Zábori et al., Atmos. Chem. Phys. Discuss., 15, 5079–5128, 2015

Response to reviewer 2

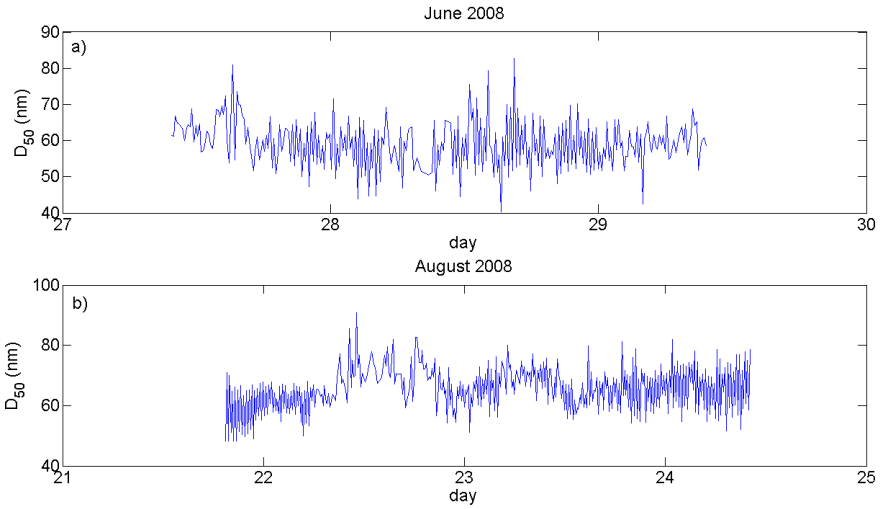
Thank you very much for taking the time to review our manuscript and pointing out areas of improvement. In the ingress the reviewer is asking if these results move Arctic CCN science forward. We believe that the reviewer actually agrees with us that the discrepancy, between κ values derived using different techniques, also highlighted by the reviewer, is indeed a significant insight. The novel results of size dependent Arctic CCN properties presented towards the end of the manuscript is important knowledge when attempting to generalize properties based on other much larger datasets. As pointed out by the reviewer the cases are not extremely extensive, and the role of material leading up to these cases are aimed at setting them into a context. This is done both in terms of climatology and the ambient weather situation for these two periods. Also important is the thorough presentation of previous CCN measurements conducted in a more traditional way of operating the CCNC instrument. The fact that we present two short cases in this study call for more background material than if we had a full annual cycle of our observations, for instance. Below follow a point-by-point response to the reviewers comment.

Specific comments:

1) It is good that our ambition to set the cases into a greater context is appreciated by the reviewer, we can agree that this material is extensive compared to the duration of the case periods. The fact that the study have this "case" design made it even more important for us to make the background description very thorough. With a much more extensive dataset, the need for this background description would have been less. As for other size resolved CCN measurements conducted at other locations worldwide, these measurements are explicitly addressing the size dependent properties which relates to the particle potentially being a cloud nuclei. The fact that measurements are conducted at other locations is not so helpful in understanding the Arctic environment. Since these cases represents the first look into size dependent CCN properties in the Arctic, any result would have been an advance in understanding. In our case the different kappa values that are derived based on different methods point us in one direction. If our results simply had corroborated previous results, the conclusion had been pointing us in a completely different direction. The results from this study shows clearly that it is absolutely imperative that future research include the size dependent particle characteristics in order to assess natural as well as anthropogenic influences on aerosol-cloud interactions. Having large datasets of bulk properties is clearly not sufficient to make generalized statements or implement derived relations based on these into models.

2) We do not agree with the reviewer that the 0.4%SS unrealistic as an upper range (e.g. Shaw; 1986). Practical considerations forced us to use a constant SS during these experiments. The choice fell on an upper level SS, but still in the mid-range of what is typically scanned by CCNC instrument. The much more extensive scanning data (several seasons) will be analyzed and presented elsewhere, but will have to be interpreted with the results from this study in mind.

3) The suggested analysis analogues to Su et al. (2010) and Padró et al. (2012) is not possible with our data as it requires scanning both D_p and SS to get a 2-dimensional data set. Because of the low particle number density in the Arctic we choose to fix the SS at 0.4% in order to get a balance between the time resolution and counting statistical uncertainty. Despite the trade-off from using a fixed SS, we still choose to average the results for the complete periods. As reference, we include here the time series of the estimated D_{50} for each scan. As expected the data presents large variability



4) This is a good idea to look at the temporal evolution. Unfortunately, as the variability suggests, we are struggling with counting statistical problems. Hence the case averages.

5) We agree with the reviewer and have completely redone the trajectory analysis. We also took inspiration from the suggested reference Yeung et al., 2014. We shortened the extent of the trajectories to 5 days, and we calculated one trajectory for each hour. We combined figures 2, 3, 4, and 5, and combined figures 7, 8, 9, and 10 but chose a slightly different layout than Yeung et al., 2014. We also chose to make each subplot a little larger for clarity and used color coding for the trajectory altitude.

6) The LIDAR data is used merely as a cloud detector and the level 1 relative backscatter provided on the MPL webpage is sufficient for this purpose. We do not need optical properties with physical values to characterize the local cloud situation near the station. The original figures included unnecessary information in the vertical and the figures have been adjusted to show the atmosphere up to 10 km altitude.

7) The means and SD are the geometric values, now changed in the text. The figure caption of Figure 14 was changed to:"Geometric means of size-resolved particle density measurements and resulting CCN concentrations for the measurement period in a) June and b) August 2008. Measurements were conducted at 0.4 % SS. Error bars indicate the geometric standard deviation.". The sentence "The presented arithmetic means and SD were calculated by assuming a logarithmic distribution of particle concentrations within different particle sizes to abate the influence of extreme concentrations on the average concentration."(p 5097, lines 19-21), was deleted. The sentence: "The upper panel shows the activated particle concentration measured by the CCNC compared to total particle (CN) concentration measured by the CPC for the measurement period during June 2008 (Fig.14a)" (p 5097, lines 15-17), was changed to:" The upper panel shows the geometric mean of the activated particle concentration measured by the CCNC compared to the geometric mean of the total particle (CN) concentration measured by the CPC for the measurement period during June 2008 (Fig.7a)". On p 5097 in line 26 "SD" was replaced by "geometric SD". The variability in figures 2 and 7 are real and to have brief periods of burst of particles in the summer is not unusual. Whether to plot the ordinate scale as logarithmic or linear is typically a matter of taste. For this dataset we have chosen linear scale for some panels and logarithmic scale for others.

8) Since there are no previous measurements in the Arctic of the kind presented in this study, we chose to compare the results with the "next best thing". Whether the Pallas station represents Arctic conditions is not the question we try to answer. Pallas is located in the Arctic if the

Arctic Circle is used as definition. It is not uncommon for e.g. remote sensing averages to include everything north of 60 degrees. Since it is about 1500 km between the Zeppelin and Pallas we do not need to spend much text on the fact that they are different, but indeed Pallas are at times taken to represent Arctic or at least sub-Arctic conditions (defining Arctic in the sense of Arctic climate). Hence, there is an interest in comparing the observations from these two sites.

9) Based on the model output from the Hysplit trajectory calculations, the integrated precipitation over the five day duration was calculated for each hourly trajectory. Over all there was little precipitation during the investigated periods with a median of less than 3.7 mm for the June case and less than 1.7 mm for the August case. The maximum integrated precipitation is an isolated event for a trajectory arriving 0600 on 27 June. For this trajectory the integrated precipitation was 18.5mm. From this we can conclude that precipitation was not a dominant factor in shaping the aerosol properties over the last five days of travel. This information was added to the manuscript text.

10) The Zeppelin station is located in such way that we can safely say that the enhancement of particles towards the end of the June period is not from local contamination. Also the enhanced particle concentration appears to be part of a much larger structure that can be seen starting towards the end of 28 June. The conditions for new particle formation and enhanced particle growth appear to be met only around midday 29 June. The trajectories suggest a systematic lowering in altitude during this period with enhanced particle concentrations. It is possible that this change in transport pattern resulted in more moisture supply to the air mass which helped promote particle formation and growth when the sun was at its highest. The information gained from the new trajectory plots was added to the manuscript.

11) We would like to avoid making this into a discussion about the usefulness about various length of trajectories and we are fully aware about the limitations of air mass trajectories. However, its usefulness depends much on what the intended use is. We know from previous experience that there is significant statistical information even for trajectories that extend over rather long periods of time. We have successfully used 10 day back trajectories together with aerosol data obtained from the Zeppelin station previously (Tunved et al., 2013), but then in a more statistical sense. Without giving it any deeper thought, we used 10 day trajectories again for this study. To harmonize with the reviewers comment we have now used 5 days back trajectories and we have calculated one trajectory for each hour of the two periods investigated.

12) The sentence on page 5093 lines 2-3 was for clarification replaced by the following sentences: “During the time period of 2:00 and 24:00 on the 21 of August, the Zeppelin research station was according to the Lidar measurements very likely unaffected by clouds. The trajectories of the 21 August show that air masses originate from the mid-latitudes and lower their height when reaching Zeppelin research station (Fig. 4). Therefore, it is likely that the peak in the particle number size distribution for particles with diameters between 100 nm and 200 nm is a result of particles being transported from the mid-latitudes to the Arctic and the processes taking place during transport rather than particles are being produced locally.” It is true that the station was not all the time unaffected by clouds, but this was the case when the particles characterized by a dry diameter of 100-200 nm peaked.

13) The κ of a substance is very much dependent on the method used to derive it. Koehler et al. (2006) refer to a κ of 0.72 determined by Köhler theory and a κ of 0.33 determined by HTDMA measurements for ammonium sulfate (Table 3). The assumption of a mean κ of 0.53 is presented in Petters & Kreidenweis (2007). However, as we assume that the “bulk κ” is biased due to the chemistry of particles >400 nm, the sentence: ”In any case, the results suggest that the aerosol resembles ammonium sulphate in its hygroscopic properties and CCN-activation”. (p5101, lines18-20), was deleted.

14) Fig. 12 and 13 was redone and combined in a new figure, which saves some space. Other figures were renamed accordingly and the text was adjusted to the new figure names. Each presented data point is the median value measured at a certain SS during one scan over the SS spectra. Since median concentrations for a given SS is essentially our primary variable here, the scatter among the data points is a direct visualization of the scan-to-scan variability in the data. Adding additional uncertainty bars in the figure would only obscure the figure. Therefore, the variability between different scans is shown by the presented medians per SS. The text in the paper is adapted according to the new calculated coefficients.

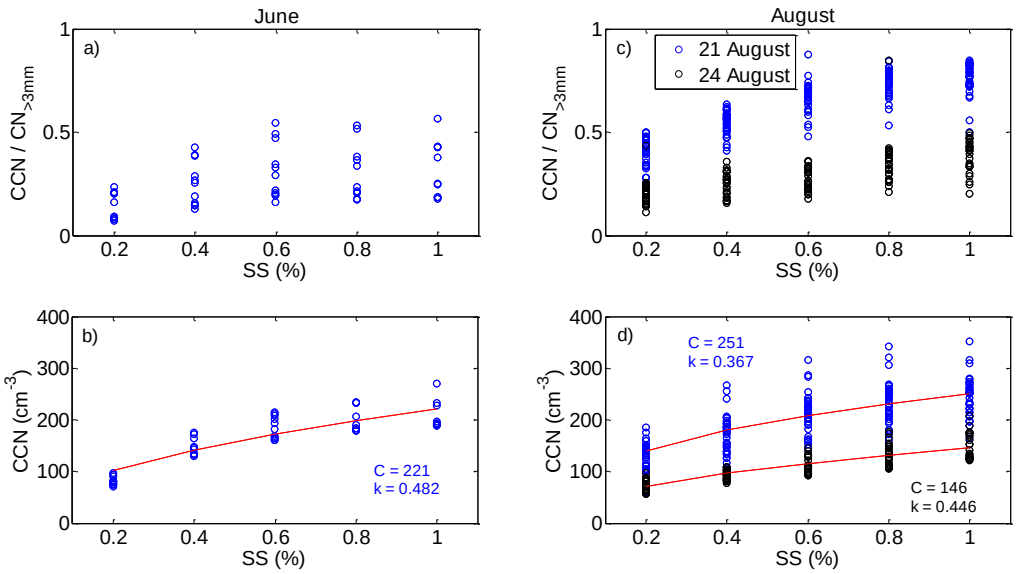


Figure 12. a) Ratios of the medians for each SS scan between CCN and particles with diameters > 3nm ($CN_{>3nm}$) for June 2008 as a function of SS. **b)** Medians for each SS scan of the total numbers of CCN as a function of SS for June 2008. **c)** Ratios of the medians for each SS scan between CCN and particles with diameters > 3nm ($CN_{>3nm}$) for 21 and 24 August 2008 as a function of SS. **d)** Medians for each SS scan of the total numbers of CCN as a function of SS for 21 and 24 August 2008. The red curves represent power-law function fits to the data with the coefficients *C* and *k*.

Minor comments:

Pg. 5081, Lines 3-4: What is meant by "active feedback mechanism"?

- *The sentence was corrected to: "[…] (i) very sensitive to changes in radiative forcing owing to a **direct** feedback mechanism […]."*

Pg. 5082, Line 7: I don't think the word "unfavourable" is right here. Sulfate and nitrate salts are highly water soluble and known to act as CCN. Sea salt (NaCl) is indeed more hygroscopic, but it's not clear to me that the difference in kappa between 0.6 and 0.8 is meaningful in the context of Arctic clouds.

- *That was the conclusion by Silvergren et al. (2014), which we summarised.*

Pg. 5082, Lines 8-10: What are the range of kappa values that are being cited here?

- *The sentence was changed to: "Both the growth factor and the values of the hygroscopicity parameter κ **ranging approx. between 0.7 and 1** were determined to be highest in October […]."*

Pg. 5082, Lines 16-17: Weird line break

- *The weird line break was deleted.*

Pg. 5082, Line 21: Do you mean "wet scavenging" instead of "cloud formation"?
- *The sentence was corrected to: "The authors suggested that this occurred as a result of wet scavenging."*

Pg. 5082, Lines 27-28 and throughout: Check your significant figures here. Two decimal places on a particle concentration is inappropriate!
- *The values were transcripts from the referenced studies, but we have changed the values to more appropriate numbers.*

Pg. 5085, Lines 23-25: If the results should be interpreted with caution due to the limited number of samples then why are these results being referenced here?
- *The fact that data is associated with a caution, does not mean that data is disqualified.*

Pg. 5085, Lines 6-7: What is meant by the term "clear proof"? Is this sentence motivating the current study? If so, then the authors should discuss how this paper "provides clear proof".
The sentence is rephrased saying that the mechanism for increasing CCN with altitude is still unclear.

Pg. 5091, Lines 9-25: Please add these percentile ranges to Figure 2b, so they can be more directly compared with the present measurements.
- *The 25th and 75th percentile from the long term data were added to Fig. 2b and Fig. 7b.*

Pg. 5094, Line 10: These should be geometric means and should have error bars reflecting the geometric standard deviation.
- *We do not agree about the need for geometric means in these figures. Please check our reply on comment regarding additional information on uncertainty in the figure.*

Pg. 5096, Line 28: Clarify where Silvergren et al. made these measurements.
- *The sentence on page 5096, lines 22-24 is changed to:" Silvergren et al. (2014) presented CCN number concentrations as a function of SS and as a function of the month from September 2007 to August 2008, calculated based on* ***aerosol collections on filters*** *at Zeppelin research station."*
- *The sentenced on page 5096, line 28 is changed to:" For August 2008, Silvergren et al. (2014) calculated CCN number concentration of approximately 65 particles cm^{-3} at 0.4% SS for the Zeppelin research station, which is although lower than the concentrations of 179 and 97 particles cm^{-3} calculated from the presented data in Fig. 12d for 21 and 24 August 2008."*

Pg. 5100, Lines 10-13: What is the justification for this assumed surface tension?
- *The water surface tension is used in the Köhler equation as a simplification (cf. Petters & Kreidenweis, 2007).*

Table 2: What are inorganics composed of? Why is a kappa value of 0.53 assumed here?
- *We do not know what the inorganics found in the particles 15 nm to 400 nm are composed of. However, in Zieger et al. (2010) and Rastak et al. (2014) ammonium sulfate (estimated κ of 0.53) was found to very well represent the scattering characteristics of particles being present during summer 2008 at Zeppelin Research Station. In the heading of Table 2 it is mentioned that for the inorganic fraction, properties of ammonium sulfate were used.*

Figures 2 and 7:
1. The size distribution color scale should be log

- *The color scale was changed to log.*

2. Please set some non-zero lower limit for the size distribution coloring and make values below this lower limit transparent. This will help the reader see the shape of the distribution given the very low values
- *Due to the change of the color scale from linear to log the shape of the distribution is well defined for the low values.*

3. It is hard to see the tick marks in part A.
- *This was corrected.*

4. Make the ordinate axis in part B log.
- *Whether to plot the ordinate scale as logarithmic or linear is typically a matter of taste. For this dataset we have chosen linear scale.*

Figure 6: Is this figure discussed anywhere in the text?
- *The information containing in Fig. 6 is used on p.5089, lines 8-12.*

Figure 13: Change CCN/CN scales to be 0 to 1. What is the meaning of that stray point in August?
- *The scale was changed as proposed. The stray point has no physical meaning and is disregarded.*

Figures 14-15: Note that the CCN spectra in these figures are for 0.4% supersaturation.
- *The caption of Figure 15 was changed to: "Activation ratio as a function of dry particle diameter (D_p) for the measurement period in June 2008 and August 2008. Obtained from measurements at a SS of 0.4%. Error bars indicate SD. The grey area indicates the for further analysis omitted data." In the caption of Figure 14 it is already stated that measurements were conducted at 0.4% SS.*

Figure 15:
1. What does the shaded region denote?
- *The grey area indicates data omitted from further studies. The figure caption was changed accordingly.*

2. Change the ordinate scale to be more reasonable (maybe 0 to 1.1?)
- *It was a conscious decision to show all the data points, hence the scale cf. p.5098 lines 3-12.*

3. Compute geometric mean and standard deviation s rather than arithmetic.
- *The presented values are based on the geometric means presented in Figure 14. This has been clarified in the figure caption.*

4. How many activation spectra went into these averaged curves?
- *The averaged curve for June consists of about 290 activation spectra whereas the averaged curve for August consists of about 374 activation spectra. This information was added to the "Experiments" section: "The measurement period for the first case study lasted from around 9.40 a.m. on 27 June to around 10.15 a.m. on 29 June during which about 290 size-resolved CCN scans were conducted. The measurement period for the second case study began at around 7.30 p.m. on 21 August and ended at around 10.50 a.m. on 24 August, resulting in about 374 size-resolved CCN scans." (p. 5087, lines 22 ff.)*

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