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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of cloud condensation nuclei concentration from aerosol optical quantities: influential factors and uncertainties" *by* J. Liu and Z. Li

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Responses to comments by Dr. Ann Jefferson

The authors appreciate very much your insightful and constructive comments about this study.

1. Much of my concerns about this paper centers on incorrect use of the data. The CCN edited and corrected data starts May 2007 not as stated 2006.09 in the paper. Given this error I wonder if the other data sets used final edited and corrected or raw data. The final datasets are either b1 or c1 data in the ARM archive. The nephelometer

data during GVAX had an incorrect calibration value in the a1 data due to a bad tank of CO2 gas. This calibration was repeated at the end of the field campaign and applied to past data in the b1 data set. Similar instrument problems are prevalent in all the data sets as is typical with remote measurements in regions with limited resources. Caution is needed when using the data to consult the data quality and monthly system reports for further removal of suspect data.

Response: We have noticed that there are several versions of data from the Aerosol Observing System (AOS) in the ARM archive. Data from all sites were double-checked thoroughly. The "sgpnoaaaosC1.b0." and "sgpnoaaaosC1.b1" data streams for the SGP site were used. The former is from 2 July 1996 to 17 May 2007 and the latter is from 19 May 2007 to March 2013. Because there are no CCN data available before 17 May 2007 in the "sgpnoaaaosC1.b0." data stream (-9999 in the data file), bad data prior to this date were automatically excluded. We matched several data sets based on the observation time, so any period void of valid data for any single dataset is excluded from subsequent analyses. As a result, the effective date range of the matched data is shorter than the date range listed in Table 1. We clarify this in the revised manuscript. For data during the GVAX period, we used the quality-assured "pghnoaaaosM1.b1" data stream. Likewise, we used the "grwnoaaaosM1.b1" data stream for the GRW site, the "fkbnoaaaosM1.b1" data stream for the FKB site, and the "nimnoaaaosM1.b1." data stream for the NIM site.

2. The DMT CCN instrument counts all particles in the size range of 1.0 microns and larger as droplets or activated CCN. In dust regions such as NIM the reported CCN counts, particularly at low %ss values are likely dust. The higher %ss values are likely a combination of inactivated dust and activated droplets. The CCN data needs to be analyzed bin by bin with the size distribution at the lowest %ss subtracted from those of higher %ss values. The first minute of every 5-minute %ss interval of the CCN needs to be discarded as the instrument temperatures and signal is unstable during this time. Thus the CCN measures 4-minute averages every 5 minutes.

Response: We checked the ARM data archive and only data streams called "nimaosccnM1.a1" contain CCN concentrations in 21 bins ranging from 0.75 um to 10.5 um. However, these data are raw data, i.e., data which have not been quality controlled in any way. In addition, as mentioned above, the data used in the study is the final version of data, which has passed all quality controls.

3. Dust at NIM was usually episodic and prevalent in the dry season. At other times, local biofuel and trash burning dominated the surface aerosol. The local burning had a distinct diurnal signature and can be readily identified. These factors complicate analysis of the NIM data and need to be mentioned in the paper. Aircraft measurements from the site over two different seasons identified smoke aerosol from biomass burning in Nigeria. The elevated aerosol layers will weaken the correlation between surface and remote measurements.

Response: We agree. The special observing periods during the AMF deployment sampled absorbing aerosols from desert dust and biomass burning. The small values of mean Angstrom wavelength indicate that dust aerosols dominated this area during our study period. However, it is still necessary to mention that biomass burning aerosols have an influence in this region and may complicate the analysis of NIM data. The correlation between surface and remote measurements is definitely influenced by aerosol vertical distribution. We added this discussion to section 3.1.1 of the revised manuscript.

4. A two-point fit to a power law distribution isn't a valid way to analyze the scattering hygroscopic growth as the error is large enough as to make the calculated values meaningless. The Jeong et al. paper used aircraft data with 2 nephelometers at set RH values that weren't scanned over a wide range. Note that the power law fit is only valid over a limited RH range for metastable particles and that data below 40% RH shouldn't be used in the fit. The RH values from inside the TSI nephelometers have a large error, as these sensors aren't calibrated. Estimated errors for these particular TSI nephelometer sensors are on the order of 10%. The RH inside the nephelometers

needs to be calculated from the dew point value of the Vaisala sensors either upstream or downstream of the nephelometer. The calculated average fRH using a least-square power law fit of the data from GRW gives an average fRH (40-85% RH) of 2.14 +/-0.21 for sub 10-micron aerosol and 1.88 +/-0.31 for sub micron aerosol. These value are more in line with a marine sea salt aerosol and quite different than the reported values in this paper of 1.36 and 1.31. The average fRH for SGP reported by Sheridan et al., JGR, 2001 was 1.83, quite different than the value reported here of 1.54.

Response: As mentioned in our manuscript, two nephelometers were deployed with one serving as the "reference" measuring dry aerosol scattering coefficients. The other is connected to a humidity scanning system to measure changes in aerosol scattering coefficient with variable RH (from $\sim$40% to $\sim$90%). Based on scattering coefficients measured at low and several high RH levels, a two-parameter empirical fit was used to describe the RH dependence of aerosol scattering coefficients. The parameters in our study are obtained from the fitting of scattering coefficients at several RH levels from low to high, based on explanations given in the AOS handbook (Jefferson, 2011). This differs from what was done by Jeong et al. As such, the citation of their work here was misleading. The sentence is now changed to "... a and b are determined from σsp measured at varying RH levels (Jefferson, 2011)." Regarding your comment on "The RH values from inside the TSI nephelometers have a large error, as these sensors aren't calibrated, Estimated errors for these particular TSI nephelometer sensors are on the order of 10%": We admit our error concerning uncertainties in the estimation of aerosol hygroscopic growth. In the revised manuscript, the fRH data stream called "noaaaosfitrhM1.b1" is used at all sites except the SGP site. This data stream is developed by the ARM program with quality controls applied to it. Based on this data product, we re-analyzed the CCN-scattering coefficients relationship. Correlation coefficients in the revised version are somewhat different than those in the old version, but our main conclusions are not altered. No ARM "fRH" products after September 2009 are available for the SGP site, so calculated values used in the old version of the manuscript are kept. These fRH (85%/40%) values and the ARM product for the

same period (Sep. 2006 to Sep. 2009) are comparable. The mean value of fRH (85%/40%) from ARM products is 1.54±0.46 and 1.57±0.45 for sub-micron and sub 10-micron aerosol particles at 450 nm, respectively. Corresponding mean values from our data are 1.60±0.23 and 1.56±0.22, respectively. Considering that bin-averaged fRH (85%/40%) derived from a very large set of data samples are used in our analysis, the effect of estimated fRH (85%/40%) uncertainties on the results are very small.

5. You need to specify how the correlations in Figure 4 were created. Did the scattering coefficient correlate to the same average interval as the CCN or were they hourly averages? Was each average of the aerosol scattering corrected to ambient RH based on the fit parameter for that hour or did you use an average RH fit value over the entire data set? How did you calculate the RH of the dry scattering coefficient from which you derived the ambient scattering?

Response: Figure 4 shows correlation coefficients of the relationship between surface-measured aerosol scattering coefficients at ambient RH conditions and CCN concentrations as a function of wavelength. The CCN bins used are the same as in Figure 2. Ambient scattering coefficients are averaged in each CCN bin. We matched aerosol scattering coefficient measurements, CCN concentrations, ambient RH measurements, and calculated aerosol hygroscopic growth factors. The aerosol hygroscopic growth factor is calculated at 1-hour intervals. Scattering coefficients corrected for ambient RH have a temporal resolution of one minute and are matched with the closest hourly value of aerosol hygroscopic growth factor. Note that the RH for dry aerosol scattering coefficients is not exactly 40%. At such a low RH, however, dry scattering coefficients do not change significantly. In the revised manuscript, fRH data are replaced by the ARM product as mentioned above. We matched measurements of CCN, dry aerosol scattering coefficient, ambient RH and fitted fRH parameters. Aerosol scattering coefficients are then corrected to the ambient RH condition based on the fRH function. We have specified this in our revised manuscript.

6. Aerosol hygroscopic diameter growth depends strongly on the aerosol chemical

composition. Scattering/extinction hygroscopic growth is highly sensitive to changes in the aerosol size and as well as composition. As composition at SGP doesn't vary dramatically (organic composition ranges between 60-80% of the mass), the fRH may be more dependent on changes in the aerosol size (see Hegg et al., JGR 98, 18435,1993). A strong relationship was observed between gRH and CCN with SGP aerosol (Gasparini, JGR 2006).

Response: We agree. As mentioned in our manuscript, fRH depends significantly on aerosol chemical composition. Although the chemical composition at the SGP site does not vary dramatically, fRH may be more dependent on changes in aerosol size. The mean aerosol Angstrom exponent changes slightly in each fRH bin, which may indicate that there are differences in their composition or that the Angstrom exponent may not completely describe the aerosol particle size, especially for small particles, which are not optically sensitive but are fRH-sensitive. As mentioned in the manuscript, it is still unclear whether growth characteristics are useful for inferring CCN properties. A probable reason is given in the manuscript. We have added the discussion about this in paragraph 3 of section 3.3.

7. Analysis of trends using binned data is fraught with problems, especially when the variable binned is the one with the highest variability. Binned data can obscure biases in the data, include outliers and combine bimodal distributions often prevalent with aerosols. The correlation coefficient will increase as the number of bins decreases, making a goodness of fit to the trend ambiguous. Binned data implies a normal distribution to the data, which may not be the case. A case in point is the plot of scattering vs CCN concentration at 0.4% ss attached below. This is subset of the data from February to May of 2009. Though only a subset it should capture most of the variability of CCN properties at SGP. Data are 30-minute averages for scattering and 4 minute averages for the CCN. While the fit parameters are quite different, the data are not normally distributed about the fit line, especially at low CCN. With such large datasets there needs to be a metric of how well the binned data captures the data trends without including

biases. This may include plotting the trend on top of a density plot of the hourly data or plotting the distribution of points about each bin number. You can use an algorithm that optimizes the derivative of the chi-square spread of points with bin number, i.e. at what optimal number of bins does the point spread not change substantially. You need to justify the number of bins used in the fits for Figures 2-8. Does a higher number of bins fit a different trend line?

Response: We agree with your comments about the limitation of using binned data in the analysis of trends. But the use of bin statistics to analyze such large datasets is unavoidable. It is true that fitting parameters vary with the choice of bins used. Likewise, differences in the fitted parameters may result from the use of all samples without binning, if different sample sizes are used in the analysis. In response to the comments, we conducted sensitivity tests regarding the use of different numbers of bins. The following figure shows the fitting parameters and the correlation coefficients derived from linear regressions of scattering coefficients and CCN concentrations using different numbers of CCN bins. Correlation coefficients and fitting parameters change slightly over the range of bin numbers chosen (5, 10, 20, and 50). If CCN=1500, calculated scattering coefficients are 43.43, 43.29, 44.03 and 44.42 Mm-1 for CCN bin numbers of 50, 20, 10, and 5. This suggests that the influence of the number of bins used is not significant, thanks to the large set of data samples. It is a very good suggestion to list the number of bins used and the number of samples in each bin. This information is given in Tables 3-5 of the revised manuscript.

Responses to comments by Prof. Ghan

We appreciate very much the constructive comments by Dr. Ghan. All comments are addressed as detailed below.

1. General: This is a welcome addition to the literature on the relationship between CCN concentration and aerosol optical properties. Previous manuscripts have ad-

dressed the relationship, but not with such a large set of data spanning diverse regions of the world. It has practical value for satellite remote sensing studies of aerosol effects on clouds.

Response: Thank you for noting the value of the study and its distinction from previous work.

2. The figures are quite clear, but the English needs much work.

Response: We have edited the language in the revised paper as carefully as possible.

3. Specific: Page 3, lines 16-17. Ghan et al. (2006) is an odd reference here, as that manuscript does nothing to establish the sentence preceding it.

Response: We have removed the citation and have added it to a more suitable place in the manuscript.

4. Page 4, lines 23-24. You could mention here that Ghan and Collins (2004) and Ghan et al. (2006) tried to account for the influence of RH.

Response: We have added the sentence “Previous attempts have been made to try to account for the influence of RH (Ghan and Collins, 2004; Ghan et al., 2006), but few systematic investigations have been conducted (Andreae, 2009), due partially to the dearth of measurements available at the time.”

5. Page 10, line 12. Note that at NIM the correlation between CCN and AI even increases with wavelength. Can you explain this?

Response: The slight increase in the correlation between CCN and AI at the NIM site may be due to dust aerosols that dominate this area. Compared with other pollution aerosols, dust aerosol optical depth shows a slightly decreasing trend with increasing wavelength.

6. Page 10, lines 24-25. Why is the correlation between CCN and scattering lowest at Grasiosa?

Response: The GRW site is dominated by sea salt aerosols. Because of their large sizes, their scattering may be strong relative to the low number concentration of large particles that are converted into CCN.

7. Page 11, line 3. You should reference the Ghan papers here, which tried to determine vertical profiles. It is also worth mentioning here that the ARM program is producing vertical profiles of CCN at its sites using the Ghan algorithm. I do NOT ask that you compare the surface CCN measurements with the values from the Ghan algorithm, because it uses the surface CCN as input.

Response: We have added the following passage to the manuscript: "Note that the ARM program has adopted the method of Ghan et al. (2006) to produce vertical profiles of CCN at its long-term sites. The method is based on aerosol extinction profiles, as well as the surface CCN measurements. While their method and ours differ significantly because they rely on different types of scattering received by active and passive sensors, the fundamental principle is the same, namely, making use of optical measurements to derive CCN."

8. Page 16, line 5. GCMs do NOT use such relationships. The relationships are most commonly used in cloud-aerosol interaction studies with satellite data. See e.g., Quaas et al., Atmos. Chem. Phys., 2009.

Response: We have changed this statement to "Aerosol loading has often been used as a proxy or predictor of CCN in cloud-aerosol interaction studies due to the dearth of CCN measurements."

9. Technical: Page 2, line 26. do not understand what the authors are trying to say. Get help with the English.

Response: The sentence has been changed to "Parameterized relationships are developed for estimating CCN which account for RH, particle size, and SSA."

10. Page 15, line 13 and 25. Are these correlations for data from all sites?

Response: They are just for the SGP site.

11. Page 17, line 18. Cite the Ghan papers here.

Response: Done.

Response to comments by referee #2

The authors of the present manuscript acknowledge the reviewer for carefully reading and providing constructive comments that lead to an improved paper.

1. General comments: However, my major concern is that the paper did not clearly outline "what are the scientific assumptions of analysis and data selection?" For example, CCN concentration used in this paper is S=0.4%, which is commonly used for convective clouds and may not atmospherically relevant to each sampling site. Because the goal of this paper is to provide more general/practical relationship between CCN and aerosol optical quantities, the data analysis at S=0.4% only will hinder the effort. In addition, it is good to be consistent with other researches, but author should provide more legitimate reasons for data selection.

Response: We agree that the CCN concentration at S=0.4% is more relevant to convective clouds and less pertinent to general atmospheric conditions. There were two reasons why we used data at S=0.4%. First, CCN measurements were made at variable S at different sites and different periods. Data at S=0.4% are commonly available and have been used by others, for example, in the highly cited study by Andréa (2009). Using CCN data at this S thus allows us to compare our findings with his. Second, the primary objective of this study is to investigate the impact of various factors on the CCN-AOD relationship. As far as this objective is concerned, our findings are not radically affected by the S value. It is important to obtain as many samples as possible so that meaningful statistical relationships can be established. Such relationships have a high practical value, even though its usage may be limited by the potential dependence

of the relationship on aerosol type that is yet to be exploited when richer information on aerosol composition becomes available. Having stated this, we do appreciate the comment about conducting more investigations using S=0.1%. Results from that analysis have been added to section 2.2 and section 3.5 in the revised manuscript.

Added text in Section 2.2: "CCN measurements were made at different values of S, and AOD at different wavelengths, but to easy compare our finding with the study by Andreae (2009), the data used here were made at S=0.4% and at 500 nm, respectively. Note that S=0.4% is more representative of convective clouds, but is too high a value for stratiform clouds. To compensate for this, low S values (S=0.1%) were considered in deriving the general aerosol optical quantities-CCN relationship for practical applications, as presented in section 3.5."

Added text in Section 3.5: "As mentioned in section 2.2, a CCN parameterization is also given for CCN concentrations at S=0.1%. Using AOD500, the parameterization is CCN0.1=3.4eˆ4·[AOD500·α]ˆ2.4752 0.85<SSA<0.95 , (8) where R2=0.90 and RE=0.91. Using σsp at 450 nm, the parameterization is CCN0.1=0.7591·[σsp·α]ˆ1.5621 0.85<SSA<0.95 , (9) where R2=0.99 and RE=0.20."

2. Minor comments: Page 23030, Line10-15: The largest mean alpha were observed at FKB site, where AOD and alpha were retrieved from MFRSR. How does the retrieval contribute to the uncertainty of alpha value? Are the comparisons among those sites fair?

Response: AOD was retrieved from Cimel sunphotometers whose quality and consistency is rigorously maintained by the AERONET at all sites but the FKB site. At that site, an MFRSR is used to retrieve AOD. Note that all ARM instruments are intercompared and well maintained. The retrieval of AOD from the MFRSR is also a mature technique that has been widely used. The consistency between AOD retrieved from a Cimel sunphotometer and an MFRSR was investigated in an earlier study (Lee et al. 2010, JGR). A good agreement was found at all wavelengths except for a somewhat

larger disagreement at 415 nm. Measurements made at 415 nm were not used in the estimation of the Angstrom exponent. The FKB site is located in an agricultural and forested area full of rich biogenic aerosols with little anthropogenic and dust aerosols. The relatively large mean α at the FKB site suggests that fine particles dominate in this area. We have made changes in the second paragraph of section 2.1 to reflect these statements.

3. Page 23031,Line 12, section 3.1.1: The section discussed the well correlation between AOD and CCN0.4 for SGP, GVAX and FKB, but did not mention the poor correlation for GRW site at all. Why is the correlation coefficient so low for GRW site? Is it because data selection? Analysis assumption? Or marine environment? Again in section 3.1.2 and section 3.2, lower correlation coefficients are also observed in Fig2 and Fig 4 for GRW site. Further discussion or explanations are needed.

Response: We have added a discussion about the low correlation between CCN and aerosol optical quantities at the GRW site. The primary challenge of estimating CCN concentration from aerosol optical measurements is the discord between aerosol attributes that dictate CCN and those that affect visible light extinction. The low correlation at the GRW site may be attributed to sea salt aerosols. Because of their large sizes, their scattering may be strong relative to the low number concentration of large particles that are converted into CCN.

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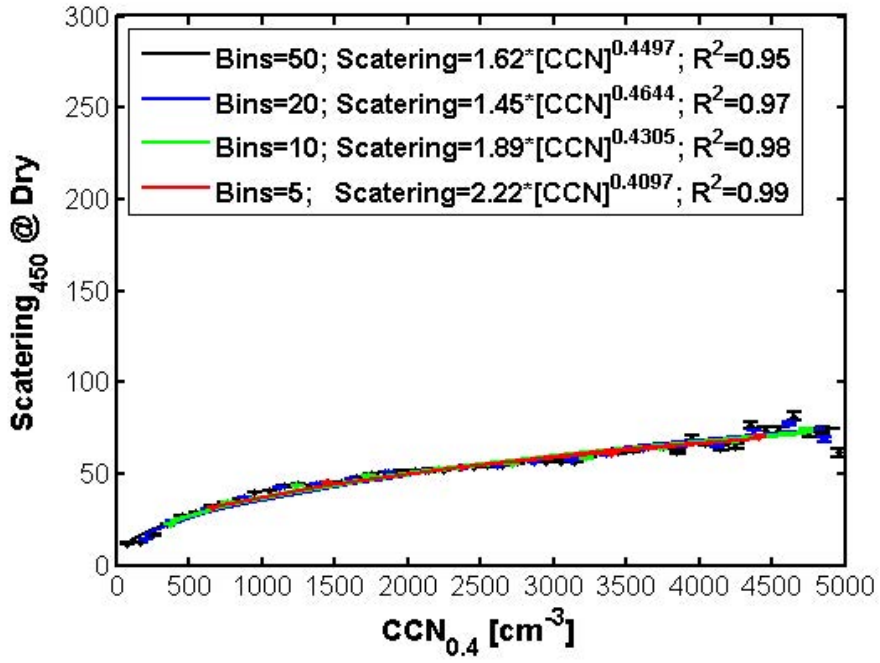

Fig. 1. Relationship between scattering coefficients and CCN for different numbers of CCN bins