Atmos. Chem. Phys. Discuss., 11, C10897–C10900, 2011
www.atmos-chem-phys-discuss.net/11/C10897/2011/

Atmospheric Chemistry and Physics 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.

K. M. Cerully et al.

nenes@eas.gatech.edu

Received and published: 27 October 2011

Referee #2

We thank this referee for the positive feedback and constructive comments. Replies to the questions and suggestions raised are found below.

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.

1) The rapid SS cycling employed in the sampling . . . 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).

To improve confidence and increase counting statistics, concentrations were accumulated over a period of 16 s (this has now been clarified in section 2.2 of the revised text), and the DMA sheath:aerosol flow ratio was maintained at 4.5:1. To ensure Poisson counting statistics errors are small, sigmoids were discarded for which average CCN counts were lower than 50 (corresponding to a CCN concentration of $\sim$*4 cm*$^{-3}$*).*

The error bars in Figures 6, 7, and 8 represent the standard deviation of the total values found for each time period during the entire study. Day-to-day variations are greater than parameter uncertainties, and are now presented in the main manuscript (for κ*) and appendix (for* $\sigma(\kappa)$*). The error calculations do not consider Poisson counting statistics alone because there are other uncertainties present (primarily from the experimental setup, instrument operation, and temporal variability in aerosol properties). As a result, we have done the error analysis using a traditional fitting and error analysis approach where equal weights are assumed for each point in the activation curve and parameter uncertainty limits are calculated from residual after the fitting.*

2) 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 recognized 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

ACPD
11, C10897–C10900, 2011

considerations on the best fit spectra calculated in figure A1 were discussed briefly in the appendix.

The transfer function was indeed assumed ideal (i.e., non-diffusing and triangular as a function of particle mobility). The non-ideal transfer function is wider than the ideal function, so this would have an effect in increasing the $\sigma(\kappa)$ offset. For this reason we conducted the sigmoidal analysis with the activation of calibration single-component aerosol, as an (empirical) upper limit of non-ideality effects.

As explained above, sigmoidal fits were not weighted by counting uncertainties, so the expected values of $\sigma(\kappa)$ (or the slopes of activation curves) are independent of the Poisson error. Standard deviations of $\sigma(\kappa)$, however, depend on the variance of the data, but again, observed variance was used in the error estimates instead of the prescribed Poisson error (because of how measurements are carried out). Error analysis for $\sigma(\kappa)$ will be added to Appendix A.

3) 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 . . . 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.

Our response intended to state that subsaturated κ values were found to be independent of RH for this data set (the correlation of κ values take nearly simultaneously at

ACPD

11, C10897–C10900, 2011

RHs of 71% and 87% showed that the average difference in values was only -0.004 ± 0.029). Also, we did not mean that subsaturated and supersaturated κ values should be similar. Several explanations for the possible differences are given in the beginning of Section 3. The text for Fig. 5 has been further clarified to state that observedκ are not suprisingly large, but can be explained by the different saturation ratios. HTDMA-derived κ values for 50 nm dry size have been added to Fig. 7c and linear fits to the HTDMA-derived 50 nm κ values are added to Fig. 9.

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