Atmos. Chem. Phys. Discuss., 11, C8042–C8046, 2011
www.atmos-chem-phys-discuss.net/11/C8042/2011/

Atmospheric
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Discussions

Interactive comment on “Aerosol hygroscopicity and CCN activation kinetics in a boreal forest environment during the 2007 EUCAARI campaign” *by* K. M. Cerully et al.

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Received and published: 20 August 2011

The authors would like to thank the reviewer for the valuable comments that have improved the manuscript. Please find below a point by point answer to the comments. The answers are displayed in italics.

Specific Comments:

My main criticism of this manuscript is that it does not advance science too significantly given that the techniques to collect and analyze the data are all previously documented and that the conclusions are not very surprising. While the paper is interesting and the techniques appropriate, it appears that this is another point in a global database of

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Discussion Paper

hygroscopicity measurements. This in itself is important, but it is not terribly exciting.

We thank the reviewer for the comments and discussion. As this methodology is new and more straightforward than other approaches, we wanted to show that we were able to reproduce what is thought to be representative of a well studied environment. Additionally, this work provides several interesting CCN characteristics (e.g., chemical dispersion and droplet growth kinetics) not reported in the relatively few CCN studies of this environment (e.g., Aalto and Kulmala, 2000; Hämeri et al., 2001; Sihto et al., 2010).

1) Page 15038, line 15. What was the purpose of switching from 90% RH to alternating between 71% and 87% on May 3? Does this not make the data from the two sections of the campaign inconsistent? What was the basis for the selection of 71%, 87%, and 90%? Without justification, it seems random.

RH was switched in the HTDMA to see the sensitivity of κ to the humidity level. RHs were chosen so that the highest and lowest RHs are above and below the deliquescence RH of ammonium sulfate ($\sim$80%), respectively. The final RH values were dependent on the instrument and the time needed to reach a stable RH. As κ is expected to be independent of RH, the data during the two sections of the campaign should not be inconsistent. These conditions are often changed during field campaigns.

2) Page 15042, lines 19-22. Why were different sizes selected for analysis in the CFSTGC and HTDMA? If the voltage on the (first) DMA is operator specified, why were these measurements not coordinated? While I doubt that comparing 50 nm to 60 nm induces much error, it would have been more appropriate to have the two instruments measuring the hygroscopicity/activation of the same size particles.

A miscommunication led to the slightly different sizes. As pointed out by the reviewer, this is not expected to influence the results by much.

3) Page 15043, line 1. Please clarify how non-hygroscopic particles affect the deter-

mination of kappa. If I understand correctly, the GF is determined by comparing the modes of the wetted and dry particles – do the non-hygroscopic particles affect the ability of the hygroscopic particles to uptake water, thereby causing a shift in the mode of the size distribution of the grown particles? This is a little bit outside my area of expertise, so please correct me if I am wrong.

While non-hygroscopic particles do not affect the ability of hygroscopic particles to uptake water, there are differences between how non-hygroscopic particles impact the calculation of κ *using the HTDMA and CFSTGC. In the HTDMA, non-hygroscopic particles are indeed seen (i.e., GF=1) and will therefore affect calculations of* κ*. In CCN measurements, non-hygroscopic particles show themselves through the change in the maximum activated fraction, E. As a result, the slope of the sigmoid used to calculate critical supersaturation, and thus* κ*, is unchanged and will not reflect non-hygroscopic particles. This is particularly important when particles are externally mixed and there is a large fraction of insoluble particles, but this is not the case in Hyytiälä, because E, the maximum activated fraction, is typically close to one as shown in Figure 6a*

4) Page 15043, line 17. On Figure 5, it looks as if a significant fraction of the data points lies above the 30% line, while relatively few lie below the -30% line. Is this because values for 60 nm are being compared to those for 50 nm? Or are there other reasons that this is the case? What fraction of the data actually lies above the +30% line?

60 and 50 nm data are expected to fall within the same hygroscopic mode and display similar κ *values. This slight shift is most likely from the difference between measurements at supersaturated and subsaturated conditions as solution non-ideality, phase separation, and partial solubility of constituents may be more pronounced in subsaturated HTDMA measurements and decrease the observed* κ *compared to supersaturation conditions. Additionally, the impact of non-hygroscopic particles on* κ *calculations in the HTDMA and CFSTGC (as discussed in the response to question 3) could result in slightly lower HTDMA-derived* κ *values than CFSTGC-derived* κ *values This issue has been clarified in the text.*

5) Page 15045, line 3. Would particle growth rate data/particle size distributions help in this discussion of 40 versus 60 versus 80 nm? That is, would it be helpful to know if particles had grown significantly in situ and over what time scale in terms of understanding chemical dispersion?

While particle size distributions may be helpful to see whether or not particles have grown from nucleation events, they would not give much information regarding external mixing. It is likely that increased daytime mixing is more important than nucleation events, but this would be difficult to assess without single particle chemical composition data.

6) Section 4.4 seems like a lot of information to get to the punch line at the end that organics do not appear to affect activation kinetics. Perhaps this can be shortened?

We chose to keep most of the discussion in order to show several methods of evaluating droplet growth kinetics because of their complexity A portion of the text was removed to shorten the section.

7) On line 24, define SMCA.

This is previously defined in section 2.2, but a reference has been added to the end of the sentence

8) Figure 1 seems extraneous as no specific analyses corresponding to the dates shown are performed. It seems clear that clean air would not come from the European continent.

This was meant to simply illustrate the diversity of air masses seen in Hyytiälä We have decided to leave this figure in the text, though it has been clarified that the backtrajectories serve only as an example.

9) The inset on Figure 10c is almost impossible to read.

The Figure has been enlarged to correct this issue.

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Typographical Errors

Page 15040, lines 15 and 22. Subject-verb and plural errors. Data were filtered and fit to a spectrum.

Page 15050, line 26 – a small fraction is and has, not are and have

Page 15045, lines 18 and 21. Who not whom.

Page 15050, line 17, extra as?

These issues are now corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 15029, 2011.

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