage.

- page 22670, section 3: This section needs to be streamlined and Fig. 2 should be explained properly and in more detail. This should include a description of what is shown (e.g. 'totals ?') and a thorough discussion of the differences of the AMS and impactor measurements. I consider this a key figure in understanding the results presented later.

An additional sub-section dealing with the composition measured by the AMS and the impactor is added to streamline section 3.

A more detailed description of the comparison between the AMS and DMPS derived volumes is added. Stating that the AMS masses can be converted to volumes assuming densities.

The legend in the Figure is rewritten clearly stating what is shown.

- It should be explained in more detail how the DMPS determined volume is used here. page 22671, section 3.1: I think a figure would be nice here. Maybe a frequency distribution of growth factors would be useful.

A figure showing the growth factor probability density distributions is added

- page 22672, section 3.2: It should be stated more clearly that figure 3 represents a comparison of measured (HTDMA) and AMS+modelling derived growth factors and κ s.

OK, "predicted from the AMS composition is added"

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- page 22672, line 20 and page 22673, line 7 (and similar also for Period 3 on page 22673): How can DMPS derived volume and AMS measured total mass be compared? Rewording required.

Again to be clear, we adopt the approach to compared the volume estimated from the AMS (assuming densities for the ions) to the volume estimated from the DMPS in all cases.

- page 22672, line 25 and 26: How can values be in agreement but have no strong correlation? Rewording required.

This is reworded to state: "The variability resulting from the uncertainty makes it difficult to say whether there is a correlation, but that there seems to be overall agreement."

- page 22673, line 25: Why is χ^2 only given here? I suggest you give the respective values for all periods.

This follows from above point. As the statistic is not necessarily meaningful in each case it is no longer reported for any of them for consistency.

- page 22674, line 10: I suggest to also include 'error bars' indicating the variability in fig. 5.

The variability if kappa in now shown in Fig 5.

- page 22674, line 18: I wonder where the multiple charges went! Some of sizes investigated are located left of the size distributions maximum, i.e., the measurement should be affected by multiple charges. The authors should at least comment on that and/or give an estimate.

They are corrected for in the data analysis therefore there is not a tail on the activated fraction plots.

- page 22676, line 6ff: This far too vague. As this might be an important result of this paper more discussion is needed.

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OK, did want to over emphasise this point for some of the reasons outlined below, could add some more specific numbers to this e.g. the negative gradients of the number size distributions above the number mode amplifies the difference in predicted potential CCN and measured potential CCN number at low supersaturations. For the Marine case this means that at 0.2% SS the fraction of CCN predicted from the HTDMA and AMS compared to the measured CCN number (X CCN) is $\sim 70\%$ and decreases linearly to $\sim 30\%$ at 0.05% SS. Although it is inevitable that any difference in D_{thres} however small will lead to a larger and larger difference in potential CCN when SS becomes smaller, the fact that this difference remains quite large up until 0.2% SS in the marine case could be important... In the African X CCN decreases from 80% at 0.1% SS. However we note that at these low supersaturations the change X CCN becomes sensitive to small changes in κ within the measurement variability. In the continental case X CCN also drops below 80% at 0.1% SS. These results highlight the need to obtain accurate and precise CCN measurements at low supersaturations in the marine environment.

- page 22676, section 3.3 Here the authors should be more explicit. As this is a quite important section, they should clearly explain and discuss their reasoning and methods. In my opinion a reference is not enough. Figure 7 is somewhat unclear.

Some more details of the fitting procedure are added. Figure 7 is redrawn to focus on the measured RHs rather than also showing the erroneous values outside of the measurement region which could have caused the confusion.

- page 22677, line 4

Changed

- page 22677, line 16: These surface tensions seem a little low for such a 'simple' aerosol. Authors should discuss the derived values and or the uncertainties of the method used to derive them. I personally consider this an artefact, but maybe I'm wrong !?

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See our response to the same point made by reviewer #1. Briefly: we do don't believe surface tension reduction could account for the entire discrepancy. This would appear physically unrealistic. This is clarified in the text.

- page 22686, figure 1: I suggest to introduce 'error bars' to indicate the variability.

These have been added.

- page 22688, figure 3: This figure is far too small. Furthermore - a,b, c should replace by period 1, 2, 3 - x and y axis labels should be consistent - GFs should be given in the upper and kappas in the lower panels - text, caption, and labels should be consistent - proper and consistent presentation of average kappas

Graph and caption altered accordingly. The figure will also be expanded and can be presented in landscape.

- page 22690, figure 5: The lower panels of this figure are too small. Furthermore, I suggest to switch upper and lower panels, and indicate variabilities.

The panels have been swapped and the lower ones enlarged. The whole figure can also be enlarged and presented in landscape.

- page 22691, figure 6: This figure is far too small !

It will be enlarged when typeset.

- page 22692, figure 7: Personally I'm not able to deduce anything from this figure. I highly recommend to make the readers' life a lot easier here.

See the same point reviewer makes on section 3.3 and the response.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 22659, 2009.

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