



Size-resolved cloud condensation nuclei concentration measurements in the Arctic

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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 Zepelin 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 mid-latitude air masses. CCN supersaturation (SS) spectra obtained on the 27 June, before size-resolved measurements were begun, and spectra from the 21 and 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 in the bulk aerosol samples.

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1 Introduction

The Arctic represents a region of special interest for atmospheric research due to the following reasons: (i) very sensitive to changes in radiative forcing owing to an active feedback mechanism, (ii) expecting greater anthropogenic activity from increased ship-
ping 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 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. R. H. Moore et al. (2011) 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 mea-
surements from ships and followed by aircraft 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 some tens per cm^{-3} .

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Silvergren et al. (2014) presented chemical and physical properties of aerosols measured 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 were determined to be highest in October, which was noted as the month with the most favourable cloud forming potential.

Bigg and Leck (2001) reported the results from CCN measurements conducted on an icebreaker at latitudes higher than 80° N, from 15 July to 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

$15\text{--}50 \text{ cm}^{-3}$, 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 air was being transported from the open ocean to the pack ice. The authors suggested that this occurred as a result of cloud formation. 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 $14.01 \pm 10.96 \text{ cm}^{-3}$, which increased up to $46.99 \pm 37.43 \text{ cm}^{-3}$ 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

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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.

5 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, and an increase in CCN concentration, approximately from 100 to 400 cm⁻³, was recorded to accompany an increase in altitude, from 1.75 to 4 km. More-
 10 over, 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 kma.s.l., 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
 15 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
 20 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
 25 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

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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 and 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 $76 \pm 29 \text{ cm}^{-3}$ 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 $250 \pm 41 \text{ cm}^{-3}$. 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 $N = C \cdot \text{SS}^k$, 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.

R. H. Moore et al. (2011) 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

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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.

Latham et al. (2013) presented results of CCN measurements conducted during research flights from 26 June to 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.

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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 the dominant processes causing this increase. 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 and 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 a.s.l.), 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.

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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 s, followed by a lag time of 5 s 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 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.

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 s 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 a.m. on 27 June to around 10.15 a.m. on 29 June, and 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. 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 29 June. However, during the study in August, CCN spectra were determined for approximately 17 h on 21 August, before the CCN size-resolved experiment began, and for approximately 13 h on 24 August after the experiment ended. In addition, 10 day backward trajectories were calculated using the online web version of the NOAA HYS-PLIT Model (Draxler and Rolph, 2013; Rolph, 2013), 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 27 to 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 27 June, when particles with diameters of approximately 70 nm dominate the particle concentration, (ii) from midnight to approximately midday of 28 June, when particle number concentrations are highest for particle diameters of approximately 20 nm and (iii) from approximately midday on the 29 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 (Figs. 3–5). All the air masses reaching Zeppelin station on the 27, 28 and 29 June 2008 have a clear Arctic origin. Air masses reaching the station at 8 a.m. on 27 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 28 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 29 and

30 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.

In addition to the trajectory analyses, Lidar measurements from Ny-Ålesund, part of the Micro Pulse Lidar Network (<http://mplnet.gsfc.nasa.gov/>, accessed 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 27 June 2008 in Ny-Ålesund showed only high clouds (altitude > 5 km), from approximately 9 a.m. Before 9 a.m., cloud-free conditions predominated at the measurement site. High clouds and cloud-free conditions alternated during 28 and 29 June 2008; therefore, Zeppelin station can be regarded as cloud-free during this time (cf. Fig. 6).

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 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 27 to 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), which show transport only within the Arctic. In addition, the Lidar data from the period from

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27 to 30 June 2008 does not indicate any cloud processing of the aerosols in the lower atmosphere boundary layer at the measurement site.

During the early morning hours of 28 June 2008 it is likely that new particle formation took place. A sharp increase in total particle number concentration is observed (Fig. 2b), and the highest concentration of particle numbers is found for particles with dry diameters of less than 20 nm (Fig. 2a). 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 18 April to 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 predominantly begin to increase at around noon; this is in contrast with our observations, where the concentrations start to increase at around midnight. 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. Fig. 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 %.

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From midday on 29 June 2008 until approximately 10.30 p.m. on that day, the total particle number concentrations of particles with diameters greater than 3 nm increased approximately from 400 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 that the air mass originated at the Alaskan coast (Fig. 5). The reasons for such high number densities are unclear, but it is possible that the high number densities indicate particle formation occurring over the Arctic basin some days before arriving at Ny-Ålesund.

To place the period in which these 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 27 to 30 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, the median total particle number concentration is around 40 % higher than the averaged June data from 2001 to 2010. This can be partly explained by the different size ranges that are included in the calculation.

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Particle number size distributions from 21 to 25 August 2008 are presented in Fig. 7a. 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 a.m. to 7.30 p.m. on 21 August and for short periods on 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). 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): (i) the final hours of 21 August, when particle number concentrations were highest for particles with diameters between 100 and 200 nm, (ii) the early morning hours of 23 August, when particle number concentrations were relatively low for all measured sizes (cf. Fig. 7b) and (iii) during the first half of 24 August, when total aerosol concentrations were relatively high for the period, but no particular size range clearly dominated. Calculated 10 day backward trajectories for the 21 August at 9 p.m. and for the 23 August at 4 a.m. indicate that the air masses originated at lower levels and from rather low latitudes (Figs. 8 and 9). However, the 10 day backward trajectory for the 24 August at 8 a.m. indicates the origin to once again be from higher latitudes (Fig. 10).

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

From around 8 to 11 p.m. on 21 August 2008, particles with dry diameters between 100 and 200 nm dominate the particle number size distribution. According to the Lidar measurements, the Zeppelin station was very likely unaffected by clouds, and the air masses were passing northwards, originating from mid-latitudes (Fig. 8). Therefore, it is likely that this peak in the particle number size distribution for particles with diameters between 100 and 200 nm is a result of particles being transported to the Arctic and the processes taking place during transport, rather than 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 23 August 2008, air masses arriving at Zeppelin station also originated in the mid-latitudes (Fig. 9), but resulted in relatively low total particle concentrations, compared to the concentrations observed between 8 and 11 p.m. on 21 August 2008. At 8 a.m. on 24 August, the air masses once again originated in the Arctic Ocean region (Fig. 10) and resulted in total particle number concentrations of approximately $330 \text{ particles cm}^{-3}$, for particle diameters greater than 10 nm, and approximately $500 \text{ particles cm}^{-3}$ for diameters greater than 3 nm (Fig. 7b). This observation, in addition to the Arctic origin of the air mass, suggest locally produced aerosols and is consistent with the observations of Arctic aerosol characteristics during summer stated by Tunved et al. (2013).

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 %

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higher total particle number concentration during the period in which the CCN size-resolved measurements were conducted, compared to the long-term average.

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 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 27 June 2008 was obtained before the size-resolved CCN measurements were begun. The data presented in this section comprise arithmetic means 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 ($CN_{>3\text{ nm}}$) for 27 June 2008 is shown in Fig. 12a. A significant increase in the ratio of CCN to $CN_{>3\text{ nm}}$ 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 27 June 2008 in Fig. 12b. 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, $N_{\text{CCN}}(\text{SS}) = C \cdot \text{SS}^k$, describing the number of CCN (N_{CCN}) with the coefficients C and k and SS, was fitted to the data shown in Fig. 12b and giving values for the coefficients of $C = 222$ and $k = 0.477$. Ranges in the parameters C and k depend on the type of air mass, and the values for 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 and 13 h observation periods on 21 and 24 August 2008, respectively, are shown in Fig. 13. The ratios between CCN and $CN_{>3\text{ nm}}$ as a function of SS are shown in Fig. 13a for the two different days. For 21 August, a significant increase in the CCN to $CN_{>3\text{ nm}}$ ratio with an increase in SS was observed in all cases. For 24 August, the increase in ratio was significant for all increases in SS, ex-

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cept for the increase from 0.4 to 0.6 % SS. The absolute number of CCN for 21 and 24 August, as a function of SS, is shown in Fig. 13b. 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 27 June 2008 (cf. Fig. 12b), a power-law function of the same form was fitted to the data from 21 and 24 August 2008, as denoted by the red lines in Fig. 13b. The fittings resulted in $C = 252$ and $C = 146$ and $k = 0.366$ and $k = 0.452$ for 21 and 24 August 2008, respectively.

Rogers and Yau (1996) demonstrated that the coefficients for maritime air vary, with $C = 30\text{--}300\text{ cm}^{-3}$ and $k = 0.3\text{--}1.0$, while for continental air the values vary between $C = 300\text{--}3000\text{ cm}^{-3}$ and $k = 0.2\text{--}2.0$. The coefficients C and k that are given by the fitted power-law function applied to the measurements during June and August 2008 (cf. Figs. 12b and 13b) are consistent with the ranges that Rogers and Yau (1996) proposed for maritime air masses. Pruppacher and 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 and 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 21 August 2008, and at the upper limit of that observed on 27 June and 24 August 2008 (cf. Figs. 12a and 13a). 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 $< 1.6\text{ km}$ 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. Figs. 12b and 13b). The average

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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 27 June, 21 and 24 August 2008.

Yum and 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 cm^{-3} , 232 and 131 cm^{-3} for 27 June, 21 and 24 August, respectively. For 27 June only high clouds (altitude > 5 km) were present, while for 21 and 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 and Hudson (2001) for non-cloudy conditions. The CCN/CN ratio calculated by Yum and Hudson (2001) is 0.65 at a SS of 0.8 %, which is higher than the average ratio determined for the CCN spectra during 27 June and 24 August 2008. The arithmetic means of the CCN/CN ratio at a SS of 0.8 % are 0.32, 0.74 and 0.34 for 27 June, 21 and 24 August 2008, respectively. Yum and 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 27 June, 21 and 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 and 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 at Zeppelin research station. For June 2008, a CCN number concentration of around $100 \text{ particles cm}^{-3}$ 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 and a SS of 0.4 %, which results in a CCN number concentration of $143 \text{ particles cm}^{-3}$. Silvergren et al. (2014) calculated a CCN number concentration of approximately $65 \text{ particles cm}^{-3}$ at 0.4 % SS for August 2008, which is

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also lower than the concentrations of 180 and 96 particles cm⁻³ calculated from the presented data in Fig. 12b for 21 and 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) lies between the two different spectra of August 2008 (Fig. 13b). Comparing backward trajectories (calculated in a similar way to those shown in Figs. 3–5 and in Figs. 8–10) arriving at Zeppelin on 27 June at 3.00 a.m., 21 August at 10.00 a.m. and 24 August at 7 p.m., corresponding to the times when the CCN spectra were measured, shows that the air masses' residence location for 27 June and 24 August is to the east of Svalbard, but to the south of Svalbard for 21 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. 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). The lower panel shows the correspondent data for the measurement period in August 2008 (Fig. 14b). 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. 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 higher SD during August compared to June. As the CN number concentration, the CCN concentration is characterized by

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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). 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. To obtain completeness, all data points are shown; however, the sizes where $CCN/CN \geq 1$ 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 a study that was conducted at the Finnish Pallas-Sodankylä Global Atmospheric Watch station that measured the ratio between cloud droplet number concentration and total particle concentration while the station was in clouds as a function of dry particle size. By comparing CCN concentrations at a fixed SS of 0.4 % with cloud droplet number concentrations, it was concluded that during the cloud events the “effective” maximal SS was likely to be approximately 0.4 % in most cases. During the five periods when the station was in clouds, D_{50} varied between 80 and 102 nm on average. A comparable study at the same measurement site resulted in D_{50} between 110 and 140 nm for maximal SS between 0.18 and 0.26 % (Anttila et al., 2009). Komppula et al. (2005) calculated D_{50} by comparing a particle number size distribution measured at a site in clouds with a nearby measured size distribution obtained at a station under cloud-free conditions. D_{50} was estimated to be 80 nm on

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average and varied between 50 and 128 nm. Unfortunately, the SS is unknown. Due to the uncertainty in SS, it is not possible to compare present study to the studies conducted at the Finnish stations directly. However, due to SS lower than 0.4 % reported by Anttila et al. (2012) and Anttila et al. (2009), larger activation diameters in these studies compared to this study are expected, which is in line with the presented results. Jaatinen et al. (2014) report activation diameters for measurements conducted during the same field campaign as reported in Anttila et al. (2012). However, compared with Anttila et al. (2012), in the present study, activation diameters were calculated differently and for a shorter period. The critical diameter was calculated by interpolating between the size bin at which the integrated particle number size distribution was equal to the amount of total measured CCN and the previous size bin. This resulted in a critical diameter of 98 ± 16 nm for 0.4 % SS (Jaatinen et al., 2014).

Besides SS, the chemical composition and mixing state determines the ability of particles to become activated to cloud droplets (Frosch et al., 2011; M. J. K. Moore et al., 2011; Ervens et al., 2010; Sullivan et al., 2009). Kreidenweis et al. (2005) summarize results of predicted and experimentally determined critical diameters of ammonium sulphate and sodium chloride particles. Predicted critical diameters for sodium chloride particles vary between 44.6 and 39.4 nm (Kreidenweis et al., 2005 and references therein) and the experimentally determined diameter for a SS of 0.4 % was reported to be 40 ± 6 nm (Corrigan and Novakov, 1999 in Kreidenweis et al., 2005). Ammonium sulphate particles had larger predicted activation diameters at SS of 0.4 %, i.e., from 62.6 to 49 nm (Kreidenweis et al., 2005 and references therein). Experimentally determined critical diameters of ammonium sulphate were 51 ± 8 and 59 ± 9 nm (Corrigan and Novakov, 1999 and Pradeep 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. Pradeep Kumar et al. (2003) experimentally determined the activation diameter of oxalic acid to be 65 nm at a SS of 0.40 %.

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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 ($D_{p, \text{act}}$) and SS derived from κ -Köhler theory (Asa-Awuku et al., 2010) was applied to the experimental CCN data:

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

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 kg m^{-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

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

where ε_i and κ_i are the volume fraction and hygroscopicity parameter of each component i , respectively (Petters and 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 PM₁₀ inlet. The inorganic fraction was determined after sampling the aerosol with an open face system without a PM₁₀ 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).

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 with diameters > 400 nm were also able to reach the filter; thus, the composition was 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

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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 km 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. 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

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measurements were conducted. A power-law function of the form $N_{\text{CCN}}(\text{SS}) = C \cdot \text{SS}^k$, 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 $C = 222$ and $k = 0.477$. Coefficients for August were $C = 252$ and $k = 0.366$ before the size-resolved measurements were conducted and $C = 146$ and $k = 0.452$ 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 $\text{CCN}/\text{CN} = 0.5$) 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 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.

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taining the Lidar measurements at Ny-Ålesund. The CRAICC project and the Bolin Center for Climate Research are as well acknowledged.

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

	Jun Activation diameter (nm)	κ	Aug Activation diameter (nm)	κ
$\text{CCN}/\text{CN} = 0.25$	49	0.7	56	0.5
$\text{CCN}/\text{CN} = 0.50$	60	0.4	67	0.3
$\text{CCN}/\text{CN} = 0.75$	72	0.2	78	0.2

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

Component	Mass fraction (%) Jun	Mass fraction (%) Aug	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

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Table 3. Total κ values calculated based on the chemical and physical properties presented in Table 2.

	Jun	Aug
Total κ	0.5	0.5

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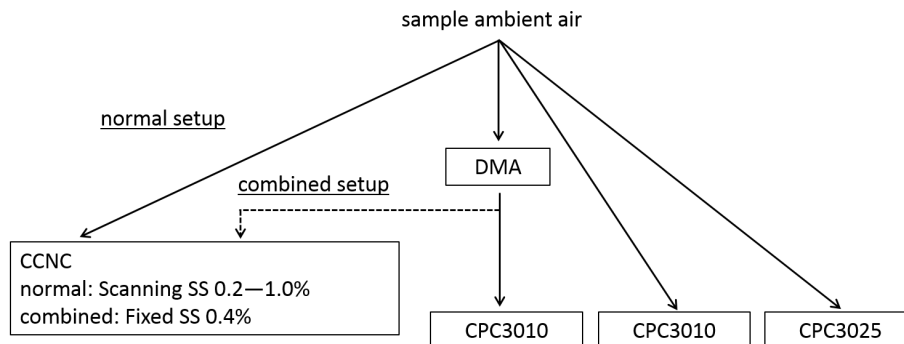


Figure 1. Scheme of the two different measurement modes for the cloud condensation nuclei counter (CCNC). When CCN size-resolved number concentration measurements took place, the CCNC was connected behind the Differential Mobility Analyzer and the supersaturation was set to 0.4 %. During normal operation,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

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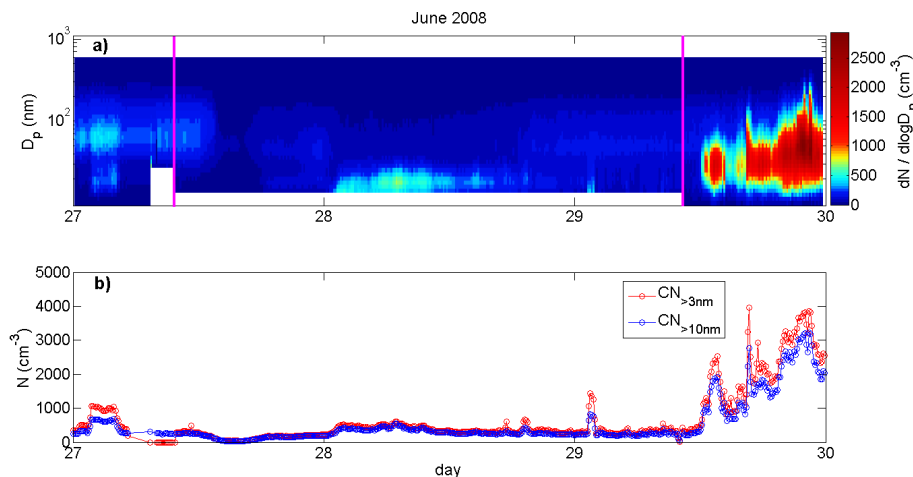


Figure 2. (a) Particle number size concentration measured before, during, and after the size-resolved CCN concentration measurements were conducted in June 2008. Purple vertical lines indicate the start and end time of the CCN size-resolved concentration measurements. (b) Time series of the 8 min medians from CPC measurements for the same period in June 2008.

NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 27 Jun 08
GDAS Meteorological Data

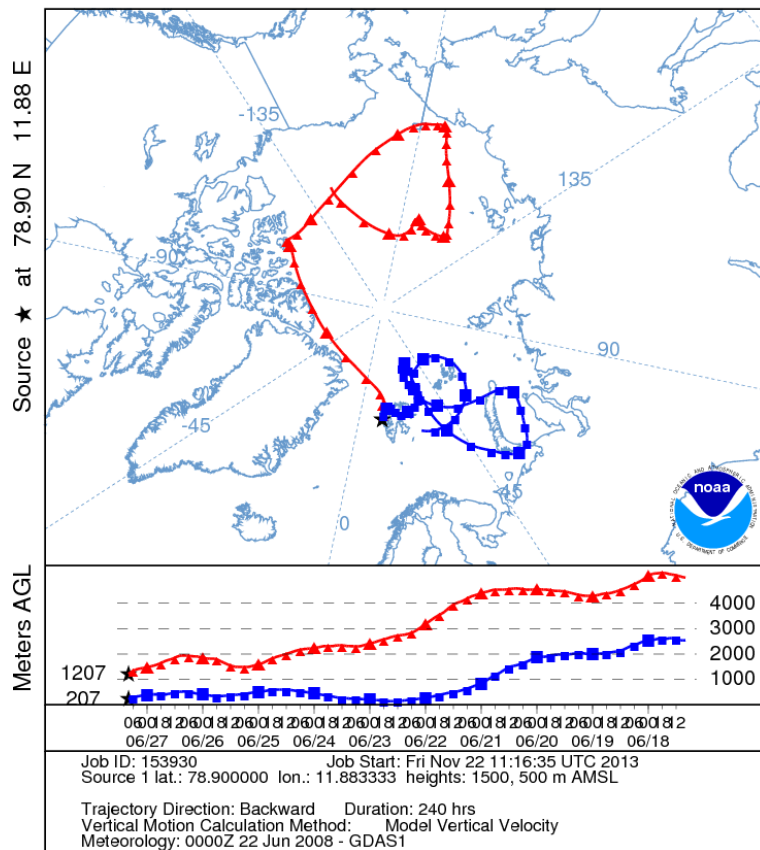


Figure 3. 10 day backward trajectory reaching Zeppelin research station, 27 June 2008 at 8.00 a.m.

NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 28 Jun 08
GDAS Meteorological Data

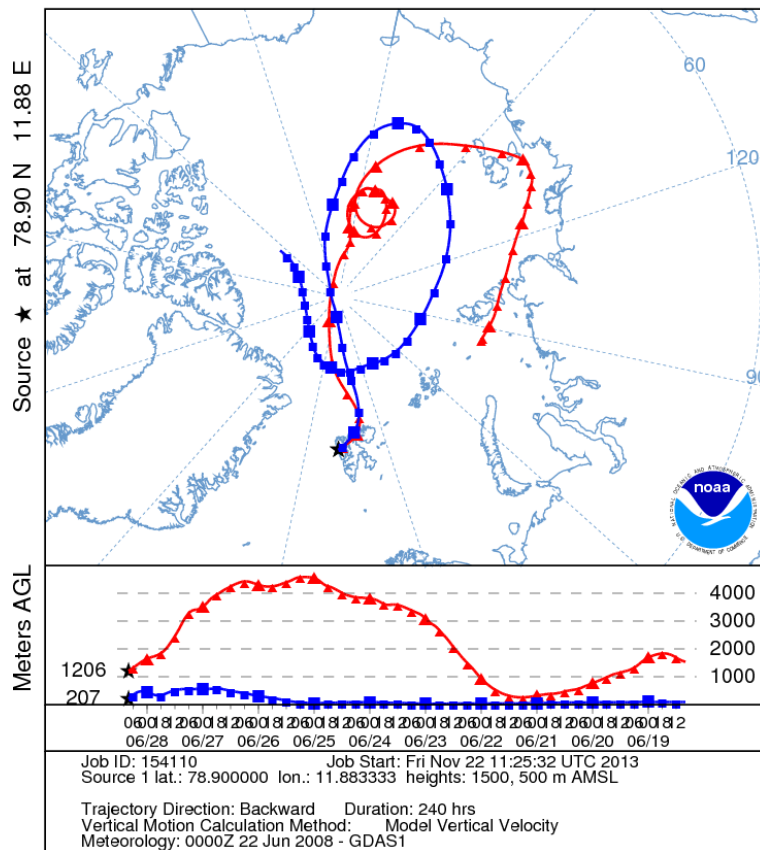


Figure 4. 10 day backward trajectory reaching Zeppelin research station, 28 June 2008 at 8.00 a.m.

NOAA HYSPLIT MODEL
Backward trajectories ending at 2200 UTC 29 Jun 08
GDAS Meteorological Data

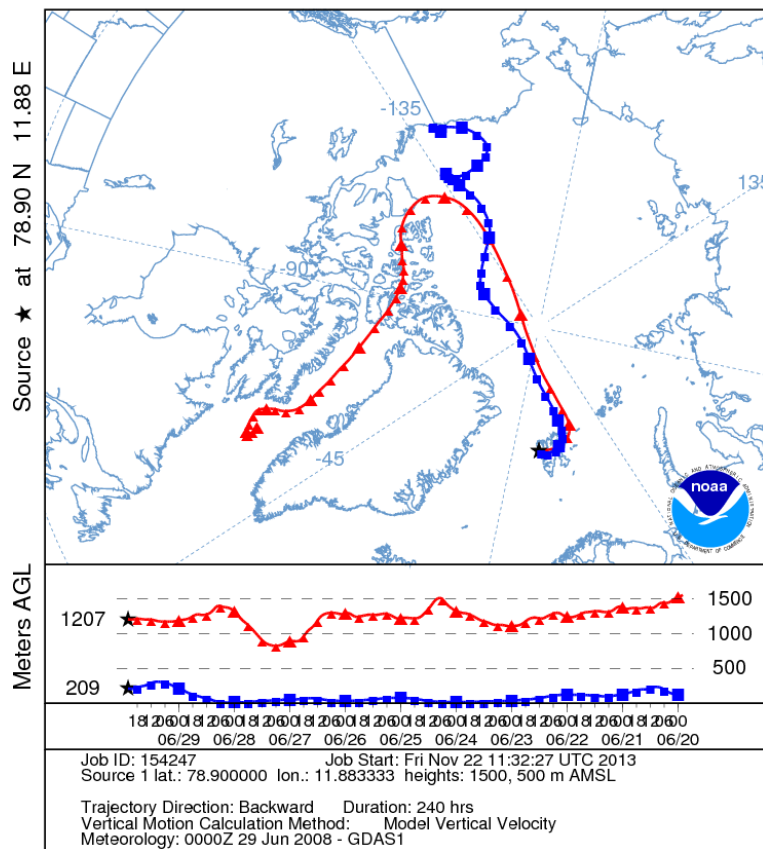


Figure 5. 10 day backward trajectory reaching Zeppelin research station, 29 June 2008 at 10.00 p.m.

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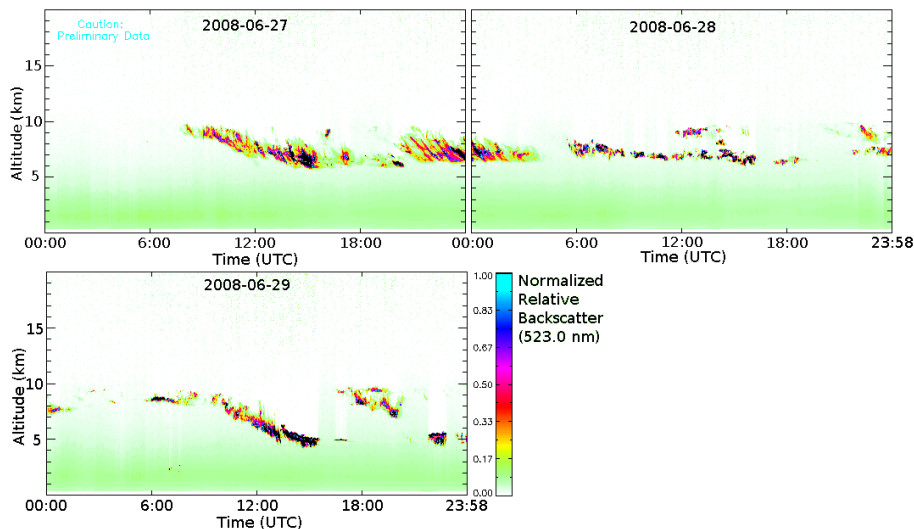


Figure 6. Normalized relative backscatter based on Lidar measurements at Ny-Ålesund recorded during the period 27–29 June 2008 (modified from <http://mplnet.gsfc.nasa.gov/>).

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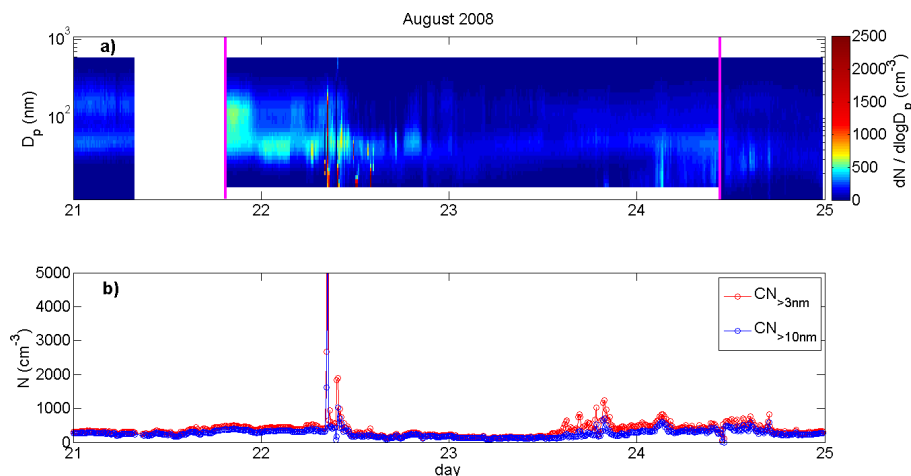


Figure 7. (a) Particle number size concentration measured before, during, and after the size-resolved CCN concentration measurements were conducted in August 2008. Purple vertical lines indicate the start and end time of the CCN size-resolved concentration measurements. (b) Time series of the 8 min medians from CPC measurements for the same period in August 2008.

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Figure 8. 10 day backward trajectory reaching Zeppelin research station, 21 August 2008 at 9.00 p.m.

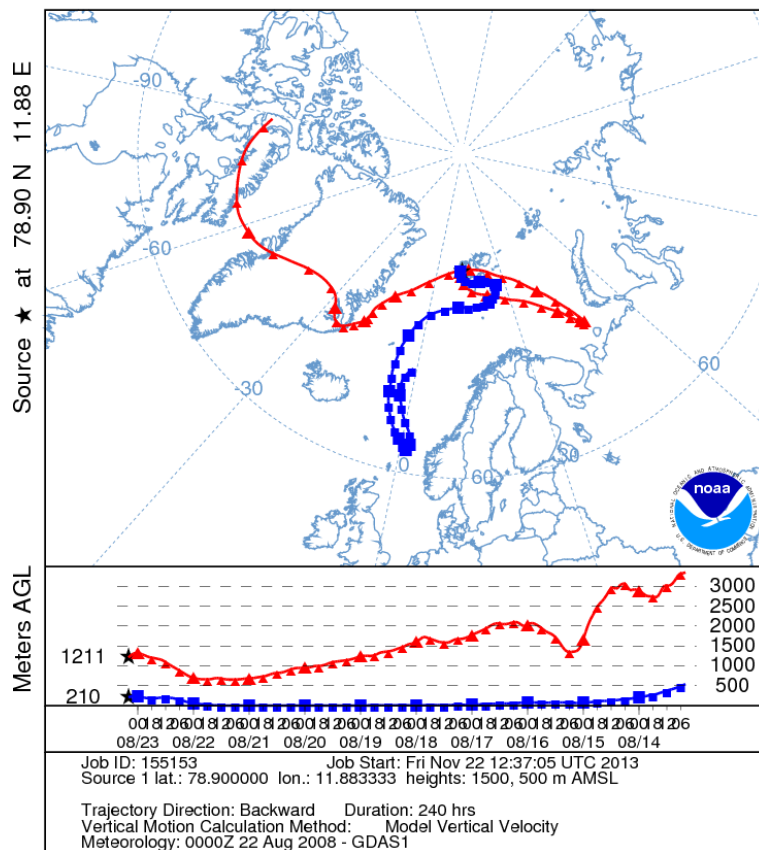


Figure 9. 10 day backward trajectory reaching Zeppelin research station, 23 August 2008 at 4.00 a.m.

NOAA HYSPLIT MODEL
Backward trajectories ending at 0800 UTC 24 Aug 08
GDAS Meteorological Data

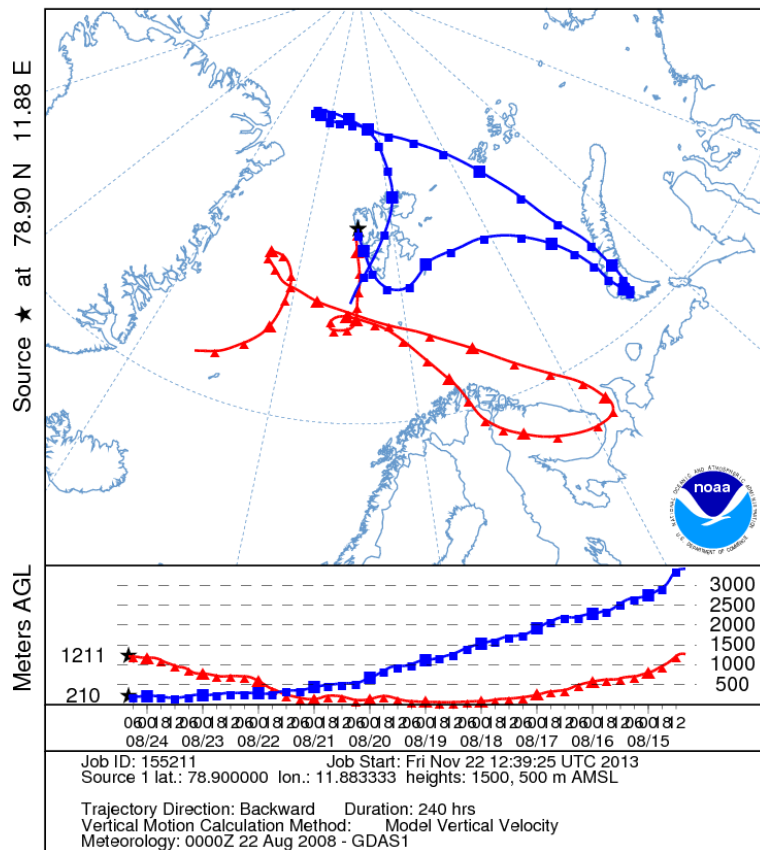


Figure 10. 10 day backward trajectory reaching Zeppelin research station, 24 August 2008 at 8.00 a.m.

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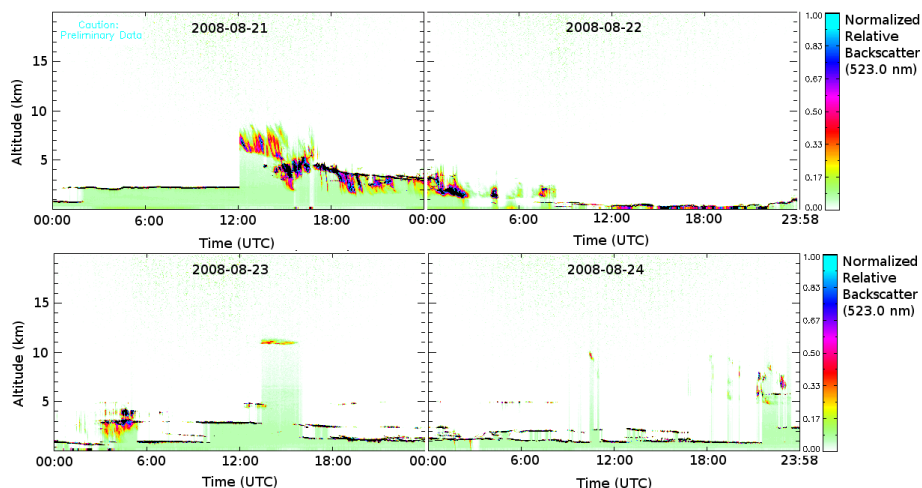


Figure 11. Normalized relative backscatter based on Lidar measurements at Ny-Ålesund recorded during the period 21–24 August 2008 (modified from <http://mplnet.gsfc.nasa.gov/>).

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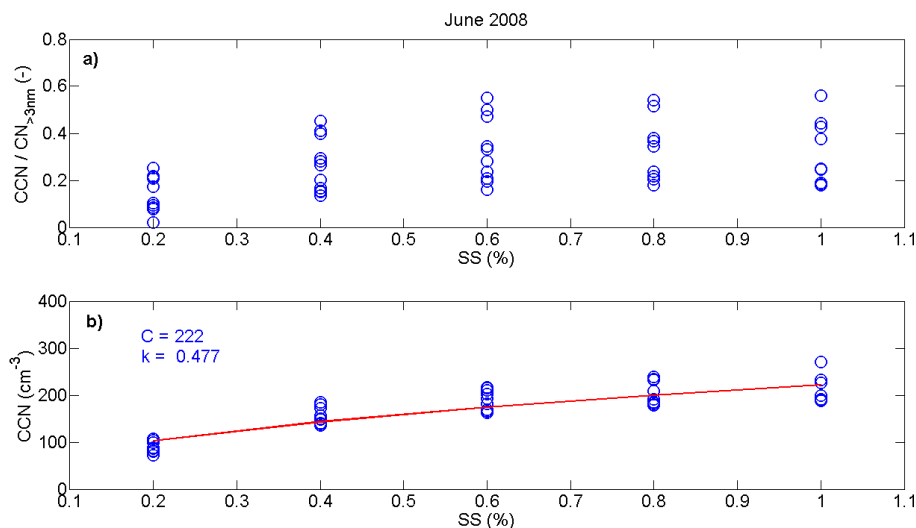


Figure 12. (a) Ratios between CCN and particles with diameters > 3 nm ($CN_{>3nm}$) for June 2008 and as a function of SS . (b) Total numbers of CCN as a function of SS . The red curve represents a power-law function fit to the data with the coefficients C and k .

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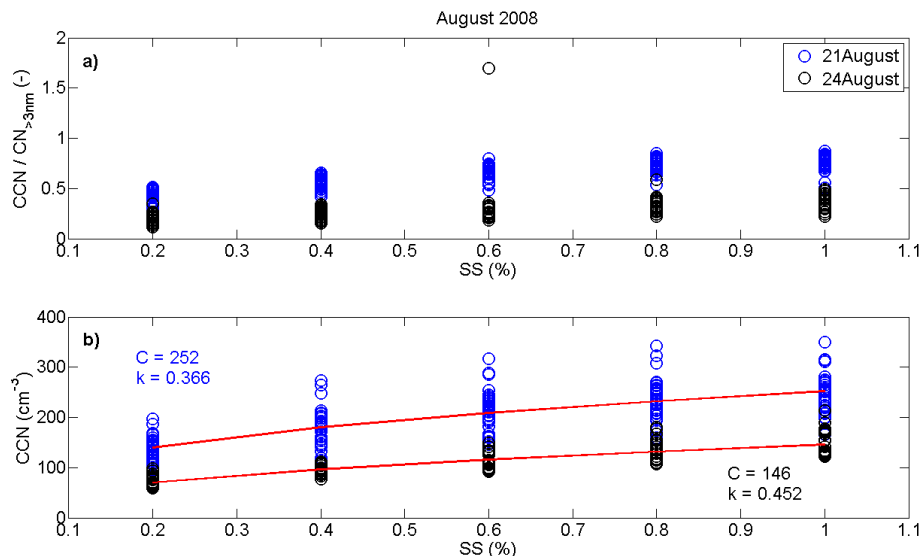


Figure 13. (a) Ratios between CCN and particles with diameters > 3 nm ($CN_{>3nm}$) for 21 and 24 August 2008 as a function of SS. **(b)** Total numbers of CCN for 21 and 24 August 2008 as a function of SS. The red curves represent power-law functions fit to the CCN data of 21 and 24 August, respectively. The resulting coefficients C and k are presented in addition.

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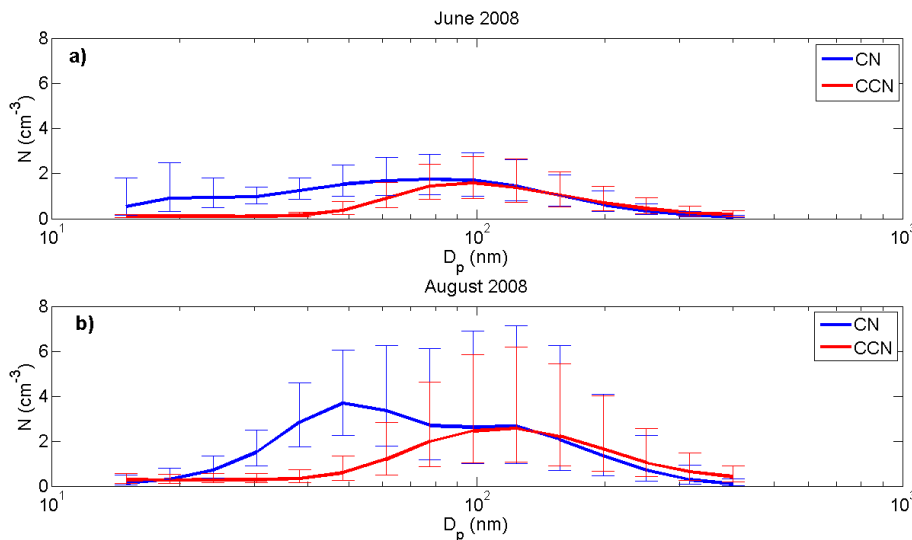


Figure 14. Arithmetic mean of size-resolved particle density measurements and resulting CCN concentrations for the measurement period in (a) June 2008 and (b) August 2008. Measurements were conducted at 0.4 % SS. Error bars indicate SD.

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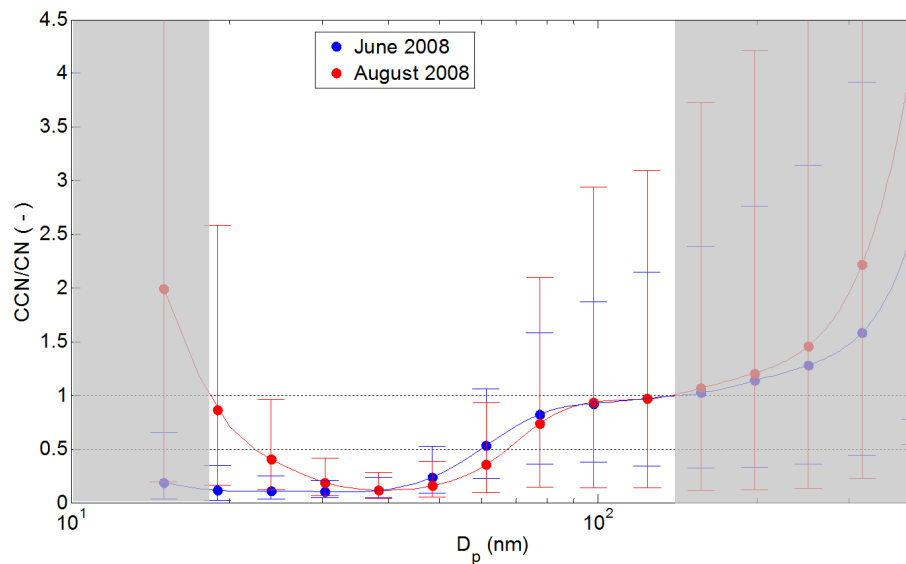


Figure 15. Activation ratio as a function of dry particle diameter (D_p) for the measurement period in June 2008 and August 2008. Error bars indicate SD.