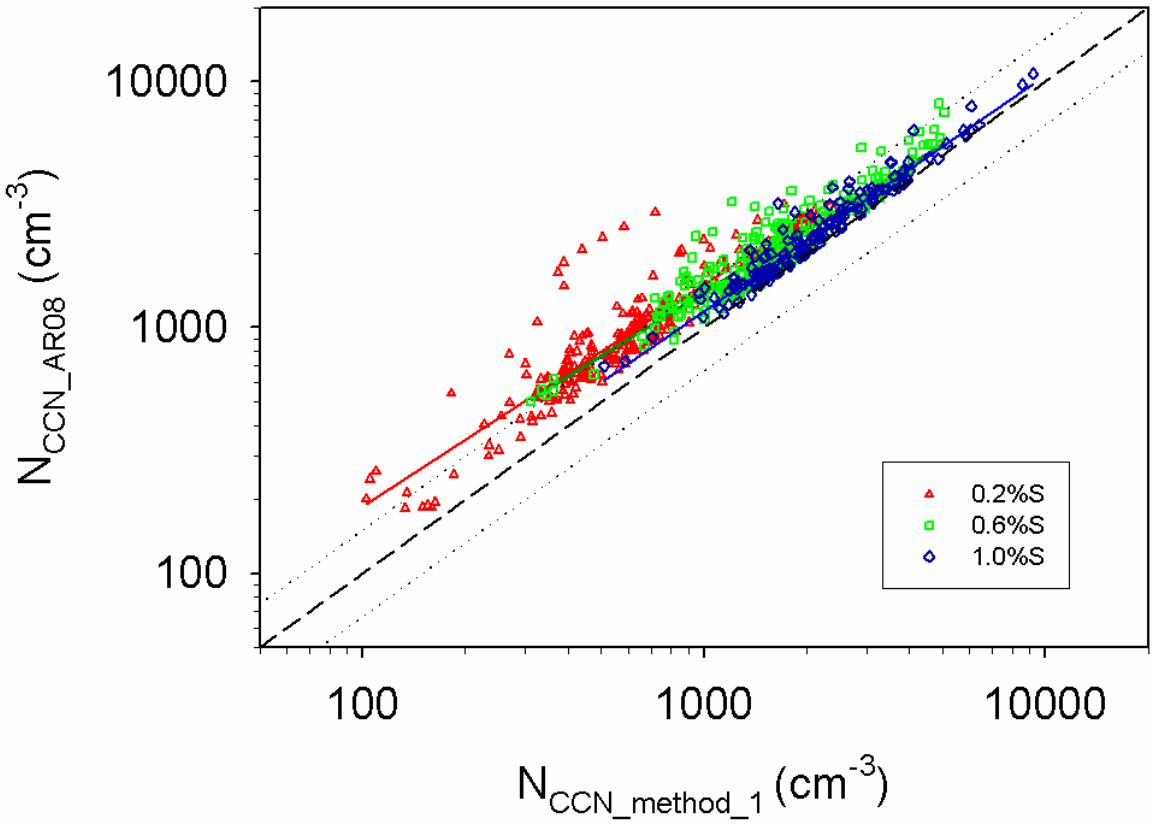
Response to the first referee

General comments

1) The authors compare four different methods for CCN closure and discuss that Method-2GF leads to the best CCN closure whereas the other methods that either ignore size-resolved information show larger deviations. While the data in Table 5 show the deviations for each method from 'truth' (i.e. measurements) a comparison would be useful that shows how much the individual methods differ among each other. I.e., given that aerosol/cloud modelers look for simplicity, how 'bad' would the CCN closure be if e.g. constant hygroscopicity vs size/temporal resolved one is assumed..I recognize that this information could be derived from the data given here, but an additional figure would make it clearer that shows (CCN-predicted by Method 1, 2 or3) versus CCN(predicted using Method 4). The results of this comparison should be briefly discussed in light of discussions that compare CCN effects on cloud droplets e.g., (Rissman et al., 2004; Cubison et al., 2008).

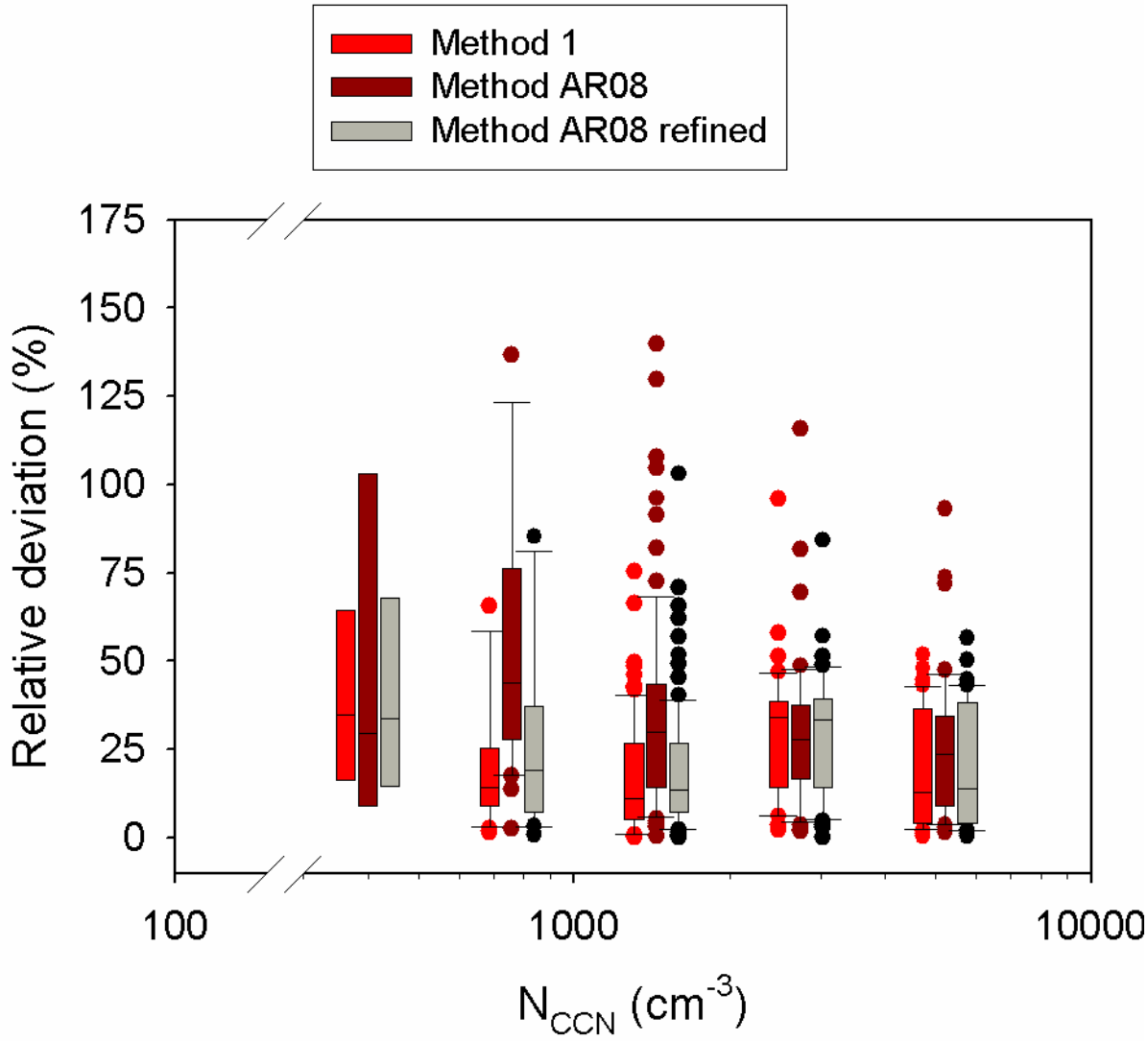
The comparison between the CCN prediction by Method 1 and that by Method 4 (renamed as AR08 following the reviewer's suggestion further below) is shown below and will be added in the revised manuscript as Fig. 12h. Assuming the constant kappa of 0.3 resulted in overprediction by 64±58%, 33±27% and 14±14% for 0.2, 0.6 and 1.0% S, respectively, compared to Method 1. The degree of disagreement between the two methods is larger for lower S. This implies that using the N_{CCN} predicted by AR08 as input to a model that calculates cloud droplet concentration (e.g., Cubison et al., 2008) could result in higher cloud droplet concentration than using the N_{CCN} predicted by Method 1. Eventually this could result in overestimation of the aerosol indirect effects by falsely making cloud droplets smaller and therefore reducing precipitation efficiency. The above statements will be included in the revised manuscript.



2) The suggestion by Andreae and Rosenfeld (AR08) to use kappa = 0.3 for continental aerosol is definitely a useful one and gives 'reasonable' results. As the authors show it could be improved since obviously smaller particles are less soluble. Since it is not feasible to take into account (due to lack of data) or implement size-resolved hygroscopicity data on a global scale into models, could you come up with a refinement of the AR08 recommendation? Does a fixed hygroscopicity (kappa < 0.3) for particles <100 nm give robust and good results?

Based on our data set, we found that prescribing kappa value of 0.17 for particles <100 nm and of 0.30 for particles >100 nm could produce almost as good agreement as Method 1 as shown in the revised Fig. 13 below for Gosan 2008 campaign. Here the red and brown colors are identical to the original Fig. 13 (except for the very minor correction for miscalculation) and the gray color is the result with refinement (kappa=0.17 for particles <100 nm and 0.30 for particles >100 nm). Such finding illustrates the need for the correct hygroscopicity information for particles smaller than 100 nm, which we have already suggested in the manuscript. We understand the need for the refinement but we would not claim that the refinement suggested here can be applied to all continental environment. To do that, we think that more observation data should be accumulated. Revised figure and the relevant statements

will be included in the revised manuscript.



Specific comments

p. 19684, l. 12: Are these the minimum/maximum values or ranges of standard deviation?

They are minimum/maximum values of the averages shown in Table 1. We do not think adding description in the manuscript is necessary because it will become obvious as we read the manuscript.

p. 19684, ,l. 19 (and throughout the text): Use 'CCN activation' instead of 'cloud activation' since (i) activation occurs for an individual particles (CCN) and (ii) cloud formation would include many additional processes that are not taken into account by CCN studies.

Suggested correction will be made in the revised manuscript.

p. 19685, l. 8-10: This statement is somewhat misleading. It is true that the aerosol indirect effect has been identified the largest uncertainty in understanding radiative forcing. However, the role of CCN is only one of many issues that is encompassed by the term 'aerosol/cloud interactions'. So, your sentence should be reworded.

The statement will be re-worded in the revised manuscript as "…realization that the aerosol indirect effects that are initiated by the anthropogenic emission of CCN are imposing the greatest…"

p. 19685, l. 25: Vapor is continuously condensing on particles; however, in equilibrium it also evaporates at the same rate. To be correct, you should say that above Sc, the former rate is much greater than the latter leading to continuous and efficient growth.

The sentence will be modified in the revised manuscript to "… vapor collapses and the vapor condensation rate exceeds the evaporation rate, leading to continuous growth of the particle."

p. 19686, l. 3: I don't think that Sc has ever been measured in clouds. The typical supersaturation in clouds is < 1% with significant temporal and spatial variability. Thus, measuring it in 'real' clouds seems impossible.

Here we are referring to the S_c measurement of size-selected ambient particles. To make it clear, the sentence will be modified in the revised manuscript to "…. available for the S_c measurement of size-selected ambient particles…."

p. 19690, l. 19: Be more specific here and replace 'some important statistics of the aerosol physical properties' by something like 'Average number concentrations and standard deviations of the aerosol populations. . .'

Suggested change will be made in the revised manuscript.

p. 19691, l. 15: A brief summary of the method by Swietlicki would be helpful.

Added in the revised manuscript will be "In short, the method assumes that the relationship between GF and RH for ambient particle is similar to that of ammonium sulphate particle of the same wet diameter."

p. 19692, l. 7: Can you really say based on a single GF that particles were internally mixed? Without any further composition information, I think, the only statement you can make is that they had very similar hygroscopicity.

This sentence will be rephrased in the revised manuscript to "….aerosols were mostly internally mixed or at least they had very similar hygroscopicity."

p. 19692, l. 4: what does 'which is located at the dominant upwind region' refer to?

We presume that you meant l. 29. It refers to Beijing. To make it clear, in the revised manuscript the sentence will be modified to "… dominant upwind region of the measurement sites."

p. 19693, l. 7: Why could the curve not be constructed?

It turned out that the measurement at S as high as 1.5% or even 2.0% should be available to construct the sigmoid curve for determination of S_c for 50 nm particles. But during Gosan 2008, 0.8% was the highest S. We did increase the upper S limit during BCMO 2009. In the revised manuscript, the sentence will be modified to "….. could not be constructed due to limited S range."

p. 19693, l. 24/25: Be more specific: What is it in the studies by Mocida that supports your results?

The result that aerosols from Chinese sources tend to have lower S_c compared to those from Korean/Japanese sources. We will state it more clearly in the revised manuscript as follows:
"From a field study during spring 2008, Mochida et al. (2010) reported that aerosols reaching Cape Hedo from China and the Pacific had higher GF and lower D_{p_act}, compared to those from Korea and Japan, which may be supportive to our suggestion that aerosols from Chinese sources tend to have lower S_c compared to those from Korean/Japanese sources."

p. 19694, l. 13-16: I don't understand why the Ddry-scan and the S-scan should give different results in terms of CCN activity as both are based Kohler equation. Are you saying that the S-scan is often done for the whole aerosol population and thus is biased if hygroscopicity varies with size?

Yes. Since D_{dry}-scan method is based on the N_{CCN}/N_{CN} ratio measured for various sizes for a given S value, it has to assume that the hygroscopicity is the same for all sizes. On the other hand, S-scan method uses the N_{CCN}/N_{CN} ratio measured for a single size and does not require such assumption.

p. 19695, l. 15/16: How relevant is S > 0.2% for atmospheric clouds? Stratocumulus clouds, the most abundant cloud type, might have S smaller than that. It has been shown that CCN predictions for much lower S is associated with greater uncertainties (e.g., Kammermann et al., 2010 and references therein). Some words on these issues should be added.

Yum et al. (1998) and Yum and Hudson (2002) estimated the effective supersaturation (S_{eff}) in clouds by comparing the measured cumulative CCN spectrum below cloud base and the measured cloud droplet

concentration in clouds and finding the S at which the cumulative CCN concentration matched with the cloud droplet concentration. Estimated S_{eff} amounted to be 0.2% or higher for stratocumulus clouds, depending on what cloud droplet concentrations (average or maximum) were used for comparison. S_{eff} was higher for continental cumulus clouds (0.34%). Therefore CCN measurement up to 1% S may cover most of the S ranges that can occur in real clouds, although recent work by Hudson et al. (2010) suggested that S_{eff} could be even greater than 1% for very clean stratus clouds. This will be stated in the revised manuscript.

About the uncertainty of CCN prediction for lower S, we have quiet different view. Kammermann et al.'s work was about predicting N_{CCN} based on aerosol size distribution measurement. Such predictions are sensitive to selecting critical diameter, as Kammermann et al. themselves pointed out. However, in this study we are not predicting N_{CCN} but just describing what we have measured, using the power law curve fitting. So we do not think parameters for lower S are subjected to larger uncertainties compared to the ones for higher S.

p. 19696, l. 15 ff: This is not conclusive: On the one hand you say that the solubility of organics cannot fully explain the differences in kappa derived from HTDMA and CCN measurements (l. 17/18); on the other hand you are saying the high organic fraction of the Gosan and BCMO aerosol could have caused this effect.

Currently we cannot fully explain the difference between HTDMA and DMA-CCN measurements. What we are suggesting here is that the high organic fraction may have partially contributed to the difference. To make it clear, the relevant sentence is rephrased to "…. BCMO (Kim et al., 2009b), which may have partially contributed to causing such effect."

p. 19607, l. 22: Is this sentence basically just repeating that you assume an internal mixture?

Yes.

p. 19697, l. 25 ff: (i) You show in Table 3 that kappa derived from CCN and HTDMA differ quite a bit but nonetheless you use the HTDMA-derived value for your CCN closure (due to the lack of data from CCN-derived kappas) and add a fair amount of discussion on the importance of differences in kappa for CCN closure. This seems to be some-what inconsistent and the uncertainties as introduced by using a 'wrong' (too small) kappa ~~and~~ should be discussed in light of the resulting differences in CCN closure for the different methods assumed here.

We must admit that the disagreement between DMA-CCN and HTDMA and the closure results from DMA-CCN being worse than those from HTDMA reveal our weakness on modeling aerosol hygroscopicities. However, we believe addressing the importance of kappa is a separate issue. Although

we have no sufficient explanation on the difference between the kappa values from HTDMA and DMA-CCN, predicted N_{CCN} shows that kappa from HTDMA is better. Considering that the ultimate goal of calculating kappa was to predict N_{CCN} in the first place, we do not think that it is wrong to use HTDMA-derived values as more reliable results.

Throughout this study, we have repeatedly emphasized the importance of knowing kappa for small particles, say, smaller than 100 nm. We believe the statements and the revised figure in the respond to the 2nd general comment, which will be included in the revised manuscript, will illustrate the uncertainty arising from using a wrong (I presume you were trying to say 'too high' instead of 'too small'?) kappa value.

(ii) I got confused about the number of the different methods. I count five different ones (1-GF, 2-GF, 2-Sc, 3-GF, and '4') whereas 3-GF was only applicable to part of the data. In order to avoid the confusion, I suggest renaming Method 4 to something like 'Method constant-kappa'. In addition, I suggest adding a table (or information to Table 5) summarizing briefly the characteristics of each method.

As shown below, Table 5 in the revised manuscript will add the description of each method and Method 4 is renamed as AR08. Also noted in the revised table is whether underprediction, overprediction or balanced was dominant in each closure result.

Table 5. Average and standard deviation of relative deviation, defined as $|N_{CCN\_pred} - N_{CCN\_meas}|/N_{CCN\_meas}$, for different CCN closure methods using the GF and S_c data. The values are given in the unit of percent and the word in the parentheses indicates whether the closure results are dominated by underprediction (under-), overprediction (over-) or balanced.

Method	Description	0.2% S	0.6% S	1.0% S
1-GF	Time varying and size segregated κ(GF) are used	28±20 (under-)	25±52 (balanced)	19±15 (balanced)
2.GF (small)	Time varying average κ(GF) for 50 and 100 nm are used for all sizes.	32±17 (under-)	25±51 (balanced)	19±14 (balanced)
2-GF (large)	Time varying average κ(GF) for 200 and 250 nm are used for all sizes.	25±24 (under-)	31±57 (over-)	22±17 (balanced)
3-GF	Size segregated but temporally averaged κ(GF) are used for all time.	28±38 (under-)	26±39 (over-)	23±29 (balanced)
2-S_c	Time varying κ(S_c) for 100 nm is used for all sizes.	38±42 (over-)	42±57 (over-)	30±37 (over-)

3-S_c	Temporally averaged $\kappa(S_c)$ for 100 nm is used for all sizes and time.	50±68 (over-)	47±61 (over-)	34±42 (over-)
AR08	Fixed κ value of 0.3 is used for all sizes and time.	30±51 (over-)	41±56 (over-)	28±35 (over-)

p. 19700, l. 6: Are there any data sets that suggest that the hygroscopicity varies over seasons and years? Table 5: Add information of different Methods here or refer to (an additional) table or to the respective text section.

We do not have such dataset yet. However, there is a clear seasonal change of air masses in East Asia due to monsoon and therefore aerosol hygroscopicity may differ from season to season, which is a subject of future work.

Reference

Hudson, J. G., Noble, S., and Jha, V.: Stratus cloud supersaturations, Geophys. Res. Lett., 37, L21813, 2010.

Yum, S. S., Hudson, J. G., and Xie, Y.: Comparisons of cloud microphysics with cloud condensation nuclei spectra over the summertime Southern Ocean, J. Geophys. Res., 103, D13, 16625-16636, 1998.

Yum, S. S., and Hudson, J. G.: Maritime/continental microphysical contrasts in stratus, Tellus, 54B, 61-73, 2002.