**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Empirical predictions of CCN from aerosol optical properties at four remote sites" *by* A. Jefferson

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Response to reviewer #3:

I appreciate the comments from the reviewer, especially with regard to the focus on data uncertainty. Trying to reduce and quantify data uncertainty is an ongoing effort that is especially difficult with remote, long term monitoring of CCN. The DMT CCN instrument is relatively new and learning to operate this instrument remotely with limited resources is challenging. I hope to address most of the reviewer's concerns.

1) "Could the goodness of fit statistics referred to in Section 3 and/or some example spectra be included so that the reader can evaluate how well this parameterization is able to capture the CCN(s) variability at each site."

I added another table to the paper that lists the "k" fit parameter, the slope of the fit line and the r2 correlation coefficient of the fit to the measure CCN. The data are all the fits with $\sim 0.2 < \%SS < 0.8$, with the exception for HFE data with a minimum %SS of 0.4. I edited the power law fits to exclude fit with exceptionally high chi-square goodness of fit parameters as these usually coincided with noisy data. A much smaller subset of data was used for the model fits to the backscatter fraction, BSF, and single scatter albedo, SSA that had boundary conditions for the BSF, SSA and range of acceptable scattering coefficients. This table represents only the fit quality of the power law fit to the CCN data over a limited %SS range. The high correlation of the power law fits to the CCN data qualifies the "C" and "k" fit parameter as valuable data products that can be further used to characterize the CCN for seasonal, long-term trends.

2) "There is much support from this study as well as past field studies that small mode particles may have only a minimal contribution to CCN formation. Smaller particles tend to have a higher fraction of absorbing material that may limit their ability to act as CCN... What size range is referred to here, and can some data from these sites or a couple of relevant citations be added in support of these assumptions?"

See the reply to the 1st reviewer as he had a similar comment. I removed this sentence and rewrote the paragraph and instead discussed the possible role of organics on CCN.

3) "Pg. 9001, Line 15: "%SS value at _0.2 was not used in the analysis. The CCN instrument may have insufficient water or too short a sample residence time for the aerosol to activate under high aerosol concentrations and low %SS." This could easily be confirmed by examining the droplet size distribution measured by the CCNC optical particle counter, in which case, one would expect the droplet distribution to have shifted close to the _1 um size threshold necessary for droplet detection."

This comment came out of a CCN workshop where we discussed issue of high CN concentration and activation time in the instrument. There is a nice graph in the DMT operation manual, which I reference, that shows the drop in CCN/CN for NaCl aerosol

of a given size at high CN. For insufficient activation from not enough time or water in the instrument at high CN the resulting droplet size distribution may shift to smaller sizes. The algorithm for editing the high CN data based on a droplet size distribution is unclear as I would need to compare high CN data size distribution to a low CN size distribution and make assumptions about the aerosol size, composition and activation properties. For editing large datasets with months or years of one-minute data it's easier to notice that the outliers happen to be times with high CN and remove them from the analysis as they skew the fits. For more detailed work with a smaller data set it would be interesting to include the high CN data and look to see if this is a problem with the instrument or a limitation of the empirical fit method.

4) "Figure 5 shows the calculated CCN from the empirical correlation plotted vs. the measured CCN. Are these the same measurements that were used to train the empirical fit or were the data resampled in some way?"

The data in Figures 1 through 6 use the same data. Note that the line across the data at 1.0 for Figure 6 was placed incorrectly for the HFE data at below 1.0. This will be fixed in the revised version of the paper.

5) "Please report the measurement uncertainties of s, BSF, SSA sigmas_sp, and CCN concentration with the instrument descriptions in Section 2. How sensitive is the calculated CCN concentration obtained from the empirical model to variability in s, BSF, SSA, and sigma_sp over the range of measured values? How do these sensitivities compare to the measurement uncertainties? "

I added the following lines to the Measurements section of the paper. "Discussion of uncertainty in the nephelometer total and backscatter coefficients can be found in Anderson et al., 1996 and Heintzenberg et al., 2006. Studies of the uncertainty in the psap absorption coefficient can be found in Sheridan et al., 2005 and Virkkula et al., 2005. "

Based on the above references the uncertainty in the backscatter fraction and single

scatter albedo are both 0.01 about the average values of 0.90 for single scatter albedo and 0.12 for back scatter fraction. Uncertainty in the CCN concentration will depend on the instrument operation predominantly on the lower size threshold for counting. If the lower threshold is too low then the CCN will count inactivated aerosol and if too high it will miss the smaller activated CCN. The threshold in all of these measurements was set to 1.0 micron. Any leak in the instrument will likely result in undercounting of the CCN. The counting efficiency is subjective. For these studies the ratio of CCN/CN number concentration periodically fell within 0.9 to 1.0 at the highest %SS. For GRW this ratio was greater than 0.9 for most every scan at the highest %SS. As a check on the instrument operation I liked to see the ratio exceed 0.9 at least once a week for a prolonged period of at least half a day at SGP and FKB. The Chinese Metereorological Association (CMA) prohibited export of the HFE data until a month after the campaign had ended, so I was unable to check the instrument operation during the campaign. The CMA turned off some of the aerosol measurements after August of 2008, which limited the data set. For the HFE data the CCN counting efficiency is unknown as the CCN periodically exceeded the CN concentration. This is because the CN counter quickly became clogged due to the high pollution levels and wasn't cleaned often enough to give good CN concentrations.

The ARM system doesn't have a portable DMA to calibrate the CCN instrument, which is my reason for using a Lance et al. model to calculate the %SS. Even with a DMA calibration this calibration will change with instrument flow and pressure and therefore isn't appropriate for long-term monitoring studies. The model uses the average thermal resistance of 3.7 K/W given in the Lance et al. reference. I'm aware that the thermal resistance of each instrument needs to be calibrated on a regular basis. As a routine part of every service visit the CCN column is purged with a weak bleach solution to mitigate the buildup of residue on the column that would change the thermal properties. At SGP the CCN is operated next to an HTDMA. The HTDMA produces a NaCl aerosol of known size once a day for calibration of its relative humidity sensors. We have just started a procedure to divert the calibration salt aerosol from the HTDMA to the

CCN for daily checks of the CCN %SS values. Once we have the algorithm written I plan to use the daily checks of the %SS calibrations to monitor the CCN column thermal resistance and record the drift in the value over the course of a year as well as the sensitivity of %SS to changes in the column thermal resistance. This work will culminate in a detailed study of the model calculations of the CCN %SS. Until I'm able to analyze the data from the salt aerosol calibrations at SGP I can't evaluate the uncertainty in the instrument %SS. Calibration of the %SS in the CCN instrument that is part of the ARM Mobile Facility will be logistically difficult, but will need to be done for future deployments.

This paper is a preliminary analysis of this empirical method. Detailed work that includes sensitivity and uncertainty analysis will come with study of laboratory aerosol of known composition as well as a detailed study at SGP that includes the aerosol size distribution and composition. My motivation is to present the method in order to generate collaborations, funding and future research. Because the work is unfunded the analysis is limited. Hopefully with better uncertainty analysis and calibration of the %SS the spread in the data will decrease and more precise fit values can be obtained.

6) "Frequency distributions (# instances vs. distance from the mean/line) would be very helpful for seeing how the data are distributed for the parameters given in Table1 (especially) and also for the scatter of the data about the fitted lines in Figures 3-6. Perhaps these could be added to the current figures as small insets?"

I added a figure with the probability distribution of the scattering coefficient to my response. I don't think more probability graphs helps to understand the empirical fit better. Differences in the aerosol properties between the sites can be understood from the average and standard deviation values given in Table 1. One can visually see the spread of data about the fit lines in graphs 3-6. A more relevant value than the probability distribution is the relative ratio of the calculated/measured of the fit with CCN concentration. Figure 6 gives information as to the goodness of the fit over a concentration range and gives insight as to the aerosol loading required for such a method to

work well.

7) "Page 8998, Lines 24-30: It's hard for me to tell from the text whether the model of Lance et al. was used to calibrate supersaturation in the CCN counter or whether measurements of $(NH_4)_2SO_4$ or NaCl aerosol (following Rose et al.) were used? If the former, did you use an instrument-specific thermal resistance for each site or one specific value? If the latter, which Köhler theory model (from Rose et al.) was used?"

The Lance et al. model was used to calculate the %SS. The model uses calibrated values of the pressure, temperature and flows to calculate the %SS. What's missing, as discussed in the response above to question 5, is a salt calibration of the column thermal resistance. Instead we use the average column thermal resistance given in the Lance et al. reference. I have plans to implement a calibration of the column thermal resistance in the near future.

Minor Comments:

Please add the citation for Ervens et al., 2007 from Pg. 9000, Line 14 to the bibliography. - Page 9000, Line 7: Remove "a" from "are essentially a non-activating." - Please add 1:1 lines in Figure 5. - I find the meaning of the sentence on Pg. 8996, Line 11 unclear: "The fit quality declined at low CCN concentrations in a region with higher data uncertainty". Please reword. - Page 8996, Lines 19-20: Change "computational intensive" to "computationally-intensive"

All suggested changes in "minor comments" implemented.

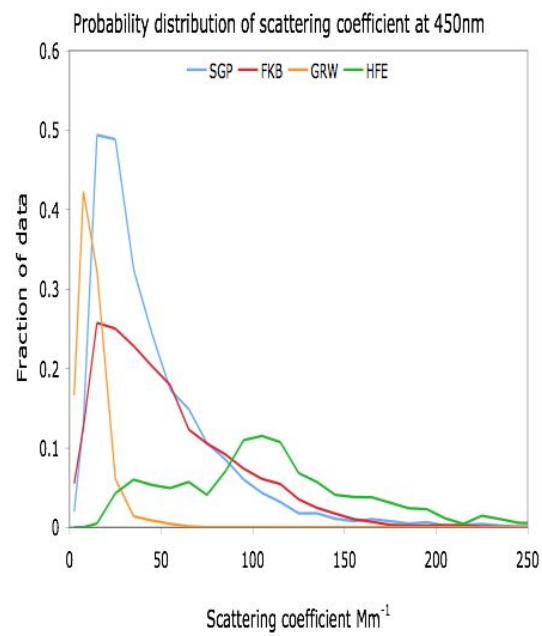Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8995, 2010.

Probability distribution of scattering coefficient at 450nm

Fig. 1.

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