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

Response to reviewer 1:

The authors thank anonymous reviewer 1 for comments on the manuscript.

The reviewer opposes publication until we have shown what is new between our results from two limited time periods of CCN measurements and the more extended data set of CCN analysis from Silvergren et al. (2014), both based on aerosol sampling at the Zeppelin research station, Svalbard.

There is a paramount difference between how the results from these two studies examining particle activation in the Arctic are obtained. In the case of Silvergren et al. (2014) aerosols of essentially all available sizes (PM10) in the Arctic air were sampled on high volume filter substrates (12 sampling periods over each about one month). The filters were taken to the lab and the material collected on the filters were dissolved into a liquid. The extract was analysed in various ways among the particle activation using a cloud condensation nuclei counter was measured. This procedure leads to a very strong averaging of chemical properties of the aerosol as well as over time. On the other hand the data cover a full annual cycle.

Our approach is very different as it is size dependent on-line measurements at the Zeppelin observatory. Very importantly, this means very little averaging over time and even more importantly no chemical influence from large particles that constitute most of the particulate mass. This difference between the two studies is fundamental.

Hence, despite the relatively short periods of intense observations, these data present a unique insight into the size dependent CCN activation in the Arctic atmosphere. Moreover, to our knowledge, these are the first and only data of this kind presented for the Arctic aerosol.

Based on the argument mentioned above, the from our measurement approach calculated hygroscopicity values κ can be understood to not be biased by the chemistry of large particles.

However, obviously the novelty of the data set was not enough stressed in the conclusions of the article. Therefore the chapter “Summary and conclusions” (p 5103) is extended. Moreover a paragraph is added pointing out the need for size-resolved CCN measurements over a longer time span.

Changes in the manuscript:

After the sentence ”For the June 2008 measurement period, D_{50} was 60 nm, while for the August 2008 measurement period, D_{50} was approximately 67 nm.”, it is added and modified in line 12 on page 5104:“For the first time κ values for the Arctic were calculated based on activation diameters obtained from in-situ size-resolved CCN measurements, meaning the κ values are based on a conserved chemistry of the

particles. Values of the hygroscopicity parameter *κ* were calculated to be 0.4 and 0.3 for June and August, respectively."
Moreover it is added after the sentence "Therefore, the *κ* values based on in-situ measured size-resolved CCN measurements and growth factors are probably more meaningful in characterizing the ability of an aerosol population to become activated to cloud droplets.", in line 19, page 5104: "In future, it is needed to establish long term size-resolved CCN measurements in the Arctic to study the size dependent activation of particles for different seasons. An analysis of the difference in resulting *κ* values with *κ* values resulting from long-term chemistry analysis of the particles is needed to quantify and explain the reason for the differences and to point out possible differences to κ to the cloud model community." The last sentence of the chapter was deleted.

References

Silvergren S. Wideqvist U., Ström J., Sjogren S., and Svenningsson B. (2014): Hygroscopic growth and cloud forming potential of Arctic aerosol based on observed chemical and physical characteristics (a 1 year study 2007-2008), J. Geophys. Res. Atmos., 119, 14,080 – 14,097, doi:10.1002/2014JD021657.

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Response to reviewer 2

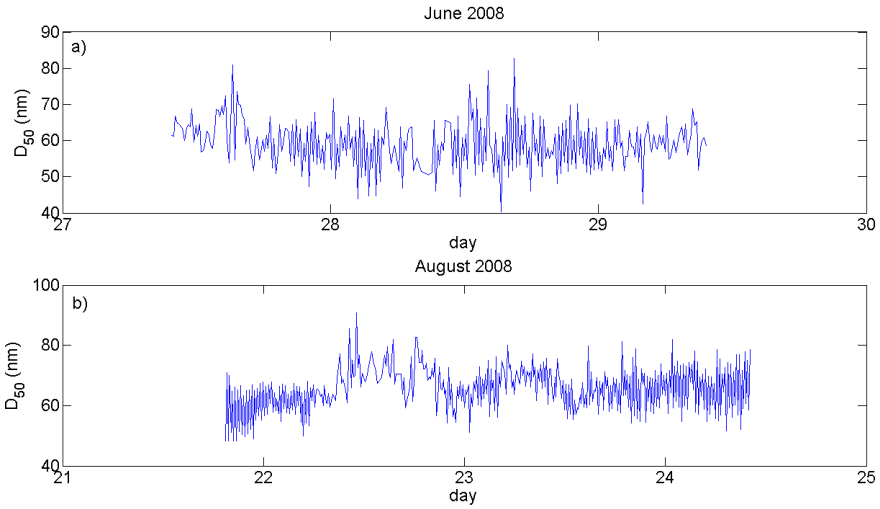
Thank you very much for taking the time to review our manuscript and pointing out areas of improvement. In the ingress the reviewer is asking if these results move Arctic CCN science forward. We believe that the reviewer actually agrees with us that the discrepancy, between κ values derived using different 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.

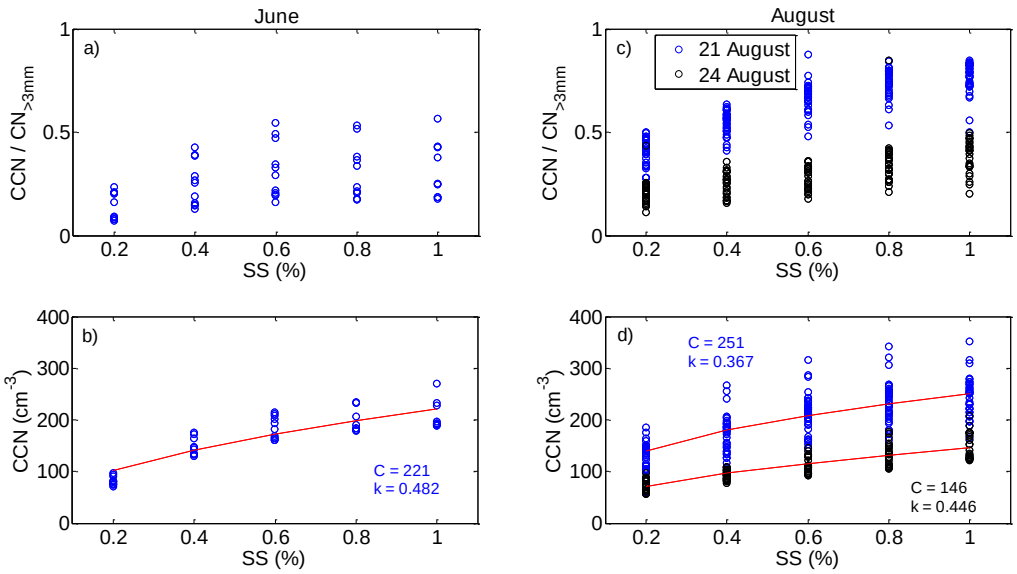


Figure 12. a) Ratios of the medians for each SS scan between CCN and particles with diameters > 3nm ($CN_{>3nm}$) for June 2008 as a function of SS. **b)** Medians for each SS scan of the total numbers of CCN as a function of SS for June 2008. **c)** Ratios of the medians for each SS scan between CCN 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.)*

References

Koehler, K. A., Kreidenweis, S. M., DeMott, P. J., Prenni, A. J., Carrico, C. M., Ervens, B., and Feingold, G.: Water activity and activation diameters from hygroscopicity data – Part II: Application to organic species, Atmos. Chem. Phys., 6, 795–809, 2006, http://www.atmos-chem-phys.net/6/795/2006/.

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Petters M. D. & Kreidenweis S. M. (2007): A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971.

Rastak N., Silvergren S., Zieger P., Wideqvist U., Ström J., Svenningsson B., Maturilli M., Tesche M., Ekman A.M.L., Tunved P., and Riipinen I. (2014): Seasonal variation of aerosol water uptake and its impact on the direct radiative effect at Ny-Ålesund, Svalbard, Atmos. Chem. Phys., 14, 7445-7460.

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Su H., Rose D., Cheng Y. F., Gunthe S. S., Massling A., Stock M., Wiedensohler A., Andreae M. O., and Pöschl U. (2010): Hygroscopicity distribution concept for measurement data analysis and modeling of aerosol particle mixing state with regard to hygroscopic growth and CCN activation, Atmos. Chem. Phys. 10, 7489–7503.

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Zieger P., Fierz-Schmidhauser R., Gysel M., Ström J., Henne S., Yttri K. E., Baltensperger U., and Weingartner E. (2010): Effects of relative humidity on aerosol light scattering in the Arctic, Atmos. Chem. Phys. 10, 3875–3890.

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Response to reviewer 3:
The authors thank anonymous reviewer 3 for comments and suggestions for the improvement of our manuscript.

General comments:

1. The literature overview in the introduction and the sections later when discussing about the results is representing the major part of the paper. I would emphasize more on the CCN results of this study, and maybe prepare a Table with all the other studies and the results of this paper.

- *The quite extensive introduction presenting studies of CCN in the Arctic had three main reasons:*

a) To ease literature research for scientists who deal with CCN research in the Arctic.

b) To show the limited amount of studies conducted on CCN in the Arctic and therefore emphasize the importance of this study, in general.

c) To show that (to our knowledge) no size-resolved CCN study was ever conducted in the Arctic environment and therefore emphasize the importance of this study.

2. Related to the comment above, the introduction is quite long and heavy to read. It would benefit from not presenting all the values from each single study (at this point the reader does not know their relevance to this study). I would extend the last paragraph of the introduction with relevant info and references from the literature review paragraphs and move the single values from different studies in a comparison Table or present them later when discussing about the results.

- *Actually, most of the results from studies presented in the introduction are not used for further comparison with our study as they do not present size-resolved measurements from the Arctic region. But still we think it is important to present the studies, because of the reasons named above. To reduce the "heaviness" of the introduction, section titles "Land-based measurements", "Ship-based measurements" and "Aircraft measurements" are added.*

Specific comments:
Methods:

1. Section 2.2: Please provide information on the inlet/sampling system for the CCN, DMPS & CPCs. Are there notable losses?

Behind line 10 on page 5087 it is added: "The shared inlet of the DMPS, TSI CPC

3025, and TSI CPC 3010 was precipitation protected with an estimated cut-off size of 5 µm." In line 6 on page 5087 the sentence is modified to: "A commercially available DMT CCN counter connected to a 1/4" stainless steel tubing inlet registered CCN concentrations at SSs of 0.2, 0.4, 0.6, 0.8 or 1 %." In the CCNC the flow inside is maintained as laminar flow in the saturation column, and the particle evaporation in the following OPC is negligible because of the minimal temperature difference (smaller than 2 degree Celsius, ref: factory manual). The particle loss in the flow can be regarded as negligible (or at least, equal to all) since we are using a common inlet in the setup.

2. Page 5087, lines 16-19: Add the resulting time resolution for the size resolved CCN cycle. 10 min per cycle?

- *One cycle (up and down scan over the 15 different particle sizes) lasted for 20 minutes. However, instead of the duration of one cycle, the total number of used activation spectra is 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.)*

Results and discussion:

1. It would be very informative and interesting to know also the temperature during the cases.

- *We have added the range of temperatures observed during the two events, which were (3.8-9.4 and 2.3-5.9, respectively.*

2. Page 5090, lines 15-19: New particle formation has been observed throughout the day, thus most frequently around noon. Having only one case here I would not state your results are in big contrast with previous results of Tunved or others. On the other hand you observe particles of 15nm at midnight, which means the actual NPF at 1nm has occurred many hours earlier on the previous day, probably in the afternoon (the growth does not seem to be very fast). Re-think this statement.

- *We agree the word "contrast" came out wrong. It was not the intention to highlight a difference, rather compare our observations with what indeed is the most common observation at this location. The section is changed to accommodate for this.*

3. Page 5091, lines 23-29: Similar comment to the one above. The case value is 40% higher than the average but still well inside the 75 percentile, so it is

quite a normal value. Maybe comparison to data from 2008-2010 would be better if the cut-off size is considered to make the difference.

- *As for the case above, our intention was not to stress a difference, just to try be quantitative of how the data sets compared. We removed the "40%" which makes the comparison more subtle. We agree that the values are quite normal.*

4. Page 5092, line 3-5, & Fig.7: Are the bad data for 22 Aug also removed from Figure 7? It seems in the Figure that only 21 Aug has some data missing.

- *The figure was redone and the data excluded when not used in the analysis.*

5. Page 5093-5094, lines 29-2: Also here the case value is well inside the 75th percentile, so it is an exceptional value.

- *The sentence on page 5093-5094, lines 29-2 was changed to: "Although, the total particle number concentration during the period in which the CCN size-resolved measurements were conducted is about 80 % higher than the long-term average, the particle number concentration still falls within the 75th percentile.*

6. Page 5099, lines 13-29: Please add a sentence (at the end) to connect this paragraph to this study. The paragraph info is used later in chapters, but at the moment it feels bit loose.

- *To connect the paragraph to the following section, a sentence on the end of page 5099 is added: "In the following section, the obtained information of the activation diameter, as well as the chemical information about the aerosol at the Zeppelin research station from another study are used to calculate the hygroscopicity parameter κ."*

7. Page 5101, lines 15-18: The particles >400 nm in the filters will make a major part of the total mass and probably is the main reason for the possible discrepancies.

- *We agree and reformulated the sentences on page 5101, lines15-18 to: "This could be due to the overestimation of the inorganic fraction in the "bulk κ", as particles with diameters >400 nm were also able to reach the filter. The chemical composition was therefore probably not accurately representative of the CCN-sized particles."*

Summary and conlusions:

1. The section is more of a summary at the moment. Few compact sentences of the main results, their importance and prospects for future work would be good.

- *After the sentence "For the June 2008 measurement period, D_{50} was 60 nm, while for the August 2008 measurement period, D_{50} was approximately 67*

nm.", it is added and modified in line 12 on page 5104:"For the first time κ values for the Arctic were calculated based on activation diameters obtained from in-situ size-resolved CCN measurements, meaning the κ values are based on a conserved chemistry of the particles. Values of the hygroscopicity parameter κ were calculated to be 0.4 and 0.3 for June and August, respectively." Moreover it is added after the sentence "Therefore, the κ values based on in-situ measured size-resolved CCN measurements and growth factors are probably more meaningful in characterizing the ability of an aerosol population to become activated to cloud droplets.", in line 17, page 5104: "In future, it is needed to establish long term size-resolved CCN measurements in the Arctic to study the size dependent activation of particles for different seasons. An analysis of the difference in resulting κ values with κ values resulting from long-term chemistry analysis of the particles is needed to quantify and explain the reason for the differences and to point out possible differences to κ to the cloud model community.

Tables

1. Table 3: This table is very small and is not needed. The values are stated in the text. They could be added also in Table 1 if wanted.

- *The values of Table 3 were added to Table 2.*

Figures

1. Interpretation of Figures 2 and 7 would benefit from plotting also the mean size distributions for the periods: before-during-after.

- *5 day-backward trajectories were calculated on an hourly basis and combined with Fig. 2 and Fig. 7., to be able to better interpret the changes in particle number size distributions with a change in air mass patterns.*

2. Maybe the trajectories for each case could be compiled in same Figure as done with the lidar plots (see Figs 6 & 11).

- *Based on comment 5) of reviewer 2, 5 days-back trajectories were calculated on an hourly basis and combined with Fig. 2 and Fig. 7., to be able to better compare air mass transport patterns with occurring particle number concentrations and particle number size distribution characteristics.*

3. Figure 14: Is the y-axis particle concentration, N? By summing up the CN channels the Ntot would be only around 20-30 cm-3 for the June case and just bit more for the August case. Or is this just me misunderstanding the plot?

- *Yes, the y-axis is the number particle concentration N, however the data are raw data and used to calculate the ratios, shown in Fig. 15.*

Technical corrections

1. Page 5096, line 25, 28, 29: Concentration values “XXX particles cm^{-3}”, delete “particles” to be consistent throughout the paper.

- *Done.*

Size-resolved cloud condensation nuclei concentration measurements in the Arctic: two case studies from the summer of 2008

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Abstract

The Arctic is one of the most vulnerable regions affected by climate change. Extensive measurement data are needed to understand the atmospheric processes governing this vulnerability. Among these, data describing cloud formation potential are of particular interest, since the indirect effect of aerosols on the climate system is still poorly understood. In this paper we present, for the first time, size-resolved cloud condensation nuclei (CCN) data obtained in the Arctic. The measurements were conducted during two periods in the summer of 2008: one in June, and one in August, at the Zeppelin research station (78°54'N, 11°53'E) in Svalbard. Trajectory analysis indicates that during the measurement period in June 2008, air masses predominantly originated from the Arctic, whereas the measurements from August 2008 were ~~characteristic of~~influenced mid-latitude air masses. CCN supersaturation (SS) spectra obtained on the ~~27th~~27 June, before size-resolved measurements were begun, and spectra from the ~~21st~~21 and ~~24th~~24 August, conducted before and after the measurement period, revealed similarities between the two months. From the ratio between CCN concentration and the total particle number concentration (CN) as a function of dry particle diameter (D_p) at a SS of 0.4%, the activation diameter (D_{50}), corresponding to CCN/CN = 0.50, was estimated. D_{50} was found to be 60 and 67 nm for the examined periods in June and August 2008, respectively. Corresponding D_{50} hygroscopicity parameter (κ) values were estimated to be 0.4 and 0.3 for June and August 2008, respectively. These values can be compared to hygroscopicity values estimated from bulk chemical composition, where

κ was calculated to be 0.5 for both June and August 2008. While the agreement between the two months is reasonable, the difference in κ between the different methods indicates a size-dependence in the particle composition, which is likely explained by a higher fraction of ~~sea salt~~inorganics in the bulk aerosol samples.

1 Introduction

The Arctic represents a region of special interest for atmospheric research ~~due to the following reasons~~because it is: i) very sensitive to changes in radiative forcing owing to ~~an active~~a direct feedback mechanism; ii) expecting greater anthropogenic activity from increased shipping and natural resource explorations in the near future and iii) yet poorly understood in terms of climate controlling processes, largely due to the lack of observational data. One of the most significant uncertainties in climate prediction is the role of clouds, and in particular, the influence of anthropogenic activities on clouds. In general, clouds have the ability to both cool the surface by reflecting incoming solar radiation back to space, or warm the surface by re-emitting long-wave radiation back to the surface (Boucher et al., 2013). The formation of clouds is dependent on the presence of excess water vapour in the air and on the presence of aerosol particles having cloud condensation nuclei (CCN) properties. Such particles must have sufficient size and hygroscopicity to act as sites for cloud droplet formation. In this study, two short case studies are presented, based on observations conducted in June and August 2008 at the Zeppelin station, Svalbard. These data complement the existing CCN and aerosol measurements conducted in the Arctic, but for the first time the CCN properties here are determined on-line as a function of dry particle size. Moore et al. (2011a) have provided a brief literature review of CCN measurements in the Arctic; however, to set our study in the context of other studies and to summarize the available information concerning Arctic CCN, we also present a short literature overview, including some of the most recent studies. For clarity, data are first grouped into land-based measurements, then measurements from ships and followed by aircraft measurements.

Land-based measurements

Shaw (1986) examined the CCN spectra of air masses characterized by Arctic haze during January and February 1985 in central Alaska. The maximum supersaturation (SS) was found

to be around 0.33%, and the dominant CCN consisted of soluble particles at a concentration of a few hundred per cm^{-3}, characterized by a rather large size of approximately 1 µm.

Silvergren et al. (2014) presented chemical and physical properties of aerosols ~~measured~~collected at the Zeppelin research station, Svalbard from September 2007 to August 2008. Hygroscopic growth and cloud forming potential were examined on a monthly basis. From this, it was shown that during the summer months, the SS has the greatest impact on the number of CCN. As the aerosol sulphate and nitrate mass concentrations reached a maximum between March and May, it was concluded that these months presented the most unfavourable cloud forming properties of the entire year. From September to February, sea salt was present in the highest mass concentrations. 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, which was noted as the month with the most favourable cloud forming potential.

Ship-based measurements

Bigg & Leck (2001) reported the results from CCN measurements conducted on an icebreaker at latitudes higher than 80°N, from ~~15th~~15 July to ~~23rd~~23 September 1996. They observed CCN concentrations ~~to range~~ between 1 and 1000 ~~CCN~~ cm^{-3} at a SS of 0.25% over the ~~entire~~ measurement period. Daily median CCN concentrations at the same SS were around 15–50 cm^{-3}, and over the course of a ~~single~~ day, concentrations could vary by up to one order of magnitude. A decrease in CCN concentration of approximately one order of magnitude was observed ~~in the first 36 h, during which~~when air was being transported from the open ocean to the pack ice. The authors suggested that this occurred as a result of ~~cloud formation.~~wet scavenging. However, after this 36 h-period, an increase in CCN concentration was observed, which was thought to be related to local aerosol production from bubble bursting occurring between the pack ice.

The results of CCN measurements conducted during three weeks in August and September 2008 on board the icebreaker ‘Oden’, which was drifting passively to the north of 87°N, are presented by Martin et al. (2011). A mean SS of 0.10% resulted in a mean CCN concentration of 14~~.01 ± 10.96~~ ± 11 cm^{-3}, which increased up to ~~46.99~~47 ± 37~~.43~~ cm^{-3} for a mean SS of 0.73%. In general, CCN closure within the measurement uncertainties was successful for SSs of 0.10%, 0.15% and 0.20%, assuming an internally mixed aerosol with a nearly insoluble

organic volume fraction. However, the calculated CCN concentrations for SSs of 0.41% and 0.73% overestimated the measured CCN concentrations; this is suggested to be a result of differing chemical properties of different aerosol sizes.

Aircraft measurements

Hoppel et al. ~~Results~~(1973) present results of aircraft measurements from ~~a flight campaign that took place in~~ February 1972 above Alaska, approximately 160 km north of Fairbanks~~, are presented by .Hoppel et al. (1973).~~ A strong temperature inversion was observed during the ~~measurement periods~~measurements, and an increase in CCN concentration, approximately from 100 to 400 cm^{-3}, was recorded to accompany an altitude increase ~~in altitude~~, from 1.75 km to 4 km. Moreover, ~~it was discovered that~~ the increase of CCN concentration with an increase in SS was dependent on the altitude of the measurements. At 4.3 km altitude, the CCN concentration increased approximately from 50 cm^{-3} at 0.3% SS to 600 cm^{-3} at 1% SS. In contrast, at approximately 0.3 km above sea level, an increase of CCN concentration was observed approximately from 60 cm^{-3} at 0.3% SS to 100 cm^{-3} at 0.8% SS, but no further increase was seen up to a SS of 1%. Hoppel et al. (1973) suggested that these data may indicate the production of CCN in the upper troposphere or in the stratosphere, followed by downward mixing into the lower atmospheric layers.

CCN data from aircraft measurements conducted during April 1992 over the Arctic Ocean were presented by Hegg et al. (1995). Measurements took place around 350 km from the Alaskan coast between 0.03- and 4-km altitude; they show CCN concentration to vary between 19.9 and 92.7 cm^{-3}, with a mean value of 47 ± 19 cm^{-3} measured at 1% SS. Hegg et al. (1995) also observed an increase in CCN concentration with altitude up to 3 km, but note that the data points above 3-km altitude are too sparse to reliably predict any trend. Below 1.6-km altitude the fraction of particles that act as CCN ranged between 0.002 and 0.38 with an average of 0.15 ± 0.08 at 1% SS. No reliable CN data were obtained for altitudes higher than 1.6-km due to a frozen valve. Hegg et al. (1995) concluded that their data are an indication of particle production at higher altitudes compared to lower altitudes. ~~The concentrations of particles having diameters of around 20 nm measured at 1.5- and 2.5-km altitude were more than one order of magnitude, which was greater than those measured at 0.1- and 0.03-km altitude.~~

Results of aircraft measurements made during 11 flights over Alaska in June 1995 were published by Hegg et al. (1996) and compared to measurements presented in Hegg et al. (1995). This further study concluded that the fraction of activated particles is, on average, approximately 0.10 at a SS of 1%. They therefore suggested that the number of smaller particles is higher during June 1995 than in the spring of 1992.

Yum & Hudson (2001) presented vertical CCN profiles obtained at least 500 km north of the Alaskan coast during a flight campaign in May 1998. They observed a clear increase in CCN concentration with an increase in altitude when low stratus clouds were present. However, under non-cloudy conditions, an increase in CCN concentration was only observed at heights with an air pressure lower than 700 mbar. Average CCN concentrations measured at a SS of 0.8% were 257 ± 79 and 76 ± 29 cm^{-3} above and below the stratus cloud, respectively. In contrast, the average CCN concentrations obtained at lower altitudes (comparable to the measurements beneath the clouds) during non-cloudy flights was 250 ± 41 cm^{-3}. The authors proposed that the CCN concentrations in the low cloudy boundary layer are controlled by cloud scavenging, resulting in a clearly altitude-dependent CCN density profile. Analyses of CCN spectra were also conducted, using the formula $N = C*SS^k$, where N is the CCN concentration at a given SS, C is the CCN concentration at 1% SS and k describes the slope of the function. The CCN spectra at specific height levels showed larger k values under the non-cloudy conditions, compared to when clouds were present (for example, 2.214 compared with 1.474 at 0.04%–0.1% SS and at 560–660 hPa), with the exception of the highest SS values found at lower altitudes.

Moore et al. (2011a) presented results from five research flights over the Alaskan Arctic during April 2008, beginning from Fairbanks and covering parts of the Beaufort Sea. The air masses sampled variously represented background conditions, biomass burning plumes, anthropogenic pollution and Arctic boundary layer conditions. Calculated activation curves with SS values ranging between 0.1% and 0.6% showed that at least 70% of the particles were activated for SS at around 0.2% for all air masses. It was therefore concluded that this similarity in observed activation pattern, despite the differences in chemical composition, is a result of aerosol size, which largely determines CCN activity. However, the authors pointed out that for SS between 0.3% and 0.6% it is likely that the particle chemical composition controls the maximum fraction of particles that can act as CCN.

Lathem et al. (2013) presented results of CCN measurements conducted during research flights from ~~26th~~26 June to ~~14th~~14 July 2008. The flight campaigns set-off from Cold Lake, Alberta, Canada and passed through the northeastern Canadian Arctic before heading to the west coast of Greenland. During the flights, the various air masses were characterized by biomass burning, boreal forest background, Arctic background and anthropogenic industrial pollution. Median CCN concentrations were highest for air masses influenced by fresh biomass burning, at 7778 cm^{-3} at standard temperature and pressure (STP, 1013 hPa and 273.15 K). At a SS of 0.55% the CCN/CN ratio was around 0.89 for those particles resulting from fresh biomass burning. The lowest CCN/CN activation ratio was 0.15 at SS 0.55%, observed for air masses characterized by industrial pollution, with a CCN concentration of 341 cm^{-3}. The Arctic background air mass resulted in a moderate activation ratio of 0.52 for 0.5% SS, while CCN concentration was 247 cm^{-3}.

During a flight campaign over the northern slopes of Alaska in April 2008, Hiranuma et al. (2013) collected ambient particles, dry residuals of mixed-phase cloud droplets and ice crystals. They analysed their size and chemical structure using an electron microscope in combination with various X-ray techniques. Note that the results should be interpreted with caution due to the limited number of samples. However, the limited data showed that the residuals of cloud droplets were enriched with respect to carbonate and black carbon, compared to the ambient particles. Significant mixing was also observed in the cloud droplet residuals. Additionally, during a period of high ice nucleation efficiency, residuals were enriched in sodium and magnesium salts compared to the ambient particles.

~~At least since the 1970s, several CCN measurements have been made in the Arctic.~~ The studies described above reveal the significant variability in CCN concentration across the Arctic, likely resulting from differing locations of CCN production (upper troposphere vs. lower boundary layer), production mechanisms, in-cloud processing and the origins of air masses. Several studies indicate an increase in CCN with increasing altitude in the lower half of the troposphere~~, but none provide clear proof of~~. However, the ~~dominant processes causing~~controlling mechanism for this increase is still unclear. In this study, we compare bulk CCN properties with those found in previous studies, and we also explore the size-dependence of CCN activation potential for the Arctic aerosols by combining a DMPS (Differential Mobility Particle Sizer) system with a CCN counter (CCNC). Although size dependent CCN activation has been studied worldwide (Bhattu & Tripathi, 2014; Rose et al.,

2010; Paramonov et al., 2013; Gunthe et al., 2009), according to our knowledge, this is the first study presenting size-resolved CCN activation in the Arctic.

2 Methods

2.1 Location

Measurements were made at the Zeppelin research station (78°54'N, 11°53'E, 474 m above sea level), which is situated approximately 2 km south-west of the small settlement Ny-Ålesund, in Svalbard. The station is seldom affected by local pollution and therefore can be considered to represent remote Arctic atmospheric conditions. Continuous aerosol measurements were begun in the year 2000, concerning which detailed information can be found in Tunved et al. (2013).

2.2 Instrumentation and experimental setup

Particle number size distributions were measured using a closed-loop Differential Mobility Particle Sizer (DMPS), consisting of a medium-sized Hauke Differential Mobility Analyzer (DMA) in combination with a TSI Condensation Particle Counter (CPC) 3010. Measurements were performed within 40 different size bins, with particle diameters ranging between 10 and 900 nm. Each particle size range was measured for 10 sec, followed by a lag time of 5 sec before the next size range was measured. Simultaneously, total particle number concentrations were precisely measured using a TSI CPC 3025 with a lower cut-off size of 3 nm, and by a TSI CPC 3010 with a lower cut-off size of 10 nm. A commercially available DMT CCN counter connected to ~~the same~~a 1/4" stainless steel tubing inlet registered CCN concentrations at SSs of 0.2~~%,~~ 0.4~~%,~~ 0.6~~%,~~ 0.8~~%~~ or 1~~%.~~ %."In the CCNC scanning mode, each SS level was measured for approximately 5 min before changing to the next SS level. After completing the 1% SS level, the measurements began again at 0.2% SS after at least a three min break in the measurements. The shared inlet of the DMPS, TSI CPC 3025, and TSI CPC 3010 was precipitation protected with an estimated cut-off size of 5 µm.

In the standard configuration these two instrument systems operate independently. In this study, however, we combined the two systems such that the DMA first selects a nearly mono-disperse aerosol, which is then supplied to the CCNC. For the CCN size-resolved

concentration measurements, the CCNC was connected to the DMA and SS was fixed at 0.4%. The number of size bins of the DMPS system was also reduced from 40 to 15, and the time each particle size was measured was extended from 10 to 35 sec to improve counting statistics. The lower and upper bounds of the DMPS scans were also narrowed to 15 and 400 nm, respectively. The two different setups of the CCNC are shown in Fig. 1.

2.3 Experiments

Two case studies are presented here, consisting of CCN size-resolved number concentration measurements conducted during summer 2008. The measurement period for the first case study lasted from around 9:40 ~~am~~ on ~~27th~~27 June to around 10:15 ~~am~~ on ~~29th~~29 June~~,~~ during which about 290 size-resolved CCN scans were conducted. The minimum and ~~the~~maximum temperatures for this period were 3.8 and 9.4°C, respectively. The measurement period for the second case study began at around 7:30 ~~pm~~ on ~~21st~~21 August and ended at around 10:50 ~~am~~ on ~~24th~~24 August, resulting in about 374 size-resolved CCN scans. The minimum and maximum temperatures for this period were 2.8 and 5.9°C, respectively. Before the size-resolved CCN concentration measurements were performed in June, CCN spectra were obtained from the total aerosols over a period of approximately 5 h. Unfortunately, no spectra were measured directly after the first CCN size-resolved experiment ended on ~~29th~~29 June. However, during the study in August, CCN spectra were determined for approximately 17 h on ~~21st~~21 August, before the CCN size-resolved experiment began, and for approximately 13 h on ~~24th~~24 August after the experiment ended. In addition, ~~10~~5-day backward trajectories were calculated on an hourly basis, using the online web version of the NOAA HYSPLIT Model (Draxler & Rolph, 2014; Rolph, 2014), to analyse the origins of the air masses from which the observed results presented in the next section were derived.

3 Results and Discussion

3.1 Time series analysis

Particle number size distributions observed from ~~27th~~27 to ~~30th~~30 June 2008 are presented in Fig. 2a. The vertical purple lines in this figure indicate the beginning and the end point of the measurement period for the size-resolved CCN number concentration data. Based on the particle number size distribution, at least three characteristic periods can be distinguished:

i) from midnight to approximately midday of ~~27th~~27 June, when particles with diameters of approximately 70 nm dominate the particle concentration; ii) from midnight to approximately midday of ~~28th~~28 June, when particle number concentrations are highest for particle diameters of approximately 20 nm and iii) from approximately midday on the ~~29th~~29 of June to the following midnight, when the concentration of particles with diameters approximately between 20 and 70 nm increased to more than 1000 cm^{-3} (cf. ~~Fig. 2a). For each of these three characteristic periods, 10-day backward trajectories were calculated (Fig. 3–Fig. 5). All the air masses reaching Zeppelin station on the 27th, 28th and 29th of June 2008 have a clear Arctic origin. Air masses reaching the station at 8 am on 27th June, at approximately the station altitude, had a relatively long residence time over the Barents Sea west of Novaya Zemlya (Fig. 3). In contrast, the air masses reaching Zeppelin station 24 h later on the 28th of June originated, without exception, from the central Arctic Ocean (Fig. 4). Air masses transporting high concentrations of particles with diameters approximately 50 nm to Zeppelin station during the night of 29th and 30th June originated from the northerly coast of Alaska (Fig. 5). Based on the size of these particles, it is likely that the high number densities towards the end of the period are derived from a major particle formation event over the pack ice.~~

Fig. 2a). For the period 27 to 29 June, 5-day backward trajectories were calculated for each hour (shown above Fig.2a and below Fig 2b). Air masses arriving between 0:00 and 11:00 at Zeppelin station are characterized by both, air coming from a southerly direction and air having its residence time exclusively at the high Arctic. From 12:00 on the 27 of June until midnight of the 29 of June air masses reaching Zeppelin station have a clear central Arctic origin. In addition to the trajectory analyses, Lidar measurements from Ny-Ålesund, part of the Micro Pulse Lidar Network (http://mplnet.gsfc.nasa.gov/, accessed ~~28th~~28 November 2013) were investigated for the presence of clouds or precipitation in the vicinity of the Zeppelin station. All times mentioned in these data refer to Coordinated Universal Time. Lidar measurements on the ~~27th~~27 of June 2008 in Ny-Ålesund showed only high clouds (altitude > 5 km), from approximately 9~~ am~~:00. Before 9~~ am~~:00, cloud-free conditions predominated at the measurement site. High clouds and cloud-free conditions alternated during ~~28th~~28 and ~~29th~~29 June 2008; therefore, Zeppelin station can be regarded as cloud-free during this time (cf. Fig. ~~6~~3).

The time series of particle number size distribution (Fig. 2a) is accompanied by two time series of total aerosol number concentrations for particles having a lower cut-off size of 3 and

10 nm, respectively (Fig. 2b). Although particles smaller than 10 nm are unlikely to be CCN, the combination of the two CPC instruments permit detection of particles that are a result of recent new particle formation. The combination of ~~10~~5-day backward trajectory analyses, Lidar measurements, particle number size distributions and total aerosol concentration time series gives a rounded picture of the conditions that prevailed during the experimental period.

The entire period from ~~27th~~27 to ~~30th~~30 June 2008 is characterized by a maximum of particle concentrations occurring at particle diameters below 100 nm. This is in line with the results of Tunved et al. (2013), who analysed long-term particle number size distributions at Zeppelin station during the years 2000–2010. In their study, the authors concluded that the Arctic summer aerosol number size distribution (June–August) is characterized by a dominance of particles with diameters less than the accumulation diameter. It is proposed that these aerosols are most likely formed within the Arctic itself. This explanation of local production agrees with our calculated trajectories (~~Figs. 3–5~~Fig. 2), which show transport almost only within the Arctic. In addition, the Lidar data from the period from ~~27th~~27 to ~~30th~~30 June 2008 does not indicate any cloud processing of the aerosols in the lower atmosphere boundary layer at the measurement site.

From midnight to approximately midday of 27 June, particles with diameters of approximately 70 nm dominate the particle concentration. The associated trajectory plot (Fig. 2) indicates that this pattern may result from a mixture of air masses, originating from the Norwegian Sea as well as from the Arctic Ocean.

During the early morning hours of ~~28th~~28 June 2008 ~~it is likely that new particle formation took place. A~~a sharp increase in total particle number concentration is observed (Fig. 2b~~), and the~~). The highest concentration of particle numbers is found for particles with dry diameters of less than ~~20 nm (Fig. 2a).~~ 20nm (Fig. 2a), which points together with the sharp increase in particle number concentration towards new particle formation during previous hours.The process of particle formation is not yet fully understood (Komppula et al., 2003; Yli-Juuti et al., 2011; Ortega et al., 2012), but sulphuric acid and organic compounds have been found to be the key components (Riipinen et al., 2007; Kuang et al., 2008; Sipilä et al. 2010; Kulmala et al., 2013). Most nucleation events take place during the daylight hours, which indicates the importance of photochemistry in the nucleation process. However, at some locations particle formation events are also observed at night when there is no ambient light (Ortega et al.,

2012). In Ny-Ålesund, the polar day lasts from around ~~18th~~18 April to ~~23rd~~23 August; therefore, the measurements made herein during June 2008 lie within this daylight period. Tunved et al. (2013) presented averaged diurnal particle number size distributions for June, based on observations made during 2000–2010, and found that the concentrations of particles with diameters less than ~~20 nm~~20nm predominantly begin to increase at around noon~~; this is in contrast with our observations, where the concentrations start to~~. Here presented data indicate, that an increase ~~at around midnight~~of particle concentration occured later in the day. In the Arctic environment, it has been suggested that dimethyl sulphide plays an important role as a condensing vapour for the nucleation process (Chang et al., 2011). Tunved et al. (2013) stated that another requirement of particle nucleation in the Arctic is a low condensation sink, which means a low concentration of particles in the accumulation mode. These authors showed that the particle mass is strongly related to accumulated precipitation along the transport path (cf. Figure 15 in Tunved et al., 2013), and that conditions are favourable for new particle formation during the period of midnight sun. ~~Despite this, our calculated trajectories did not indicate any precipitation en route, although calculated relative humidities at times reached approximately 90%.~~ 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 18.5 mm. From this we can conclude that recent precipitation within the last five days was not likely a dominant factor in shaping the aerosol properties during transport.

From midday on ~~29th~~29 June 2008 until approximately ~~10~~22:30 ~~pm~~ on that day, the total particle number concentrations of particles with diameters greater than 3 nm increased approximately from 400 cm^{-3} to 3860 cm^{-3} (Fig. 2b). The highest concentrations were found for particles with diameters between 30 and 70 nm. ~~The trajectory calculated for 500 m above sea level at this time indicates~~A change in the height pattern of the trajectories is seen between the midday of the 29 June and the following hours (Fig. 2). It seems that the air ~~mass originated at the Alaskan coast (Fig. 5). The reasons for such high number densities are unclear, but~~masses´ height is reduced and it is possible that ~~the high number densities indicate~~this change in transport pattern resulted in more moisture supply to the air mass which

helped promote particle formation ~~occurring over the Arctic basin some days before arriving at Ny-Ålesund~~and growth when the sun was at its highest.

To place the period in which ~~these~~the size-resolved CCN measurements were conducted in a long-term context, the median of the total particle number concentration for particles with diameters greater than 10 nm during this period is compared with the medians of the June data for the years 2001–2010 (Tunved et al., 2013). The long-term data have a time resolution of 1 h, but around 9% of these data are missing or are of poor quality and are therefore not considered in the calculation. The data are available within specific size distributions, and the total number was calculated by integrating over the distinct size ranges. From 2001 to 2005 the lowest measured size was 17.8 and the largest was 707.9 nm. From 2006 to 2007 a size bin with a lower measurement range of 13.8 nm was added. For 2008–2010 the size distribution diameter range was again broadened, to range between 10 and 790 nm. The calculations resulted in a median particle number concentration of 177 cm^{-3} for 2001–2010, with a 25th percentile of 80 cm^{-3} and a 75th percentile of 339 cm^{-3}. The median values with 25th percentile and 75th percentile for the period during which our CCN size-resolved measurements were conducted during June 2008 are 245, 195 and 292 cm^{-3}, respectively. ~~From this we can conclude that, although the data from 27th to 30th June 2008 appear relatively representative of the long-term June data, considering the low particle concentrations in the accumulation mode and the occurrence of a nucleation event,~~Although the median total particle number concentration is ~~around~~somewhat 40% higher than the averaged June data from 2001 to 2010~~.~~, it falls within the 75th percentile of the long term data. This in combination with the low particle concentrations in the accumulation mode and the occurrence of a nucleation event indicates that the case study data from June 2008 can be ~~partly explained by the different size ranges that are included in the calculation~~regarded as relatively representative.

Particle number size distributions from ~~21st~~21 to ~~25th~~25 August 2008 are presented in Fig. ~~7a~~4a. In this figure, the purple vertical lines indicate the start and end times for the CCN size-resolved concentration measurements. Difficulties with the DMPS measurements occurred approximately from 8 ~~am~~:00 to ~~7~~19:30 ~~pm~~ on ~~21st~~21 August and for short periods on ~~22nd~~22 August; these time periods are omitted from the analysis. The particle number size distribution time series represent time series of total particle number concentrations with dry diameters greater than 3 and 10 nm, respectively (cf. Fig. ~~7b~~4b). As with the measurements

from June 2008, different periods with different characteristic particle number size distributions can be distinguished during the studied time period in August 2008 (Fig. ~~7a~~4a): i) the final hours of ~~21st~~21 August, when particle number concentrations were highest for particles with diameters between 100 and 200 nm; ii) the early morning hours of ~~23rd~~23 August, when particle number concentrations were relatively low for all measured sizes (cf. Fig. ~~7b~~4b) and iii) during the first half of ~~24th~~24 August, when total aerosol concentrations were relatively high for the period, but no particular size range clearly dominated. Calculated ~~10~~5-day backward trajectories for ~~the 21st August at 9 pm and for the 23rd August at 4 am~~each hour indicate that ~~the~~ air masses ~~originated at lower levels and~~arriving on the 21 August at Zeppelin station mainly come from ~~rather low latitudes~~the southern part of the Norwegian Sea (Fig. ~~8 and Fig. 9). However,~~4). Air masses arriving from the ~~10 day backward trajectory for~~22 August until midday the ~~24th~~24 August at ~~8 am indicates the~~Zeppelin station have a more northern origin~~ to once~~, the Barents Sea Air masses arriving between midday and midnight on the 24 of August at Zeppelin station have again ~~be from higher latitudes (Fig. 10).~~an origin over the Norwegian Sea.

As with the measurement period in June 2008, Lidar data were consulted to investigate any local effects from clouds and precipitation (cf. Fig. ~~11~~5). During the ~~21st~~21 August 2008, apparently ~~non precipitating~~ clouds are present approximately between 0.7 and 9 km above the Zeppelin station. ~~On 22nd~~However, no precipitation reaching the station level could be detected. On 22 August low clouds (altitude < 2 km) were observed from approximately 9 ~~am~~:00, and precipitation started at approximately ~~12 pm~~24:00, continuing until approximately 9~~ pm~~:00. Only a few precipitation events are observed on ~~23rd~~23 August 2008 for the most part, no clouds are observed at the altitudes above the Zeppelin station. On ~~24th~~24 August, clouds were only observed in Ny-Ålesund at altitudes higher than 0.8 km.

From around ~~8~~20:00 to ~~11 pm~~23:00 on ~~21st~~21 August 2008, particles with dry diameters between 100 and 200 nm dominate the particle number size distribution. ~~According to the Lidar measurements~~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~~, and the~~ . The trajectories of the 21 August show that air masses ~~were passing northwards, originating~~originate from the mid-latitudes and lower their height when reaching Zeppelin research station (Fig. ~~8~~4). Therefore, it is likely that ~~this~~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. The accumulation mode-dominated size distribution differs somewhat from the typical summer conditions. Tunved et al. (2013) demonstrated from their long-term average, during June–August, which locally produced particles with diameters in the nucleation and Aitken mode dominate the particle number size distribution. In the ~~early~~ morning hours of ~~23rd~~23 August 2008, air masses arriving at Zeppelin station ~~also~~ originated in the ~~mid-latitudes~~Barents Sea (Fig. ~~9), but~~4) and resulted in relatively low total particle concentrations, compared to the concentrations observed between ~~8~~20:00 and ~~11 pm~~23:00 on ~~21st~~21 August 2008~~. At 8 am on 24th August, the air~~ (Fig. 2b). Air masses ~~once again~~in the morning of the 24 of August originated ~~in the Arctic Ocean region (Fig. 10) and resulted~~as well in the Barents Sea, but result in higher total particle ~~number~~ concentrations ~~of approximately 330 particles cm^{-3}, for particle diameters greater~~ than ~~10 nm, and approximately 500 particles cm^{-3} for diameters greater than 3 nm (Fig. 7b). This observation, in addition to the Arctic origin of~~ observed on the ~~air mass, suggest locally produced aerosols and is consistent with the observations~~23 of ~~Arctic aerosol characteristics during summer stated by Tunved et al. (2013).~~August.

To place our second case study data in a long-term context, we compare median values of August 2008 with the 10-year climatology presented by Tunved et al. (2013). Approximately 12% of the hourly data were excluded from calculations of the median integrated particle number concentration from 2001 to 2010 August, owing to them being either missing or of poor quality. The calculations produced a median particle number concentration of 127 cm^{-3} for August during 2001–2010, with a 25th percentile of 58 cm^{-3} and a 75th percentile of 252 cm^{-3}. In comparison, the median values with 25th percentile and 75th percentile for the size-resolved CCN measurement period in August 2008 are 226, 147 and 329 cm^{-3}, respectively. ~~This highlights a roughly 80% higher~~Although, the total particle number concentration during the period in which the CCN size-resolved measurements were conducted~~, compared to~~ is about 80 % higher than the long-term average~~.~~, the particle number concentration still falls within the 75th percentile.

Overall, the June case is similar to the long-term climatology and appears to be more representative of the summer period, with air masses of Arctic origin. In contrast, the August case differs more from the long-term climatology and shows a more significant influence of lower latitudes and higher number densities of accumulation particles.

3.2 CCN spectra

A CCN spectrum for a 5 h-long period on the ~~27th~~27 June 2008 was obtained before the size-resolved CCN measurements were begun. The data presented in this section comprise ~~arithmetic means~~medians calculated from one SS scanning cycle. The ratio, as function of SS, between CCN number concentration and the total particle number concentrations for particles with diameters greater than 3 nm ($CN_{>3nm}$) for ~~27th~~27 June 2008 is shown in Fig. ~~12a~~6a. A significant increase in the ratio of CCN to CN with an increase in SS is detectable by applying the two-sample Kolmogorov–Smirnov test, only for an increase in SS from 0.2% to 0.4%. The absolute number of CCN dependent on SS is shown for ~~27th~~27 June 2008 in Fig. ~~12b~~6b. Applying the two-sample Kolmogorov–Smirnov test to the data resulted in a significant difference in CCN numbers with increasing SS (5% significance level). The power-law function, $N_{CCN}(SS) = C*SS^k$, describing the number of CCN (N_{CCN}) with the coefficients C and k and SS, was fitted to the data shown in Fig. ~~12b~~6b and giving values for the coefficients of C = ~~222~~221 and k = 0.~~477~~482. Ranges in the parameters C and k depend on the type of air mass, and the values for ~~27th~~27 June 2008 will be discussed in a later section, in combination with the values obtained from the August 2008 data.

CCN spectra obtained during 17 h and 13 h observation periods on ~~21st~~21 and ~~24th~~24 August 2008, respectively, are shown ~~in~~ on the right side of Fig. ~~13~~6. The ratios between CCN and CN as a function of SS are shown in Fig. ~~13a~~6c for the two different days. For ~~21st~~21 August, a significant increase in the CCN to CN ratio with an increase in SS was observed in all cases. For ~~24th~~24 August, the increase in ratio was significant for all increases in SS, except for the increase from 0.4% to 0.6% SS. The absolute number of CCN for ~~21st~~21 and ~~24th~~24 August, as a function of SS, is shown in Fig. ~~13b~~6d. For both days, the increase in CCN number from one SS to the next is significant. This is based on applying the two-sample Kolmogorov–Smirnov test with a 5% significance level. As with the same data from ~~27th~~27 June 2008 (cf. Fig. ~~12b~~6b), a power-law function of the same form was fitted to the data from ~~21st~~21 and ~~24th~~24 August 2008, as denoted by the red lines in Fig. ~~13b~~6d. The fittings resulted in C = ~~252~~251 and C = 146 and k = 0.~~366~~367 and k = 0.~~452~~446 for ~~21st~~21 and ~~24th~~24 August 2008, respectively.

Rogers & Yau (1996) demonstrated that the coefficients for maritime air vary, with C = 30–300 cm^{-3} and k = 0.3–1.0, while for continental air the values vary between C = 300–3000

cm^{-3} and k = 0.2–2.0. The coefficients C and k that are given by the fitted power-law function applied to the measurements during June and August 2008 (cf. Fig ~~12b~~6b and Fig. ~~13b~~6d) are consistent with the ranges that Rogers & Yau (1996) proposed for maritime air masses. Pruppacher & Klett (2010) also presented a compilation of C and k values from different studies, alongside the CCN/CN ratio for a SS of 1% at different locations, characterized by either maritime or continental air masses. Only one study from the Arctic, influenced by a maritime air mass, is presented, providing a C value between 100 and 1000 cm^{-3}. Pruppacher & Klett (2010) did not present a k value for this study, but stated that the CCN/CN at 1% SS is 0.5. Compared to the data from Zeppelin in this study, this range is at the lower limit of that observed on 21st August 2008, and at the upper limit of that observed on ~~27th~~27 June and ~~24th~~24 August 2008 (cf. Fig. ~~12a~~6a and Fig. ~~13a~~6c). However, it should be noted that direct comparison is difficult as it is not known which size range was considered for the integrated number of CN. Hegg et al. (1995) also presented a number of C and k values and CCN/CN ratios obtained during several flight campaigns over the Arctic. Although an increase in CCN was observed with an increase in altitude for a SS of 1%, CCN concentrations for altitudes < 1.6 km were always lower than 100 cm^{-3}. This is in contrast to the measurements obtained at Zeppelin station in this study, where CCN concentrations were generally higher than 100 cm^{-3} at a SS of 1% (cf. Fig. ~~12b~~6b and Fig. ~~13b~~6c). The average CCN/CN ratio for measurements conducted at altitudes lower than 1.6 km was calculated to be 0.15 (Hegg et al., 1995), which is lower than the calculated average ratios of 0.32, 0.77 and 0.38 obtained for a SS of 1% for ~~27th~~27 June, ~~21st~~21 and ~~24th~~24 August 2008.

Yum & Hudson (2001) estimated an average CCN concentration of 76 cm^{-3} in conditions when low clouds are present and an average of 250 cm^{-3} for non-cloudy conditions at a SS of 0.8%. Estimating the CCN concentration at 0.8% SS with the power-law function results in CCN concentrations of ~~200~~199 cm^{-3}, ~~232~~231 and ~~131~~132 cm^{-3} for ~~27th~~27 June, ~~21st~~21 and ~~24th~~24 August, respectively. For ~~27th~~27 June only high clouds (altitude > 5 km) were present, while for ~~21st~~21 and ~~24th~~24 August clouds were observed at altitudes higher than 0.7 km (cf. Sect. 3.1). The CCN concentrations calculated herein using the power law relation and a SS of 0.8% for June and August clearly lies between those CCN concentrations determined by Yum & Hudson (2001) for non-cloudy conditions. The CCN/CN ratio calculated by Yum & Hudson (2001) is 0.65 at a SS of 0.8%, which is higher than the average ratio determined for the CCN spectra during ~~27th~~27 June and ~~24th~~24 August 2008. The arithmetic means of the

CCN/CN ratio at a SS of 0.8% are 0.~~32~~31, 0.~~74~~75 and 0.~~34~~35 for ~~27th~~27 June, ~~21st~~21 and ~~24th~~24 August 2008, respectively. Yum & Hudson (2001) also present altitude-dependent k values for a SS range of 0.1%–0.6%. The k values for cloudy conditions ranged between 0.27 and 0.55 and between 0.34 and 0.75 for non-cloudy conditions. Calculated k values for ~~27th~~27 June, ~~21st~~21 and ~~24th~~24 August 2008, only considering a SS range of 0.2% to 0.6%, were found to be 0.65, 0.41 and 0.37, which is similar to the results obtained by Yum & Hudson (2001).

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 ~~measurements~~aerosol collections on filters at Zeppelin research station. For June 2008, a CCN number concentration of around 100 ~~particles~~ cm^{-3} at a SS of 0.4% is shown. This concentration is lower than the calculated CCN number concentration found here using the power-law relation shown in Fig. ~~12~~6 and a SS of 0.4%, which results in a CCN number concentration of ~~143 particles~~142 cm^{-3}. For August 2008, Silvergren et al. (2014) calculated a CCN number concentration of approximately 65 ~~particles~~ cm^{-3} at 0.4 % SS for ~~August 2008,~~ the Zeppelin research station, which is ~~also~~although lower than the concentrations of ~~180~~179 and ~~96~~97 particles cm^{-3} calculated from the presented data in Fig. ~~12b~~6d for ~~21st~~21 and ~~24th~~24 August 2008.

No clear separation can be made between the two CCN spectra from August 2008 and the one CCN spectrum from June 2008. In general, the CCN spectrum of June 2008 (Fig. ~~12b~~6b) lies between the two different spectra of August 2008 (Fig.~~13b~~ 6d). Comparing backward trajectories ~~(calculated in a similar way to those shown in Fig. 3–5 and in Fig. 8–10)~~ arriving at Zeppelin ~~on 27th~~before midday the 27 June ~~at 3:00 am, 21st~~(Fig. 2) and before midday the 21 August ~~at 10:00 am~~ and ~~24th~~after midday the 24 August ~~at 7 pm,~~(Fig. 4), corresponding to the times when the CCN spectra were measured, ~~shows~~show that the air masses' ~~residence location~~origin was for ~~27th June and 24th August is to the east~~the most of the times southerly of Svalbard~~, but to the south of Svalbard for 21st August (not shown).~~. However, even those air masses with similar origins can show differences in their aerosol characteristics (Park et al., 2014).

3.3 CCN activation diameter

The size-resolved activation of particles having D_p between 15 and 400 nm at 0.4% SS is shown in Fig. ~~14~~7. 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. ~~14a~~7a). The lower panel shows the correspondent data for the measurement period in August 2008 (Fig. ~~14b). The presented arithmetic means and standard deviations 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.~~7b). The most distinct differences between the particle number concentrations of total particles measured by the CPC during the experimental period in June 2008 and August 2008 is a) a higher particle number concentration having D_p < 20 nm during June; b) a peak of particle concentration at approximately D_p 50 nm in August and c) a higher variation in particle concentration for the different size bins indicated by a higher ~~standard deviations~~geometric SD during August compared to June. As the CN number concentration, the CCN concentration is characterized by a higher variability during the measurement period in August compared to the measurement period in June.

To establish the presented study contextually with other studies, the ratio between CCN and CN as a function of dry particle diameter was calculated (Fig. ~~15~~8). Note that during June the CCN concentration exceeds the total particle concentration for D_p > 156 nm, and during August the CCN concentration is higher than the CN concentration for D_p < 19 nm and D_p > 123 nm. The experimental approach of selecting a narrow size range that can be applied to the CCNC results in very low particle concentrations in the instrument. In particular, for measurements made at either end of the size distribution, small errors can cause large changes in the ratio, as presented in Fig. ~~15~~8. To obtain completeness, all data points are shown; however, the sizes where CCN/CN ≥ 1 have been shaded and disregarded.

After applying a spline interpolation to the measurement data, the dry diameter at which 50% of the total particle number concentration was activated (D_{50}) was calculated to be 60 nm for the measurement period in June 2008 and 67 nm in August 2008. To the best of our knowledge, to date no size-resolved CCN measurements in the Arctic have been published; therefore, data are compared to results obtained in the subarctic. Anttila et al. (2012) reported

557 a study that was conducted at the Finnish Pallas-Sodankylä Global Atmospheric Watch
558 station that measured the ratio between cloud droplet number concentration and total particle
559 concentration while the station was in clouds as a function of dry particle size. By comparing
560 CCN concentrations at a fixed SS of 0.4% with cloud droplet number concentrations, it was
561 concluded that during the cloud events the "effective" maximal SS was likely to be
562 approximately 0.4% in most cases. During the five periods when the station was in clouds,
563 D_{50} varied between 80 and 102 nm on average. A comparable study at the same measurement
564 site resulted in D_{50} between 110 and 140 nm for maximal SS between 0.18% and 0.26%
565 (Anttila et al., 2009). Komppula et al. (2005) calculated D_{50} by comparing a particle number
566 size distribution measured at a site in clouds with a nearby measured size distribution
567 obtained at a station under cloud-free conditions. D_{50} was estimated to be 80 nm on average
568 and varied between 50 and 128 nm. Unfortunately, the SS is unknown. Due to the uncertainty
569 in SS, it is not possible to compare present study to the studies conducted at the Finnish
570 stations directly. However, due to SS lower than 0.4% reported by Anttila et al. (2012) and
571 Anttila et al. (2009), larger activation diameters in these studies compared to this study are
572 expected, which is in line with the presented results. Jaatinen et al. (2014) report activation
573 diameters for measurements conducted during the same field campaign as reported in Anttila
574 et al. (2012). However, compared with Anttila et al. (2012), in the present study, activation
575 diameters were calculated differently and for a shorter period. The critical diameter was
576 calculated by interpolating between the size bin at which the integrated particle number size
577 distribution was equal to the amount of total measured CCN and the previous size bin. This
578 resulted in a critical diameter of 98±16 nm for 0.4% SS (Jaatinen et al., 2014).

579 Besides SS, the chemical composition and mixing state determines the ability of particles to
580 become activated to cloud droplets (Frosch et al. 2011; Moore et al., 2011b; Ervens et al.,
581 2010; Sullivan et al., 2009). Kreidenweis et al. (2005) summarize results of predicted and
582 experimentally determined critical diameters of ammonium sulphate and sodium chloride
583 particles. Predicted critical diameters for sodium chloride particles vary between 44.6 and
584 39.4 nm (Kreidenweis et al., 2005 and references therein) and the experimentally determined
585 diameter for a SS of 0.4% was reported to be 40±6 nm (Corrigan & Novakov, 1999 in
586 Kreidenweis et al. 2005). Ammonium sulphate particles had larger predicted activation
587 diameters at SS of 0.4%, i.e., from 62.6 to 49 nm (Kreidenweis et al., 2005 and references
588 therein). Experimentally determined critical diameters of ammonium sulphate were 51 ± 8

and 59 ± 9 nm (Corrigan & Novakov, 1999 and Kumar et al., 2003 in Kreidenweis et al., 2005). Corrigan and Novakov (1999) experimentally estimated D_{50} measured at a SS of 0.4% to be 82 nm, 148 and 74 nm for succinic acid, adipic acid and glucose aerosols, respectively. It was concluded that all D_{50} match well with the D_{50} calculated theoretically, except for the less soluble adipic acid. Kumar et al. (2003) experimentally determined the activation diameter of oxalic acid to be 65 nm at a SS of 0.40%. In the following section, the obtained information of the activation diameter, as well as the chemical information about the aerosol at the Zeppelin research station from another study are used to calculate the hygroscopicity parameter κ.

3.4 Comparison of κ values obtained with different methods

The hygroscopicity parameter κ was first introduced by Petters and Kreidenweis (2007) to describe the relationship between particle dry diameter and CCN activity. In this study, κ values were calculated with two independent approaches for June and August 2008: 1) based on the CCN activation of the aerosol population; 2) based on the bulk chemical composition of the particulate mass sampled at the site.

First, the relationship between the activation diameter ($D_{p,act}$) and *SS* derived from κ-Köhler theory (Asa-Awuku et al., 2010) was applied to the experimental CCN data:

$$SS = \frac{2}{3}\left[\frac{4M_w\sigma}{RT\rho}\right]^{3/2}\left(3\kappa D_{p,act}^3\right)^{-1/2}. \quad (1)$$

where M_w (kg mol^{-1}) is the molar mass of water, T is the temperature, R is the universal molar gas constant, σ is the surface tension of the solution/air interface and ρ (kg m^{-3}) is the density of the solution. The surface tension of pure water 0.072 J m^2 and the density of pure water 1000 kgm^{-3} were applied. Temperature was assumed to be 295 K to match the temperature in the instruments. When analysing the experimental data, the activation diameter was assumed to be the dry diameter corresponding to the CCN to CN ratio of 0.5. However, we tested the sensitivity to this assumption by repeating the calculations for CCN/CN values of 0.25 and 0.75. The resulting κ values were 0.2–0.7 for June and 0.2–0.5 for August, with the best estimates (corresponding to the 50%-points in the CCN/CN ratios) of 0.4 and 0.3, respectively (Table 1).

Second, the κ values derived from the CCN activation data were compared to κ values obtained using the aerosol composition data. In this case, the total κ for the multi-component aerosol particles was calculated using the simple mixing rule

$$\kappa = \sum_i \varepsilon_i \kappa_i \cdot \quad (2)$$

where ε_i and κ_i are the volume fraction and hygroscopicity parameter of each component i, respectively (Petters & Kreidenweis, 2007). We assumed internally mixed aerosol particles, composed of four surrogate components (inorganics, more water-soluble organics, less water-soluble organics and elemental carbon, similar to Rastak et al., 2014; see Table 2 for the assumed single-component properties). The monthly mass fractions of the organic components were estimated by analysing filter samples of particles that passed a PM_{10} inlet. The inorganic fraction was determined after sampling the aerosol with an open face system without a PM_{10} inlet but shielded with a cylinder that reduced the sampling efficiency for particles larger than 10 µm (Silvergren et al., 2014). It should be noted that the properties of ammonium sulphate were used to describe the inorganic fraction. The sea salt contribution in the inorganic fraction was not considered for particles less than 400 nm (upper bound of the DMPS in this study). The resulting total κ values were approximately 0.5 for both considered months (Table ~~3~~2).

Comparison of the "bulk κ" (obtained with Eq. (2) and bulk chemical composition) with the "CCN κ" (obtained from the CCN/CN = 0.50 point with Eq. (1)) shows reasonable agreement for June but a slight overestimation for August. This could be due to the overestimation of the inorganic fraction in the "bulk κ~~." Particles~~", as particles with diameters >~~-~~400 nm were also able to reach the filter~~; thus, the~~. The chemical composition was therefore probably not accurately representative of the CCN-sized particles. ~~In any case, the results suggest that the aerosol resembles ammonium sulphate in its hygroscopic properties and CCN activation.~~ In addition, it should be noted that the data used to calculate κ with Eq. (1) are based on only 2–3 days of measurements during June and August 2008 while the calculations used in Eq. (2) are based on bulk aerosol properties over the whole month of June and August 2008.

Silvergren et al. (2014) used three different approaches to calculate κ for June 2008 and August 2008. Bulk aerosol samples on filters were obtained during June and August 2008 at the Zeppelin research station. The filters were extracted and the extract was again filtered so that only the water-soluble fraction of the aerosols remained. With the first two approaches,

particles were generated by an atomizer and then measured in a Hygroscopic Tandem Differential Mobility Analyser (HTDMA) and a CCNC. The corresponding κ values were calculated based on determined growth factors and critical SS and were estimated to be approximately between 0.4 and 0.5 on an average for both June and August 2008. In the third method, Silvergren et al. (2014) used the chemical information from the filter samples in combination with literature values to determine the activation diameter and the critical SS based on Köhler theory. The resulting κ values were approximately 0.7 for both June and August 2008 (cf. Fig. 9 in Silvergren et al., 2014). In present study, the "CCN κ" value for June was 0.4, which is in the lower end of results reported by Silvergren et al. (2014). For August, "CCN κ" was 0.3, which is lower than the results presented by Silvergren et al. (2014). The "bulk κ" values determined here are within the ranges determined by Silvergren et al. (2014) based on HTDMA and CCNC measurements.

Based on aerosol optical properties, Zieger et al. (2010) determined a mean κ value of 0.6 for the period July to October 2008 at the Zeppelin research station. The value presented by Zieger et al. (2010) is somewhat higher than the "bulk κ" estimated in this study (0.5 for both months). Conversely, the κ values calculated from the D_{50} using the DMPS-CCNC combination are clearly lower (0.4 for June, and 0.3 for August).

The main reason for the differences between the present study and both Zieger et al. (2010) and Silvergren et al. (2014) is probably related to the influence of large (>400 nm) particles in determining κ based on aerosol optical properties and the bulk chemical composition.

Anttila et al. (2012) reported a κ value of approximately 0.1 at Pallas for the same period as he presented activation diameters for (cf. Sect 3.3). The presented κ values are based on HTDMA measurements at 90% relative humidity for particles with D_d = 100 nm. Jaatinen et al. (2014) presented κ values obtained during the same measurement campaign as Anttila et al. (2012). However, the hygroscopicity parameter was determined differently and for a shorter period. The κ value derived by the critical diameter (cf. Sect 3.3) for a SS of 0.4% was estimated to be approximately 0.1 and thus is in agreement with the HTDMA-based observations of particles with D_d = 100 nm reported by Anttila et al. (2012). Conversely, the κ calculated based on Aerosol Mass Spectrometer (AMS) data collected approximately 6 kilometres from the site of the CCN and HTDMA measurements was approximately 0.3. The "CCN κ" values determined in this study are higher than those obtained by Anttila et al.

(2012) and Jaatinen et al. (2014). These differences can be explained by the different chemical composition of the aerosol population. Jaatinen et al. (2014) showed that at Pallas 47% of the measured mass concentration of the aerosols consisted of organic compounds, while at Zeppelin 90% of the aerosol mass was inorganic material and thus more hygroscopic material.

4 Summary and Conclusions

For the first time, size-resolved CCN measurements in the Arctic have been reported. Measurements were conducted at the Zeppelin research station, Svalbard during two short periods in June and August 2008. A near monodisperse aerosol having a D_p between 15 and 400 nm was selected by a DMA. The DMA was connected to a CCNC operating at 0.4% SS and in parallel to a CPC 3010. Before and after the size-resolved CCN measurements were taken, the CCNC was measuring the ambient air without previous selection of a monodisperse aerosol. During these periods, the SS in the CCNC was changed to 0.2%, 0.4%, 0.6%, 0.8% and 1%.

Trajectory analysis showed that during the measurement period in June 2008 air masses arriving at Zeppelin were dominated by Arctic air, while during August 2008 air masses ~~dominantly~~ originated from the ~~mid-latitudes~~Norwegian Sea and from the Barents Sea. A comparison of long-term June particle number size distributions with those registered during the size-resolved CCN measurements in June 2008 showed that the size distribution characterized by a nucleation event and low particle concentrations for D_p < 100 nm is representative for averaged conditions during June. In contrast, the particle number size distributions registered during August 2008 indicate long-range transport that differs from the long-term observations during August. In addition to the size-resolved CCN measurements, SS spectra were determined. In June, this was done directly before the size-resolved measurements were completed and in August directly before and after the size-resolved measurements were conducted. A power-law function of the form $N_{ccn}(SS)= C* SS^k$, with N_{ccn} as the number of CCN and the coefficients C and k, was fitted to the SS spectra. The coefficients for June were estimated to be C = ~~222~~221 and k = 0.~~477~~482. Coefficients for August were C = ~~252~~251 and k = 0.~~366~~367 before the size-resolved measurements were conducted and C = 146 and k = 0.~~452~~446 after the size-resolved measurements were conducted. The spectra measured during June lies between the two measured during August.

For a SS of 0.4%, CCN number concentrations as a function of dry particle diameter were presented. From the size dependent CCN measurements, D_{50} (particle diameter where CCN/CN = 0.5) was estimated. For the June 2008 measurement period, D_{50} was 60 nm, while for the August 2008 measurement period, D_{50} was approximately 67 nm. ~~Corresponding values~~For the first time κ values for the Arctic were calculated based on activation diameters obtained from in-situ size-resolved CCN measurements, meaning the κ values are based on a conserved chemistry of the particles. Values of the hygroscopicity parameter κ were calculated to be 0.4 and 0.3 for June and August, respectively. Estimating κ based on simplified bulk chemical properties that were observed in June and August (2008) gave a value of 0.5. The higher κ value based on chemistry is likely explained by an enhanced influence of larger and more hygroscopic particles. It should be considered that, due to their lower numbers, these larger particles are less crucial for CCN activation. Therefore, the κ values based on in-situ measured size-resolved CCN measurements and growth factors are probably more meaningful in characterizing the ability of an aerosol population to become activated to cloud droplets. ~~Size-resolved chemical observations for particles less than 60 nm are clearly advantageous to further understanding of cloud condensation nuclei properties and cloud activation~~In future, it is needed to establish long term size-resolved CCN measurements in the Arctic to study the size dependent activation of particles for different seasons. An analysis of the difference in resulting κ values with κ values resulting from long-term chemistry analysis of the particles is needed to quantify and explain the reason for the differences and to point out possible differences to κ to the cloud model community.

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Table 1. Measured diameters when CCN/CN = 0.25, 0.50 and 0.75 and corresponding calculated κ values with Eq. (1)

	June		August	
	Activation diameter (nm)	κ	Activation diameter (nm)	κ
CCN/CN = 0.25	49	0.7	56	0.5
CCN/CN = 0.50	60	0.4	67	0.3
CCN/CN = 0.75	72	0.2	78	0.2

900 Table 2. Experimentally-derived mass fractions (Silvergren et al., 2014), densities ρ and κ_i
901 values for each component used for the total κ calculations (Rastak et al., 2014 and references
902 therein). Properties of ammonium sulphate were assumed for the inorganic fraction.

Component	Mass fraction (%) June	Mass fraction (%) August	ρ (kg m^{-3})	κ_i
inorganics	88	90	1770	0.53
more water-soluble organics	10	7	1560	0.27
less water-soluble organics	2	2	1500	0.10
elemental carbon	1	1	1800	0.00

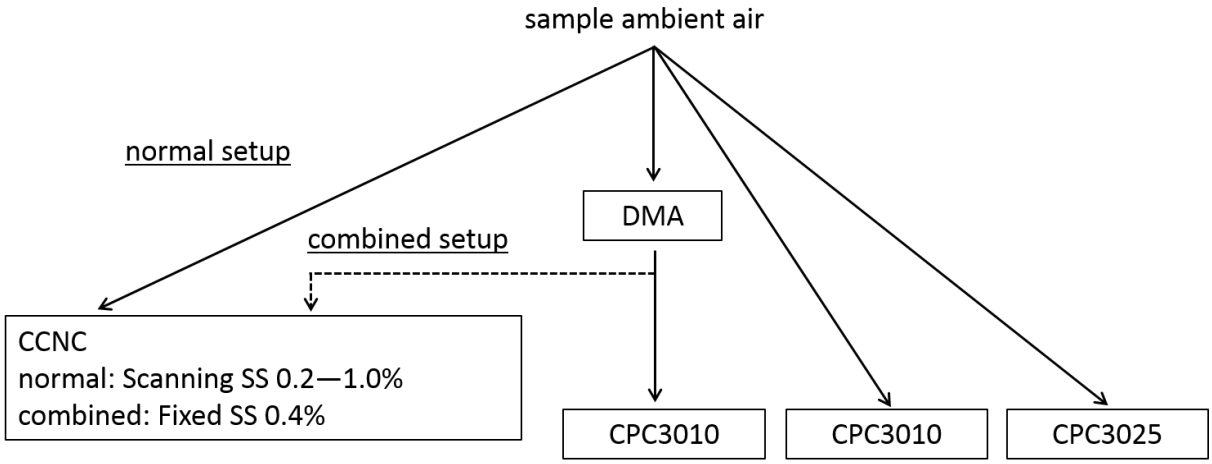
903

~~Table 3.~~

~~Total κ~~ ~~values calculated based on the chemical and physical properties presented in Table 2~~

			Total κ
	June	~~August~~	0.5
~~Total κ~~	August	~~0.5~~	0.5

907



908

909 Figure 1. Scheme of the two different measurement modes for the cloud condensation nuclei
910 counter (CCNC). When CCN size-resolved number concentration measurements took place,
911 the CCNC was connected behind the Differential Mobility Analyzer and the supersaturation
912 was set to 0.4%. During normal operation, the CCNC was connected parallel to the DMA and
913 SS alternated between 0.2% and 1.0%.

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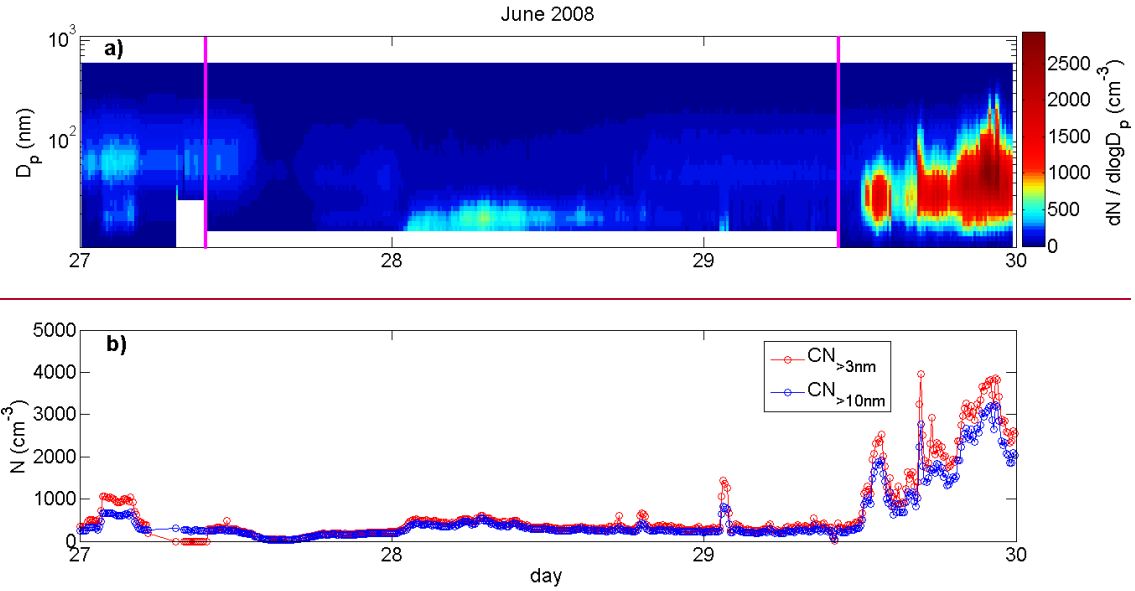

June 2008
a)
10^3
10^2
D_p (nm)
27
28
29
30
2500
2000
1500
1000
500
0
dN / dlogD_p (cm^{-3})
b)
5000
4000
3000
2000
1000
0
N (cm^{-3})
$CN_{>3nm}$
$CN_{>10nm}$
day

915

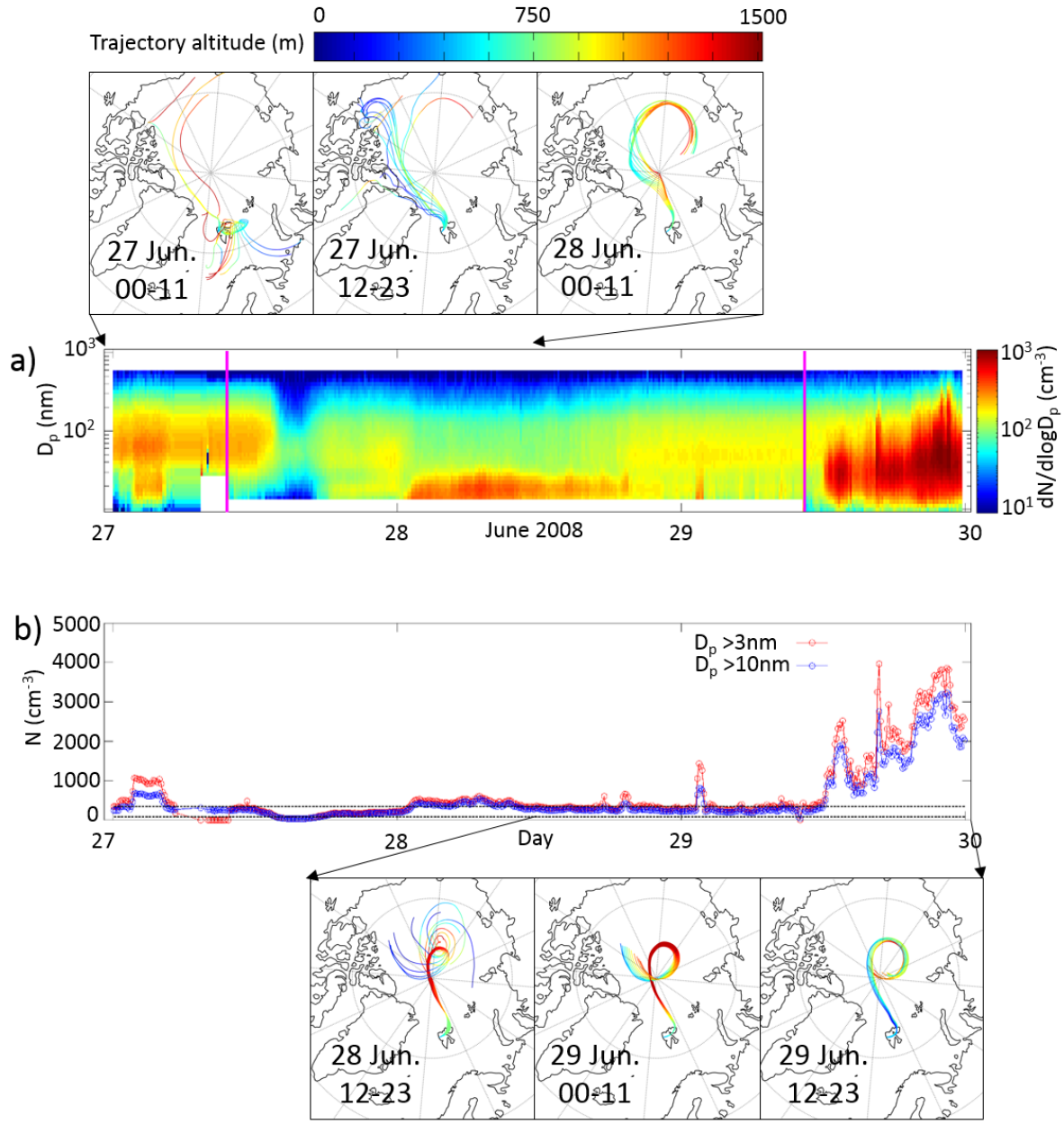
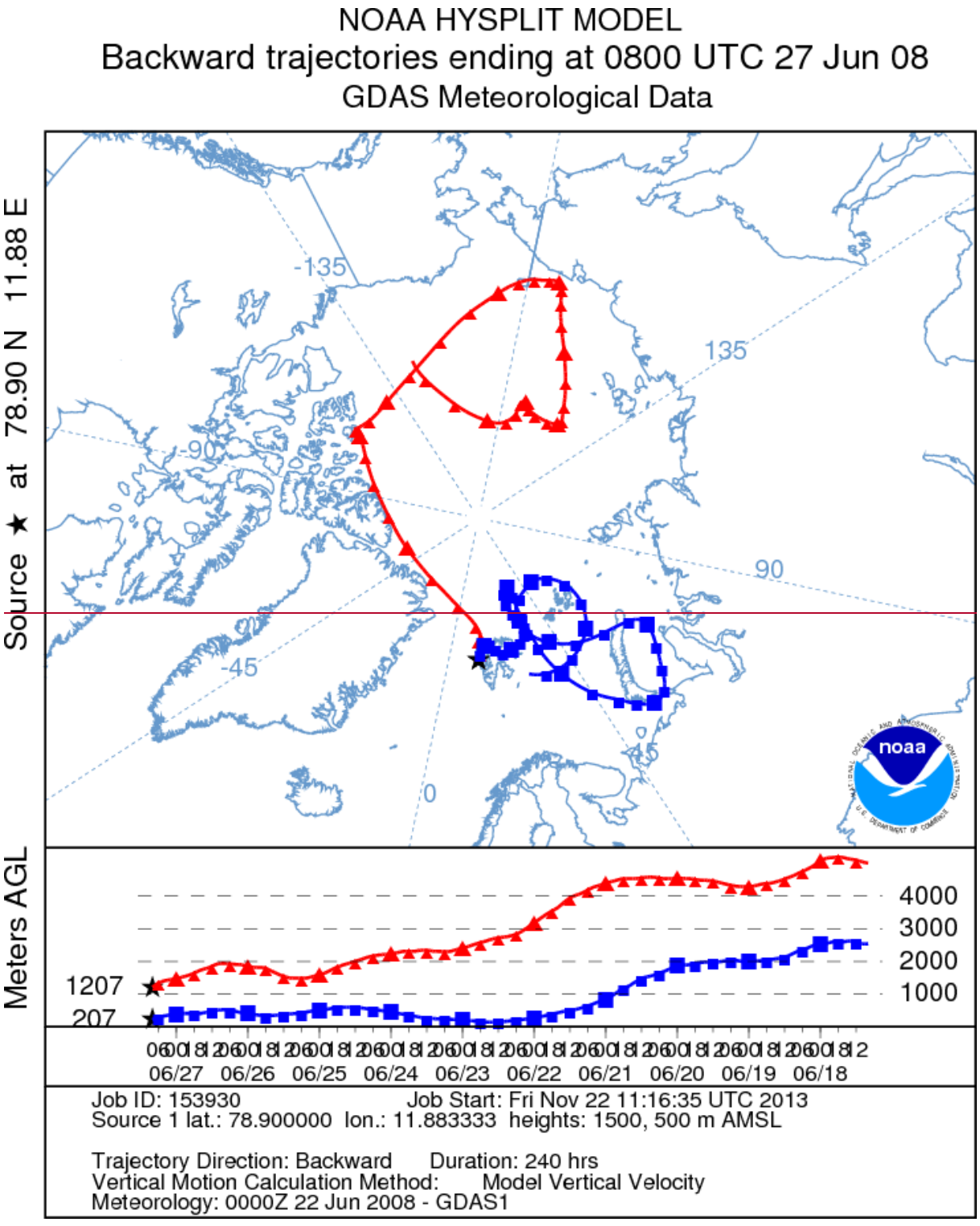


Figure 2. a) Particle number size concentration measured before, during, and after the size-resolved CCN concentration measurements were conducted in June 2008. Purple vertical lines indicate the start and end time of the CCN size-resolved concentration measurements. b) Time series of the 8-min medians from CPC measurements for the same period in June 2008. Horizontal dashed lines represent the 25th and 75th percentile of the CN number concentration for June during the years 2001 to 2010. Trajectory plots show 5-day backward trajectories, calculated for every hour. Trajectory plots on top of panel a) show air masses arriving

between the 27 and midday of the 28 June at Zeppelin Research Station. Trajectory plots below panel b) show air masses arriving between midday of the 28 June to midnight of the 29 June at Zeppelin Research Station.


NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 27 Jun 08
GDAS Meteorological Data
Source ★ at 78.90 N 11.88 E
-135
135
-90
90
-45
0
noaa
Meters AGL
4000
3000
2000
1000
1207
207
06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12
06/27 06/26 06/25 06/24 06/23 06/22 06/21 06/20 06/19 06/18
Job ID: 153930 Job Start: Fri Nov 22 11:16:35 UTC 2013
Source 1 lat.: 78.900000 lon.: 11.883333 heights: 1500, 500 m AMSL
Trajectory Direction: Backward Duration: 240 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 22 Jun 2008 - GDAS1

929

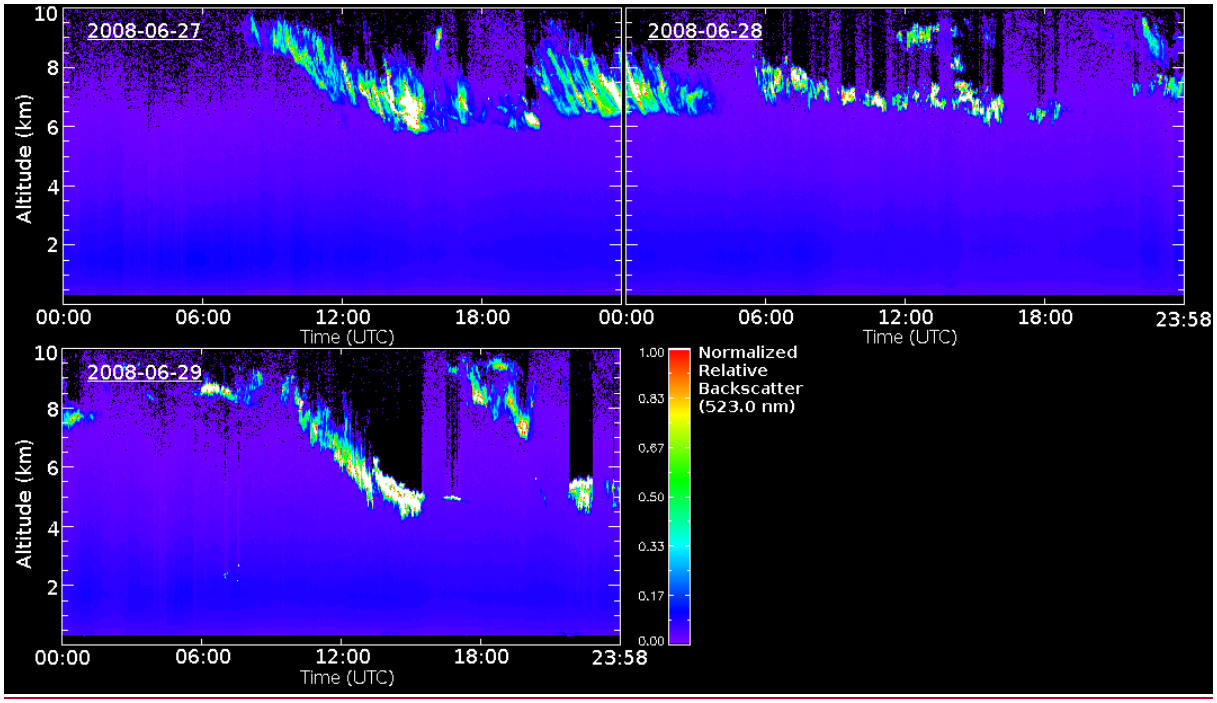
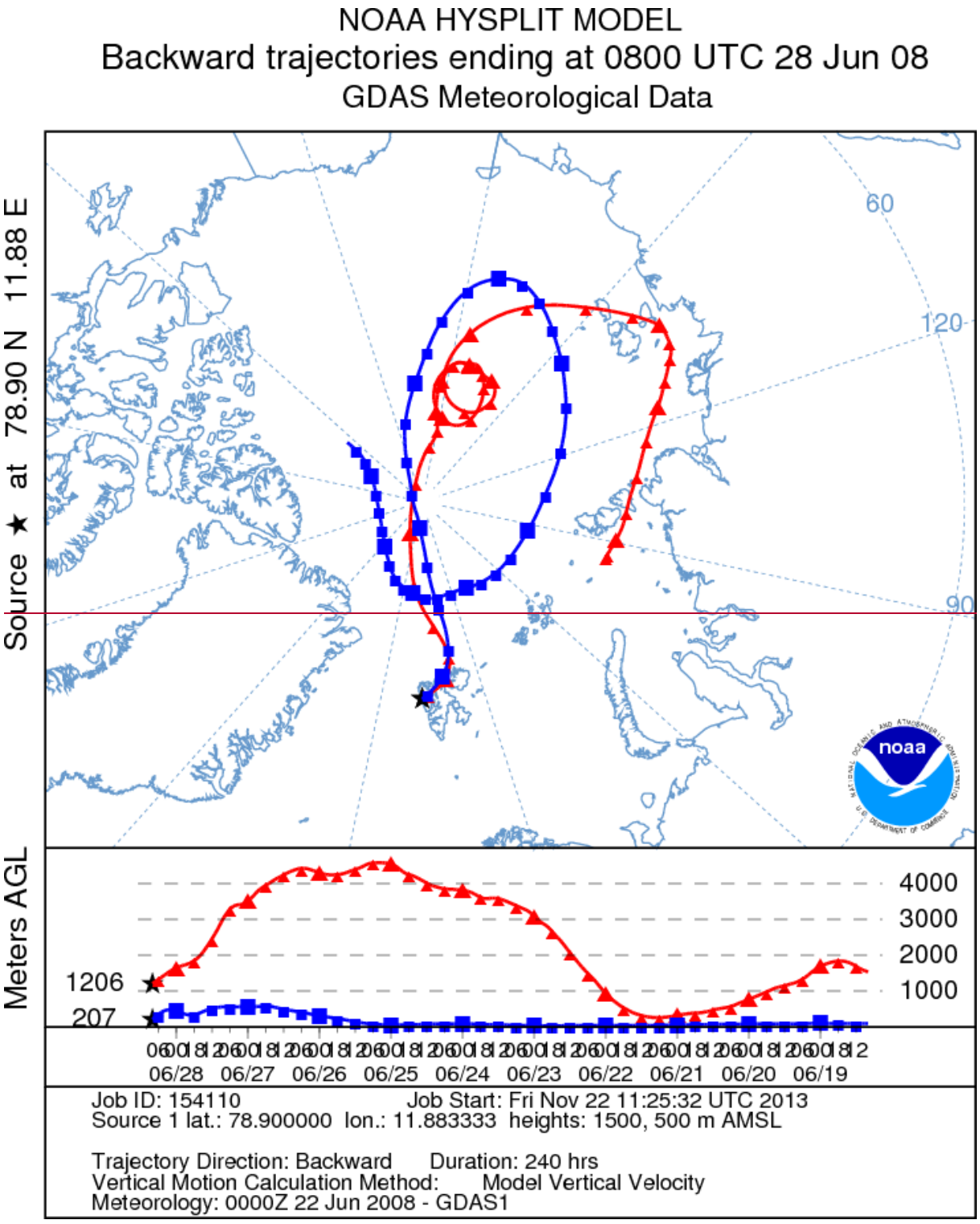
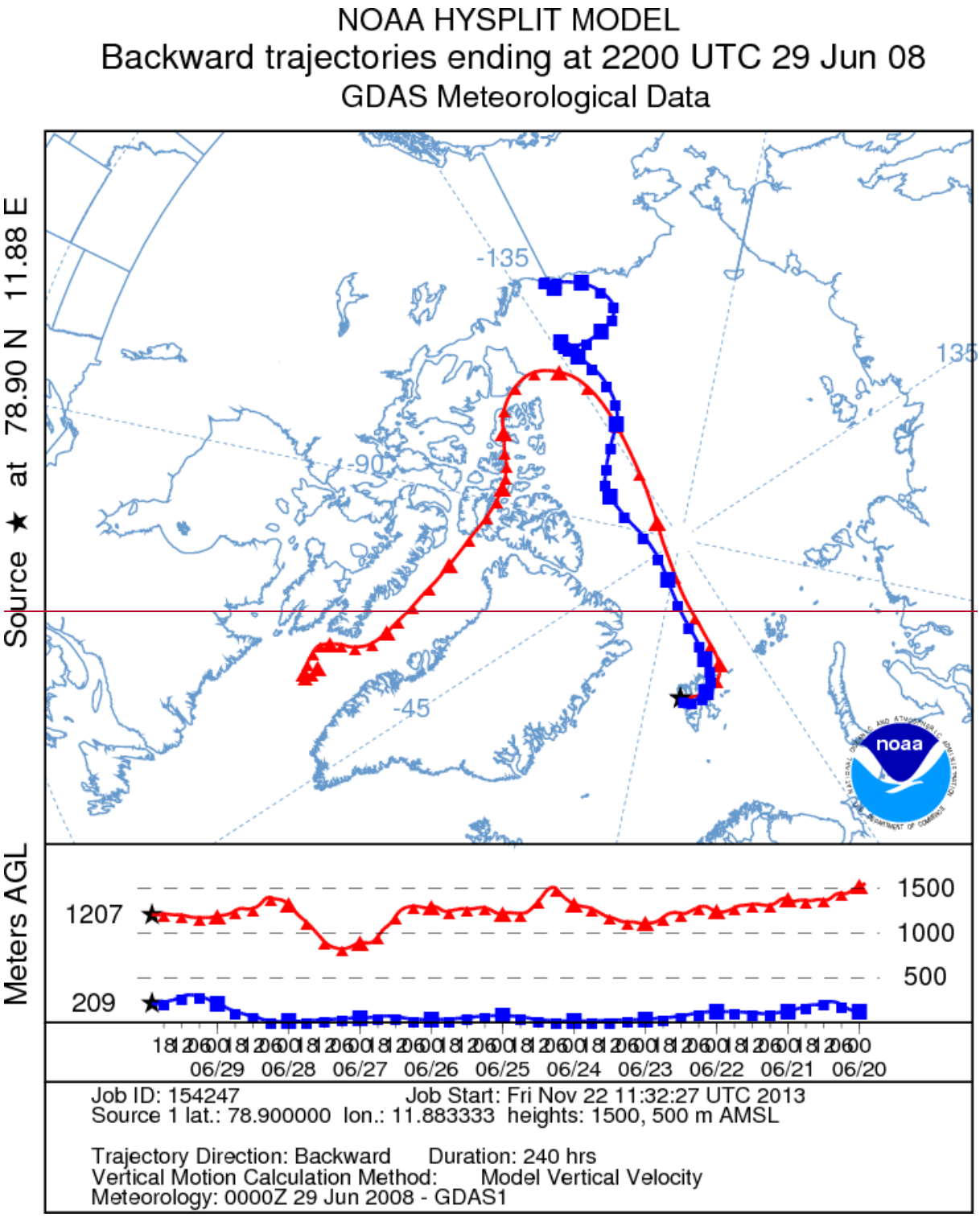


Figure 3. ~~10-day backward trajectory reaching Zeppelin research station, 27 June 2008 at 8:00 am~~



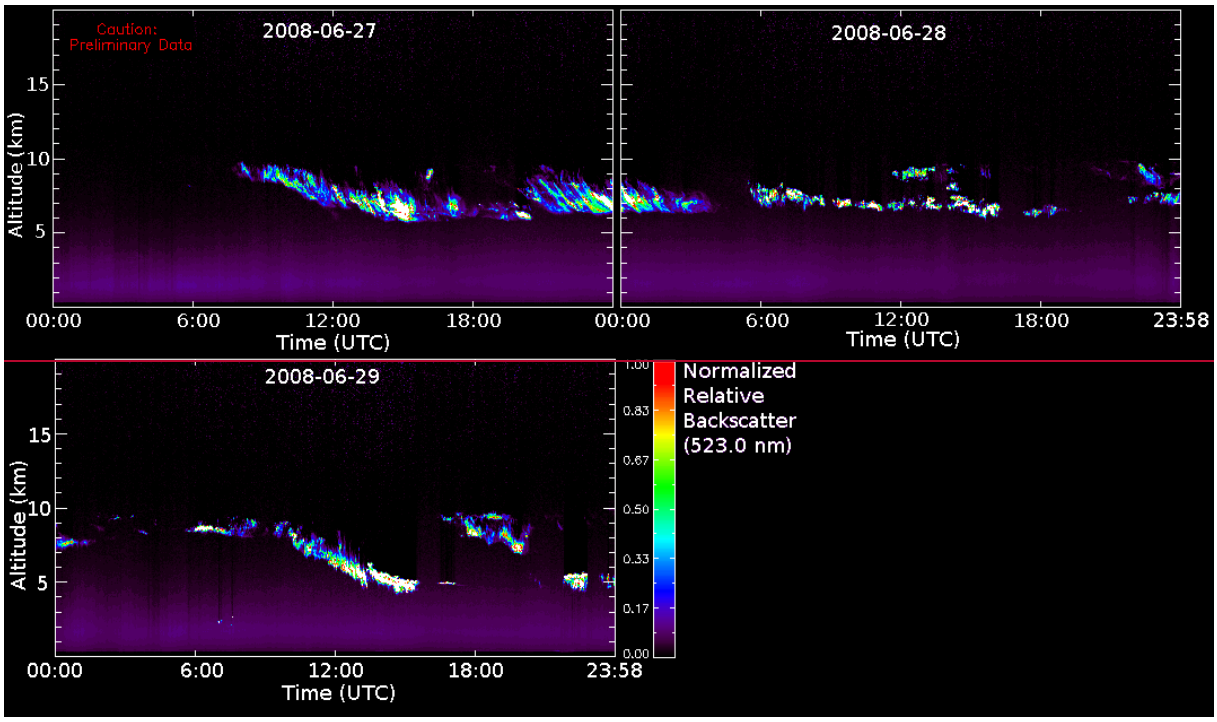
~~Figure 4. 10-day backward trajectory reaching Zeppelin research station, 28 June 2008 at 8:00 am~~



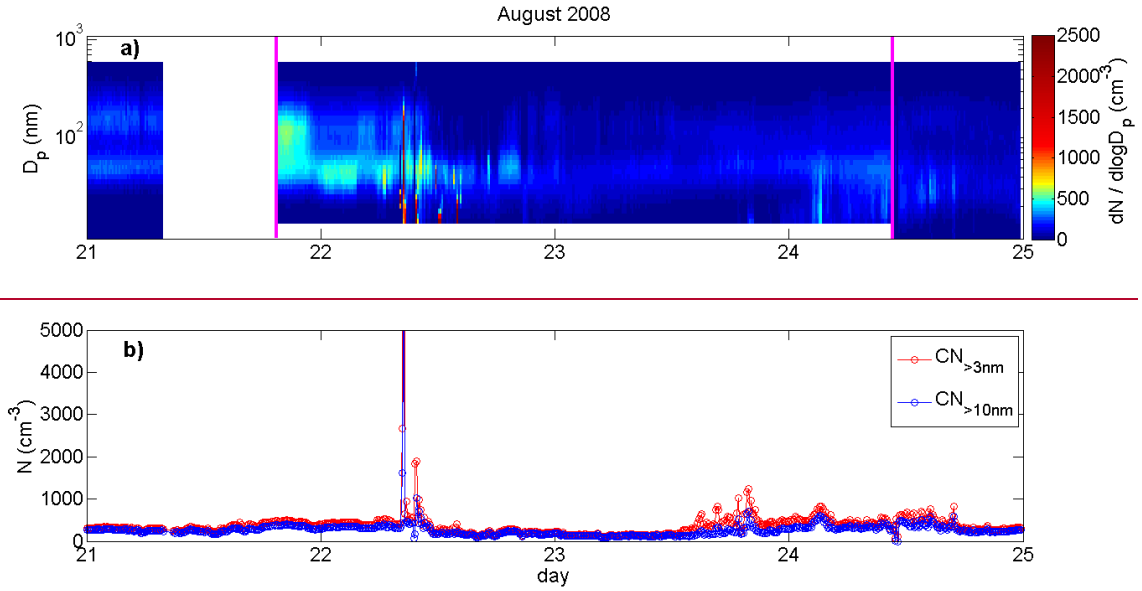
938

939 ~~Figure 5. 10-day backward trajectory reaching Zeppelin research station, 29 June 2008 at~~

940 ~~10:00 pm~~



~~Figure 6.~~ Normalized relative backscatter (Level 1.0 data) based on Lidar measurements at Ny-Ålesund recorded during the period ~~27th–29th~~ 27–29 June 2008 (modified from http://mplnet.gsfc.nasa.gov/)


August 2008
a)
10^3
10^2
D_p (nm)
21
22
23
24
25
2500
2000
1500
1000
500
0
dN / dlogD_p (cm^{-3})
b)
5000
4000
3000
2000
1000
0
N (cm^{-3})
$CN_{>3nm}$
$CN_{>10nm}$
day

946

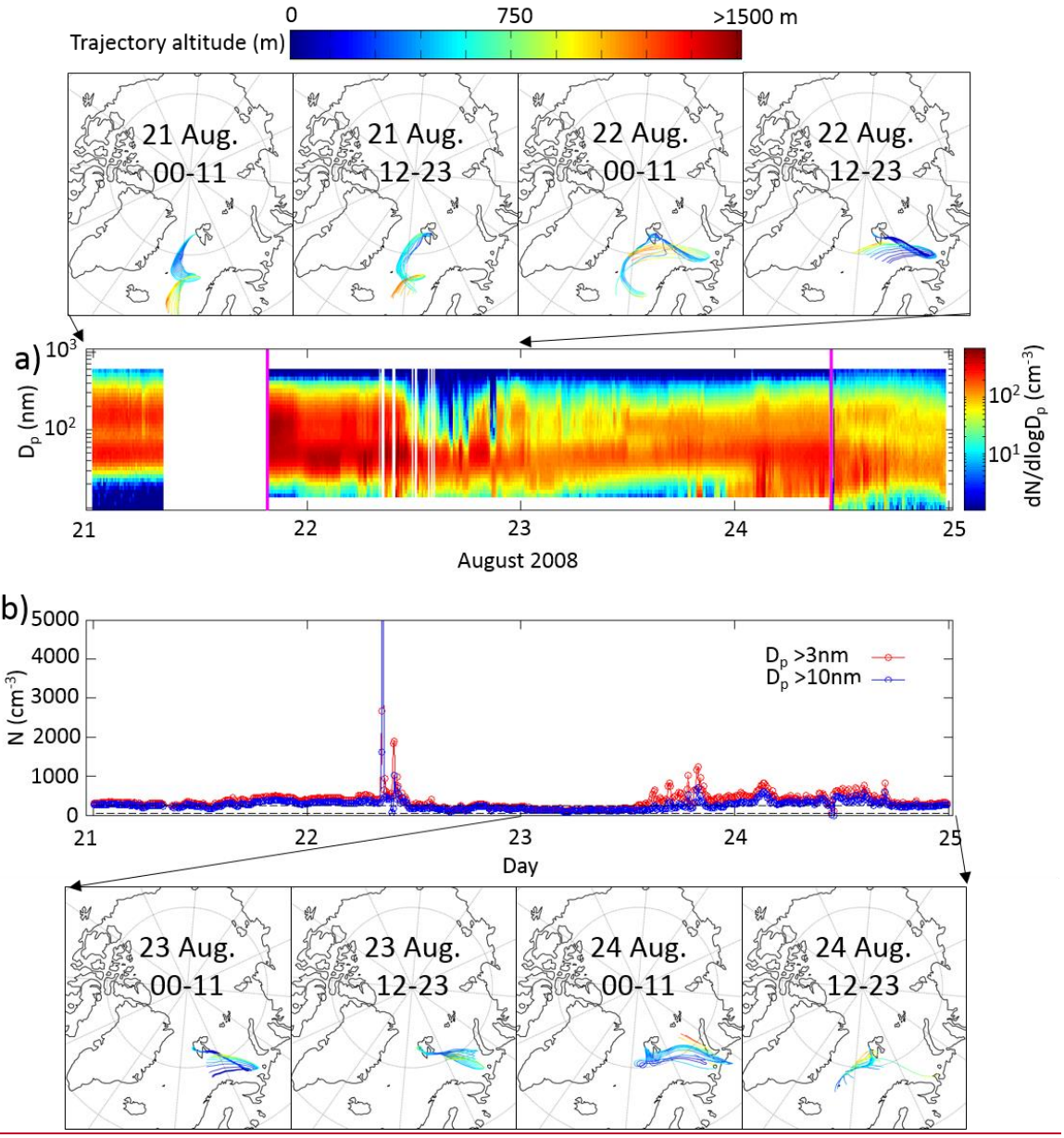
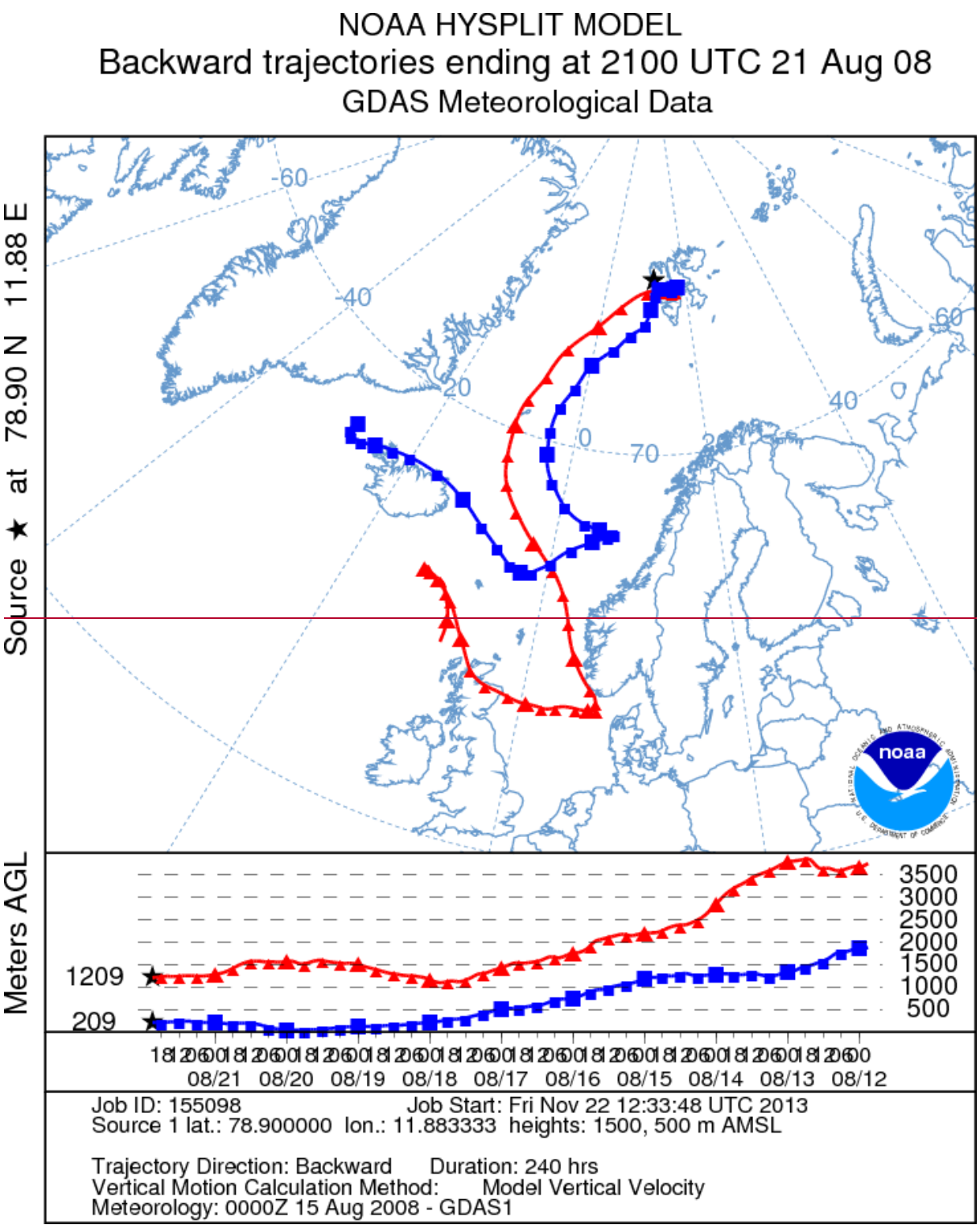


Figure ~~7~~4. a) Particle number size concentration measured before, during, and after the size-resolved CCN concentration measurements were conducted in August 2008. Purple vertical lines indicate the start and end time of the CCN size-resolved concentration measurements. b) Time series of the 8-min medians from CPC measurements for the same period in August 2008. Horizontal dashed lines represent the 25th and 75th percentile of the CN number concentration for August during the years 2001 to 2010. Trajectory plots show 5-day backward trajectories, calculated for every hour. Trajectory plots on top of panel a) show air masses arriving between the 21 and 23 August 2008 at Zeppelin Research Station. Trajectory

plots below panel b) show air masses arriving between midnight of the 23 August to midnight of the 24 August at Zeppelin Research Station.


NOAA HYSPLIT MODEL
Backward trajectories ending at 2100 UTC 21 Aug 08
GDAS Meteorological Data
Source ★ at 78.90 N 11.88 E
-60
-40
-20
0
20
40
60
70
noaa
Meters AGL
3500
3000
2500
2000
1500
1000
500
1209
209
08/21 08/20 08/19 08/18 08/17 08/16 08/15 08/14 08/13 08/12
Job ID: 155098 Job Start: Fri Nov 22 12:33:48 UTC 2013
Source 1 lat.: 78.900000 lon.: 11.883333 heights: 1500, 500 m AMSL
Trajectory Direction: Backward Duration: 240 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 15 Aug 2008 - GDAS1

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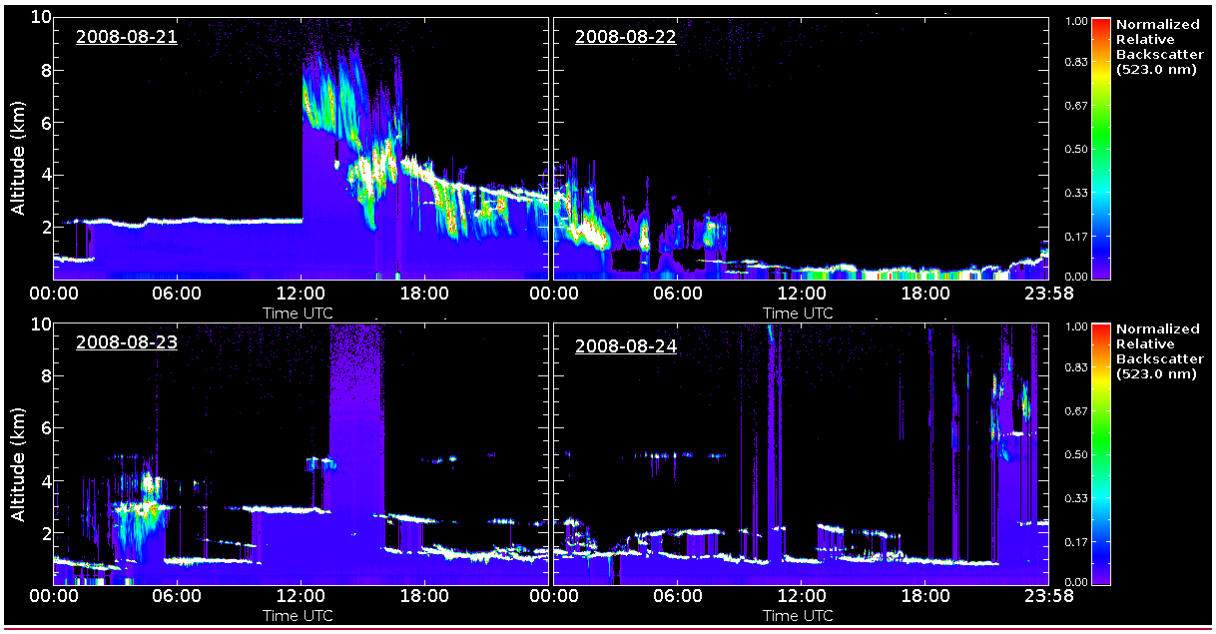
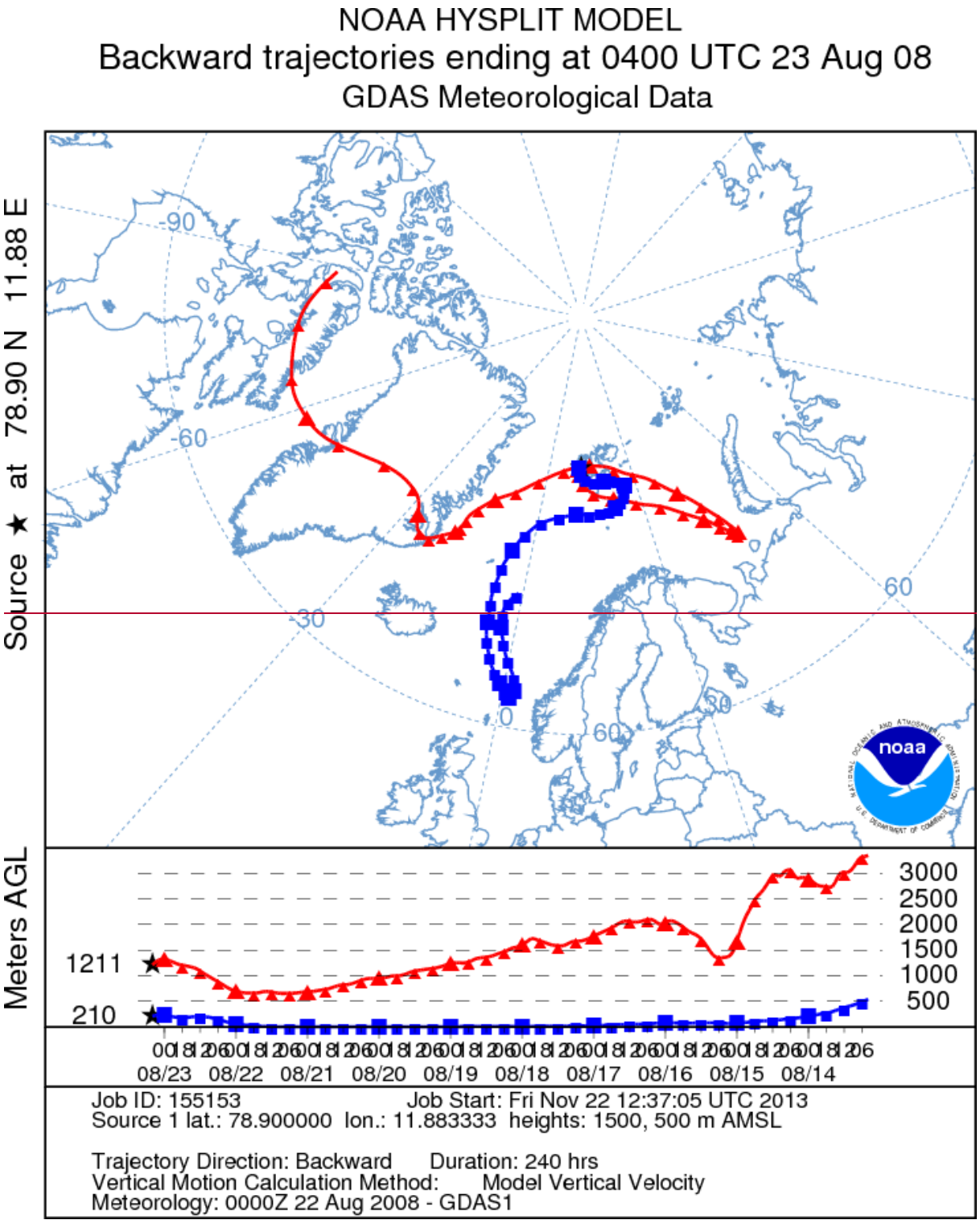
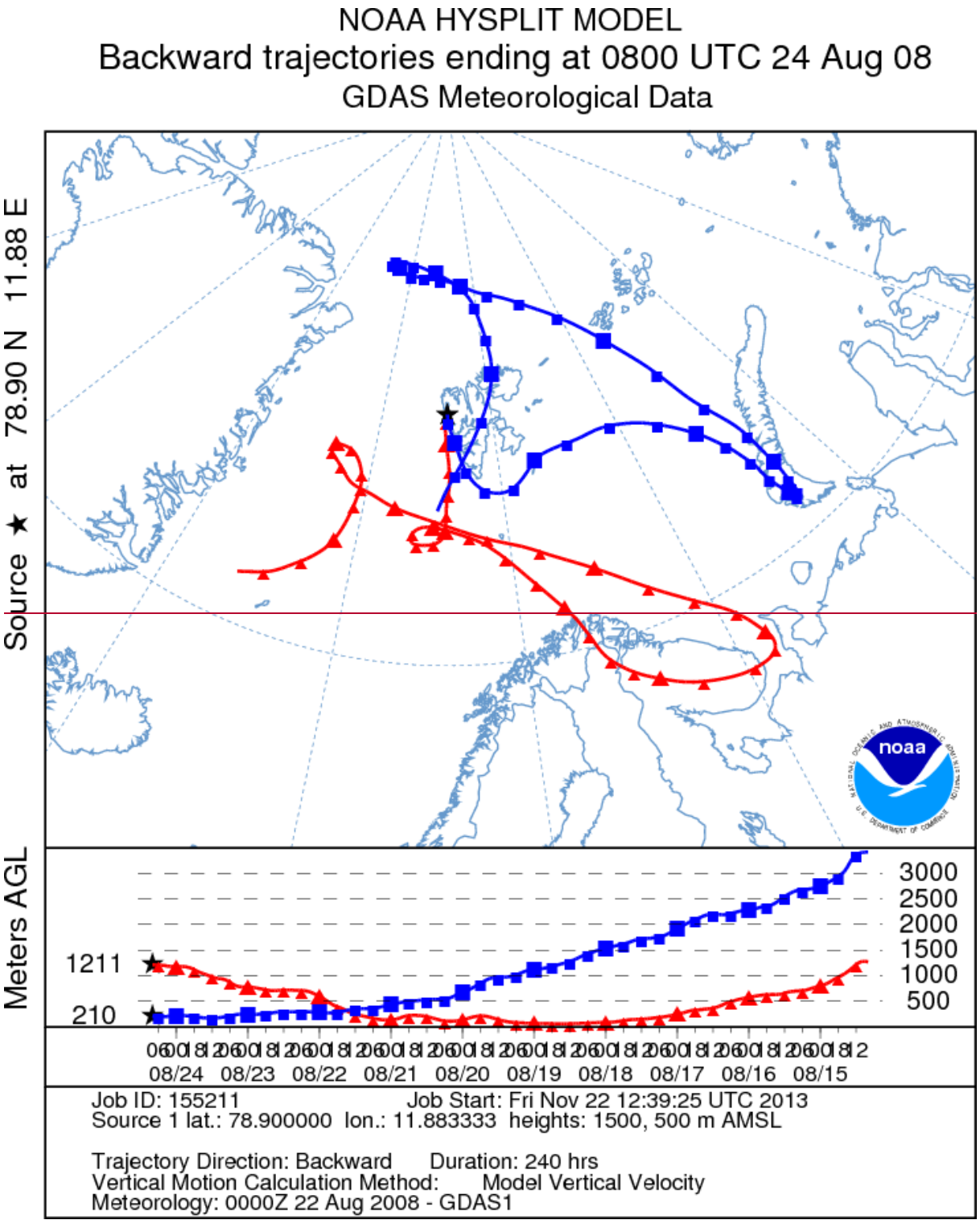


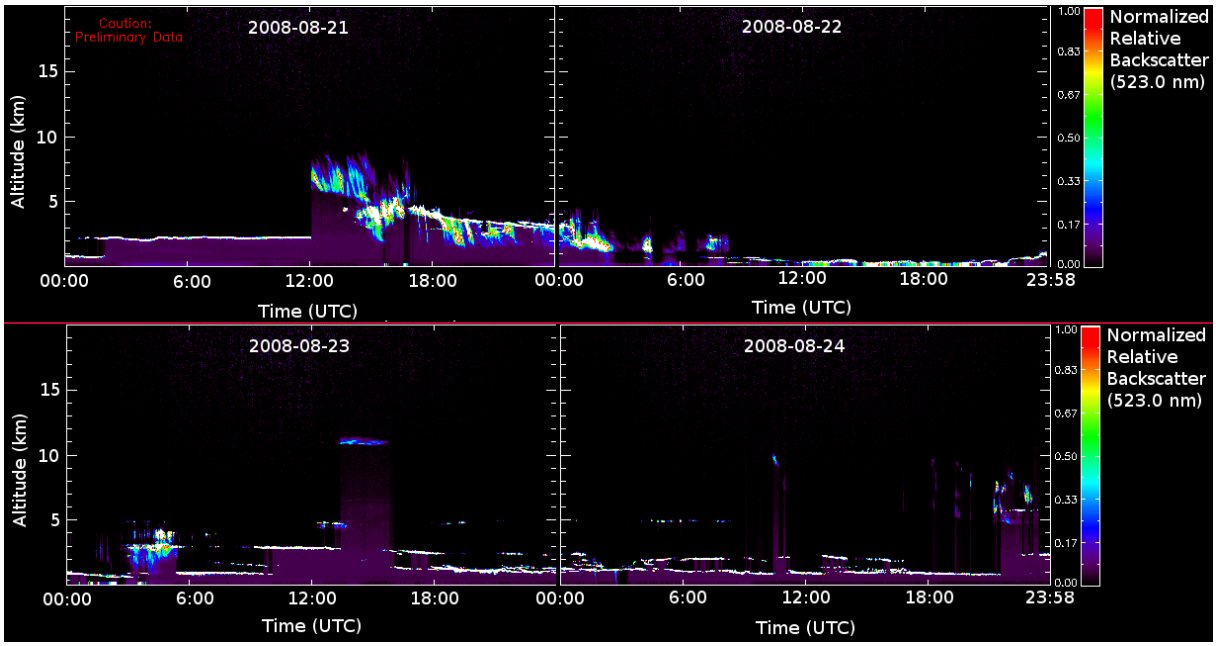
Figure ~~8. 10-day backward trajectory reaching Zeppelin research station, 21 August 2008 at 9:00 pm~~



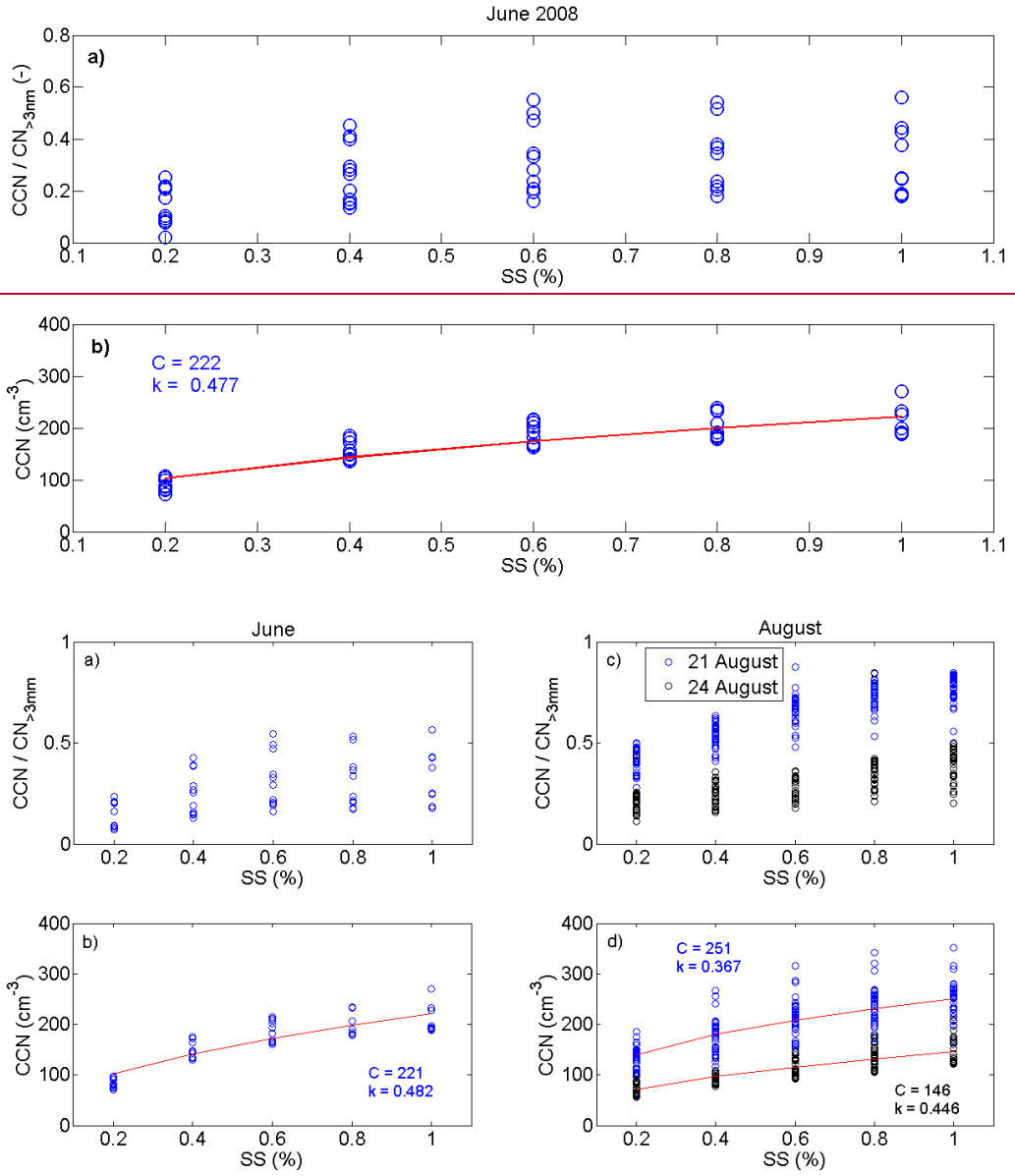
~~Figure 9. 10-day backward trajectory reaching Zeppelin research station, 23 August 2008 at 4:00 am~~



~~Figure 10. 10-day backward trajectory reaching Zeppelin research station, 24 August 2008 at 8:00 am~~



~~Figure 11~~5. Normalized relative backscatter (Level 1.0 data) based on Lidar measurements at Ny-Ålesund recorded during the period ~~21st–24th~~21–24 August 2008 (modified from http://mplnet.gsfc.nasa.gov/)



977

978

979 Figure ~~12~~6. a) Ratios of the medians for each SS scan between CCN and particles with

980 diameters > ~~3 nm~~3nm ($CN_{~~>3nm~~}$) for June 2008 ~~and~~ as a function of SS. b) ~~Total~~Medians

981 for each SS scan of the total numbers of CCN as a function of SS~~. The red curve represents a~~

982 ~~power law function fit to the data with the coefficients C and k.~~

983

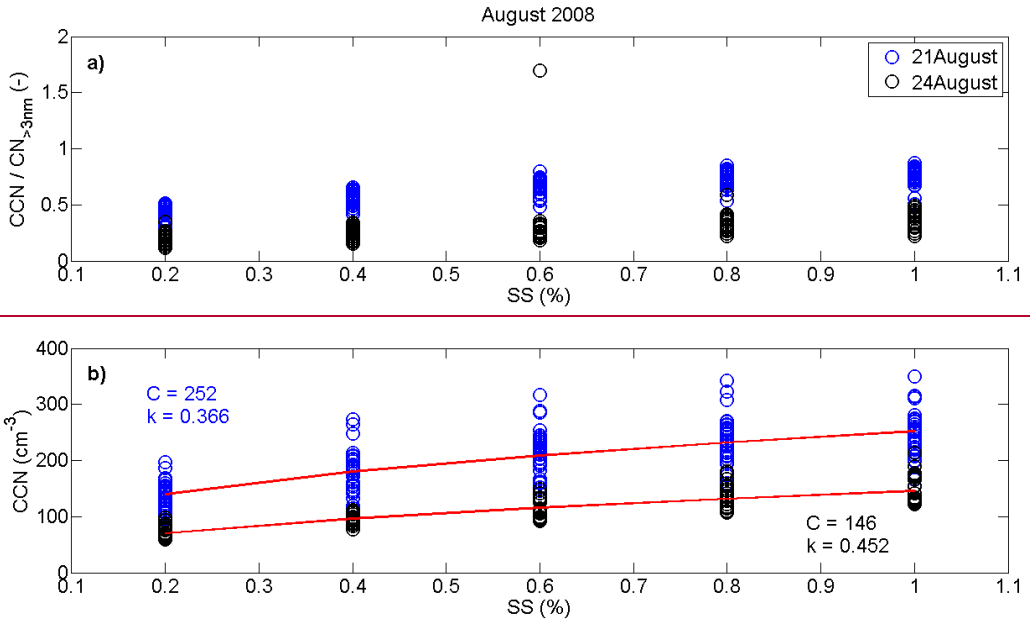


Figure 13. a for June 2008. c) Ratios of the medians for each SS scan between CCN and particles with diameters > 3 nm3nm ($CN_{>3nm}$) for 21st21 and 24th24 August 2008 as a function of SS. b) Totald) Medians for each SS scan of the total numbers of CCN for 21st and 24th August 2008 as a function of SS. for 21 and 24 August 2008. The red curves represent power-law functions fitfunction fits to the CCN data of 21st and 24th August, respectively. The resultingwith the coefficients *C* and *k* are presented in addition.

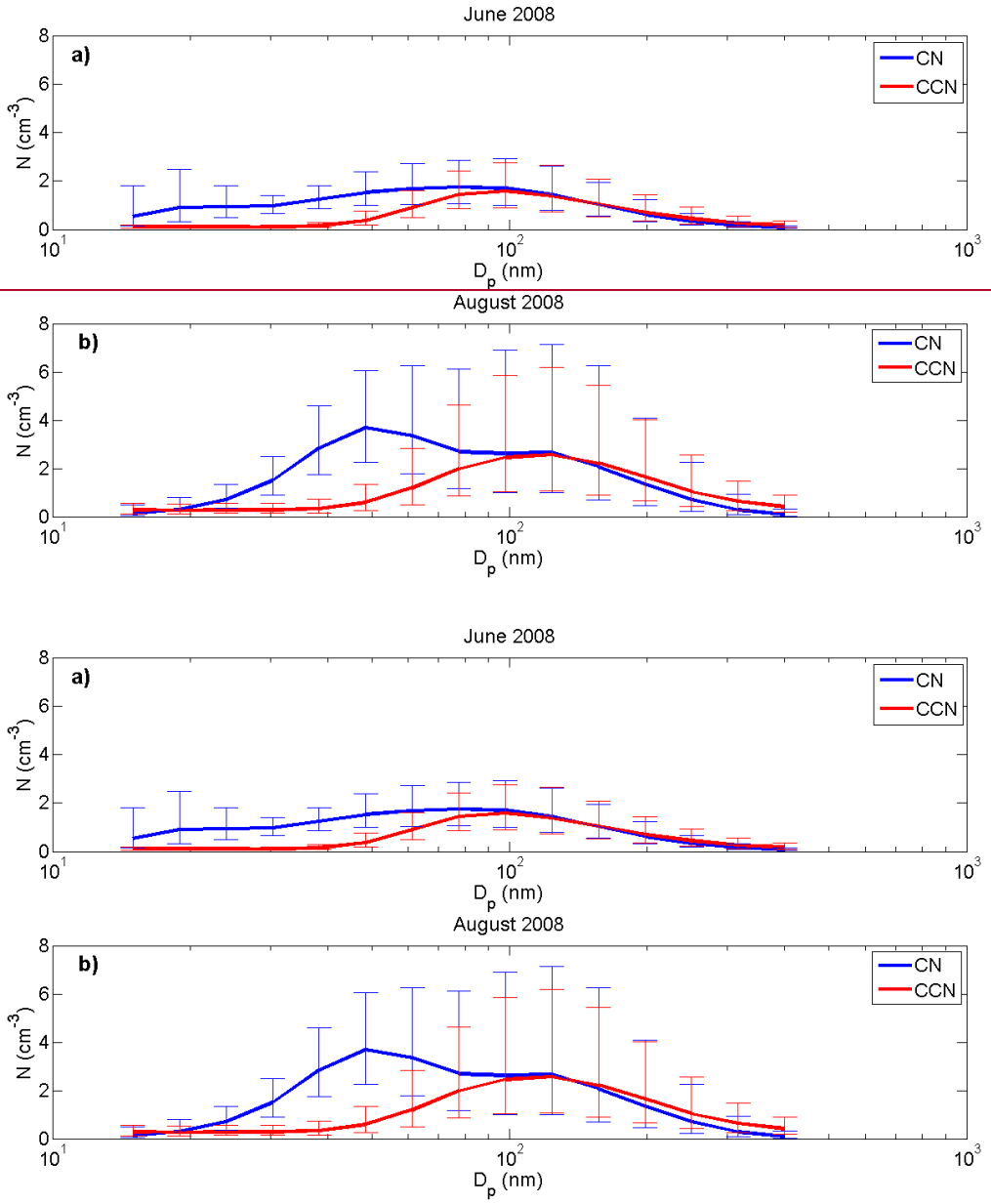
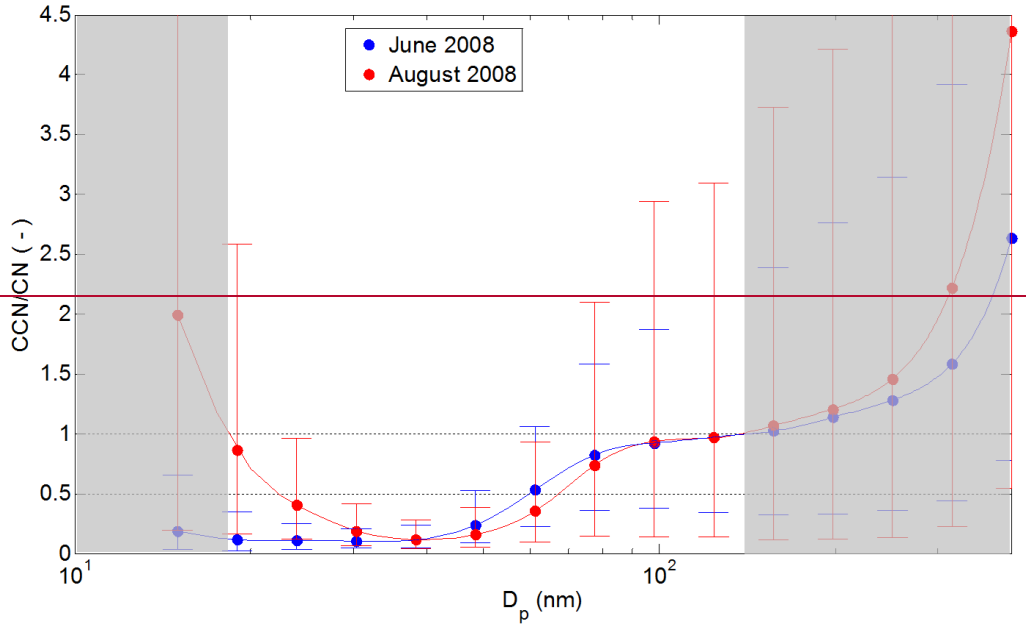


Figure ~~14. Arithmetic mean~~7. Geometric means of size-resolved particle density measurements and resulting CCN concentrations for the measurement period in a) June 2008 and b) August 2008. Measurements were conducted at 0.4% SS. Error bars indicate the geometric standard deviation.



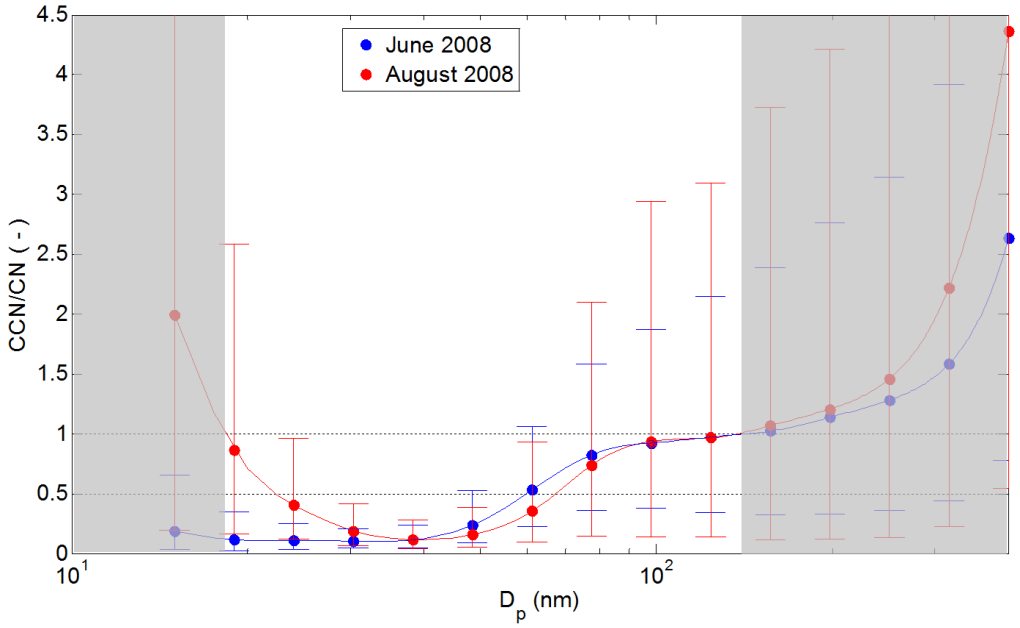


Figure ~~158~~. Activation ratio as a function of dry particle diameter (D_p) for the measurement period in June 2008 and August 2008. Obtained from measurements at a SS of 0.4%. Error bars indicate ~~standard deviations.~~ SD. The grey area indicates the for further analysis omitted data.