Atmos. Chem. Phys. Discuss., 9, C11016–C11020, 2010
www.atmos-chem-phys-discuss.net/9/C11016/2010/

Atmospheric
Chemistry
and Physics
Discussions

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

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Received and published: 21 February 2010

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

General comments:

Section 2.6 has been rewritten, more clearly stating how kappa was derived from the AMS measurements.

Size dependence occurs because the AMS measured the bulk (all particle sizes combined) component masses the measurement will be weighted towards the mode of the aerosol mass size distribution. Thus, the AMS derived composition is perhaps more

likely to accurately reflect the non-refractory aerosol composition in the range of 100-200nm dry diameter. Therefore if the inorganic ratio changes significantly with particle size, this will not be captured. We also note the presence of ions not detected by the AMS (defined as originating from refractory compounds). The impactor analysis performed shows the ratio of ions which are believed to contribute refractory and non-refractory compounds increasing with particle diameter. Thus, a second consideration is that for larger particles the AMS derived (non-refractory) composition captures a smaller fraction of total aerosol mass. The HTDMA selects particles of a single mobility, thereby capturing the size dry size dependence of the aerosol water uptake. The size range of the particles probed by the CCNc is limited by the lowest supersaturation setting and the CCN activity of the aerosol sampled. The result of this means the CCN activity measured by the CCNc was limited to particles less than ~100nm.

Differences: The differences between the HTDMA and AMS derived kappa values is generally much less than 0.2. For the periods which we identified the AMS derived composition as significantly incomplete the difference is largest. The main differences appear when comparing them to the CCNc derived kappa. Which as the reviewer states is often quite large. Three mechanisms which might contribute towards an apparent or intrinsic increase in kappa from the observed (kappaHTDMA) and assumed (kappaAMS) subsaturated behaviour to the observed supersaturated behaviour are proposed. A change in the solution non-ideality as the water activity increases would cause a change in kappa. To investigate a possible change in kappa as a function of water activity the humidograms measured by the HTDMA were analysed. No significant change in kappa was indentified at RHs up to 94%. However we noted a limitation of the HTDMA measurements was that we could not measure above ~94% RH and therefore could not probe the region closer to the water activities likely at the point of cloud activation. A second possible contributor to an increase in the apparent kappa are compounds which deliquesce above the RH in the HTDMA. Again we are able to probe up to 94% RH using the HTDMA, however a step change in kappa indicative of a significant sparingly soluble fraction is not seen in any case. Once again the maximum

RH set by the HTDMA poses a limitation on our ability to constrain the behaviour at the point of CCN activation. A third factor is that the Kelvin term tends to increase in importance as the point of CCN activation is approached. Therefore if our representation of the Kelvin term is not 100% accurate the kappa we derive from the CCN measurements could be tainted by this inaccuracy. In our representation of the Kelvin term we assume the surface tension and molecular mass of water. To explore the possibility that a surface tension change might contribute to the observed discrepancy in kappa we used the AMS composition and a variable surface in the Kelvin term to fit to the measured CCN activity. We would stress that these results show the contribution from a reduced surface tension alone required to reconcile the measurements. That is we do not suppose that the discrepancy is entirely due to the surface tension reduction, but that is could be a contributory factor. .

Organics are included when deriving kappa from the AMS data. This is clarified in the rewritten section 2.6.

Minor comments

- Abstract:

This is correct, what we are stating it that we measured up to 94% RH with the HTDMA and not beyond and within this measurement range the representation is satisfactory. The point being we can say what is occurring above 94% from the HTDMA measurements performed. The sentence is rewritten as:

"Measurements made at relative humidities (RH) up to 94% showed the water activity appears satisfactorily represented by a single parameter derived at 90% RH."

- Page 22662, line 5: The position of “e.g.”

Changed to "for example Topping"

- Page 22667, lines 25-29: It is stated that in sulphate poor regimes there is excess number of moles of SO42-. Should it be opposite?

Correct. Changed to: "in sulphate rich regimes"

- Page 22668: You say that GF for organics is estimated to be 1.2. Does it change results if some other value is used?

We tested 3 organic kappa values 0.01, 0.085 and 0.2 in order to derived kappaAMS the sensitivity is also ways small. The predicted growth factor using the AMS data varies by less than ±0.03 at 90% RH in all cases across the kappaorg range. Varying the organic density between 1200 and 1600 kg m-3 had changes the predicted growth factor compared to assuming the a density of 1400 kg m-3 by less ±0.015 at 90% RH. This equates to difference in kappaAMS of less than 0.05 for a particle with a growth factor of 1.7 at 90% RH.

- Section 3: It would be useful to have a map of cruise area including the main origins of air masses.

A plot of the cruise track has been added indicating the 3 periods.

- Page 22671, line 28: Should it be “with a varying influence from organics”?

Correct. Changed

- Page 22673, line 21-22.: Here you give the total mass during period 3. For period 1 the mass was given in page 22670, but I can not find it at all for period 2. Is it actually relevant at all to know what is the mass as the only important thing is how big fraction is seen by AMS?

Agree, the important point is the fraction is this paper. Therefore the absolute value is removed and the focus of the text in each period is on the agreement.

- Page 22673: You talk about _ values between 1.15 to 1.40. How do you explain those? In Table 1 the single component values are below that.

These are the values derived from the CCNc. Table 1 only shows the compounds that are predicted to form from the AMS derived composition. There may well be other components not accounted for i,e, sodium / chloride containing aerosols which to have kappa values in this range. There is of course the possibility of a measurement bias, however the calibrations performed showed the instrument was operating reliably.

- Page 22677: line 15: Are such surface tensions reasonable based on the existing knowledge?

For surface tension alone to be the entire cause of the discrepancy between the CCN and non-CCN derived kappa values is probably beyond what can hypothesised using our existing knowledge. That is it would need to be a very powerful surfactant to reduce the surface tension of the droplet to the stated levels given the inorganic content without simultaneously removing so much soluble material that the increase in kappa is not significantly offset. Hence we do not state that surface tension is the cause, but demonstrate broadly the reduction required in order to illustrate the maximum possible effect.

- Citations: Allan et al. 2009 is already in ACP.

Indeed, it was accepted shortly after submission of this manuscript. The reference is updated accordingly.

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