Response to the third referee

General comments:

The hygroscopicity closure is far from perfect, and looking at Fig. 9, it seems as the slope of the fit is actually much steeper than what is indicated when forcing the linear fit through the origin of coordinates. Why is the GF-kappa so much more constant than the CCNC kappa? This makes me very uncertain of the quality of the data. How was the CCNC and the H-TDMA calibrated, and how often was it calibrated during the campaigns? Without this information it is hard to draw too much conclusions.

(i) H-TDMA: RH sensors were calibrated by local distributor as written in the manuscript. DMAs were calibrated using PSL or by monodisperse aerosols generated by DMA before each campaign. CPC (TSI CPC 3010) counts were compared to a separate CPC of the identical model.

(ii) DRI CCNS: During Gosan 2006, DRI CCNS was used and was calibrated every day.

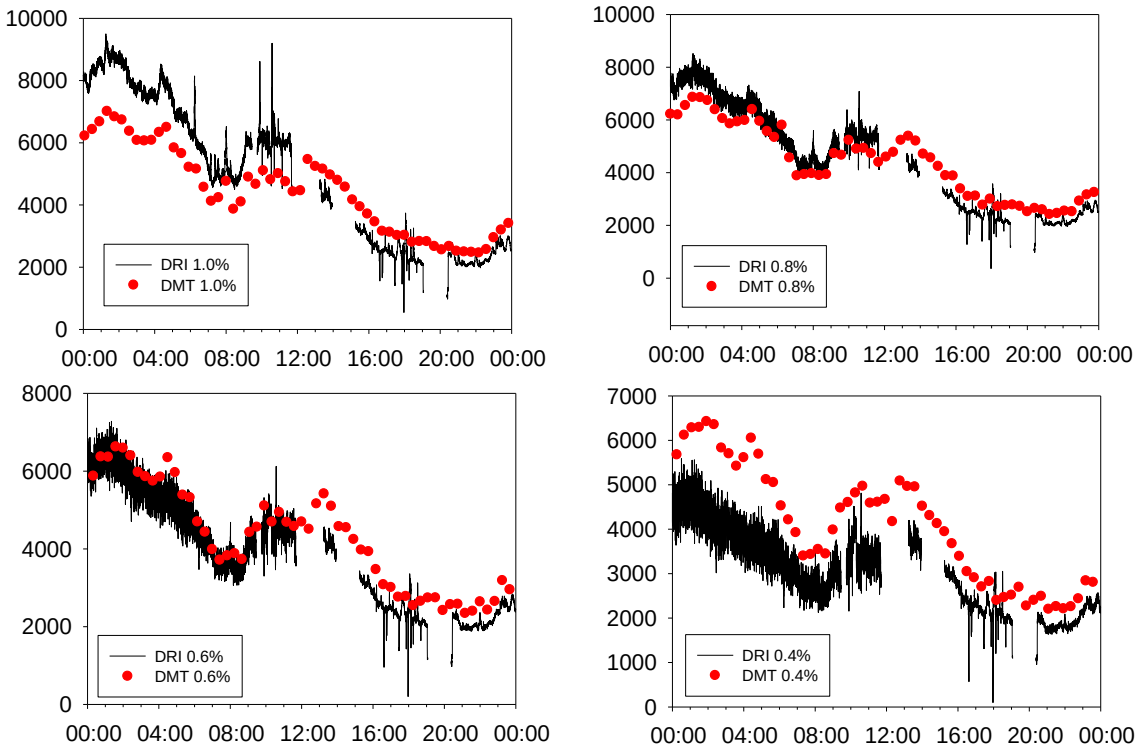
(iii) DMT CCNC: ①Supersaturation calibration: The supersaturation calibration maps temperature gradient to supersaturation field. Due to the atmosphere pressure difference between DMT located in Boulder, Colorado (840mb) and Gosan (1000mb), we performed calibration with monodisperse NaCl particles in November, 2006 and obtained supersaturation calibration curve. We applied the curve for Gosan 2007 and Gosan 2008. Before BCMO 2009, we did another experiment with monodisperse NaCl particles at the lab and found a little calibration drift. What had been 0.2, 0.4, 0.6, 0.8 and 1.0% before was 0.19, 0.42, 0.66, 0.89 and 1.13%, respectively. Note that the differences were very small for most of the measured S_c range between 0.2 and 0.4%.

②Flow calibration: The flow calibration maps voltage measured by flow sensors to actual volumetric flow and is not affected by pressure difference and therefore calibration result from the factory at Boulder can be used without further calibration at the Gosan site. The instrument was calibrated at DMT in August, 2006 and such calibration was used for Gosan 2007. The flow was recalibrated at the site on the very last day of Gosan 2008 and the result was applied to all data in Gosan 2008. Since the difference in flow could affect the supersaturation field in the instrument, we calculated supersaturation error arising from such flow difference according to Lance et al. (2006) and found that it would be smaller than 0.03% in supersaturation value for 0.85% S for thermal resistance value between 3.5 and 5.0 K W^{-1}.

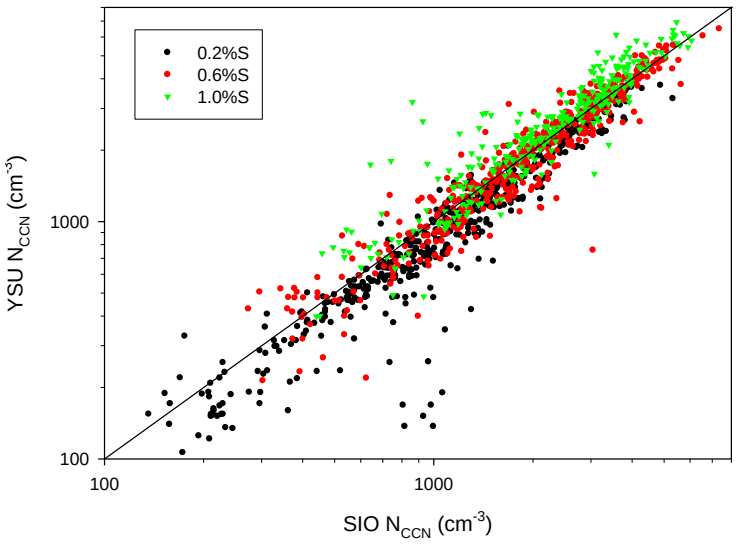
③OPC calibration: Since the droplet size information is only of second importance, we did not calibrate OPC measured size. However, we did compare CCN concentration for monodisperse NaCl particles with the CPC counts at high supersaturation when we were doing supersaturation calibration and found a good agreement.

④Comparison of the two CCN instruments: Although the N_{CCN} data from DMT CCNC was not used for Gosan 2006, we did make a comparison of the CCN concentrations simultaneously measured by DMT

CCNC and DRI CCNS for a day and the result is shown in the figure below.

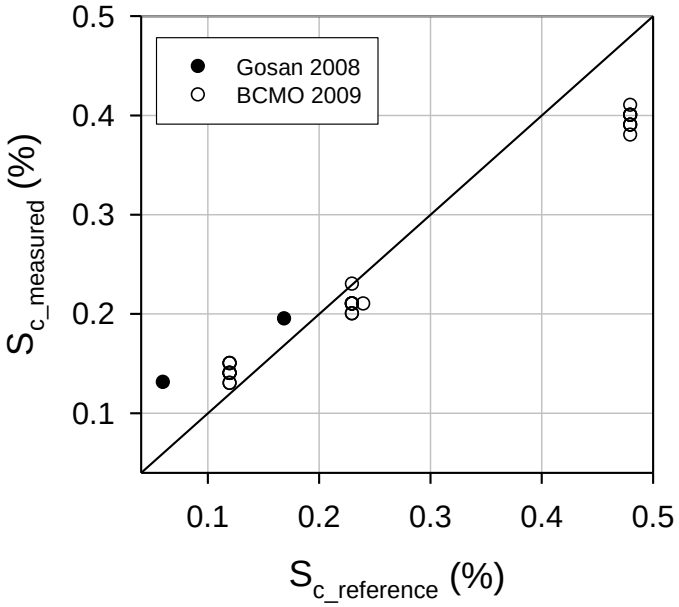


During Gosan 2008, another DMT CCNC unit was deployed by a research group from SCRIPSS Institution of Oceanography (SIO) and shared the same inlet. The figure below shows the comparison between the two instruments. On the y axis label 'YSU' indicates our instrument from Yonsei University.



(iv) Critical Supersaturation (S_c) measurement: To confirm the validity of our S_c measurement method,

we measured the S_c of ammonium sulphate and sodium chloride particles of known sizes during Gosan 2008 and BCMO 2009, respectively, and compared with the theoretical values. For the calculation of the theoretical values, Van't Hoff factor of 2.2 was used for ammonium sulphate, the shape factor of 1.08 was used for sodium chloride, and the Kohler model denoted as AA1 in Rose et al. (2008) was used. For such experiment during Gosan 2008, N_{CCN} was higher than 10000 cm^{-3} so we applied vapor depletion correction according to Lathem et al. (2011). The result is shown in the figure below.



After examining all these results, the disagreement between HTDMA and DMA-CCN results cannot be attributed unduly to measurement uncertainty. Hameri et al. (2001) also found large disagreement in the soluble fractions estimated from HTDMA and DMA-CCN measurements at boreal forest. In their case, the soluble fractions from DMA-CCN showed much less variability than those from HTDMA. There are series of papers that solely discuss the disagreement between HTDMA and DMA-CCN (Petters et al., 2009; Wex et al., 2009; Poulain et al., 2010), which implies that such disagreement can arise even when the quality of the measurement is assured.

On Page 19694, row 29 you state that although the two DMA-CCN measurements were based on different concepts the difference between the datasets is too large to be attributed to methodological differences. This is quite important, and I think you leave this question too fast. I would like to see an error propagation calculation, preferably based on calibration measurements, but at least on reasonable assumptions. What is the expected uncertainty in your derived kappa values? Once you have this, you can speculate more freely on possible reasons for the difference.

First, we want to note that the difference between the two DMA-CCN settings is a separate issue from the difference between DMA-CCN and HTDMA since we only used a single DMA-CCN setting ('S scan').

As can be seen in the figure above, the measured S_c values agreed with the reference values within ±0.03% in S for measured S_c range between 0.13 and 0.23, which covers all but one average S_c values shown in Table 3 (0.33% for 81 nm, KP). For 0.33% (81 nm, KP), The deviation for S_c ~ 0.49% is about -0.08% in the figure above and therefore it would be much smaller for S_c of 0.33% (81 nm, KP). Because propagation of S_c uncertainty to kappa uncertainty depends on several factors such as S_c value, D_{dry} and temperature, it is difficult to provide a range of kappa uncertainty. In order to give you some pictures of how uncertainties in S_c may propagate to kappa uncertainty, we applied S_c uncertainty of ±0.03% to the average S_c values in Table 3 and calculated the range of kappa for the given S_c uncertainty as shown in the table below.

Campaign	Average S_c (%, from Table 3)	Corresponding kappa (range of kappa for S_c uncertainty ±0.03%)
Gosan 2008	0.19 (100 nm)	0.36 (0.27~0.51)
BCMO 2009	0.33 (81 nm, KP)	0.22 (0.19~0.27)
	0.22 (110 nm, KP)	0.20 (0.16~0.27)
	0.22 (81 nm, nC)	0.51 (0.39~0.68)
	0.15 (110 nm, nC)	0.44 (0.30~0.68)

Note that the last column is different from the fourth column in Table 3. This is because the average kappa shown in Table 3 is the average of individual kappa values and not the corresponding kappa values for the average S_c values.

In chapter 4.2 you state that some of the difference in kappa can be explained by the organic fraction and that you don't see the full hygroscopcity in the H-TDMA. But if you would use kappa values from the CCNC to predict the CCN concentration, wouldn't you over predict the CCN concentration quite a lot? (even more..?) This somehow undermines the argument of organics being the reason for the discreprency in kappa between CCN and H-TDMA, I think. Can you comment on this?

We didn't mean to claim that all of the discrepancies were due to organics. Yet, it is still true that CCN closure results based on S_c are worse than those based on GF and we do not have sufficient explanation for that at this point. We will mention it more clearly in the revised manuscript by adding the following sentences at the end of chapter 4.2:
"On the other hand, as discussed later, N_{CCN} prediction based on κ (S_c) resulted in significantly larger overprediction compared to the prediction based on κ (GF). Such result implies that sparingly soluble organics alone cannot explain the difference between the two because if that were the case, κ (S_c) should result in better prediction than κ (GF)."

Specific comments:
Page 19685, row 10: "climate change prediction". It is actually uncertainty in radiative forcing.

The sentence will be rewritten in the revised manuscript as follows:
"The necessity to gain sufficient understanding of cloud condensation nuclei (CCN) has been increasing within the scientific community due to the realization that the aerosol indirect effects that are initiated by the anthropogenic emission of CCN are imposing the greatest uncertainty in radiative forcing required for climate change prediction."

Page 19685, row 21: "was suggested as a tool". Suggested by whom?

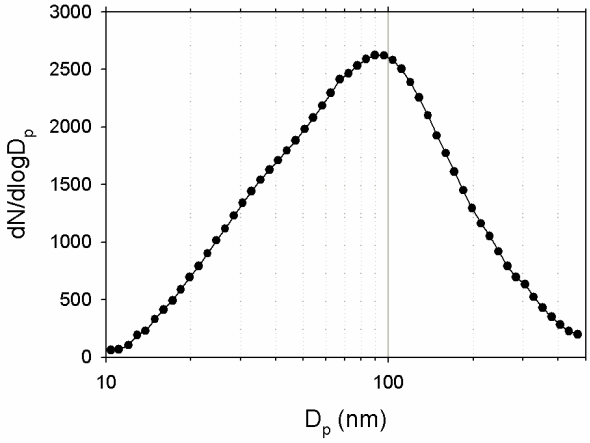
In the revised manuscript, the following references will be added at the end of the sentence:
(e.g. Brechtel and Kreidenweis, 2000; Kreidenweis et al., 2005)

Page 19685, row 26: "Provides a theoretical link..". This is too vague. Explain briefly what is included in the kappa approximation and what assumptions are made (it is basically ideal raoults law + Kelvin effect with water properties)

In the revised manuscript the sentence will be chanced to:
Recent development of a single parameter κ that incorporated the Raoult's law and the Kelvin effect with the given value of surface tension of water made the quantitative comparison between hygroscopicity at sub-saturated condition and CCN activation more feasible.

Page 19687, row 13: "10-300 nm". Does this cover most of the size distribution? It seems like a very small size span. You also state that the sampled air was not dried, but I assume that there were driers in the DMA sheath air?

There was a small mistake in describing SMPS measurement setup. The SMPS covered 10-470 nm (will be corrected in the revised manuscript) and the correct size range was used when analyzing the data. Such size range was selected because they covered most of the number size distribution and we expected particles larger than the upper limit of SMPS to be of little importance, at least for number concentration. The figure below shows the average aerosol number size distribution measured by SMPS during Gosan 2008:



No separate driers were used in DMA sheath air in order to measure actual ambient size distributions.

Page 19687, row 18: "The two CCN instruments were calibrated..". How often were they calibrated, and how much did they drift between calibrations? Same question goes for the HTDMA data.

The calibration issue is discussed in the response to general comment 1.

Page 19691, row 16: "Converted GF90 values were then classified into four categories:" These GF boundaries are tied to a specific dry size. In fact they are based on soluble volume fractions, so for different dry sizes you have to correct for the Kelvin effect. Did you do this? This also has to be mentioned in the text.

Yes, the Kelvin effect correction was applied as a part of the method illustrated in Swietlicki et al. (1999). We will mention it in the revised manuscript.

Page 19692, row 27: "in the range of 1.4 to 1.7". At which dry size? GFs must always be connected to a dry size.

For all sizes, as shown in Table 2. We will add "for all sizes" after the clause in the revised manuscript.

Page 19695, row 4: "When the results from Wiedensohler et al. (2009) is...". Should be "If the results from Wiedensohler et al. (2009) are. . .".

Ok.

Page 19695, row 15: "linear behavior". This is exponential and not linear.

Yes, this is exponential and we will correct the sentence in the revised manuscript.

Page 19696, row 19: "that significant" should be "that a significant". However, I am not sure how that it is possible to explain the difference just by strange behavior of the organics. I would like to see a test calculation, based on some "extreme" but still realistic assumptions regarding the particle properties (basically it is solubility and surface tension effects that comes to mind).

This issue is discussed in the response to general comment 1.

Page 19697, row 9: "measured Nccn are compared" should be "measured Nccn were compared". Stick to one tense (in this case past tense).

Ok.

Page 19697, row 11: "data, CCN" should be "data, the CCN".

Ok.

Page 19697, row 12: "because size" should be "because the size".

Ok.

Page 19697, row 12: "All aerosols were assumed to be internally mixed". How do you define internally mixed? Describe in the methods your way of evaluating the H-TDMA data. There are a number of different ways to do this.

Multi-mode lognormal Gaussian curves were fitted to the H-TDMA data. When only a single mode was found all of the sample were considered to have very similar GF and classified as internally mixed.

Page 19699, row 8: "the temporal variation of the size distribution was taken into account as in "Method 3"". Isn't the temporal variation of the size distribution taken into account in all approaches?

Yes. What we were trying to say is that no other temporal variation was taken into account, just as Method 3. To avoid confusion "as is Method 3" will be deleted in the revised manuscript.

Page 19699, row 12: Delete "(not shown)".

Ok.

Page 19699, row 18. Delete "goodness". "accuracy" is a better word.

Ok.

Page 19700, row 24. "Kammermann et al. (2010) have found that" should be "Kammermann et al. (2010) also found that"

Ok.

Page 19702, row 10: "size-resolving" should be "size-resolved".

Ok.

Page 19702, row 10. I think successfully predicted is quite a strong statement. An error of 28% is neither great nor terrible, at least if you compare to old closure studies (see e.g. Kammerman et al. 2010).

In the revised manuscript the sentence will be written with the deletion of the word 'successfully' as follows:
The temporally varying and size-resolved HTDMA hygroscopicity data predicted N_{CCN} with the average relative deviations of 28±20%, 25±52% and 19±15% for 0.2, 0.6 and 1.0% S, respectively.

Page 19702, row 16. You should state if it is an under-prediction or an over-prediction of the CCN concentrations. This must also be clear in table 5, as well as in all other places of the manuscript (e.g. the abstract)

Such information will be added to the revised Table 5 (shown at the end of this document), abstract and conclusion.

Page 19702, row 20. "as global" should be "as a global".

Ok.

Figure 4. Explain what the error bars represent.

The error bars represent standard deviations of each measurement. We will explain it in the revised figure caption.

Figure 5. Explain what the error bars represent

The error bars represent standard deviations of N_{CCN}. We will explain it in the revised figure caption.

Figure 6. This figure can be deleted. Time series are very hard to interpret, and should only be used if there is a special feature that should be highlighted, and I do not see what that is in this case.

We have a different view and think that the figure is worth presenting. A time series can provide much more information than one or two statistical values such as average and standard deviation. This figure shows how aerosol physical properties varied temporally and how pollution affected such properties.

Figure 7. The staples do not seem to add up to 1, which they should, why is this?

The staples do add up to 1 when relative frequencies for all hygroscopicity classes are added for a given dry diameter.

Figure 8. can be deleted on the same note as Figure 6.

Again we have a different view on time series. We think that the discussion on Figure 8 (p19693, line 5-9) is important.

Figure 10. Explain what the error bars represent.

The error bars represent standard deviations of κ(GF). We will explain it in the revised figure caption.

Figure 11. Explain the box-plot, what limits are assigned to the boxes and what are considered outliers?

The upper and lower ends of the box represent 75 to 25 percentile, respectively, and the horizontal bar within the box indicates the median value. The upper and lower whiskers outside the box represent 90 and 10 percentile, respectively. The data outside 10~90 percentile range are considered as outliers. This will be explained in the revised figure caption.

Figure 13. The x-axis' are CCN concentrations from the CCN counter I presume (not modeled values)? Please clarify this in the caption.

Yes. Throughout the manuscript 'NCCN' without any attachment of subscript represent measured CCN concentration. However, to avoid confusion, the x-axis label will be modified to 'N_{CCN_meas}.' In the revised figure.

I would like to see a table which explains what CCN models include what assumptions. This would be very helpful as a compliment to the text when looking at the figures and trying to interpret them.

Table 5 will be revised as shown at the end of this document.

References:

Brechtel, F. J., and Kreidenweis, S. M.: Predicting particle critical supersaturation from hygroscopic growth measurements in the humidified TDMA. Part I: Theory and sensitivity Studies, J. Atmos. Sci., 57, 1854-1871, 2000.

Hämeri, K., Väkevä, M., Aalto, P. P., Kulmala, M., Swietlicki, E., Zhou, J., Seidl, W., Becker, E., and O'Dowd, C. D.: Hygroscopic and CCN properties of aerosol particles in boreal forests, Tellus, 53B, 359-379, 2001.

Kammermann, L., Gysel, M., Weingartner, E., Herich, H., Cziczo, J., Holst, T., Svenningsson, B., Arneth, A., Baltensperger, U., Subarctic atmospheric aerosol composition: 3. Measured and modeled properties of cloud condensation nuclei, Journal of Geophysical Research, vol 115, D04202, doi:10.1029/2009JD012447, 2010.

Kreidenweis, S. M., Koehler, K. A., DeMott, P. J., Prenni, A. J., Carrico, C. M., and Ervens, B.: Water activity and activation diameters from hygroscopicity data - Part I: Theory and application to inorganic salts, Atmos. Chem. Phys., 5, 1357–1370, 2005.

Lance, S., Medina, J., Smith, J. N., and Nenes, A.: Mapping the operation of the DMT continuous flow CCN counter, Aeros. Sci. Techno., 40, 242-254, 2006.

Lathem, T. L., and Nenes, A.: Water vapor depletion in the DMT continuous-flow CCN chamber: Effects on supersaturation and droplet growth, Aeros. Sci. Technol., 45, 604-6015, 2011.

Petters, M. D., Wex, H., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and CCN activation for secondary organic aerosols – Part 2: Theoretical approaches, Atmos. Chem. Phys., 9, 3999-4009, 2009.

Poulain, L., Wu, Z., Petters, M. D., Wex, H., Hallbauer, E., Wehner, B., Massling, A., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and CCN activation for secondary organic aerosols – Part 3: Influence of the chemical composition on the hygroscopic properties and volatile fractions of aerosols, Atmos. Chem. Phys., 10, 3775-3785, 2010.

Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and CCN activation for secondary organic aerosols: Part 1 – Evidence from measurements, Atmos. Chem. Phys., 9, 3987-3997, 2009.

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 units 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 κ(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-)