Interactive comments on "Size-resolved cloud condensation nuclei (CCN) activity and closure analysis at the HKUST Supersite in Hong Kong," originally "Cloud condensation nuclei (CCN) and HR-ToF-AMS measurements at a coastal site in Hong Kong: size-resolved CCN activity and closure analysis," by J. W. Meng et al.

J. W. Meng et al.

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We sincerely thank the reviewers for the suggestions and comments. Here we provide point-by-point responses to those comments.

Reviewer #1

General comments:
Comment 1:
The paper "Cloud condensation nuclei (CCN) and HR-ToF-AMS measurements at a coastal site in Hong Kong: size-resolved CCN activity and closure analysis" by J. W. Meng, et al. 2014 provide a good data set and the results are very valuable for understanding the aerosol impacts on Pearl River Delta area. The figures are clear. However, the author did not report any daily variation on the data, especially for foggy and hazy days and did not explain the assumptions or appropriate reasons to average the data in each selected period.

Response:
In Fig. 2 of the original manuscript, we reported the time series of N_{CCN}, N_{CN}, their ratios at different SS% as well as the NR-PM1 AMS compositions and total mass during the campaign, including the foggy and hazy periods. Unfortunately, CCN and AMS data were available only for one day during the foggy period and for three days during the hazy period. Shown in Fig. 3 of the original manuscript are the average size-distribution of AMS mass concentrations, the average size-distribution of volume fractions, κ_{CCN} and κ_{AMS} for the foggy, hazy and non-episode days for easy comparison.

The division of the month of May in 2011 into foggy, hazy, and non-episode periods was based on differences in their meteorology, such as RH, temperature and cloud cover, and mass concentration and the O:C ratio (last paragraph, page 9077). During the foggy period, the average RH was 91.1% and the bulk NR-PM1 was as high as 30

μgm^{-3}. The hazy period saw the highest mass concentration of NR-PM1 species recorded during the whole campaign, but the RH was relatively low (66.6%). The highest degree of oxygenation of organic aerosol (OA) with O:C ratio = 0.51 and fraction of organic at m/z 44 = 0.15 occurred during the hazy period. For detailed information please refer to Li et al. (2013). The above information was given in the last paragraph on page 9077.

Changes to manuscript:
The last paragraph on page 9077 manuscript now reads

"There were two periods of particular interest during this campaign: one was a foggy period (15 May) and the other was a hazy period (28–30 May). The division of the month of May in 2011 into foggy, hazy and non-episode periods was based on differences in meteorology, such as RH, temperature and cloud cover, and mass concentration and the O:C ratio. On average, the foggy period had a high RH (91.1%), a low temperature (23.3 ^{o}C) and a high percentage cloud coverage (89.7%) and a high liquid water content (LWC) in fine particles (47.5 $\mu g\ m^{-3}$) as shown in Li et al. (2013). The hazy period had a much lower RH (66.6%), a higher temperature (26.2 ^{o}C) and a much lower percentage cloud coverage (43.3%) and LWC (17.5 $\mu g\ m^{-3}$). The slowing surface winds and the establishment of a well-defined land-sea breeze with a gradual daily reversal of wind direction contributed to the accumulation of local and regional pollutants coming from the PRD due to the persistent northerly and northwesterly air masses (Lee et al., 2013).

During the foggy period, the bulk NR-PM1 was as high as 30 $\mu g\ m^{-3}$ (Fig. 2e; Li et al., 2013). The hazy period was much less humid and it saw the highest mass concentration of NR-PM1 species recorded during the whole campaign. The highest degree of oxygenation with average O:C ratio of 0.51 was also obtained (Li et al., 2013)."

Comment 2:
In addition, the Methods listed in Table 3 are not very clear. Please clarify how to calculate kappa_AMS from bulk AMS measurements and size-resolved measurements. What is the difference? Is the difference significant for this study? If possible, please plot the time series of kappa_AMS calculated from bulk AMS measurements and size-resolved measurements.

Response:

AMS can provide both bulk and size-resolved mass concentrations for organics and inorganics. κ_{AMS} was averaged from the individual values derived from the size-resolved volume fractions of organics and inorganics assuming the densities of organics and inorganics to be 1.3 g cm^{-3} and 1.75 g cm^{-3} (given in section 2.3.2) respectively. Also, it was assumed that $\kappa_{inorg} = 0.6$ for the whole campaign, $\kappa_{org} = 0.2$ for the hazy period and $\kappa_{org} = 0.1$ for the foggy and non-episode periods.

The bulk κ_{AMS} (κ_{AMS_B}) was calculated from the bulk volume fractions of organics and inorganics derived from the bulk mass concentrations using the densities of organics and inorganics shown above. The time-series hygroscopicities derived from bulk and size-resolved AMS measurements are shown in the newly added Fig. S2 (Fig. 1 near the end of this response). κ_{AMS_B} are larger than size-resolved κ_{AMS} (κ_{AMS_SR}) in all four SS because bulk AMS compositions were biased towards the inorganics. Their difference increases as SS increases because the corresponding D_{50} decreases and these smaller particles have a larger difference in organic fraction than the bulk.

Changes to the manuscript:
Page 9076, line 16-22 the paragraph now reads

"The hygroscopic parameter κ_{AMS} can be obtained from AMS measurements using

$$\kappa_{AMS} = \kappa_{org} \times f_{org} + \kappa_{inorg} \times f_{inorg} \qquad (3)$$

where f_{org} and f_{inorg} are the organic and inorganic volume fraction derived from AMS measurements. Bulk κ_{AMS} (hereafter κ_{AMS_B}) and size-resolved κ_{AMS} (hereafter κ_{AMS_SR}) are obtained from the corresponding bulk and size-resolved volume fractions of organics and inorganics, respectively. Also, it was assumed that $\kappa_{inorg} = 0.6$ for the whole campaign, $\kappa_{org} = 0.2$ for the hazy period and $\kappa_{org} = 0.1$ for the foggy and non-episode periods.

The time-series hygroscopicities derived from bulk and size-resolved AMS measurements are shown in Fig. S2. κ_{AMS_B} were larger than κ_{AMS_SR} in all four SS because bulk AMS compositions biased towards the inorganics as discussed below. Their difference increases as SS increases because the corresponding D_{50} decreases and these smaller particles have a larger difference in organic fraction than the bulk has."

Comment 3:
It also seemed the AMS part is used to mainly support CCN analysis in this paper. If

authors plan to report AMS in a separate manuscript, please consider revise the title.

Response:

The current ms focuses on the analysis of the CCN data in May 2011 for which we have both AMS and CCN data. Details of the analysis of AMS data for May 2011 have been published by Li et al. (2013) and Lee et al. (2013). We feel that the title is appropriate but would be happy to remove the term AMS in the title. We also propose to change the title to highlight the fact that the measurements were made at the HKUST Supersite.

Changes to the manuscript title:

The title is now "Size-resolved cloud condensation nuclei (CCN) activity and closure analysis at the HKUST Supersite in Hong Kong".

Specific comments:

Comment 1:

Page 9068, Line 10-12: Based on figure S5, the average size-resolved CCN activation ratio at each SS, it seemed the variation on the D_{50} is around 10% of D_{50}. That will lead to 0.1 variation on the determined kappa_CCN. Is the variation significant for this study?

Response:

The variations in D_{50} and κ_{CCN} are mainly due to the changes in the composition of the measured aerosols. The variations in D_{50} and κ_{CCN} are comparable to those in previous studies in the PRD region (e.g. Rose et al., 2011). In Figure 4, we plotted κ_{CCN} against f_{org} to determine κ_{inorg} and κ_{org}. These variations are useful in our closure analysis.

Comment 2:

Page 9071, Line 5-10: Again, please clarify how to calculate kappa_AMS?

Response:

Please see our response to Comment 2 (General Comments) above.

Comment 3:

Page 9073, Line 14-20: Each SMPS scan is 6 mins, and at SS=0.15%, the system will finish 3 scans (18 mins) and wait 4 mins for stabilization, and the total will last 22 mins. However, at next SS, it lasted 12 min and also need 2 min to stabilize. Does that mean it should have lasted 14 mins? Please clarify the description of CCN running

sequence.

Response:

Simultaneous measurements of SMPS and CCN were made. SMPS measurements were made continuously with each scan taking 6 minutes. The sequence of CCN measurements is more complicated to allow time for changing and stabilizing SS settings. The table below shows the time sequence of CCN measurements. The highlighted time segments indicate the amounts of time for which the previous SS setting was changed to the subsequent new setting. Longer time durations were used for low SS settings to obtain better statistics. The cycle was repeated for continuous measurements. However, the sequence may be too complicated for general readers. Hence we have decided not to change the manuscript.

time (min)	SS%	total time at SS (min)
6	0.15	22
6	0.15	
6	0.15	
4	0.15	
2	0.35	12
6	0.35	
4	0.35	
2	0.5	12
6	0.5	
4	0.5	
2	0.7	12
6	0.7	
4	0.7	
2	0.15	22
6	0.15	
6	0.15	
6	0.15	
2	0.15	
4	0.35	12
6	0.35	
2	0.35	
4	0.5	12
6	0.5	
2	0.5	
4	0.7	12
6	0.7	
2	0.7	
4	0.15	22
6	0.15	
6	0.15	
6	0.15	
6	0.35	12
6	0.35	
6	0.5	12
6	0.5	
6	0.7	12
6	0.7	

Table I The time sequence of SMCA measurements

Comment 4:

Page 9075, Line 12: What is signal to noise ratio and particle lens transfer efficiency of AMS at the size range less than 200 nm in this study? Please consider to provide it in supplement materials. Because the average activation size in this study is less than 120 nm in mobility size, which suggests the AMS data in the similar range is very important.

Response:

In the CCN closure analysis, D_m ranged from 42 nm to 200 nm, corresponding to D_{va} of 71 nm to 340 nm. The signal-to-noise ratio of the AMS data concerned, i.e. the ratio of mass concentrations for the measurement period to that for the filter period, was higher than 6 for this particle size range.

Changes to the manuscript:

The above sentence has been added in SI where the uncertainty of κ_{AMS} is discussed.

Comment 5:

Page 9076, Line15-23, equation 3: what does author mean "f_i the size-resolved volume fraction"? The subscript "i" stands for each species. Is f the volume fraction of averaged fraction from individual sizes or the bulk fraction of all size? If it is the averaged fraction, please provide time series of the averaged fraction and the bulk fraction.

Response:

Since we only separated the species into organics and inorganics in this paper, Equation 3 was deleted. The original Equation 4 was adequate and is renamed as Equation 3 in the revised ms.

Changes to the manuscript:

Page 9076, lines 15-20, Equation 3 was deleted.

Comment 6:

Page 9077, section 3.1: what is the meteorology information during the sampling period? Does that explain the aerosol sources difference on foggy day or hazy day?

Response:

The last paragraph on page 9077 has some related information but it can be elaborated and rearranged.

Changes to the manuscript:
The first few sentences (lines 22-26, page 9077) are now replaced by:

"There were two periods of particular interest during this campaign: one was a foggy period (15 May) and the other was a hazy period (28–30 May). The division of the month of May in 2011 into foggy, hazy, and non-episode periods was based on differences in meteorology, such as RH, temperature and cloud cover, and mass concentration and the O:C ratio. On average, the foggy period had a high average RH (91.1%), a low temperature (23.3 ^{o}C) and a high percentage cloud coverage (89.7%) and a high liquid water content (LWC) in fine particles (47.5 μg m^{-3}) as shown in Li et al. (2013). The hazy period had a much lower RH (66.6%), a higher temperature (26.2 ^{o}C) and a much lower percentage cloud coverage (43.3%) and LWC (17.5 μg m^{-3}). The slowing surface winds and the establishment of a well-defined land-sea breeze with a gradual daily reversal of wind direction contributed to the accumulation of local and regional pollutants coming from the PRD due to the persistent northerly and northwesterly air masses (Lee et al., 2013).

During the foggy period, the bulk NR-PM1 was as high as 30 μg m^{-3} (Fig. 2e; Li et al., 2013). The hazy period was much less humid and it saw the highest mass concentration of NR-PM1 species recorded during the whole campaign. The highest degree of oxygenation with average O:C ratio of 0.51 was obtained (Li et al., 2013)."

Comment 7:
Page 9077, Line 17 and Figure 2: How does author calculate the bulk volume fraction of NR-species? What assumptions are used for the density?

Response:
The bulk volume fractions of organics and inorganics are derived from the bulk mass concentrations which are in turn obtained from AMS measurements assuming the densities of organics and inorganics to be 1.3 g cm^{-3} and 1.75 g cm^{-3} respectively. Also, see our response to Comment 2 above.

Comment 8:
Page 9078, Line 15, Figure 3: In page 9075, $D_{va}=D_m*1.7$. $D_m=285$ nm in foggy period, then the D_{va} will be around 484.5 nm. Does Figure 3(a-c) in D_m instead of D_{va}? Page 9078, Line 18-19, Figure 3(d-f): why only focused on the range of 42-200 nm (D_m or D_{va}?) of AMS chemical composition? What about the AMS chemical composition larger than 200 nm?

Response:

In Fig. 3a-c the diameter is in the form of D_m instead of D_{va}. AMS data for particles larger than 200 nm were obtained but they were not relevant to the CCN measurements and the closure analysis because they were much larger than the D_{50} of the SS we obtained. We refer the reviewer and readers to Lee et al., (2013) for the details of the size-dependent AMS compositions.

Changes to the manuscript:

Page 9078, line 15: "Hereafter, diameters shown are D_m" is added.

Comment 9:

Page 9078, section 3.2; Page 9081, section 3.3.1: I suggested using something like kappa_AMS_B and kappa_AMS_SR to represent the kappa_AMS calculated from different methods.

Response:

Thank you for the kind suggestion and we have changed the paper accordingly. See line 40 on page 2 and line 239 on page 9 of the revised ms.

Changes to the manuscript:

The size-resolved κ_{AMS} and bulk κ_{AMS} shown have been replaced by κ_{AMS_SR} and κ_{AMS_B} as suggested.

Comment 10:

Page 9079, Line 17-23: What is the O:C ratio in the foggy period and non-episode period? It is useful to include a time-series O:C ratio in the main content to support the discussion.

Response:

The average O:C ratios are 0.43 and 0.39 for the foggy period and the non-episode period, respectively, whereas that for the hazy period is 0.51. The time-series O:C ratios are available in Li et al., (2013).

Change to the manuscript:

The related sentences now read: (page 9079 line 19-23)

In the hazy period (Fig.3e), assuming κ_{org} =0.2 and κ_{inorg} = 0.6 gave better agreement between κ_{AMS_SR} and κ_{CCN}. The hazy period had a higher O:C ratio of 0.51, compared to 0.43 and 0.39 in foggy and the non-episode periods respectively (Li et al., 2013),

leading to a higher hygroscopicity of the organic aerosols (Chang et al., 2010; Lambe et al., 2011; Massoli et al., 2010; Mei et al., 2013; Moore et al., 2012b).

Reviewer #2

Major comments:
Comment 1:
The advantage of size-resolved CCN measurements is that it can provide information on both hygroscopicity and mixing state. The size-resolved N_CCN was measured in this study. However, most of the discussion is limited in kappa and D_{50}. It will be also interesting to see the variation of size-resolved activation ratio during the three episodes. Size-resolved activation ratio may also helpful in the explanation of the closure results in sect. 3.3.

Response:
The average size-resolved activation ratios during the hazy, foggy and non-episode periods at SS of 0.15% and 0.7% are shown in Fig. 7 (also shown in Fig. 2 in this response). At SS of 0.15%, the activation ratios during the hazy and non-episode periods are similar. The foggy period witnessed a higher activation ratio than the rest possibly due to its higher inorganic fraction (Fig. 3d-f) and the smaller amount of non/less hygroscopic organics (Li et al., 2013). At SS=0.70%, the CCN activation ratios of particles from 50 nm to 100 nm are lower in the hazy period than in the other two periods.

On page 9085, lines 5-13, we have:

"From the AMS measurements, the portion of non/less hygroscopic species inferred from the fractions of f_{43} and f_{57} increased as the particle size decreased (Lee et al., 2013). Because of their higher abundance, their mixing with the hygroscopic components has a higher impact at SS = 0.70% (D_{50} = 46 nm) than at low SS = 0.15% (D_{50} = 116 nm), where the reduction in the overestimation is minimal, from 10% when using the average D_{50} to 9% when using the average activation ratios. On the contrary, a difference of 19% was found when hygroscopicity increased from 0.30 to 0.39 at this low SS."

The difference in the trends at SS=0.15% and 0.70% may be due to the larger fractions of non/less hygroscopic species in smaller particles in the hazy period (Li et

al., 2013). These particles, which constitute a larger fraction of OA in the hazy period than in the other periods, likely formed external mixtures containing the aged particles of sulfate and the more oxidized (and hygroscopic) organics. Hence, a larger difference in the activation ratios between the hazy and the other periods were observed at SS=0.70% than at SS=0.15%.

Changes to the manuscript:
We have added the following paragraph in the revised manuscript after the quoted paragraph above.

"The average size-resolved activation ratios during the hazy, foggy and non-episode periods at SS=0.15% and 0.7% are shown in Fig. 7. At SS=0.15%, the activation ratios during the hazy and non-episode periods are similar but it is higher during the foggy period due possibly to the higher volume fraction of inorganics (Fig. 3d-f) and the smaller amount of non/less hygroscopic organics (Li et al., 2013). At SS=0.70%, the CCN activation ratios of particles ranging from 50 nm to 100 nm in size are lower in the hazy period than in the non-episode period. The difference in the trends at SS=0.15% and 0.70% may be due to the larger fractions of non/less hygroscopic species in smaller particles in the hazy period. These particles, which constitute a larger fraction of OA in the hazy period than in the other periods, likely formed external mixtures containing the aged particles of sulfate and the more oxidized (and hygroscopic) organics. Hence, a larger difference in the activation ratios between the hazy and the other periods could be observed at SS=0.70% than at SS=0.15%."

Comment 2:
The author showed the measurement in three cases: a foggy, a hazy and the rest. It will be better if the author can explain the results in respect of the differences of these cases (such as meteorology information, pollution condition, air mass type, etc.).

Response:
Reviewer 1 also raised a similar comment. Please see our responses to Comment 1 of Reviewer 1.

Comment 3:
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 N_{CCN} by the measured N_{CN} at each size, calculating N_{CCN} based on each activation ratio scan and N_{CN} at each size would not be meaningful. In the closure study, the averaged fit of the measured activation ratios was used for N_{CCN} 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 N_{CCN} prediction.

Comment 4:

There are a lot of figures show linear regressions of parameters. Some of them does not bring much information. For example, fig. 5, 6 and 8 do not bring any extra information compared to table 4. Please consider to merge or delete them.

Response:

Agree. We have combined them into a single figure.
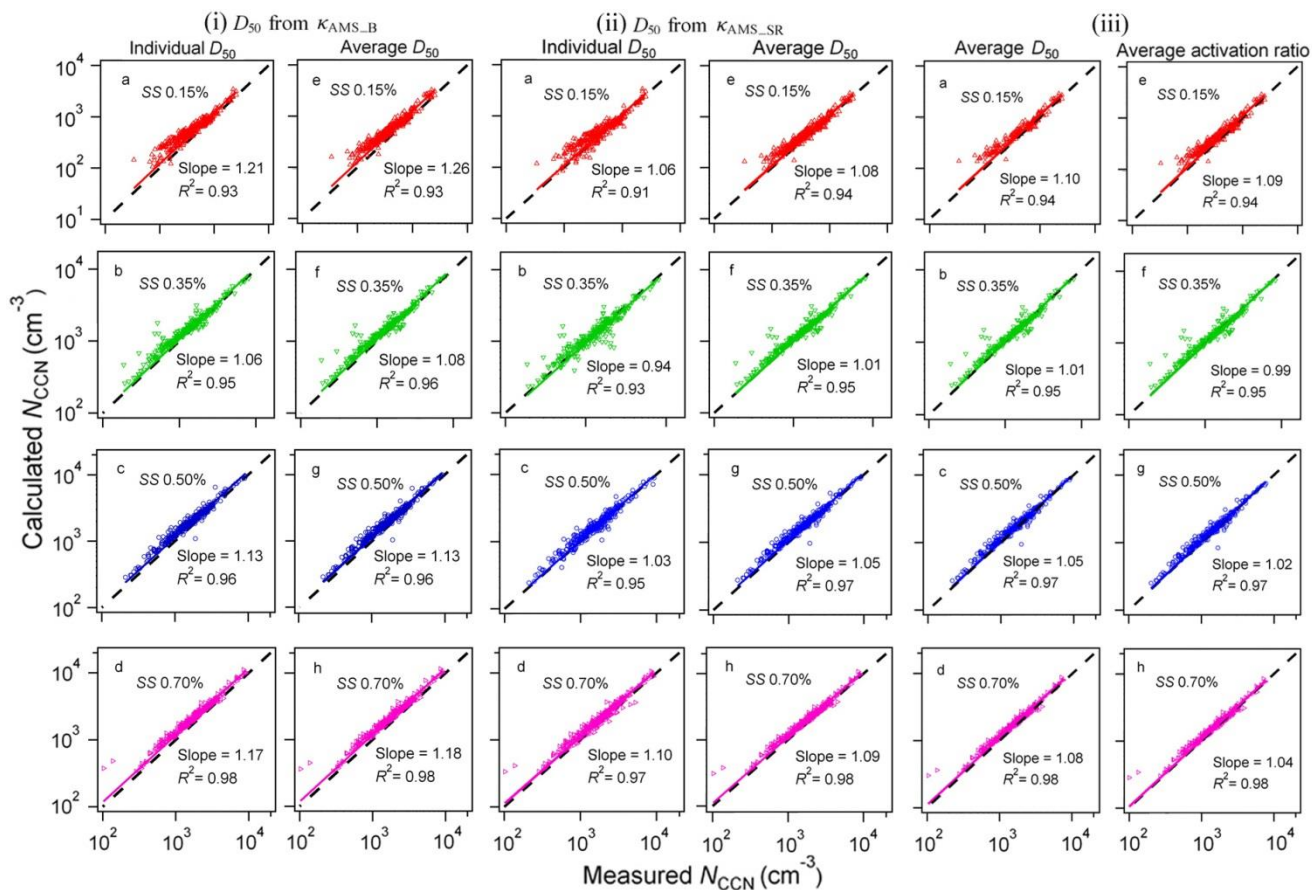
Change:

The above figure replaces the original Figs. 5, 6 and 8.

Minor comments:

Comment 1:

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 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.

Comment 2:

P.9073 line 14: Does the lasting time 22 min and 12 min include waiting time? It is mentioned that the time resolution of SMPS is 6 min. The lasting time of 22 min can be a waiting time of 4 min plus three scans. But what about the lasting time of 12 min? For me also, a waiting time of 2 min for increasing deltaT and 4 min for decreasing deltaT might be not sufficient.

Response:

Please see our responses to Comment 3 of Reviewer 1. Below shows an example of the temperature control results. T1, T2, and T3 are the measured temperatures at the top, middle, and bottom control zones of the column in the CCN, respectively. A Boolean output channel indicates whether T1, T2, or T3 varies by > 0.4 ℃ from their set point values. A value of 1 indicates all three are close to their set points. It can be seen from the figure that a time slot of 4 min for the decreasing deltaT and 2 min for the increasing deltaT is generally adequate for temperature stabilization.
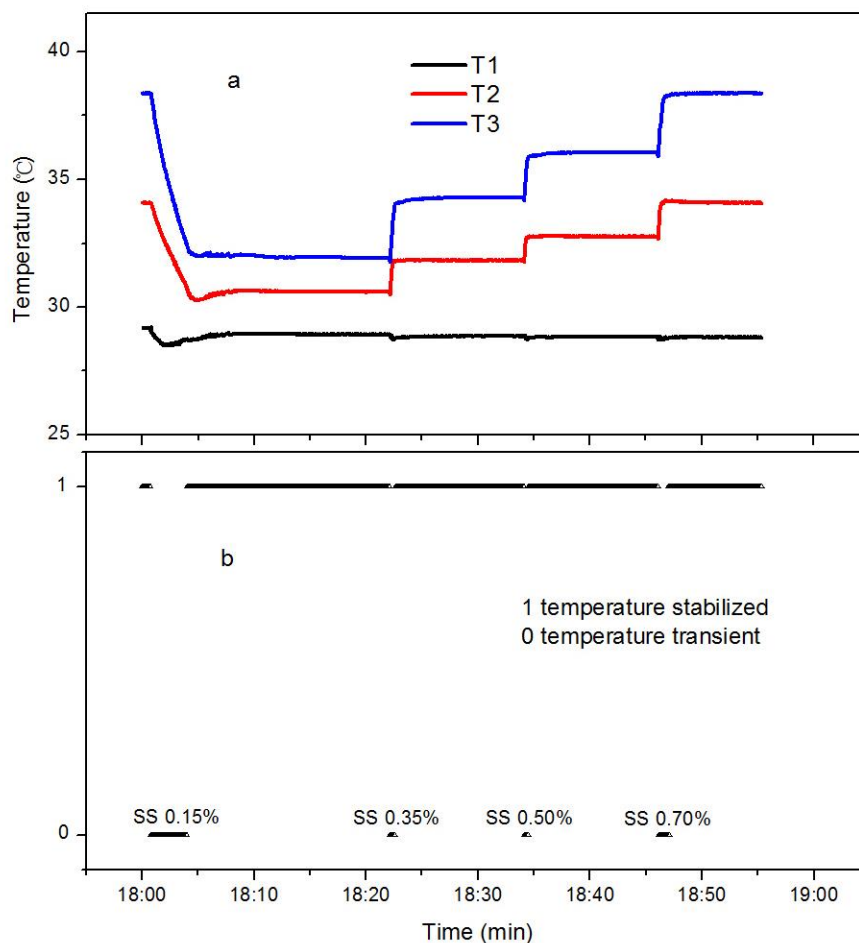
Comment 3:

p.9074 line 18: The EC mass mainly concentrates at small size. Although accounting for only 5% of PM1 mass, the volume fraction of EC can be high at around 100 nm, which is the size range of D_{50} for the SS in this study. It would be good if the EC was taken into account in the calculation of kappa. If size resolved EC information is not available, at least the author should include this issue when explain the bias in CCN closure.

Response:

Size-resolved EC measurements were not available. We agreed that EC might also have caused the overestimation in N_{CCN} prediction.

Changes to the manuscript:

In section 3.3.1, page 9082, line 16-19, we have added a sentence as shown below

"They contribute little to N_{CCN} by themselves but the assumption of internal mixing allows them to contribute to CCN due to their mixing with more hygroscopic species and leads to an overestimated N_{CCN} (Rose et al., 2011; Wang et al., 2010). Size resolved EC was not available and EC might also have caused the overestimation in N_{CCN} prediction."

Comment 4:

p.9076 eq. (1): Equation is not correct. Should be $1/(1+(D_p/D_{50})^c)$.

Response:

Thank you. This was a typo. The equation: $N_{CCN}/N_{CN} = B/(1+(D_p/D_{50})^c)$ is from the SMCA manual available at http://nenes.eas.gatech.edu/Experiments/SMCA.html and Padró et al. (2012). "B" accounts for the plateau (maximum activation ratios), which may not always be unity due to the measurement uncertainty and/or the existence of the non-hygroscopic species.

Change:

The equation 1 now is: $N_{CCN}/N_{CN} = B/(1+(D_p/D_{50})^c)$.

Comment 5:

p.9077 line 8 and table (1): It does not make much sense to provide the statistics of bulk N_CCN, since this value is mainly dominated by the aerosol number size distribution. It would be more valuable to give the statistics of D_{50} in table 1. Are the

statistics in table 1 based on 6-min data?

Response:
We think it is useful to provide a general idea of N_{CCN} and N_{CN} and their ratios measured at this coastal site. Both N_{CN} and hygroscopicity of aerosol, a major focus of this work, play important roles in determining N_{CCN}. The statistics of D_{50} and the associated kappa values are available in Fig. 3d-f and Fig. S2. Yes, these statistics were based on the 6-min data sets.

Comment 6:
p.9077 line 24: A fog event with average RH of 91%? I think a low visibility event can be named "fog" only if some of the particles are activated at supersaturation, otherwise it should be called "heavy haze"?

Response:
We differentiated the foggy period from the hazy period based on the relative humidity and liquid water content (LWC) of aerosols (Li et al., 2013). The average LWC and NR PM1 concentrations in the foggy, hazy, and non-episode periods are 47.5, 17.5 and 13.2 μg m^{-3}, and 19.8, 32.2 and 11.9 μg m^{-3} respectively (Li et al., 2013).

Comment 7:
p.9078 line 8: Again, I cannot get any idea from these N_CCN values, since these values are mainly determined by N_CN. It is better to give bulk N_CCN/N_CN or D_{50} here.

Response:
The statistical and the time-series bulk N_{CCN}/N_{CN} values at the four SS are given in Table I and Fig. 2 respectively. We have now added the bulk N_{CCN}/N_{CN} value as suggested.

Changes to the manuscript: (page 9078 line8-10)

The original sentence
"The mean bulk N_{CCN} ranged from 1100 cm^{-3} at SS = 0.15% to 5300 cm^{-3} at SS = 0.70% during the hazy period, while it ranged from 300 cm^{-3} at SS = 0.15% to 2700 cm^{-3} at SS = 0.70% during non-episode periods"

is changed to

"During the hazy period, the mean bulk N_{CCN} ranged from 1100 cm^{-3} with bulk N_{CCN}/N_{CN} of 0.22 at SS = 0.15% to 5300 cm^{-3} with bulk N_{CCN}/N_{CN} of 0.72 at SS = 0.70%. During non-episode periods, the mean bulk N_{CCN} ranged from 300 cm^{-3} with bulk N_{CCN}/N_{CN} of 0.14 at SS = 0.15% to 2700 cm^{-3} with bulk N_{CCN}/N_{CN} of 0.61 at SS = 0.70%".

Comment 8:
p.9081 sect. 3.3.1 and table 4: It seems that to use individual D_{50} does not bring a better result in the closure than to use average D_{50} for the whole period. Could the author give any explanation?

Response:
Agree. In fact, using the average D_{50} can give a slightly better correlation coefficient than that using the individual D_{50} in some cases. One possible reason might be that the uncertainty in D_{50} increases as SS and N_{CCN} decrease. An example is shown in Fig. 5ii (Fig. 6 in the original manuscript), where N_{CCN} (Fig. 5ii a) at low SS based on individual D_{50} are more scattered than those based on average D_{50} (Fig. 5iie), especially at lower N_{CCN} values. On the contrary, the average calculation of D_{50} might offset uncertainty in closure studies at low N_{CCN}, but will also introduce higher uncertainty for high N_{CCN} compared with the individual D_{50}.

Comment 9:
p.9084 line 9-13: It would be much clearer if the equations with which the N_CCN is calculated are given here

Response:
We apologize for the confusion.

Changes to the manuscript: (page 9084 line 11-13)
The sentence "The second method involves integrating the product of the measured size-distribution of N_{CN} and the average size-resolved CCN activation ratio in each particle size bin" is replaced by
"The second method involves integrating the product of the measured size-distribution of N_{CN} and the average size-resolved N_{CCN}/N_{CN} activation ratio in each particle size bin."

Comment 10:

Fig. 3(d-f): right y-axes: it is better to use "kappa" rather than "hygroscopicity".

Response:

Agree and changed.

References

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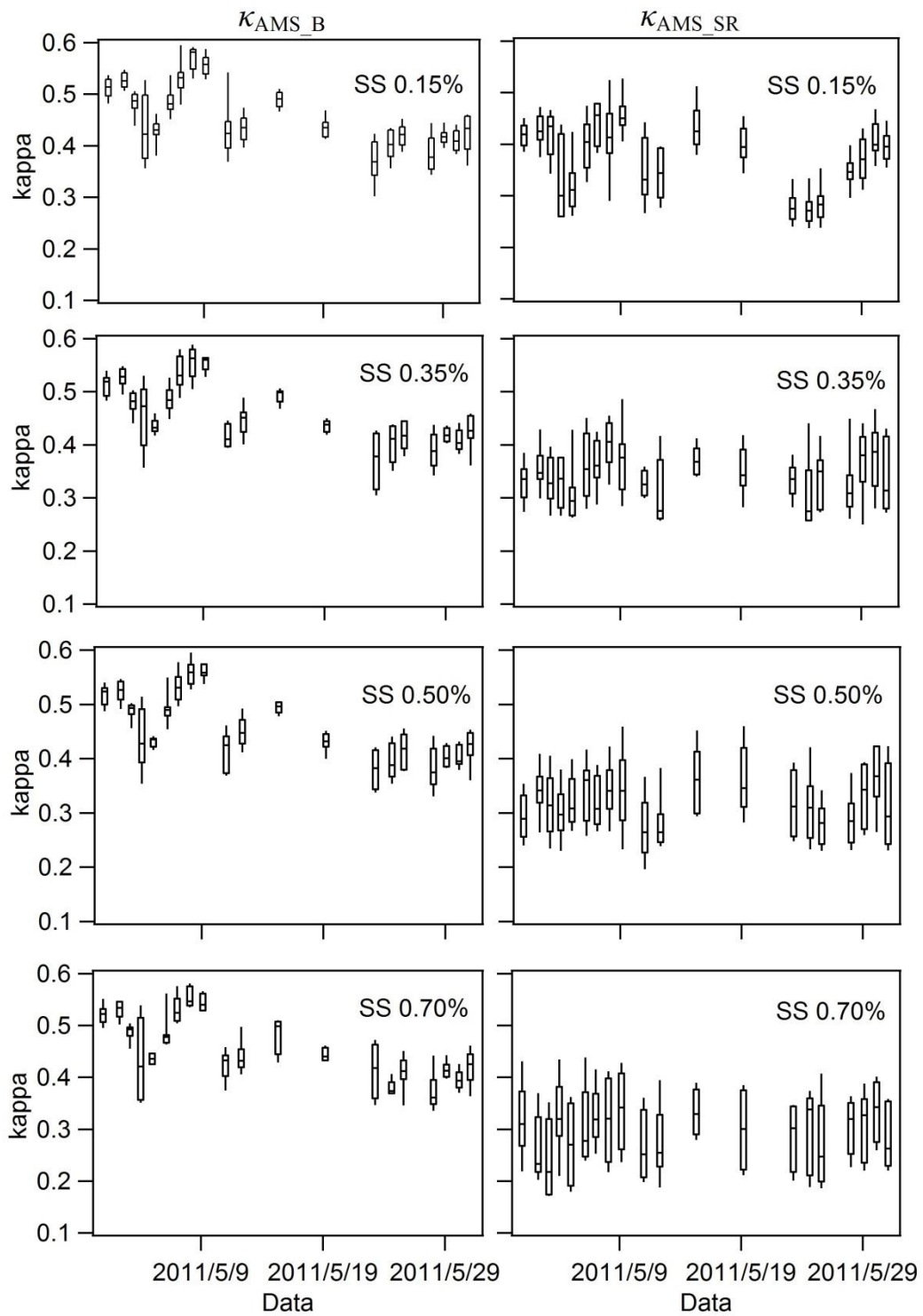
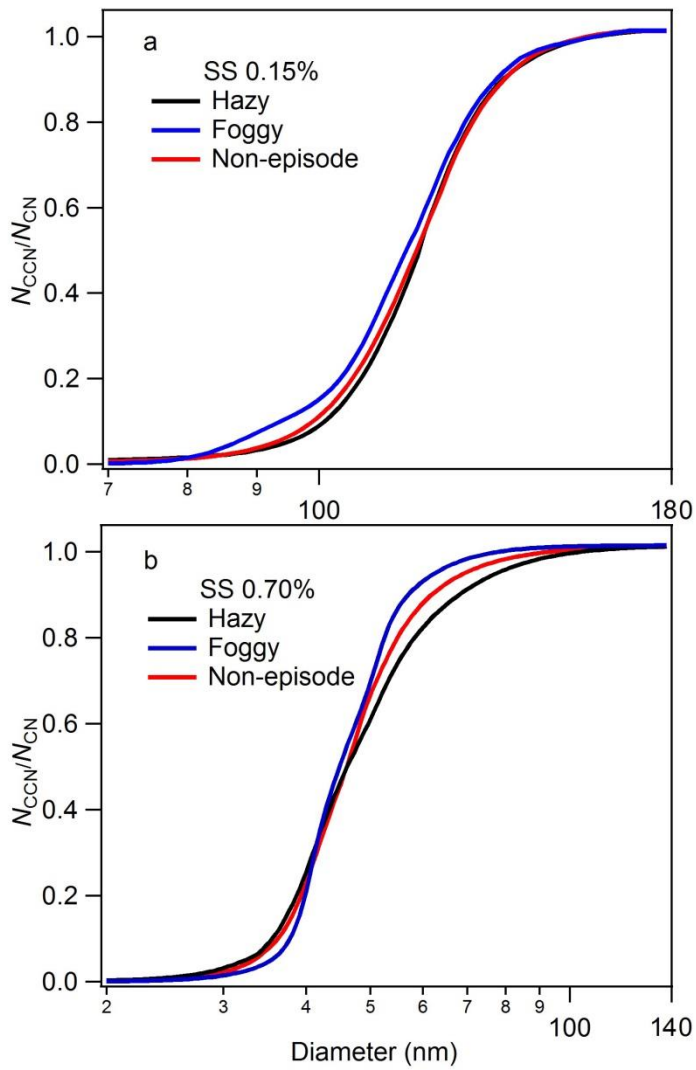Fig. 1 $\kappa_{\text{AMS\_B}}$ and $\kappa_{\text{AMS\_SR}}$ derived from AMS measurements.

Fig. 2. The average size-resolved CCN activation ratio at *SS* of (a) 0.15% and (b) 0.70% during the hazy, foggy and non-episode periods.