

Responses to reviewers' comments on "Size-resolved cloud condensation nuclei (CCN) activity and closure analysis at the HKUST Supersite in Hong Kong" by J. W. Meng et al.

(Please note that we have changed the title of the paper to highlight the CCN study. This paper was originally called "Cloud condensation nuclei (CCN) and HR-ToF-AMS measurements at a coastal site in Hong Kong: size-resolved CCN activity and closure analysis.")

We sincerely thank the suggestions and comments from the reviewer and the editor. Here we provide point-by-point responses to those comments.

Re-review comments from reviewer

Comment 1

The author did not answer my major comment 3: How does the fit result represent the measured activation ratio? To my experience, size resolved activation ratio curves are usually not rotational symmetric. Sometimes a sigmoidal function such as eq.(1) can not represent well the original curve. The representative of the fit results cannot be proved by the closure study with the average fit, since the closure results are mainly influenced by the temporal variation of the activation curve.

Also, it seems that only average fit was used in the closure study. Why not use the average measured activation curve for the closure? Now the bias in the closure results stem from a mixture of the uncertainty of measurements, temporal averaging and fitting. By using the average measured curve, at least the uncertainty in the fit can be avoided.

Response :

Highlighted below are the original comment and our response in last review.

Original Comment:

In p.9075 line 2 it is mentioned that "Then, the size-resolved CCN activation ratio was obtained by fitting the activation fraction with the sigmoidal function described by Eq.(1). Is the fit result of activation ratio used in the calculation of N_{CCN} ? How does the fit result represent the measured activation ratio? Why not using measured activation ratio in the calculation? It will be also interesting to see the result of N_{CCN} calculated with individual measured size-resolved activation ratio.

Response:

Since the measured activation ratios were obtained by dividing independent measurements of NCCN by the measured NCN at each size, calculating NCCN based on each activation ratio scan and NCN at each size would not be meaningful. In the closure study, the averaged fit of the measured activation ratios was used for NCCN prediction to see how well it represents the individual activation ratios in terms of overall

closure. Furthermore, we compared the results based on the curve fit with the results based on the average D_{50} to examine the role of mixing state in NCCN prediction.

We have misunderstood the original comment in last review. We did not use the actual measured CCN data directly because it would involve a circular argument of using the measured CCN data to predict the CCN after some manipulation. Furthermore, using fits to represent the size resolved activation ratio data, instead of directly reading the specific median data point, is a common practice to obtain D_{50} for representing the general activation characteristics of aerosols.

As the editor has suggested, we computed N_{CCN} directly from individual S curve fit and compare the N_{CCN} values with the measured ones. Most of the data points have differences of the calculated and measured values of less than 15%, which is considered the results of a good fit of CCN data according to Padró (2012).

As stated in our original response, our goal is to see how well the averaged fit can be used to predict N_{CCN} . It would serve a better and consistent comparison to use the fitted S curve accordingly to examine the role of mixing state on N_{CCN} prediction, in comparison with a single parameter D_{50} that implicitly assumes internal mixing. The S curve fit represent general activation characteristics of aerosols well as integrating the fitted S curve reproduce the measured N_{CCN} within 9% (Fig.5 iii e-h).

We have added the following sentence in section 3.3.3:

Since D_{50} was obtained from the sigmoidal fits, those fits instead of actual data points were also used in the second method for better comparison.

Comment 2

The answer to my minor comment 1 can not really convince me. I do not think a concentration of 1000 cm⁻³ is too low for CCN measurement and can cause such a large uncertainty in measured CCN number concentration.

Response

Below shows the original comments and our responses

Original Comment:

p.9073 line 10 and fig. S1: Does fig. S1 include the measurement at all the four SS? The slope of the fit of N_{CCN} from column A and B is quite close to 1. But the correlation is so weak comparing with other studies (e.g. Deng et al., 2011). Does the author have any explanation?

Response:

Fig. S1 includes the measurements at all four SS. A possible reason for the weak correlation is the relatively higher uncertainty in N_{CCN} measurements because of the lower CCN number concentrations in this study. The average N_{CCN} was ~ 500 to 2000 cm^{-3} at the four SS (Table I) while Deng et al. (2011) reported N_{CCN} of ~ 2000 to $13,000 \text{ cm}^{-3}$ at SS from 0.056 to 0.70%, as discussed in SI. Another possible reason is that N_{CCN} for column A is the integrated concentrations during each size-resolved particle scan (which took 6 min) but N_{CCN} in column B is the averaged bulk N_{CCN} measurements made every second. The mismatch of the measurements might also have led to the weak correlations.

We still believe that the low concentration we obtained contributes to the weaker correlations. As shown in Table 1, the N_{CCN} concentrations were low, especially at SS=0.15%. $N_{CCN} = 512 \pm 452 \text{ cm}^{-3}$.

Comment 3

I still cannot understand the time table (table 1 in your response) of SMPS and SS scan. Does it mean some SMPS scans are located in two different SS? What kind of SMPS scan in table 1 was finally used in your study?

Response

SMPS scans were continuously made while N_{CCN} were measured. Table 1 is for the N_{CCN} measurements. We apologize for the confusion.

Comment 4

Line 263 and fig. 2 in the manuscript v3: in fig. 2 the max of NR-PM1 is about 60 ug m^{-3} but not 72 ug m^{-3} as state in the text. Please check the figure and the data again.

Response

Thank you for identifying the typo. It should be 62.4 ug m^{-3} and is corrected.

Comment 5

Fig. 3 in the manuscript v3: the right y-axes label should be Greek letter “kappa”.

Response

Corrected, now it is “ κ ”.

Comment 6

Section 3.3.3: the word “hygroscopicity” used in this section is not appropriate. Hygroscopicity is a property of aerosol. The hygroscopicity of aerosol population is determined by both chemical composition and mixing state. Some expression sounds strange or confusing, such as “non/less hygroscopicity species”, “the relative importance of mixing state and hygroscopicity”, “when hygroscopicity increased from 0.30 to 0.39”, “the sensitivity of the N_{CCN} prediction to hygroscopicity”, “ N_{CCN} prediction is more sensitive to mixing state at high SS than hygroscopicity”, etc.

Response

We agree with the comments made by the reviewer that hygroscopicity depends on mixing state and compositions. In this paper, hygroscopicity refers to the hygroscopicity of the chemical components/species (or the Kappa values). In the revised manuscript, we have clarified this at the end of the introduction.

“Hygroscopicity is technically a property of aerosols and it is determined by their chemical composition, mixing state, and size distribution. In this paper, we refer hygroscopicity as a property of the components, assuming internal mixing, in aerosols for the discussions below.”

Comment 7

The comparison between the N_{CCN} calculated with average D_{50} and average AR curve is unreasonable. Large temporal variations can be found in fig. S6 for AR curve. In fig. 5iii, the bias of the slope in subplots e-h mainly stem from the temporal variation of AR (maybe also the uncertainties in the sigmoidal fit of measured AR); while the bias of the slope in subplots a-d mainly stem from the disadvantage of D_{50} (i.e. stepwise AR) and the temporal variation of D_{50} . The influence of the temporal variation is unknown, thus the comparison between those biases of slopes can not support the conclusion “ N_{CCN} was found to be more sensitive to hygroscopicity than to mixing state at SS = 0.15% but the reverse is true at SS = 0.70%”. To see the importance of aerosol mixing states to N_{CCN} prediction, one should use real-time D_{50} and AR. Actually, fig. 5ii a-e can give hints to the role of assumption of mixing states. In these subplots, x-axes can be considered as the N_{CCN} calculated with real-time PNSD and size-resolved AR, and y-axes is the N_{CCN} calculated with real-time PNSD and D_{50} . From those subplots it can be seen the N_{CCN} calculated with real-time PNSD and D_{50} is 6%, -6%, 3% and 10% higher than N_{CCN} calculated with real-time PNSD and size-resolved AR for at SS of 0.15%, 0.35%, 0.50% and 0.70%, respectively. This result does not coincide with the conclusion of section 3.3.

Response

We thank the reviewer for the thoughtful comments.

However, we are not sure if the reasons on the bias of Figure 5iii are due to the temporal variation of the AR and D_{50} . Temporal variation exists even if there is perfect closure as the ambient aerosol changes its characteristics, i.e., chemical composition, mixing state and aerosol hygroscopicity, all the time. When we compared Figure 5iii (a-d) and 5iii (e-h), in which both columns of data are affected by temporal variation, we aimed at comparing the effects of (a-d) assuming internal mixing as via a D_{50} value and (e-h) using actual AR fit that reflects the actual extent of mixing state. From $SS=0.35\%$ to 0.70% , the difference of the slopes between the two approaches widen. Hence, we propose that the role of mixing state increases as SS increases. This is intuitively consistent with the discussions later in the manuscript that at high SS , activation will be less selective and hence component hygroscopicity becomes less important. Furthermore there is no obvious reason why temporal variation would yield trends of the observed difference of the slope as a function of $\%SS$ shown in the figure.

The discussion of Figure 5ii raised by the reviewer is useful to improve our understanding of the results. However, comparing y axes and x axes is more on the results of a closure analysis of using size resolved κ_{AMS} than the comparison of chemical composition and mixing state.

Having said that we do agree that the discussion on comparing the role of mixing state and chemical composition is not as solid as we wanted because the differences in the trends between 5iii (a-d) and 5iii(e-h) are rather small and that the data at $SS=0.15\%$ was compromised by lower particle counts. We have softened the tone in these discussions in the revised ms.

Changes to manuscript:

The sentence on lines 491-493, page 19, now read:

This comparison supports that N_{CCN} prediction is likely more sensitive to mixing state than to hygroscopicity at high SS and vice versa at low SS .

Line 515-516, page 19 now read:

N_{CCN} appears to be more sensitive to hygroscopicity than to mixing state at $SS = 0.15\%$ but the reverse is true at $SS = 0.70\%$.

References:

Padró L. T., Moore, R. H., Zhang, X., Rastogi, N., Weber, R. J., and Nenes, A.: Mixing state and compositional effects on CCN activity and droplet growth kinetics of size-resolved CCN in an urban environment, *Atmos. Chem. Phys.*, 12, 10239-10255, doi:10.5194/acp-12-10239-2012, 2012.