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Atmospheric
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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.

Anonymous Referee #2

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The material presented by the paper is appropriate for publication in ACP and is, in general, well-written and presented. Many of the points I would raise have already been addressed by the previous comments and responses. There are one or two further considerations that the authors might like to address prior to publication.

The rapid SS cycling employed in the sampling is rather surprising (as mentioned by Dr Rose) both because of supersaturation settling and the low counting statistics encountered during monodisperse CCN measurements in a relatively clean environment. Each of these will require relatively long sampling times to improve confidence in the measurements (through settling of the instrument and improved signal to noise respec-

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tively). Given the low number of counts likely to be obtained in each scan (a raw CN and CCN spectrum as f(Dp) could usefully be provided), can the authors comment on how the uncertainty bars in each of the derived quantities in figures 6, 7 and 8 represent in any way the uncertainty in precision attributable to the Poisson distribution of counts. The treatment of the error propagation leading to the magnitude of the uncertainty bars should be described (particularly the propagation of the Poisson error in the activated fraction through the fitting error to obtain critical supersaturation).

This leads to another question - Figure 3 very clearly attributes the slope in the activation spectrum to the degree of heterogeneity of activated particles. As is recognised in the appendix, there will be a contribution to this slope from the breadth of the DMA transfer function as modified by multiple charging. It is noted that the non-multiple charging related component of this spread is practically time and size invariant. However, was the transfer function assumed ideal in calculating the 64-80% contribution to the $\sigma(\kappa)$ offset? If so, how would the likely deviation from an ideal function as is frequently exhibited in DMAs affect the heterogeneity offset. If not, how was the transfer function measured. Obviously, the Poisson error in the number of counts (which would need to be propagated through the multi-charge correction) will contribute to the error in the slope as well as in activated fraction. It would be informative if the effects of these considerations on the best fit κ spectra calculated in figure A1 were discussed briefly in the appendix.

Finally, the authors' response to the other referee's comment below seems peculiar:

"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 (c.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."

and raises some concerns about the applicability of a single parameter hygroscopicity parameter to represent behaviour across the full range of RH. It is rather contentious to state that κ should be expected to be independent of RH. Notwithstanding earlier (almost consensual) acceptance of the concept of a simple single parameter hygroscopicity, some previous and more emerging work suggests that the non-ideality differences at different RH lead to discrepancies in κ derived from different RH measurements (even if these conditions are often and routinely changed in field campaigns). Care should be taken when stating that the measurements at different RH should be consistent. Indeed, it is quite clear from Figure 5 that κ derived from HTDMA and CCN do not agree. Amongst the many other reasons for disagreement, it is very possible that they should not agree because of differences in ideality at sub-saturated RH and at the point of activation. The contribution to the discrepancy in κ because of non-ideality should be clearly stated. There is too much of a tendency in the literature to tune surface tension to obtain agreement, or to brush aside discrepancies of the magnitude shown in Figure 5 (e.g. "These differences are well within the range of uncertainty often seen for differences in HTDMA and CFSTGC-derived κ values (e.g....). For simplicity, κ values derived from CFSTGC data will be used throughout the rest of this study"). Supplementary material should be provided containing the figures repeated using κ values derived from HTDMA measurements so that the sensitivity of any conclusions to the source of κ can be readily examined.

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