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Interactive comment on "Hygroscopic properties of the ambient aerosol in southern Sweden – a two year study" by E. O. Fors et al.

Anonymous Referee #1

Received and published: 5 May 2011

The manuscript presents long-time hygroscopicity measurements on ambient aerosols. The manuscript is well written, the topic is highly interesting. I suggest the paper for publication in ACP after some revisions.

1. The use of two water activity parameterization is unnecessary and simply confuses the reader. The difference between the kappa_R and kappa_P model is simply that in kappa_P the correction term for the non-ideal behaviour is included. In the other case the non-ideality of a model salt is taken into account. How can you be sure that the non-ideality of the measured ambient aerosol is similar to the one of ammonium sulphate? This inserts an unnecessary extra uncertainty in the closure, even if as you state this is a small effect. I suggest using only one water activity parameterization (kappa_P) throughout the manuscript.

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- 2. Page 6611, description of the closure: I think this paragraph has to be rewritten. I could only follow the text until line 10 ("The particles from one particular DMPS spectrum were then distributed over this surface function"). Then it is not clear anymore how you made your calculations. E.g.: line 11, after which the new surface function was integrated... Along which variable? Between what limits? I believe/hope that the made calculations are correct, I think it has to be simply better/correctly explained. For this reason I would like to see the exact equations that you used. One equation tells much more than many sentences.
- 3. Section 2.5 (Parameterisation of the CCN(s) concentrations): This paragraph for me is a mystery. The title suggests that some kind of parameterization of the CCN concentration will follow. Even though the only thing that is explained is that how one can fit the size distribution with a log-normal function and how then the cumulative log-normal distribution looks like. And then one sentence follows about the definition of the critical dry diameter, where you say if it is known along with the size distribution the CCN concentration can be calculated. For me this paragraph does not make any sense, why do you even need a fit for the size distribution if you have a measurement of it?
- 4. The discussion about the CCN data is weak. One could do much more with that data, calculating seasonal/diurnal variations of the CCN concentration or dry activation diameter. Or other CCN properties. I understand if this is out of the scope of this paper and you plan to publish the CCN data elsewhere and you only wanted to show the closure here. Just it would be a pity if the CCN data was not published anywhere.

Specific comments:

Page 6602 line 8: Define CCN

Page 6602 line 24: what do you mean by CCN parameterization?

Page 6603 line 8-9: the direct aerosol climate effect includes both scattering and ab-

sorption

Page 6604 line 13: the sentence suggests that this is the only or most common classification, please rephrase!

Page 6604 line 14: nearly hydrophobic: this name is misleading. Hydrophobic material is known to repel water. An aerosol particle with GF=1 at high RH means simply that the particle is non soluble at this RH, so it is non-hygroscopic. There is no way to detect hydrophobicity with the H-TDMA technique. Therefore I suggest to use "nearly non-hygroscopic" instead of "nearly hydrophobic" phrase. And be consistent with it throughout the whole manuscript.

Page 6605 line 10: Please define OA!

Page 6605 line 12: Delete "that"

Page 6605 line 20-23: monodisperse CCN measurements could give information on the mixing state, reformulate the sentence.

Page 6607 line 7: 1:10 is a ratio and has no unit.

Page 6607 line 17: Please mention the residence time in the humidifier already here!

Page 6608 line 5-11: The theoretical values from Tang and Munkelwitz are old and based on a single measurement I suggest to use newer values from e.g. AIM or AD-DEM models.

Page 6608 line 20 and 21: flow ratio has no unit.

Page 6609 line 15: typo "thesvalues"

Page 6609 line 19: " \sim 30 m", change "m" to "min"

Page 6609 section 2.3: how, and how often was the CCNC calibrated?

Page 6610 equation 3: change "a" to "3"

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Page 6612 line 2-4: Please rewrite the sentence, I do not understand it.

Page 6610 line 11: "cot"?

Page 6614 line 19: what is FNL?

Page 6615 line 17-18: Is the shift of the more hygroscopic mode at larger diameters only because of the Kelvin effect or is the hygroscopicity changing as well? Page 6615 line 23-24: How clear is this clear increase in hygroscopicity if you exclude the Kelvin effect? It would be actually much easier to interpret the results if you would show kappa-PDFs, I suggest doing so, or including it as a second panel in figure 3.

Page 6618 line 6-7: Have you thought about a chemical composition, hygroscopicity closure for that month when AMS measurement is also available?

Page 6618 line 1:Did the Fourier analysis reveal only a daily cycle?

Page 6618 line 18: why did you use for the averaging the volume weighted growth factors?

Page 6618 line 28: How do you define the amplitude? Difference between minimum and maximum or average?

Page 6619 line 7: "three smallest particles", change particles to sizes

Page 6619 line 5-8: for me it seems like, that the 35nm particles are more hygroscopic than the 50 and 75 nm ones. Is this significant, what could be a reason for that?

Page 6619 line 15: the aerosol particles are getting more hygroscopic not the GF-PDF

Page 6619 line 20-22: and you actually see the opposite, the amplitude of the diurnal cycle is higher for the smaller particles.

Page 6619 line 27-29: it does not matter what kind of organics you evaporate from your particle as long as the overall hygroscopicity of the particle is higher than the hygroscopicity of that organics (which is the case if I am correct). Please correct the

sentence!

Page 6620 line 2-3: for the hygroscopicity the mass (volume) ratio is relevant not the mass itself, so does this diurnal cycle also present if one looks at the mass/volume ratios? Section 3.6: The correct calibration of the CCNC is crucial, and this could be also responsible for the not perfect agreement between measured and calculated CCN concentration.

Page 6621 line 8-12: you argue that the maximum CCN(s) values at 1% supersaturation reach the values of the integrated DMPS number concentration, and therefore the counting efficiencies of the different instruments cannot be responsible for the observed closure discrepancy. I cannot agree fully with this argument. If someone looks at the points where everything should be activated, due to unavoidable measurement uncertainties some points would be above the one-to-one line and some below. Therefore it is expected that the maximum CCN(s) values are actually higher than the corresponding DMPS number concentration values.

Page 6622 line 26-29: Could you give an estimate what kind of surface tension value would explain the observed discrepancy? Page 6623 line 14-15: Why do you need a fit if the direct measurement can be used for the calculation? The fitting introduces only unnecessary error. As it can be seen in Figure 9, the fit is not perfectly matching the measured data.

Page 6624 line 19-21: How often?

Figure 1: Please add the information on at which dry sizes (or RH) were the measurements performed, or were both varying during the calibration experiments? Please include the number of measurement points at each water activity value.

Figure 2: increase the size of the figure annotation, I suggest to include it only once (but bigger) since it is the same for all panels.

Figure 4: Adding the monthly average GFs to the graph would be good. And please C2785

use more colors for this plot not only 8.

Figure 6: this figure contains a lot of points and therefore significant part of the plot is just grey. One cannot really see where the highest density of them is. It would be much more informative if you would not show every single point, but rather the density of points per area, or something similar. Do it for all graphs where you show similar amount of points.

Figure 7: Too many overlapping points here as well. I suggest to show rather the different percentages of the ratio distributions.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 6601, 2011.