Kim et al. present a comprehensive data set of hygroscopicity and CCN activity measurements which were performed during several field campaigns on two islands of Korea in summer 200 2009. Parallel measurements of the total aerosol mass concentration and concentrations o several gaseous species indicated that the two islands were under anthropogenic influence from the Asian continent. These data sets allowed classification into pollution and non-pollution days though no significant differences between those days could be observed for the particle hygroscopicity.

In a closure study, the measured total CCN concentrations were compared with the value predicted by several different methods using the dry particle size distribution and the κ-Kohler model by Petters and Kreidenweis (2007), in which the hygroscopicity parameter κ is derive from either the hygroscopic growth factor or the critical supersaturation. Similar as other earlier studies, Kim et al. found that using size-resolved and temporally varying κ values provide better agreement between predictions and measurements than using a constant average κ o 0.3. They conclude that the hygroscopicity of particles smaller than 100 nm must be known for accurate prediction of the CCN concentration.

The current study yields another important data set of size-resolved hygroscopicity and CC activity measurements in Asia, which may help to understand how the emission anthropogenic aerosol particles change cloud microphysical and radiative properties. The results of this study confirm and complement the findings of earlier studies and provide important knowledge on how the CCN activity can be described and implemented in numerical models. The manuscript therefore represents a substantial contribution to scientific questions It is within the scope of ACP and I recommend its publication after the following questions comments and suggestions for correction/improvement have been addressed.

General comments

1) The closure study was performed only for the Gosan 2008 campaign since this was the only one with dry particle size distribution measurements. Your HTDMA results show increasing hygroscopicity with particle size (e.g., Tab. 2), but your CCN results show the opposite (e.g., Fig 11, and 8; larger κ for 50 nm than for 100 nm). Also, an earlier study by Kuwata et al. (2008) presented slightly increasing κ with increasing size and generally higher κ for the same location (Tab. 3). I am wondering if this discrepancy might be due to an artifact in the actual supersaturation of your CCN counter. As you write in Sect. 2.1, the CCN counter was calibrated with ammonium sulfate or sodium chloride using the Kohler model denoted as AA1 in Rose e al. (2008). As shown in Fig. 11 of that paper, however, the AA1 model gives significantly higher critical supersaturations for the same particle diameter and composition than the more precise (and recommended) Kohler model AP3. Moreover, the relative deviation between AA1 and AP is strongly increasing with decreasing critical supersaturation.

That means, when your CCN counter measures the CCN concentration at a low supersaturation (e.g., 0.2%, determined with AA1), the actual supersaturation according to model AP3 would be ~18% smaller (i.e., 0.16%). Assuming that from your CCN measurements you observe for 100 nm particles a critical supersaturation of 0.16% (AP3) instead of 0.2% (AA1), you would calculate according to Petters and Kreidenweis (2007), a κ value of 0.54 (AP3) instead of 0.34 (AA1) Similarly, at a higher supersaturation (e.g., 0.55%, determined with AA1), the actual supersaturation according to AP3 would be ~14% smaller (i.e., 0.47%). Assuming that you observe for 50 nm particles a critical supersaturation of 0.47% (AP3) instead of 0.55% (AA1), you would calculate a κ value of 0.5 (AP3) instead of 0.36 (AA1). So, only by using a different Kohler model, your results will change a lot. It would be good if you could discuss this issue upon revision of your manuscript and estimate the possible influence on your further results (CCN closure, etc.)

Before answering your comments, we must note you that we were not able to determine S_c higher than 0.61% during Gosan 2008 due to limited S range (≤ 0.8%), which was briefly explained in the manuscript (p19693 line 5-7). It turned out that about half of 50 nm samples were expected to have S_c higher than the limit and therefore were discarded, so it is possible that $\kappa(S_c)$ for 50 nm shown in Fig. 11 may be biased toward higher κ value. For the samples that produced S_c lower than 0.61% for both 50 and 100 nm, the κ difference between the two sizes decreased a little, although κ for 50 nm was still higher than that for 100 nm.

As you suggested, it may be interesting to see the effects of using different Kohler models. But this seems not as simple as you approached, since the calibration affects all S used during the measurement (5 S values used to obtain the calibration curve and 11 S values used to obtain every single S_c value) and not just the final S_c value. From the supplement data provided by Rose et al. (2008), we find that the difference between AA1 and AP3 increases nonlinearly with increasing S. Also the curve fitting procedure shown in Fig. 3 is nonlinear as well. One needs to carry out the full procedure in order to test the errors that might arise from using different Kohler models.

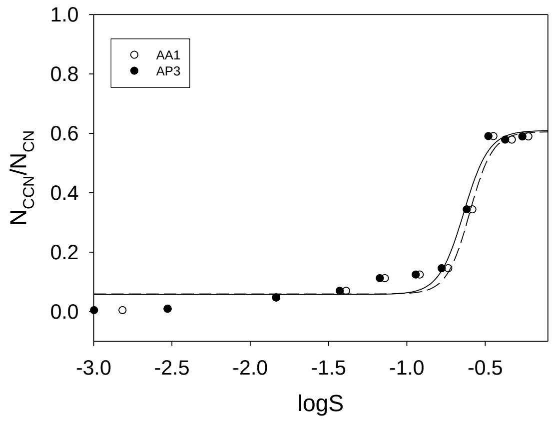
To apply AP3 model, we first obtained exponential regression between AA1 and AP3 from the supplement data for sodium chloride in Rose et al. (2008). The r^2 was higher than 0.9999. Then we applied such relationship to our original calibration curve (the curve that maps temperature gradients to supersaturations or vice versa) where the 5 S values were used and obtained a new calibration curve. Then we applied the new calibration to the 11 S values used for measuring S_c. We took three measurement samples for both 0.20% (100 nm) and 0.55% (50 nm) obtained by applying AA1 and compared them with the resulting S_c obtained by applying AP3 as shown in the table below.

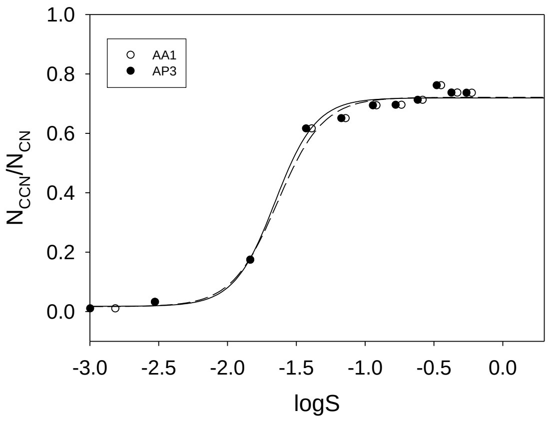
	100 nm (AA1→AP3)	50 nm (AA1→AP3)
Sample 1	0.1953 → 0.1903	0.5479 → 0.5253
Sample 2	0.1978 → 0.1953	0.5479 → 0.5253
Sample 3	0.2004 → 0.1953	0.5479 → 0.5313

In the table above, we extended the value to the fourth digit below the decimal point to show more detail although we think our instrument does not stand to such precision. The difference is much smaller (≤0.03% in S) than you would expect.

The reason for the much smaller difference may be due to the following. First, we used sodium chloride to calibrate CCN instrument. Ammonium sulphate was used for the calibration of the DRI CCN spectrometers during Gosan 2006, the procedure of which is completely different. We will state it more clearly in the revised manuscript. Second, we found that the curve fitting procedure moderates most of the differences between the Kohler models.

The data in the table above are derived from the figures below. The left figure is for 50 nm particle with 0.55% S_c and the right one is for 100 nm with 0.20% S_c. Since we do the curve fitting over log S space, the difference of S for different Kohler models is very small near the point where the sigmoid curve begins to rise. Furthermore the effect of using different Kohler models is reduced even more by the modulation of curve shape, which is more pronounced for 100 nm (right figure).





As discussed in the response to 3rd referee, we verified that our instrument reproduces S_c within ±0.03% for most of the S range in consideration. We think the uncertainties that may stem from using different Kohler models are within such limit.

2) For better comparison of your data set with other studies it would be helpful if you could give more general averages of the observed parameters (e.g., not only the average GF for the individual hygroscopicity categories, but also an overall average GF (weighted by the number fraction in each class); average S_c and κ for individual particle sizes during each campaign).

The revised Table 5 shown at the end of this document now contains such information. The average S_c and κ for individual particle sizes for each campaign are already shown in Table 3.

Specific comments

P 19684, L 2: better write “total particle concentration”

The first word ‘aerosol’ is supposed to be applied to “total concentration”, too. Moreover, the meaning of total concentration is explained in the parentheses next to the words. Therefore it would seem redundant to include ‘particle.’

P 19684, L 22: If you include “κ” after “Hygroscopicity parameters” one can expect already from the abstract that you used the terminology according to Petters and Kreidenweis (2007).

Ok.

P 19685 and elsewhere: Be more careful in the use of the word “aerosol”. Rather use “aerosol particle” or just “particle” when you explicitly talk of the particulate matter (e.g., L13: “particle within the droplet”, L 25: “vapor … condenses onto the particle”, etc.), since “aerosol” denote the suspension of particles in a gas.

Thank you for the clarification of the distinctive usage of the terms ‘aerosol’ and ‘particle.’ We will correct them in the revised manuscript following your suggestion.

P 19685, L 18: Asmi et al. (2011) gave a comprehensive overview of particle size distributions in Europe.

Thanks. This information will be added at the end of the sentence as “….(Kulmala et al., 2004) and the effort to systematically combine surface measurement sites is only at its earliest stage in the European region (Asmi et al., 2011).”

P 19686: Please consider adding a few recent publications in the introduction, e.g., L 4: Cerull et al. (2011), L 13: Rose et al. (2011), L 19: Gunthe et al. (2011).

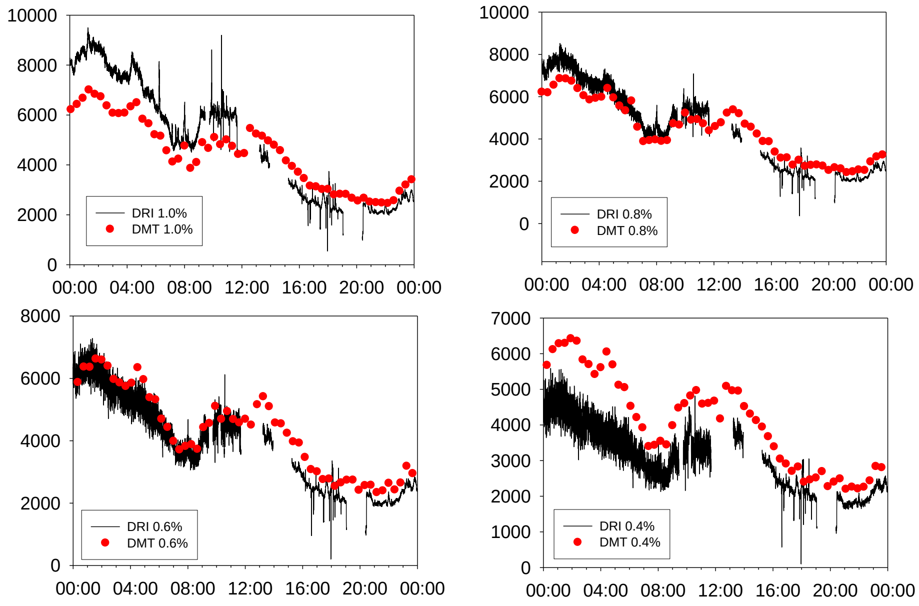
Rose et al. (2011) and Gunthe et al. (2011) scanned diameter instead of supersaturation and measured activation diameter (D_{p_act}) instead of critical supersaturation (S_c). In this sentence only the studies that measured S_c are introduced. We will introduce Cerull et al. (2011) in the revised manuscript.

P 19687, L11-14: Was the CPC integrated in the SMPS or operated separately? How was the total particle number concentration measured? By the CPC only (measuring the polydisperse concentration) or by integrating the size distribution measured by the SMPS? Please clarify.

Total particle number concentration was measured by a separate CPC in all four campaigns. We will clarify it in the revised manuscript.

P 19687, L 14-17: Can you please say something on the comparability of the two different CCN spectrometers. Did you perform any direct comparison of the 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. They agree reasonably well.



P 19688, L 6-9: Do you mean the total CCN concentration with “N_{CCN}” or the concentration of the monodisperse particles? How did you measure the total CCN concentration? Please describe the second path of the scheme in Fig. 2. Did you regularly switch between a polydisperse and monodisperse measurement? What kind of dryer did you use and what was the maximum relative humidity of the aerosol?

Throughout the manuscript N_{CCN} refers to the total CCN concentration. When the second (lower) path of the scheme in Fig. 2 was selected, the sample air bypassed the dryer, neutralizer and DMA, and was directly fed to CCNC. We regularly switched between polydisperse and monodisperse measurements. We measured sample loss rate for polydisperse measurement and applied them to obtain N_{CCN}.

We used diffusion driers with silica gel as desiccant during Gosan 2008 and nafion dryers during

BCMO 2009. We did not measure RH of sample air exiting the drier as a part of the DMA-CCN measurement but we were able to estimate their general performance from the RH data at the exit of the drier for HTDMA. For diffusion drier, RH was below 30% for most of the period and the maximum was still below 40% during Gosan 2006. During Gosan 2007 and Gosan 2008, RH was always maintained below 30%.

During BCMO 2009, nafion dryers were used for both HTDMA and DMA-CCN settings. Since all the nafion driers shared the same vacuum source for purged air, the same RH would be resulted provided that the sample flow is the same. The RH measured at the exit of nafion drier at HTDMA was always below 30%. The sample flow rate for DMA-CCN was slightly larger (1.1 lpm) compared to HTDMA (1.0 lpm) but such difference would result in negligible effect on RH.

This detailed information would be too much to be presented in the revised manuscript.

P 19688, L 9-11: For how long did you measure at each supersaturation? What was the settling time of the supersaturation (time between setting a new supersaturation level and starting measurement at this level)? How long did it take to finish one complete spectrum of supersaturations?

The DMA-CCN setting in Fig. 2 was set to maintain a constant S for 350 seconds for Gosan 2008 and 265 seconds for BCMO 2009. Those times are partitioned as follows:

Apply new S and wait until it stabilizes	150 s
Measure total CCN concentration	30 s
Change 3-way valve direction and apply 1st voltage (50 nm) and wait for DMA to stabilize	55 s
Measure monodisperse CCN concentration	30 s
Apply 2nd voltage (100 nm) and wait for DMA to stabilize	55 s
Measure monodisperse CCN concentration	30 s
Apply 3rd voltage (200 nm) and wait for DMA to stabilize (2008 only)	55 s
Measure monodisperse CCN concentration (2008 only)	30 s

After finishing the last S of the CCN measurement cycle, we gave extra amount of time (6 minutes) for S to stabilize because the S change between the last of the cycle (1.0% or 1.3%) and the first of the cycle (0.07%) was large and also because there was no chiller within the OPC and we had to wait until the temperature within the OPC dropped to an appropriate level by natural cooling.

During Gosan 2008, we measured 11 S and 3 sizes (50, 100 and 200 nm), although the data for 200 nm were not used in this study because their reliability was questionable due to too low S_c. In total it took 5145 seconds (1 hour and 25 minutes and 45 seconds) to obtain a complete cycle. During BCMO 2009, we also measured 11 S but only 2 sizes and it took 4560 seconds (76 minutes) for a complete cycle.

P 19688, L 20-21: The HTDMA setup is not shown in Fig. 2.

That is correct. Our intention was to write “The CCNC configuration in Fig. 2, TSI CPC 3010 and HTDMA were again used” and will be corrected as such in the revised manuscript. We used a typical HTDMA setting so we did not show it.

P 19689, L 21-23: Please change to “BCMO 2009 had the lowest PM2.5 … concentrations, but it SO2…“

Ok.

P 19695, L 11 ff: Do you have any reference for other studies using the same power law approximation? Please compare your results with Rose et al. (2010) who used a similar approach for a data set in south China.

Other studies using power law approximation: Pruppacher and Klett (1997) (p. 289) provides compilation of such works. More recent works include Hudson et al. (1998), Hudson and Yum (2001) and Yum and Hudson (2001).

When compared to Rose et al.'s (2010) Guangzhou measurement, their average N_{CCN} at 0.27% (6640±3975 cm^{-3}) was even higher than all of our average N_{CN} (not N_{CCN}!) values in this study. Detailed comparison is shown in the table below, which also includes Gunthe et al.'s (2011) measurement at Beijing, another mega city.

		Gosan 2006	Gosan 2007	Gosan 2008	BCMO 2009	Guangzhou[1]	Beijing[2]
N_{CN} (cm^{-3})		4697±1823	4217±1514	3890±1808	5117±1880	18150±7991	16510±9000
N_{CCN} (cm^{-3})	1.0% S	3290±1964	4074±1857	2713±1271	4694±2567	13890**	12011**
	0.6% S	2803±1545	3527±1718	2076±989	4360±2297*	10484**	8421**
	0.2% S	1550±659	1952±1286	1043±646	3051±1310	5724**	3923**

1 Guangzhou measurement from Rose et al. (2010)
2 Beijing measurement from Gunthe et al. (2011)
* N_{CCN} and N_{CCN}/N_{CN} for 0.59% S.
** Interpolated (for 1.0% and 0.6%) or extrapolated (for 0.2%) N_{CCN} value from measured N_{CCN} values except the lowest S which is below 0.2% by exponential curve. r^2 was higher than 0.99 and 0.96 for Guangzhou and Beijing, respectively.

However, it should be noted that the measurements of Rose et al. (2010) and Gunthe et al. (2011) were conducted at a site very close to the city boundary while our measurement was conducted at the two islands that are at least several hundred kilometers away from big cities in Korea. In short, the purpose of those works was to characterize CCN near the source while we are focusing on CCN found at rural islands. Therefore we think providing such comparison in the revised manuscript can be misleading.

P 19696, L 26: include "could" before "be used for"

Ok.

P 19696 L 25 - P 19697 L 2: A modeling study by Pringle et al. (2010) also suggested an average κ = 0.3 for aged continental aerosol.

We will add the following sentence at the end of the paragraph in the revised manuscript:

"A pioneering study to investigate global distribution of κ using global numerical simulation also reached similar value of κ, 0.27±0.21, for continental aerosol (Pringle et al., 2010)."

P 19697 L 3-5: I agree that in your study the particles are mainly of continental origin but since you measured on an island I would expect also marine influence, i.e. higher hygroscopicity than for aged continental aerosol (i.e., on average κ > 0.3). Can you comment on that? As already mentioned in my general comments, Kuwata et al. (2008) did observe higher κ for the same location. What is the distance (km) between Jeju Island and the mainland?

The time period when the 5 day HYSPLIT (Draxler et al., 2002) backtrajectories for 500, 1000 and 1500 m experienced no contact with land surfaces were classified as maritime.

Such periods were found only during Gosan 2006 and Gosan 2008: for Gosan 2006, 15:00, Aug. 21~03:00, Aug. 22 (Maritime 1) and 21:00, Aug. 26~09:00, Aug. 27 (Maritime 2); for Gosan 2008, 15:00, Aug. 10~12:00, Aug. 11 (Maritime 3), 21:00, Aug. 12~06:00, Aug. 14 (Maritime 4) and 21:00, Sep. 14~03:00, Sep. 15 (Maritime 5).

During such periods, N_{CN} and N_{CCN} are commonly lower than the half of the campaign average values. Even among the maritime air masses, there was clear difference between the trajectories that came from the South China Sea or the Philippine Sea (Maritime 1, 3 and 4) and from the North Pacific Ocean that lies east of Japan (Maritime 2 and 5). The N_{CN} and N_{CCN} were much higher for the former implying that the former air mass is still influenced by emissions from southern Asia even after 5 days.

The N_{CCN}/N_{CN}, $\kappa(GF)$ and $\kappa(S_c)$ averages for each maritime period are compared with the campaign averages in the two tables below. $\kappa(GF)$ was mostly larger than the campaign averages except for 50 nm during Maritime 1. $\kappa(S_c)$ was mostly larger than the campaign averages except for 50 nm during Maritime 3 and 5. N_{CCN}/N_{CN} during the maritime periods are not notably different from the campaign averages. This may be related to the fact that N_{CCN}/N_{CN} ratio depends also on the sizes of aerosol population, which differs for each maritime period as different D_g values in the table indicate.

Gosan 2006	Maritime 1	Maritime 2	Campaign avg.
N_{CN}	2096±994	1473±1225	4697±1823
N_{CCN} (0.2%)	806±201	607±203	1550±659
N_{CCN} (0.6%)	1112±316	887±333	2803±1545
N_{CCN} (1.0%)	1223±363	1033±520	3290±1964
N_{CCN}/N_{CN} (0.2%)	0.46±0.14	0.41±0.08	0.40±0.06
N_{CCN}/N_{CN} (0.6%)	0.62±0.16	0.61±0.11	0.67±0.12
N_{CCN}/N_{CN} (1.0%)	0.67±0.16	0.71±0.1	0.77±0.16
D_g	74±15	N/A	98±25
κ(GF, 50nm)	0.25±0.01	0.53±0.06	0.34±0.13
κ(GF, 100nm)	0.54±0.02	0.67±0.04	0.51±0.10
κ(GF, 150nm)	0.59±0.00	0.76±0.06	0.58±0.13
κ(GF, 200nm)	0.63±0.03	0.77±0.08	0.58±0.12

Gosan 2008	Maritime 3	Maritime 4	Maritime 5	Campaign avg.
N_{CN}	1364±1565	1439±617	470±123	3890±1808
N_{CCN} (0.2%)	218±89	582±184	117±36	1043±646
N_{CCN} (0.6%)	435±121	989±195	239±43	2076±989
N_{CCN} (1.0%)	654±167	1288±535	376±29	2713±1271
N_{CCN}/N_{CN} (0.2%)	0.18±0.11	0.42±0.15	0.25±0.05	0.29±0.15
N_{CCN}/N_{CN} (0.6%)	0.41±0.2	0.69±0.13	0.56±0.11	0.60±0.25
N_{CCN}/N_{CN} (1.0%)	0.77±0.24	0.87±0.17	0.93±0.16	0.83±0.31
D_g	63±13	112±23	57±7	81±24
κ(GF, 50nm)	N/A	N/A	0.21±0.01	0.18±0.04
κ(GF, 100nm)	N/A	N/A	0.21±0.01	0.16±0.03
κ(GF, 150nm)	N/A	N/A	0.33±0.00	0.21±0.04
κ(GF, 200nm)	N/A	N/A	0.30±0.00	0.22±0.05
$\kappa(S_c$, 50nm)	0.53±0.06	0.74±0.23	0.47±0.05	0.53±0.36
$\kappa(S_c$, 100nm)	0.54±0.18	0.47±0.16	0.45±0.05	0.40±0.07

The closest distance between Jeju Island and the Korean Peninsula is about 90 km and Gosan is about

120 km south of the southern end of the Korean Peninsula.

The above tables can be provided as supplemental data to the revised manuscript.

P 19702, L 2 and 10: write “size-resolved” instead of “size-resolving”

Ok.

Tab. 2: Is it possible to include a column with the overall average GF at each particle size?

The revised Table 2 shown at the end of this document now contains such information.

Tab. 3: Why do you list only CCN results for Gosan 2008 and BCMO although you write in Sect 2.1 that you did CCN measurements during all campaigns? For Gosan 2008 also the results for 50nm are missing.

As stated in section 2.1, during Gosan 2006 and Gosan 2007, we measured only total N_{CCN} and did not measure S_c. Therefore we listed only HTDMA results for these two campaigns in Table 3.

As mentioned in the response to the first general comment, we were only able to measure S_c lower than 0.61% which is equivalent to κ being higher than 0.28 for 50 nm. Only about half of the 50 nm samples satisfied such constraint. Therefore we thought that the result for 50 nm could be significantly biased toward low S_c and that it was not suitable to present the values as the campaign average.

Such issue was briefly explained in the manuscript (p19693 line 5-7).

Tab. 5: It might be good to mention here also whether the prediction over- or under-estimates the measurement (i.e., is there a positive or negative bias in the prediction) rather than giving only the relative deviation.

The revised Table 5 shown at the end of this document now contains such information.

Fig. 2: What was the setup like during the other Gosan campaigns?

During the other Gosan campaigns, we did not have DMA-CCN setting and only measured total CCN concentration.

Fig. 3: Please indicate that the supersaturation at h/2 is S_c.

Ok.

Fig. 4 and 5: What are the error bars?

The error bars in Fig. 4 represent the standard deviation of the measured values in each campaign. Likewise the error bars in Fig. 5 represent the standard deviation of the measured N_{CCN} at selected S values in each campaign. This will be explained in figure captions in the revised manuscript.

Fig. 4: Please change the caption to something like “Average concentrations of PM mass and of various gaseous species…”

Ok.

Fig. 9: What are the parameters of the regression lines and what is the correlation coefficient?

	r^2	slope	y-intercept
(a) 2008 Gosan, 50 nm	0.27	1.64	0.16
(b) 2008 Gosan, 100 nm	0.56	1.67	0.15
(c) 2009 BCMO, 110 nm	0.35	1.01	0.05

These values will be shown as a part of Fig. 9 in the revised manuscript.

Fig. 11 and 13: Are these plots showing the statistical distribution (median, percentiles, etc.) of κ? If so, please indicate which line of the box and the bar belong to which percentile.

Figures 11 and 13 are the schematic plots that show the statistical distribution of different parameters. Following the typical way of presenting a schematic plot, the horizontal bar within the box indicates the median value and the upper and lower ends of the box represent 75 and 25 percentiles, respectively. The upper and lower whiskers outside the box represent 90 and 10 percentiles, respectively. The data outside 10~90 percentile range are considered as outliers and each data point is marked with a symbol. This explanation will be added in the figure caption in the revised manuscript.

Fig. 13: Should the x-axis label be “N_{CCN_meas}”? Is “$_{\_meas}$” also missing in the caption? Why and how do you merge the data in individual bins? A simple scatter plot (rel. dev. vs. N_{CCN_meas}) would show the same I guess.

We tried to be consistent by denoting N_{CCN} as the actual measured CCN concentration, but it seems not clear enough. In the revised Fig. 13, the x-axis label will be changed to $N_{CCN\_\_meas}$.

We merged the data by log-uniform bins: 289-550 cm^{-3}, 550-1047 cm^{-3}, 1047-1991 cm^{-3}, 1991-3787 cm^{-3} and 3787-7203 cm^{-3}. We preferred bin because the number of data in different bins vary greatly and therefore a scatter plot did not effectively visualize the trend we wanted to emphasize. The number of data in each bin was 8, 35, 97, 86 and 10, respectively. To note is that there was a small mistake when treating the last two bins and the figure should be changed slightly, which negligibly affected the result. The corrected figure will be presented in the revised manuscript.

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Yum, S. S., Hudson, J. G.: Vertical distributions of cloud condensation nuclei spectra over the springtime Arctic Ocean, J. Geophys. Res., 106, D14, 15045-15052, 2001.

Revised Table 2. Average and standard deviation of GF90 and cumulative number fraction (NF) of each hygroscopicity class; class 1, 2, 3 and 4 corresponds to ‘nearly hydrophobic’, ‘less hygroscopic’, ‘more hygroscopic’ and ‘very hygroscopic’, respectively.

Period	D_{dry} (nm)	GF90_1	GF90_2	GF90_3	GF90_4	NF_1	NF_2	NF_3	NF_4	Number weighted GF90
Gosan 2006	50	1.00±0.06	1.40±0.11	1.59±0.03	-	0.29±0.31	0.61±0.32	0.10±0.19	-	1.30
	100	1.05±0.04	1.50±0.05	1.66±0.06	-	0.22±0.23	0.33±0.38	0.45±0.33	-	1.47
	150	1.04±0.03	1.52±0.02	1.71±0.09	1.87±0.03	0.14±0.22	0.22±0.37	0.59±0.40	0.05±0.16	1.58
	200	1.02±0.02	1.51±0.03	1.67±0.08	1.87±0.02	0.10±0.15	0.15±0.21	0.66±0.30	0.09±0.16	1.60
	250	-	-	-	-	-	-	-	-	-
Gosan 2007	50	-	-	-	-	-	-	-	-	-
	100	1.07±0.04	1.48±0.06	1.58±0.03	-	0.05±0.08	0.66±0.23	0.30±0.25	-	1.50
	150	1.05±0.03	1.48±0.05	1.57±0.01	-	0.05±0.11	0.70±0.26	0.25±0.24	-	1.48
	200	1.04±0.05	1.47±0.04	1.57±0.01	2.17*	0.03±0.10	0.67±0.25	0.30±0.25	0.04*	1.57
	250	1.08±0.04	1.48±0.05	1.58±0.02	-	0.03±0.06	0.66±0.25	0.32±0.23	-	1.51
Gosan 2008	50	1.07±0.04	1.32±0.07	-	-	0.22±0.25	0.78±0.25	-	-	1.27
	100	1.08±0.05	1.32±0.06	-	-	0.14±0.25	0.86±0.25	-	-	1.29
	150	1.06±0.04	1.41±0.06	1.57±0.01	-	0.09±0.19	0.87±0.22	0.05±0.14	-	1.40
	200	1.08±0.02	1.43±0.05	1.58±0.02	-	0.04±0.10	0.88±0.23	0.09±0.21	-	1.44
	250	1.06±0.02	1.44±0.04	1.57±0.01	-	0.03±0.10	0.85±0.25	0.12±0.24	-	1.44
BCMO 2009	53	1.07±0.03	1.47±0.08	1.65±0.04	-	0.04±0.08	0.24±0.31	0.72±0.31	-	1.58
	113	0.99±0.02	1.50±0.04	1.62±0.04	-	0.01±0.03	0.25±0.32	0.74±0.32	-	1.58
	163	1.06±0.02	1.42±0.16	1.74±0.06	1.87±0.03	0.01±0.04	0.04±0.11	0.87±0.20	0.09±0.19	1.75
	225	1.08±0.07	1.51±0.04	1.68±0.06	1.92±0.05	0.04±0.10	0.07±0.15	0.85±0.21	0.04±0.13	1.65

* Only a single measurement was available.

Revised 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-)