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Discussions

# *Interactive comment on* "Influence of common assumptions regarding aerosol composition and mixing state on predicted CCN concentration" *by* Manasi Mahish et al.

**Anonymous Referee #2**

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This paper describes several years of growth factor, CCN data, and chemical composition data collected at the SGP site. CCN concentrations are calculated from aerosol size distributions and constrained assumptions about aerosol hygroscopicity. Sensitivity calculations to the assumptions mixing state are presented.

The presented work analyzes an impressive amount of data in the form of a traditional CCN closure study. The data and modeling results are interesting and relevant to the readers of ACP. Taking the analysis at face value, the results are what one would reasonably expect. However, important details are missing and the manuscripts requires significant clarification. In addition, the manuscript is poorly written.

I concur with the points raised by referee #1. As written, it is not clear how the manuscript is on par, or advances the field relative to the cited studies in the introduction.

The most significant weakness is the complete absence of details on the instrumentation, measurement uncertainties, data quality, complexity of the data set, and data processing used. The description of the methods is insufficient to understand what the authors did, and what the uncertainties are. This prevents a critical evaluation of the manuscript. In it's current form, the manuscript is not suitable for publication.

After several attempts, I cannot make sense of section 4. It is unclear what the authors mean by an ideal solution. The derivation of kappa from experimental data makes no claim about ideality or not. In practice, kappa serves as an activity coefficient. Perhaps the authors seek to express that they use the kappa measured at RH = 90% to predict CCN activity? If so, the assumption is that the activity coefficient at RH = 90% is the same as at the composition of droplet activation (which also is often at non-ideal compositions). There is abundant set of studies that show prove that this is a good assumption and an equally abundant set of studies that show that this may be a bad assumption. Another possibility is that the authors refer to ideality as ZSR mixing. In either case, I do not understand how the data in Figure 2 relate to ideality. A better explanation is needed here.

Equation 2 is presumably derived from the ZSR mixing rule. However, the derivation and origin of that equation is unclear. A kappa_org is derived through averaging over mixing state within the population at a single size, then averaged over all sizes, and the parsed through several unceratin quantities, including OA and BC mass fraction, mass absorption efficiency, OA and BC density, and an assumed kinorg. Given these assumptions, it would be important to define some uncertainty on the derived estimate. How relevant is the volume weighted average kappa to the 40-80 nm kappa that likely drives CCN closure? Figure 6 in Mahish and Collins shows an increase from ∼0.1 to ∼0.2 between 50 and 400 nm. What size does the volume weighted average kappa most correspond to (i.e. what is the volume weighted mean diameter)?

Justification for splitting up the distribution shown in equations (4) and (5) is unclear. I can follow the algorithm mathematically. However, how to these three partial size distributions for inorg, org, and insoluble relate to the actual mixing state? Does this algorithm reproduce the measured kappa distributions at the different dry diameters? If so, this needs to be shown. If not, then the algorithm seems a semi-arbitrary decomposition into partial size distributions.

In general, the experimental description lacks important details. While data downloaded from an archive may be partially quality controlled, the required analysis for this type of work must go well beyond the standard Q/A procedures. Well described quality controlled data sets are a prerequisite to closure calculations.

(a) ACSM: The manuscript doesn't even provide a reference. to the ACSM. No lower size limit of the aerodynamic lens is provided. The potential role of species that is not measured by the ACM (black carbon, dust) in contributing to composition is ignored. Other experimental uncertainties in deriving mass fractions are not mentioned. (E.g. was derived sulfate compared to a PILS? What are the calibration factors applied to data? What is the statistical uncertainty in the fractions?) The ACSM is used in equation (2). Is there mass closure/volume closure between the ACSM and the SMPS volume? How relevant is ACSM data for CCN activation and growth factors at small sizes?

(b) GF: No GF data were presented, nor is even a brief description about the HTDMA included. Some technical details about flow rates, flow ratios, data inversion, humidity calibration, data inversion, and data reduction can be gleaned from two cited references (Collins, 2010, and Mahish and Collins, 2017). However, after reading Collins 2010, it lists data quality flags. How were these used in the processing? The reader should have sufficient information that they can download the data and repeat the calculations as presented in the paper. Currently this is not possible.

(c) CCN: It's surprising that no information is provided about the CCN data at all in the paper, given that the main focus is one of CCN closure. How was the supersaturation calibrated? How were the data quality controlled other than removing data points with Nccn>Ncn? (Why is the CCN broken in this case, and not the CPC?) How was the instrument operated (other than the brief info in Table 1)? Absolute temperature and pressure changes affect the instrument supersaturation, were those accounted for in the processing? Was stability of the temperature gradient monitored? Was the droplet size distribution used to monitor data quality? Was flow stability verified and calibrated? Where inlet losses accounted for?

(d) The origin of equation (2) is unclear. Giving the expression the benefit of the doubt, how was BC measured? An instrument to measure BC is not listed in Table 1. Which instrument in Yang et al. (2009) was used? Or is it cited for the mass absorption efficiency? Furthermore, what is the uncertainty in kappa_org using this approach? It should be evaluated using error propagation. Also, the seasonal cycle shown here should be distinguished from, and compared with, the seasonal cycle from their previous paper (Mahish and Collins, 2017), where the authors show the size resolved seasonal cycle in kappa over more years from the same dataset.

The closure approach to replicate absolute CCN concentrations from size distribution data. To do this well a myriad of non-trivial experimental issues must be addressed with care, which is not obvious from this paper. The DMT CCN instrument is prone to significant particle loss at D < 100 nm. For example, Figure 2 in Hodas et al. (http://www.atmos-chem-phys-discuss.net/acp-2016-236/) shows a gradual decline in activated fraction between 100 and 40 nm for ammonium sulfate, even though 100% are expected. The decline is due to losses in the inlet. Therefore, the absolute CCN number needs to be loss corrected before comparing to an integrated number from a size distribution. It needs to be verified that number closure between the SMPS and CPC is achieved. Furthermore, it needs to be verified that the SMPS sizing is stable and accurate, and preferably, the supersaturation calibration should done on the same

SMPS system than the one used for closure to avoid absolute biases. (A 5% sizing difference between the calibration SMPS and sample SMPS results in a significant bias in CCN closure). Getting all of this right is challenging for a short-term campaign and much more challenging in multi-year semi-autonomous sampling site. Discussion of these effects, uncertainty analysis, and long-term stability analysis are needed.

Discussion of the results in the context of past CCN closure attempts should be included, beyond the cursory mention in the introduction. There is a clear evolution of these studies over time, with more recent studies generally showing a higher success rate, in part due to the improved understanding of the experimental difficulties. How does the closure attempt here compare to previous results? What new insights are gained from this dataset?

Other comments

"In several studies, particles composed entirely of organic species have been reported to be largely ineffective in droplet formation (Abbatt et al., 2005; Prenni et al., 2007)"

I am not sure what ineffective in droplet formation means. Prenni et al. Report data for SOA having kappa = 0.1, which corresponds the kappa_org derived here and elsewhere. Are the the aerosol here ineffective in droplet formation?

"in Figure 6 and for all of 2011 in Figure S3"

change to Figure 7 and S3

Eq. 6, Table 3: what is the interpretation of NRSME values? Obviously smaller is better. What other utility does that metric provide in weighing the different approaches? Please specify.

Fig. 7 I suggest to add +/- 20% or similar error lines to the scatterplots.

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