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Discussions

New particle formation in the Svalbard region 2006 - 2015

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Abstract

Events of new particle formation, (NPF), were analyzed in a ten-year data set of hourly particle size distributions recorded on Mt. Zeppelin, Spitsbergen, Svalbard. Three different types of NPF-events were identified through objective search algorithms. The first and simplest algorithm utilizes short-term increases in particle concentrations below 25 nm, (PCT-events). The second one builds on the growth of the sub-50 nm diameter-median, (DGR-events), and is most closely related to the classical "banana-type" of events. The third and most complex, so-called multiple-size approach to identifying NPF-events builds on a hypothesis suggesting the concurrent production of polymer gel particles at several sizes below about 60 nm, (MEV-events).

As a first and general conclusion we can state that NPF-events are a summer phenomenon and not related to Arctic haze, which is a late winter-to-early spring event. NPF-events appear to be somewhat sensitive to the available data on precipitation. The seasonal distribution of solar flux suggests some photochemical control that may affect marine biological processes generating particle precursors and/or atmospheric photochemical processes that generate condensable vapors from precursor gases. Whereas the seasonal distribution of the biogenic methanesulfonate, (MSA), follows that of the solar flux it peaks before the maxima in NPF-occurrence.

A host of ancillary data and findings point to varying and rather complex marine biological source processes. The potential source regions for all types of new particle formation appear to be restricted to the marginal ice and open water areas between Northeastern Greenland and Eastern Svalbard. Depending on conditions yet to be clarified new particle formation may become visible as short bursts of particles around 20 nm, (PCT-events), longer events involving condensation growth, (DGR-events), or extended events with elevated concentrations of particles at several sizes below 100 nm, (MEV-events). The seasonal

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distribution of NPF-events peaks later than that of MSA and, DGR and in particular of MEV-events reach into late summer and early fall with much open, warm, and biologically active waters around Svalbard. Consequently, a simple model to describe the seasonal distribution of the total number of NPF-events can be based on solar flux, and sea surface temperature, representing environmental conditions for marine biological activity, and condensation sink, controlling the balance between new particle nucleation and their condensational growth. Based on the sparse knowledge about the seasonal cycle of gel-forming marine microorganisms and their controlling factors we hypothesize that the seasonal distribution of DGR and more so MEV-events reflect the seasonal cycle of the gel-forming phytoplankton.

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1. Introduction

In the late 1970ies and early 1980ies the interest in the Arctic atmospheric aerosol widened from the well-identified winter phenomenon of Arctic haze (Rahn and Shaw, 1977; Heintzenberg and Leck, 1994) to summer conditions in this northernmost remote region. In the pristine Arctic summer air the so-called background aerosol (Junge, 1963) was expected to be most clearly visible, far away from the northern hemispheric anthropogenic emission centers at lower latitudes. Episodic and localized occurrences of high concentrations of ultrafine particles, (here defined as particles with diameters < 100 nm), in the summer Arctic were explained by rare import of polluted air from lower latitudes (Flyger and Heidam, 1978; Heintzenberg and Larssen, 1983) or hypothetical anthropogenic sources in the Arctic (Jaenicke and Schütz, 1982).

With the advent of sensitive condensation nuclei counters (Agarwal and Sem, 1980) and differential mobility analyzers (Knutson and Whitby, 1975a, b) more details became visible in the Arctic sub-micrometer aerosol. High numbers of ultrafine particles were observed in connection with fog passages (Lannefors et al., 1983) and chemical aerosol information indicated a regional – possibly biogenic – particle sources in the summer Arctic (Heintzenberg, 1989). The high molar ratios of methane sulfonate, (MSA), to non-sea salt sulfate, ($nssSO_4^{2-}$), of 0.28 in the Arctic summer aerosol found by Heintzenberg and Leck (1994) substantiated the biogenic source of the particles.

The establishment of long-term Arctic aerosol monitoring at the fringes of the pack ice in Alaska (e.g., Polissar et al., 1999), Canada (e.g., Norman et al., 1999; Willis et al., 2016), and on Spitsbergen (e.g., Tunved et al., 2013) revealed more details of potential sources of the summer aerosol, in particular their connection to the marine biosphere in the Arctic. The unique series of systematic aerosol studies in the central Arctic north of 80°N onboard the Swedish icebreaker *Oden* in 1996 led to the formulation of a new hypothesis concerning a

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specific process of marine biogenic particle formation (Leck and Bigg, 1999). The marine biogenic particles involved behaved as polymer gels and originated in the surface microlayer (SML) of the ocean, (Orellana et al., 2011b), from the activity of sea-ice algae, phytoplankton and, perhaps, bacteria. The new particle events were reported to occur as simultaneous enhancement of particle number concentrations in the whole size-range below 50 nm, and not with the prototypical "banana growth" (Kulmala et al., 2004). Two more *Oden* cruises in 2001 and 2008 yielded results that were partly contradicting (Held et al., 2011b; Held et al., 2011a), partly supporting the SML hypothesis (Leck et al., 2013; Karl et al., 2013; Orellana et al., 2011b; Leck and Bigg, 2010). The synopsis of the results of four *Oden* cruises of Heintzenberg et al. (2015) identified geographic regions of new particle formation (NPF) in the inner Arctic while stressing the importance of recent open water and related biological activity in the sea in transects by air masses with new particle formation over the central Arctic.

Two years of aerosol size distributions from Mt. Zeppelin, Spitsbergen and Alert, both located at the fringes of the central pack ice, were analyzed by Croft et al. (2016a) with a global aerosol geophysics model. They discuss classical new-particle nucleation, coagulation scavenging in clouds, scavenging by precipitation, and transport in order to explain the annual cycle of the Arctic aerosol. Croft et al. (2016a) find two seasonal maxima in their modeled particle nucleation rates, one in March, and one in July. In spring, their simulated NPF occurs mainly in the free troposphere, whereas in summer, it occurs also in the planetary boundary layer. More recently, Croft et al. (2016b) state that ammonia from seabird-colony guano is a key factor contributing to bursts of newly formed particles, which are observed every summer in the near-surface atmosphere, at least at Alert, Nunavut, Canada. Earlier, the results of studies with another global aerosol model by Browse et al. (2014) suggested that the potential increase in NPF in the Arctic with potential increases in cloud condensation nuclei is compensated by wet scavenging. They also state that scavenging by pre-existing large particles suppresses NPF-events.

Based on three years of data from the two Arctic sites Thule and Ny-Ålesund (gruvebadet) Becagli et al. (2016) examined the sources and environmental factors controlling the biological aerosol component MSA. Their analysis included satellite-derived Chlorophyll-*a* (an indicator of phytoplankton biomass), oceanic phytoplankton primary productivity, (PPP), and sea ice. Whereas they found good correlations between MSA, PPP and sea ice, (the latter two being closely related), their data did not allow any statements on NPF processes.

To date the longest record of sub-micrometer number-size distributions of the Arctic aerosol down to 5 nm particle diameter and below has been accumulated on Mt. Zeppelin, Spitsbergen (Tunved et al., 2013; Heintzenberg and Leck, 1994). For the ten years from 2006 through 2015 a total of 63936 quality-controlled hours of aerosol data are available, i.e. during 73% of all hours of the ten years. In the present study we exploit this formidable data set in a search for processes forming new particles. An important first step in this work was formulating completely objective criteria for the identification of events. In the relatively clean Arctic environment we do not expect the classical nucleation and growth events as frequently observed over the continents, (cf. Kulmala et al., 2004), to dominate. Thus, we refrained from applying the objective search algorithm formulated by Heintzenberg et al., (2007) for this "Banana-type" of events. Instead we formulated new objective search algorithms allowing several potential types of new particle formation events or formation processes. With a host of complementary atmospheric and surface physical, chemical, and biological information a large number of NPF-events identified with these algorithms will be analyzed in the following chapters.

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2. Database

The Mt. Zeppelin observatory

Situated at the top of Mt Zeppelin, Svalbard (78° 56'N, 11° 53'E), the Zeppelin observatory offers a unique possibility to study the characteristic features of Arctic atmospheric constituents such as trace gases and aerosol particles. At a height of 474 m a.s.l. the station is located near the top of the local planetary boundary layer and represents remote Arctic conditions. The closest source of pollution, the small community of Ny-Ålesund, is located ~2 km north of the station (http://www.esrl.noaa.gov/psd/iasoa/sites/default/files/stations/nyalesund/nyalesund_site.jpg). However, the elevation difference and typical wind patterns largely prevent pollution from nearby sources to reach the Zeppelin Observatory. The dominating wind pattern is east-southeast katabatic flow from Kongsvegen glacier or from northwesterly directions as channeled by the Kongsfjord (Beine et al., 2001; Heintzenberg et al., 1983). The station itself was initially established in 1991, and is owned by the Norwegian Polar Research Institute (NP). The Norwegian Institute for Air Research (NILU) is responsible for the coordination of the scientific program.

2.1 Physical aerosol data

After a period of continuous aerosol measurements by the Department of Meteorology, Stockholm University in the early 1990ties, (Heintzenberg and Leck, 1994), the Department of Analytical Chemistry and Environmental Science, Stockholm University, initialized observations of the aerosol number size distribution in mid-2000. Originally, the system consisted of a single differential mobility analyzer system, (DMPS), consisting of a medium-size Hauke-type differential mobility analyzer, (DMA), together with a TSI 3760 condensation particle counter, covering diameters between 20 and ~500nm. From 2006 on the particle size range was widened covering particle sizes between 10 and 790 nm. In 2005, the rain-cover over the inlet was replaced. Initially, the instrument inlet was of a PM10 type, removing particles or hydrometeors with diameters >10 µm from the sampled air stream. During a substantial renewal of the Stockholm University equipment in 2010-2011, both inlet and DMPS system were replaced.

Since then, the DMPS-system utilizes a custom-built twin DMA-setup comprising one Vienna-type medium DMA coupled to a TSI CPC 3010 covering sizes between 25-800 nm and a Vienna-type long DMA coupled with at TSI CPC 3772 covering sizes between 5-60 nm. The size distributions from the two systems are harmonized on a common size grid and then merged. Both systems use a closed-loop flow setup. The current inlet hat is of whole air type, complying with EUSAAR[1] standard for high altitude or Arctic sampling conditions. In the current setup, the inlet is operated with a flow of ca. 100 liters per minute, (lpm). Laminar flow conditions apply throughout the sampling lines. Outside of the station, the inlet temperature is kept above 0°C using active heating. Inside the station the temperature increases gradually to room temperature (maximum temperature reaches ca. 25 °C, but remains typically around 20°C). Relative humidity, (RH) and temperature are internally monitored and measurements are maintained at dry conditions with RH < 30%. The system is regularly checked with latex spheres and flow controls. The recorded data are manually screened and crosschecked with other available observation as in Tunved et al. (2013). If inconsistencies were found between the different datasets, further investigation was

[1] EUSAAR (European Supersites for Atmospheric Aerosol Research) is an EU-funded I3 (Integrated Infrastructures Initiatives) project carried out in the FP6 framework of the specific research and technological development gram "Structuring the European Research Area - Support for Research Infrastructures", (http://www.eusaar.net/).

performed to exclude data that were identified as affected by instrumental errors. Using the instrumental logbook, periods of local activity potentially influencing the sampling were also excluded from the dataset. During the years 2006 – 2010 no particles below ten nanometers in diameter were recorded. From 2011 on four more diameter bins down to 5 nm were included and a different diameter array was utilized. To allow for a synopsis of all years all size distributions were interpolated on the pre-2011 diameter array and all integrals of the size distribution over particle diameter were taken over the joint diameter range 10 to 631 nm. For the pre-2011 years the data at the four size channels below 10 nm were flagged as missing. However, whenever results cover the complete time series the resulting number concentrations in the four first channels covering the years 2011 – 2015 are carried along.

For the identification of NPF in terms of particle growth the parameter D50 in nanometer was calculated as the number median diameter of particles smaller than 50 nm but larger than 10 nm, i.e. 50% of all particles below that size are smaller than D50. Besides this parameter Table 1 lists nine integral particle parameters, which are utilized in the NPF-search approaches or in the interpretation of results. These aerosol parameters quantify total particle number, (NTO), and particle numbers in sub-ranges of the number size distribution such as N25, quantifying the total number of particles between ten and 25 nm.

Following the concept developed by Pirjola et al., (1999), and Kulmala et al., (2001) we calculated the condensation sink, (CS, s^{-1}), as a parameter with which the probability of new particle formation from the gas phase and the necessary amount of condensable vapor can be estimated. For this calculation we utilized from our database number size distributions, pressure and temperature.

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2.2 Chemical aerosol data

For the interpretation of NPF-events we employed chemical information derived from the analyses of high volume particle samples taken by the Norwegian Institute for Air research, (NILU). A high volume sampler (PM10) was used to collect samples for a quantitative determination of sodium, (Na^+) sulfate, (SO_4^{2-}) and MSA (CH_3SOO^-). The sampler collected material for analysis in one to three days. Blank samples were obtained by mounting the glass fiber filters at the sampling site with the same sampling period but without air passing through. Na^+ and SO_4^2 were analyzed by NILU and have been downloaded for the present study from the EBAS database (http://ebas.nilu.no), which list details about the sampling technique and the sampling protocol. To be able to apportion the measured sulfate to $nssSO_4^{2-}$ the observed concentrations of Na^+ were used as the reference element based on the assuming that all Na^+ is of marine origin, (Keene et al., 1986).

MSA was analyzed at the laboratory of the Department of Meteorology, Stockholm University. To allow for subsequent chemical determinations the ambient samples and blanks were carefully handled in a glove box (free from particles, sulfur dioxide and ammonia). At the time of the chemical analyses, still in the glove box, the substrates were extracted (in centrifuge tubes) with 60 cm^3 deionized water (Millipore Alpha-Q, conductivity 18 MΩcm). The extracts were thereafter analyzed for weak anions by chemically suppressed ion chromatography (IC, Dionex ICS-2000) using Dionex AG11/AS11 columns. In order to trap carbonates and other ionic contaminants a Dionex ATC-1 column was used before the injection valve. The injection volume was 50 μdm^3. Quality checks of the IC-analyses were performed with both internal and external reference samples (Das et al., 2011). The analytical detection limits obtained for the various ions, defined as twice the level of peak-to-peak instrument noise, was 0.0001 $\mu eq\ dm^{-3}$ for MSA. The overall analytical accuracy was better than 1.5%.

2.3 Back-trajectories and meteorological data

For every hour during the ten years 2006 through 2015 three-dimensional back trajectories have been calculated arriving at 474 m at Mt. Zeppelin. The trajectories have been calculated backward for up to ten days using the HYSPLIT4 model (Draxler and Rolph, 2003) with meteorological data from the Global Data Assimilation System with one-degree resolution (GDAS1). More information about the GDAS dataset can be found at Air Resources Laboratory (ARL), NOAA (http://ready.arl.noaa.gov), from which the meteorological data were downloaded.

During the analyzed time period meteorological records at the Mt. Zeppelin station are rather limited in quality and were frequently interrupted. In order to have an internally consistent, and unbroken meteorological record we utilized the hourly meteorological parameters at trajectory arrival times as calculated by the HYSPLIT4 model. As an additional parameter we evaluated the vertical air movement of the trajectories during the last hour before arrival by subtracting the trajectory height one hour before arrival at the arrival height of 474 meter. The resulting vertical displacement parameter, DZ, is given in meters per hour. Positive values of DZ indicate a lifting of the air.

The most important missing meteorological information concerns the local cloudiness. No direct recording was available of times during which the station was in clouds. The closest available cloud instrument is a ceilometer operated by the Alfred Wegener Institute (AWI) at their Koldewey Station in Ny-Ålesund, i.e. in the valley below Mt. Zeppelin, some 2.8 km of horizontal distance from the position of the mountain station. From the one-minute records of the ceilometer we derived hourly values of the 25% percentile of cloud base, which was used as an indicator for the Zeppelin station being in clouds. The meteorological parameters are listed in Table 1.

Precipitating clouds scavenge the planetary boundary layer and thus reduce the available particle surface for condensational uptake of particle precursors. As a consequence nucleation from the gas phase may be facilitated (Tunved et al., 2013). As in Tunved et al. (2013) we utilized the HYSPLIT-modeled precipitation along the back trajectories. Sums of precipitation, (SP, see Table 1), were calculated along each back trajectory and will be referred to as SP1 (during the last day), SP2 (during the last but one day), and SP5 (during days three to five) before arrival at Mt. Zeppelin.

2.4 Marine biological data

The biologically active marginal ice zone is a major natural source of sulfur in the Arctic summer atmosphere, (Leck and Persson, 1996b, a), and Wiedensohler et al. (1996), indicated a potentially important role of dimethyl sulfide (DMS) in regional new particle formation. DMS emissions from the sea have long been proposed to control new particle formation in the marine boundary layer, (Charlson et al., 1987), which builds on DMS_{aq} being transported via turbulence and diffusion to the sea-air interface, represented by the transfer velocity, which in turn depends on sea-surface temperature, salinity, and wind speed, (Liss and Merlivat, 1986). Once ventilated to the atmosphere DMS_g is photochemical oxidized via intermediates such as sulfuric acid and methane sulfonic acid, (Ayers et al., 1996), which eventually leads to the formation of aerosol $nssSO_4^{2-}$ and MSA.

Dimethyl sulfide in the ocean (DMS_{aq}) is produced through the degradation of its algal precursor dimethylsulfoniopropionate (DMSP) by microbial food webs, (Simó, 2001). At high latitudes, total DMSP (DMSPt) and therefore DMS_{aq}, essentially follows the seasonal cycle of phytoplankton biomass, (Lana et al., 2012). DMSPt is defined as the sum of $DMSP_{dissolved}$ and $DMSP_{particulate}$ concentration. Yet, the amount of DMSPt per unit

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phytoplankton biomass may vary depending on species composition and physiological state, (Keller et al., 1989).

The dissolved organic carbon (DOC) concentrations in surface waters of the high Arctic Ocean are up to ten times higher than in any other ocean basin and closer in range to DOC levels reported for sea ice (Gao et al., 2012). A large fraction of DOC spontaneously assembles into polymer gels: polysaccharide forming hydrated calcium bonded three-dimensional networks to which other organic compounds, such as proteins and lipids, are readily bound. The assembly and dispersion of the polysaccharide molecules can be affected by environmental parameters, such as UV-B radiation (280-320nm) dispersing or inhibiting gel formation, and/or pH and temperature inducing gel volume phase changes (swelling and shrinkage). In the study of Orellana et al., (2011b), the swelling and shrinking of the polysaccharide networks or polymer gels were also causally related by additions of nano to micromolar levels of DMS and DMSP. High DMSP concentrations have also been measured in the mucilage surrounding prymnesiophyte *Phaeocystis pouchetii* colonies in Arctic waters, representing up to 25% of the total water column DMSP pool, (Matrai and Vernet, 1997).

The findings made by Orellana et al., (2011b) were in agreement with previous findings by Orellana et al., (2011a) that high concentrations of DMSP and DMS are stored in the acidic secretory vesicles of the *Phaeocystis* algae where DMSP is trapped within the condensed polyanionic gel matrix until the secretory vesicles are triggered by environmental factors such as temperature to release gels that undergo volume phase transition and expand at the higher pH of seawater. Exocytosis of polymer gels accompanied by elevated DMS and DMSP concentrations suggests the transport of these chemical compounds by the gel matrix. Schoemann et al., (2005) report that *Phaeocystis antarctica* is particularly well adapted to low temperatures, being more competitive than *P. pouchetii for temperatures between* -2 and +2 °C. *Phaeocystis pouchetii*, however, appears to be better adapted to temperatures closer to

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5 °C. In the Arctic a higher occurrence of the *Phaeocystis pouchetii* would be expected in the northward advection of warm Atlantic water masses around Svalbard.

Here we estimated DMSPt at the sea surface using the algorithm described by Galí et al. (2015). The DMSPt algorithm exploits the distinct relationship between DMSPt and Chlorophyll-*a* depending on the light exposure regime of the phytoplankton community. The light exposure regime is defined by the ratio between euphotic layer depth and mixed layer depth (Z_{eu}/MLD). Additional predictor variables used are sea surface temperature (SST) and particulate inorganic carbon (PIC), which is used in the algorithm as a proxy for coccolitophores such as *Emiliania huxleyi*. During late bloom stages, the calcite plates that cover coccolitophore cells (called coccoliths) detach and cause an increase in seawater backscatter that invalidates satellite retrievals of Chlorophyll-*a*. Therefore, inclusion of PIC in the algorithm as a proxy for DMSPt increases data coverage. Although the algorithm was developed for the global ocean, validation results with in situ data indicate that it performs as well or slightly better in Arctic and sub-Arctic waters.

The use of remotely sensed DMSPt as a proxy for marine DMSaq emission is a significant improvement with respect to prior studies that used Chlorophyll-*a* (Becagli et al., 2016; Zhang et al., 2015). Yet, it is not ideal because (i) the ratios DMS_{aq}/DMSPt in surface seawater are variable, and tend to be higher in high solar irradiance and nutrient-poor conditions typical of summer, (Galí and Simó, 2015), and (ii) even if DMSPt is a better proxy for DMS_{aq}, the influence of meteorological and sea surface conditions (mainly wind speed and SST) on the sea-air flux of DMS_{aq} is not taken into account. Development is underway of an algorithm for the retrieval of DMS_g concentrations in air and DMS fluxes.

The DMSPt algorithm was run for the 2006-2015 period using daily composites of the Moderate Resolution Imaging Spectroradiometer on the Aqua satellite (MODIS-Aqua) at 4.64 km resolution (L3BIN, reprocessing R2014.0) downloaded from NASA's Ocean Color website (http://oceancolor.gsfc.nasa.gov). The MODIS variables used include Chlorophyll-*a*

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concentration derived with the GSM algorithm, (Maritorena et al., 2002), PIC, nighttime SST and Z_{eu}. MODIS nighttime SST was complemented with SST from the Advanced Very High Resolution Radiometer (AVHRR, https://podaac.jpl.nasa.gov/AVHRR-Pathfinder) to increase data availability. MLD was obtained from the MIMOC climatology, (Schmidtko et al., 2013), which was linearly interpolated from its original 0.5°x0.5° grid at monthly resolution to the MODIS grid at daily resolution.

Satellite remote sensing of biological activity in surface waters requires ice-free and at least part of the time cloud-free. The passive sensing methods of MODIS additionally require a minimum of solar illumination of the scenes (i.e., solar zenith angle < 70°; (IOCCG, 2015)). Consequently, the length of the satellite-observable period used to compute DMSPt means shortens from all-year-round at latitudes <45° to approximately six months (the spring-summer semester) at 80°N. In addition, the annual DMSPt map in Fig. 3 excludes all land and ice covered regions. In order to increase data coverage, daily DMSPt composites were binned to five-day periods and a 46.4 km equal-area sinusoidal grid, (10x10 bins of the original pixel size). The average distance between a trajectory point and the closest center of a MODIS pixel is 18 km.

Following the same approach as with the ice data average DMSPt from ocean color data, (OC), along each back trajectory were calculated and will be referred to as OC1 (the last day), OC2 (the last but one day), and OC5 (days three to five) before arrival at Mt. Zeppelin. In this procedure missing data were flagged as such, and were not taken into account.

2.5 Ice data

For the interpretation of events of new particle formation observed during the *Oden* cruises information on pack ice extent under the air masses reaching the sampling points proved crucial (Heintzenberg et al., 2015). Another motivation for utilizing ice data in the present study is the fact that the Svalbard region experiences large seasonal changes in pack ice cover which we expect to have strong effects on emissions of particles and their precursor gases. Thus daily ice concentrations were taken from the NSIDC database (https://nsidc.org/data). The irregularly shaped data gap around the pole caused by the inclination of satellite orbits and instrument swath was filled with 100% cover. To each hourly position and data of the back trajectories the ice information in the corresponding maps of ice concentrations were added and displayed in Fig. 2. On average the closest pixel in the ice maps was about 12 km off any trajectory point.

In the discussion of results we utilize the complement of ice cover, i.e., the amount of open water because the marine biological processes of interest predominantly take place in the open water, (Leck and Persson, 1996a). As integral parameters average open water, (OW), percentages along each back trajectory were calculated and will be referred to as OW1 (the last day*)*, OW2 (the last but one day), and OW5 (days three to five) before arrival at Mt. Zeppelin. The most solid ice cover is seen in an area reaching from Northeastern edge of Greenland via North Pole to Parry Island. A marginal ice zone extends along the east coast of Greenland to Franz-Josef-Land, and the area between Svalbard and the latter island.

2.6 ERA-Interim data of sea surface temperature

Daily Sea Surface Temperature (SST) data for our study period (2006-2015) were downloaded from the website of the European Centre for Medium-Range Weather Forecasts (ECMWF). A description of the global atmospheric reanalysis, (ERA-Interim), has been given by Dee et al. (2011), and a guide to the products and the download procedures can be found at http://www.ecmwf.int/en/elibrary/8174-era-interim-archive-version-20. Briefly, ERA-Interim is an assimilating model reanalysis of the global atmosphere and sea-surface physical parameters covering the data-rich period since 1979. SST data were downloaded at a resolution of approximately 0.56° and regridded onto the same 46 km equal-area sinusoidal grid used for DMSP and cloud fraction, (see below). Ice-covered pixels were screened out prior to the back-trajectory analysis. In the Arctic region, ERA-Interim has been shown to be a top performer among a number of atmospheric reanalyses, (Lindsay et al., 2014).

2.7 MODIS cloud fraction

Persistent cloud cover limits PPP in Arctic and Sub-arctic seas, (Bélanger et al., 2013), and, as mentioned above, irradiance at the sea surface, which is largely controlled by cloudiness, influences $DMSP_{dissolved}$-to-DMS_{aq} conversion. Boundary layer clouds are known to be additional controllers of the surface aerosol (Heintzenberg, 2012). In the summer Arctic low level clouds and fogs are widespread (Warren and Hahn, 2002). Both scavenging and new particle formation have been observed in connection with low clouds and fog passages (Lannefors et al., 1983; Heintzenberg and Leck, 1994; Leck and Bigg, 1999; Heintzenberg et al., 2006; Karl et al., 2013). Beyond the cloud base derived from the ceilometer we have no other in situ local or regional cloud information. Thus, we utilize satellite-derived cloud information.

Daily Level-3 global cloud fraction with one-degree resolution was downloaded from NASA website (http://modis-atmos.gsfc.nasa.gov, Hubanks et al., 2015) and extracted for our region of interest. Briefly, level 3 images correspond to the aggregation of all level 2 images (1 km resolution) available within the one-degree resolution grid. For a given L2 scene, each pixel is assigned a value of 1 (cloudy) or zero (clear sky), and then the individual scene values are averaged over a 24-hour period. Note that a given pixel can be revisited up to six or seven times in the course of a day at high latitudes. Finally, the daily composites were re-projected to 46.4 km pixels to match the spatial resolution of DMSPt. The average distance between a trajectory point and the closest MODIS pixel was 18 km.

The cloud fraction CF as well as other cloud properties from MODIS have been extensively used, for instance to study the global spatial and temporal distribution of clouds over the last decade (e.g., King et al., 2013). Several studies have also successfully performed validation by comparison with in situ data (e.g., An and Wang, 2015) which demonstrated the ability of the MODIS-aqua sensor to retrieve cloud cover.

Following the same approach as with the ice data and DMSPt, average cloud fractions, (CF, see Table 1), along each back trajectory were calculated and will be referred to as CF1 (the last day), CF2 (the last but one day), and CF5 (days three to five) before arrival at Mt. Zeppelin. Missing data are flagged as was done with DMSPt data.

3. Three approaches to identifying events of new particle formation

There are no definitive and no generally accepted methods to identify or predict NPF-events in atmospheric time series of aerosol data. Thus, in the present study we explored different approaches with varying degrees of complexity to identify such events. We emphasize that none of these approaches explicitly is connected to diel cycles such as in Dal Maso et al., (2005) or makes any assumptions about the time of day during which new particle formation occurs. Three objective search algorithms were written in FORTRAN to analyze the time series of hourly records of aerosol parameters in search of new particle formation:

1. The simplest approach of upper percentiles (PCT-approach), assumes that NPF-events are characterized by extremely high concentrations of small particles in terms of N25 (see Table 1). The key parameter characterizing each PCT-event was the value of N25

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averaged over a fixed number of hours, ($N25_{av}$), after the nominal start of an event, (see below). With $N25_{av}$ also a nominal length of PCT-events was defined as the number of hours after the start of an event by which N25 sank to less than half of $N25_{av}$.

2. The more specific approach of diameter growth (DGR-approach) builds on the temporal development of the particle size distribution in terms of a systematic growth of the diameter D50 (see section 4.1) to find the classical "Banana Type" of NPF-event, (Kulmala et al., 2004). The key parameter characterizing each DGR-event was the average growth of D50 during the nominal event length NUC, (see below). For this approach the nominal length of events was reached when the running two-hour average growth fell below the value one.
2. The most complex approach of multiple-size events, (MEV-approach), searches for events with concurrent appearance of concentration increases in several size classes below 60 nm diameter (Karl et al., 2013; Leck and Bigg, 2010). The key parameter characterizing each MEV-event was the relative concentration increase averaged over the chosen size classes below 60 nm during the nominal event length NUC, (see below). As with PCT-events a nominal length of MEV-events was defined as the number of hours after the start of an event by which N25 sank to less than half of $N25_{av}$.

Three time-related parameters were commonly defined for all three approaches:

1. Nominal NPF-event length, (NUC) was nine hours.
2. Pre-event periods, (PRENUC), from which increases in diameters or number concentrations were calculated, were six hours.
3. Reference periods, (REF), before PRENUC and after NUC periods were defined in order to compare event and pre-event data with non-event conditions. Each of these reference periods had the length of half the sum of pre-event plus event time periods, making the total reference time period of each event as long as that of the event itself.

Besides these common characteristic lengths individual fixed thresholds were chosen and discussed below for each approach in order to generate at least 200 unique events per approach, (see Table 2).

The aerosol data used to define the NPF-events were complemented by a large number of environmental parameters. The primary temporal resolution of the environmental parameters was between one minute (C25, cf. Table 1) and five days (DMSPt, cf. Table 1). C25 was calculated as 25% percentile on an hourly basis. The parameters with resolutions higher than an hour (OW, CF, and OC, cf. Table 1) were evaluated along the hourly back trajectories. While this procedure yielded hourly varying results even of OW, CF, and OC it has to be kept in mind that this hourly variability is the result of hourly resolved trajectories traversing the grid; the low primary temporal resolutions of the OW, CF, OC, and chemical parameters remain. For these slowly varying parameters the REF periods before and after the events were extended to one day beyond the longest primary resolution, i.e., six days.

For two reasons the three search algorithms may yield temporarily redundant results, i.e., they may identify the same events. One, they go independently through the same time series of aerosol data, possibly causing inter-approach redundancy. Two, each algorithm goes through the time series hour by hour, thus allowing for temporal overlap of events found by each approach, (intra-approach redundancy).

The three types of events were assumed to be mutually exclusive and potentially being caused by different sets of conditions for new particle formation. Thus, a FORTRAN procedure was developed to eliminate both intra and inter-approach redundancy while maintaining a maximum of identified NPF-events. To remove intra-approach redundancy the procedure identifies overlapping events within each approach. Of each ensemble of such overlapping events the one with the strongest key parameter of the respective approach (growth of D50, or concentration increases as defined above) is retained. Next, inter-approach redundancy is addressed by the procedure. However, there is no unique solution to the problem of the partly redundant three time series of events. In order to avoid any preference of one or several types of events in the tests of inter-approach overlap pairs of events of different approaches are chosen at random and compared for overlap. This random comparison is done as often as the product of the number of events of the three approaches. This rather time-consuming random test yields stable numbers of non-overlapping events within less than one percent, irrespective of the order in which the events of the three approaches were arranged for the test. By removing intra and inter-approach redundancy in the first two steps of the procedure a number of time periods will be "freed". Consequently, in a last step, the procedure tries to fill the "freed time periods" non-redundantly with events of the three approaches that had been eliminated in the first two steps. Table 2 collects total numbers and unique numbers of events for each approach. In the rest of the paper only non-redundant events will be discussed. The total number of new particle formation events will be shortened to TNPF.

3.1 The upper percentile of N25 (PCT-approach)

Events of new particle formation were identified by time periods in which N25 was consistently, i.e. on average for three hours, above a set threshold. With a threshold of the 93%-percentile (170 cm^{-3}) 4143 PCT-events were identified in the total data set, only 240 of which were unique because most of them overlapped with event or pre-event times of the other two approaches. Average N25 during these unique events was 330 cm^{-3} and the average length of events 4 ±0.9 h, (one standard deviation).

Fig. 4, (top), shows the average temporal development of the relative size distributions for the unique PCT-events as in the results in Karl et al. (2013), i.e. relative concentrations were formed by dividing the absolute number concentrations by the average total number during the six-hour pre-event time periods. The events are characterized by a nearly monomodal distribution around 20 nm that broadens somewhat around the nominal start of the events. During the last three hours before the events D50 decreased slightly and returned to the pre-event level during the nine NUC hours.

In connection with PCT-events average aerosol parameters NTO through N300 showed an average increase by a factor of 2.2 during PRENUC-periods, which was maintained on an average level of 1.5 during the events. The aerosol-chemical parameters Na^{+}, $nssSO_4^{-2}$, and MSA were on an average level of 20% of their reference value. The average environmental parameters indicate a strong increase by a factor of 14 in solar radiation and a lifting of cloud base before the events. During the events the level of solar radiation was still elevated by a factor of six above its reference value. As a consequence temperature at the station was up by 2 – 3 degrees. Precipitation 12 h before trajectory arrival time, (SP12) was a factor of five above reference levels for air arriving during NUC-periods, whereas SP35 to SP5D were below their respective reference levels. Cloud fractions were slightly raised 12 - 48 h before air arrival. Of the ocean parameters more open water was met by trajectories 12 to 24 before their arrival with ocean temperatures 12 to 48 h before trajectory arrival having been up to four degrees warmer than their respective reference values. On average DMSPt-parameters OC24 through OC5D showed were raised by a factor of two above their reference value.

In Fig. 5, (left top panel), average trajectory height profiles during PRENUC and NUC-periods are displayed. Widely varying vertical air mass paths occurred before and during PCT-events. Median vertical trajectory paths during PRENUC and NUC times indicated air coming from some 300 m above station level five days ago sinking to about one hundred meters above station level during the last two days before arrival. The upper quartiles of the PCT-height profiles point at strong subsidence before air mass arrival.

The right top panel in Fig. 5 maps average horizontal trajectory positions in 12 h steps in months having at least ten PCT-events, i.e., May - September. Filled circles around the

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trajectory positions comprise 95% of all events. The monthly average horizontal trajectory direction during PCT-events mostly was from the northwest. In June and July the trajectories reached farthest into the multiyear ice cover northeast of Greenland. Only during September the back trajectories covered ice-free and marginal ice areas in the Fram Strait. We note that the five-day back trajectories of PCT-events, (and of the other two approaches as well), stayed within some 800 km of Mt. Zeppelin.

3.2 The diameter growth (DGR) approach

The DGR-approach to identify events of new particle formation builds on the classical concept of particle growth through condensable vapors after an initial nucleation of sub-five nanometer particles that cannot be observed with the available instrumentation, so called "Banana-type" (Kulmala et al., 2004). The respective algorithm utilizes the parameter D50, (see Table 1), and requires a growth of this diameter by at least a factor of 1.5 after the nominal start of an event. With this threshold the algorithm searched through all 87646 hours of the ten-year record and found 1199 DGR-events of new particle formation. After eliminating cases of temporal overlap with the other two approaches 235 unique events of this type remained, (see Table 2). Other or more DGR-events could have been found by shortening the nominal nine NUC hours. For two reasons we refrained from discussing shorter growth periods in the DGR-approach. Maintaining common-length NUC periods facilitated the comparison of results of the three approaches. Furthermore, reducing the growth period would also make PCT and DGR-events ever more similar.

Starting with an average value of D50 ≈ 16 nm at the nominal start of DGR-events an average growth rate of 1.8 nmh^{-1} is derived, which is in the range of 1 – 2 nmh^{-1} derived by Ström et al. (2009) for new particle formation in the lower boundary over Ny-Ålesund, Spitsbergen but considerably lower than the maximum growth rate of 3.6 nmh^{-1} reported by Asmi et al. (2016) for July at the Siberian station Tiksi at the coast of the Laptev Sea. The average length of DGR-events was 10 ±1 h, (one standard deviation).

The average temporal development of the relative number size distribution during DGR-events is presented in Fig. 4, (center). After a decrease of the sub-50 nm diameter median from about 25 to 16 nm during the six hours before the nominal start of the events D50 increases systematically during the following nine NUC hours with somewhat reduced growth towards the end of the event.

During the PRENUC-periods particle number concentrations N300, and the condensation sink, (CS), decreased relative to the reference periods before and after the events. Subsequently, during the NUC periods the strongest increases was found for N60. Environmental parameters around air mass arrival showed a strong lifting of cloud base, (C25), and an extremely high increase in solar radiation, (by a factor of 11 during PRENUC and by a factor of 60 during NUC periods). However, 12 h before air arrival precipitation had been up by a factor of 2.5. Cloud fractions were down to about 70% of their reference values 24through 48 h before air arrival. Of the chemical aerosol parameters Na^{+} and $nssSO_4^{-2}$ showed an increase of 2.6 and 2.3, respectively. OC12 and OC48 were slightly higher than reference level before and during the events. Sea surface temperatures T24 were raised by nearly one degree whereas earlier SST-values, (T36 – T5D), were up to one degree below reference values.

Fig. 5, (left center panel), shows statistics of the vertical air movement before trajectory arrival during DGR-events at Mt. Zeppelin covering a wide range of vertical movements between 200 m and beyond 1500 m height. During the days when elevated DMSPt levels were noted median trajectory heights were six to nine hundred meters. Median trajectories during PRENUC times dipped down to the station level, (474 m a.s.l), about one day before arrival, albeit lifted and subsided again shortly before arrival. Vertical trajectory pathways will be discussed further in Section 4.2.

Monthly average trajectory positions and their variability in connection with DGR-events are shown in Fig. 5, (center right panel). The months April through October had at least ten DGR-events per month. As with PCT-events the general trajectory direction was from the northwest, mostly staying for several days over the marginal ice zone between northeastern Greenland and eastern Svalbard. During the earliest month of April with 14 DGR-events the back trajectories reached farthest south into the ice-free parts of the Fram Strait.

3.3 The Multiple-size approach (MEV)

Leck and Bigg (2010) and Karl et al. (2013; 2012) discussed a type of new particle formation that to date only has been reported from the summer Arctic. During these MEV-events high ultrafine particle concentrations appear concurrently in a broad diameter range reaching from under 10 to some 60 nm. We simulated this type in a search that required the concurrent increase of NTO, N20, N40, and N60, (cf. Table 1), as averaged over the first three NUC hours by a factor ≥ 1.6 over their respective averages during the six (PRENUC) hours. Over the ten years of data 1191 such events of this type were identified, 266 of which remained after removal of those overlapping with events of other approaches. During these unique events the average concurrent concentration increase was 4.7 and the average length of the events 12±0.8 h.

The bottom part of Fig. 4 shows the average temporal development of relative number size distributions before and during MEV-events. The development before the nominal start of MEV-events is more complex than during the PRENUC-periods of the first two types of events. Intermittently a mode around seven nanometers shows up that broadens and becomes more prominent about two hours before the nominal start of events. The major PRENUC-mode around 25 nm also broadens and becomes more prominent towards NUC. A weak mode exists during PRENUC around 120 nm and hardly any particles beyond 400 nm. D50 sinks from 25 to about 20 nm and stays below 25 nm through the MEV-events even though number concentrations increase during the first NUC-hours by more than a factor of five.

During NUC-periods all particle number concentrations increased, on average by a factor of 1.6. Average solar radiation also increased by about 90% above reference level during NUC-periods. Of the chemical parameters $nssSO_4^{-2}$ showed an increase by a factor of three during PRENUC and NUC-periods, and MSA a slight increase during PRENUC-periods. On one hand, precipitation 12 h, and 36 to 48 h before trajectory arrival, (SP12, SP48), were above reference levels for air arriving during PRENUC-periods. On the other hand, during PRE, SP24, SP36, an SP5D indicated dry conditions during PRENUC and NUC-periods. Only three to five days before air arrival slightly increased cloud fractions were noted. Sea surface temperatures up to five days before trajectory arrival were on average about one degree lower than their reference values. DMSPt parameters OC12 to OC36 were raised by factors of 1.3 and 1.6 during PRENUC and NUC-periods, respectively.

Percentiles of vertical trajectory coordinates prior to and during MEV-events are displayed in Fig. 5, (bottom left panel). During the events, and even stronger during the PRENUC periods median trajectories had been below 500 m for more than four days. Furthermore, the final air approach to Mt. Zeppelin mostly came from below the station level. Upper quartiles of the vertical trajectory positions are substantially lower than with DGR-events. We note, however, that a short excursion above station level occurred in the upper quartiles during the last three hours before arrival.

The bottom right panel of Fig. 5 gives the monthly average trajectory positions and their variability in connection with MEV-events. The months April through October had at least ten MEV-events per month. As with the other approaches the general trajectory direction was from the northwest, albeit with stronger swings towards the ice-free areas south of Svalbard early and late in the season, (April, May, and September). Interestingly, the trajectories of the 11 MEV-events in October were directed nearly straight north from the North Pole.

Summarizing differences and commonalities among the results of the three approaches we can state that the length of the events increases from four to ten and twelve, going from PCT to MEV-events. PCT-events are characterized by lower-than-reference aerosol-chemical parameters. Na^+ and $nssSO_4^{-2}$ show strong increases in the other two types of events: Na^+ in connection with DGR-events and $nssSO_4^{-2}$ in connection with MEV-events. Both, PCT and DGR-events exhibit strong increases in solar radiation. Precipitation before air arrival was raised at varying times in connection with the three types of events. Cloudiness bot increased and decreased at varying times before air arrival with the three types of events. Increased open water under the trajectories was strongest with DGR-events and least important with MEV events. Only in connection with PCT-events strongly raised sea surface temperature were noted before trajectory arrival. DMSPt related ocean parameters were raised to varying degrees and at varying times before all NPF-events, most strongly in connection with PCT-events and least in connection with DGR-events.

4. Discussion

4.1. Environmental setting

The discussion of the results on new particle formation in the Svalbard region builds on the variability of new particle formation and related environmental parameters on scales of months, and days. Fig. 1 gives an overview over the geographic areas which were covered by one, two, and five-day back trajectories to Mt. Zeppelin during the ten years of the present study covering the months March through October. This figure illustrates that air arriving at Mt. Zeppelin during the ten summers of the present study came from widely varying regions from the central ice-covered Arctic via the northern seas and northernmost Scandinavia to Greenland. One-day back trajectories cover a roundish area from the central east coast of Greenland via northern Scandinavia to Franz-Josef-Land, North Pole and back to the north coast of Greenland. Excluding inner Greenland this area is widened by roughly 500 km by 2-day back trajectories and by at least another 500 km by 5-day back trajectories reaching over most of Greenland and the adjacent seas west of Greenland. This is a much wider region from which air may reach Mt. Zeppelin as compared to sites in the inner Arctic as illustrated in Fig. 2 of Heintzenberg et al. (2015).

On the path of trajectories to Mt. Zeppelin quite different ice conditions were met (see, Fig. 2). On average North Atlantic open waters reached around West Spitsbergen all the way to Nordaustlandet. Drift ice was passed over by trajectories along the whole east coast of Greenland. One-day trajectories passed over the marginal ice zone from the Fram Strait to Franz-Josef Land but also over more contiguous ice close to the North Pole. At times, with five-day back trajectories, even the marginal ice regions of Baffin Bay and Beaufort Sea were reached.

The long-term geographical distribution of DMSPt in Fig. 3 reflects the water conditions for phytoplankton biomass around Svalbard. Directly at the coasts of Greenland and Eurasia increased nutrient availability in coastal and shelf waters (due to continental run-off and enhanced hydrodynamics) cause localized areas of high DMSPt values. The low DMSPt values further out along the coast of Greenland are due to sea ice reaching through the Fram Strait far south, (see Fig. 2). A prominent feature in the regional DMSPt distribution is the tongue of high DMSPt, (intense blue color), and thus high phytoplankton biomass east of this area, reaching from Spitsbergen to roughly Jan Mayen that lies within one-day back trajectories. Northward-flowing Atlantic waters, carried by the West Spitsbergen Current, and southward-flowing fresh surface waters from melting ice, and recirculated Atlantic waters, carried by the East Greenland Current (Rudels et al., 2005) are meeting. The layering created by water masses of different density stabilizes the water column and traps phytoplankton cells at well-lit depths. If sufficient nutrients are available, this can lead to the development of large phytoplankton blooms, which can result in high concentrations of DMS_{aq}, (see Fig. 2 in Leck and Persson, 1996a).

In the ten-year average cloud fractions systematic differences in cloudiness appear. Depending on transport pathways as identified by the back trajectories, cloudiness varies on the way to Spitsbergen. The ice-covered areas, (cf. Fig. 2), from the east coast of Greenland to Franz-Josef-Land exhibit somewhat lower cloud fractions than the ice-free regions southwest to east of Spitsbergen.

4.2 Seasonal variability

Seasonal changes are discussed in terms of monthly averages taken over the ten-year study period. As expected in Earth's polar regions the seasonal variability of all environmental parameters is very high as exemplified by the solar flux, (SFL), and the air temperature, (TEM), at Mt. Zeppelin in Fig. 6. Due to the seasonal change in cloudiness, (cf. Fig. 7), the seasonal distribution of SFL is not quite symmetrical about midsummer but is skewed slightly towards the cloud minimum in spring. The air temperature, however, does not peak before July and has a broad shoulder into fall and winter. The first, and partly absolute maxima, of the seasonal distributions of NPF-events in Fig. 6 coincide with that of the SFL but then drop of more slowly towards fall than solar radiation. In particular, MEV-events do so and even have their main maximum in August. The occurrence of all NPF-events drops off sharply in October. Whereas May as the first month with larger numbers of events is dominated by

PCT-events, followed by DGR and then MEV-events, the contributions of the three NPF-types are reversed in the last month with high NPF-numbers, i.e., September.

Fig. 6 clearly shows that the formation of new particles in the Svalbard region is not controlled by the late winter-to-early-spring phenomenon of Arctic haze peaking with highest sulfate-concentrations in March, (cf. Fig. 3 in Heintzenberg, 1989, and Fig. 6), which has a minimum in the total number of NPF-events. This minimum is in contrast with the maximum in new particle formation rates found by Croft et al. (2016a) with their global aerosol model. The high numbers of accumulation mode particles during the Arctic haze months in late winter and spring yield the annual maximum in condensation sink, (CS in Fig. 6), which could quench nucleation events and subsequent growth. Thus, even though photochemistry may produce significant amounts of nucleating material, the freshly formed particles will not grow to stable size before they are removed via either deposition or coagulation as discussed by Tunved et al. (2013) and others. An alternative explanation of the late onset of NPF-events in TNPF in spring lies in the marine biological processes not being activated nearby during the Arctic haze period yet, (Heintzenberg and Leck, 1994).

Fig. 7 collects the seasonal variation of environmental parameters as averaged along the back trajectories to Mt. Zeppelin. From their minimum in March-April open water conditions improve until September, after which the pack ice extent under the trajectories rapidly increases again. The widening open water areas are reflected in sea surface temperatures under the trajectories that increase until September before they drop off strongly in October. Consequently, because of its connection to marine biological activity DMSPt increases in the euphotic zone from first photosynthetic light in May until it evens out around July and drops off in October. Largest DMSPt values are reached in the vicinity of Svalbard, (cf. OC12 in July and August in Fig. 7), i.e. considerably later than MSA. The ending of DMSPt-curves in October is due to the lack of data not due to zero-DMSPt. Still, DMSPt concentrations are expected to be low at this time of the year at temperate to polar latitudes due to low phytoplankton biomass and low light exposure, (see Fig. 9 in Galí et al., 2015). In terms of the MODIS-derived cloud fraction cloudiness increases rapidly from its minimum in April and evens out on a plateau of 80 – 90% after July. The spring-minimum in cloudiness is confirmed by the maximum in cloud base as indicated by C25 in Fig. 7. This seasonal distribution of cloudiness does not correspond to the classical picture of near-surface cloudiness that exhibits near cloud-free conditions in winter and mostly overcast with Arctic stratus and fogs during the summer months (Warren and Hahn, 2002; Huschke, 1969). We explain the difference by the specific atmospheric pathways covered by the back trajectories of the present study (cf. Fig. 1). Trajectory-averaged precipitation parameters (SP12-5D in Fig. 7) have minima in the period April – May, from which they increase towards their maxima in fall and winter.

The chemical aerosol information derived from the analyses of filters samples has a relatively low temporal resolution of at least one day combined with frequent gaps of several days in between samples. Thus, it cannot directly be related to the time periods of NPF-events. The seasonal distribution of chemical tracers, however, yields important information about new particle formation. Taken over the whole year $nssSO_4^{2-}$ in Fig. 6 is largely anthropogenic, (Heintzenberg and Leck, 1994), and has its maximum during the peak of Arctic haze in March and April and its minimum in August, which does not match any seasonal distribution of NPF-events. We also plotted Na^+ in Fig. 6 as a tracer of the inorganic marine aerosol components sea salt. Na^+ decreases from its winter maximum to its summer minimum in June/July, again without similarity to the NPF-distributions. Instead, the seasonal distribution of Na^+ rather closely follows that of the trajectory-derived wind speed during the last hour before arrival, (not shown in the figure). Wind speed as driver for sea salt production is a well established phenomenon (Blanchard and Woodcock, 1957). After a steep rise in April MSA in Fig. 6 sharply peaks in May and then gradually drops off towards its minimum in October, more gradually than reported for data taken from 1991 to 2004 by

Sharma et al. (2012) and earlier than reported by Heintzenberg and Leck (1994), both at the same station. Our seasonal distribution of MSA most closely resembles that of SFL, in Fig. 6, albeit with its peak in May a month earlier that SFL and more strongly skewed towards spring. According to Leck and Persson (1996b) on average the concentrations of the marine biogenic sulfur components, (DMS and MSA), fell with a decline rate of about 30% per week approaching zero values in September explained by reduced ppp (Leck and Persson, (1996a), (consistent with Becagli et al., 2016).

As MSA is the only measured aerosol component with exclusively marine biogenic sources, we illustrate its seasonal distribution in greater detail in Fig. 8. In this figure MSA-concentrations measured on Mt. Zeppelin have been extrapolated along 5-day back trajectories, forming monthly average monthly maps of potential MSA-sources during the biologically most active months of March through October.

Fig. 8 yields several items of information that are relevant to the issue of new particle formation. Early in spring the biological aerosol sources are limited to the North Atlantic and Norwegian Sea. In April the tongue of newly opened waters between Novaya Zemlya and Franz-Josef-Land seemingly is beginning to become biologically active. In May this area widens towards the Barents Sea while the North Atlantic also becomes more active, reaching the Fram Strait. In August two wide potential source regions cover the region from Northern Greenland to the northern end of Scandinavia and the region Barents to Kara Sea. In September even the pack ice north of Svalbard becomes biologically active, (Leck and Persson, 1996a), and shows potential MSA sources, in particular, north of the northern coast of Greenland. Finally, the very weak potential MSA sources in October appear to be situated mainly over the Kara Sea and over the North Atlantic.

How do these seasonal distributions compare to those of the NPF-events identified by the three search-approaches defined in Section 3? To address this question we constrained the average seasonal distribution of environmental parameters to those hours that had been

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identified by the NPF-events of the three approaches. However, none of the individual seasonal distributions of constrained environmental parameters follows closely any of the NPF-events. In particular, the main MSA peak remains in May, thus one month earlier than any peak of the NPF-occurrences. To elucidate further potential differences in the three types of NPF-events we return to the discussion of vertical pathways of related back trajectories, (see Fig. 5). In this figure all three types of NPF-events exhibit a wide range of vertical trajectory paths. As we expect the regional sources of primary particles and particle precursors to be at or near the surface we segregated the NPF-events into subpopulations with back trajectories that remained a given time below 500 m, (roughly station level). In Fig. 9 we collected the results concerning the 93 NPF-events that occurred with trajectories under the 500 m limit, i.e., roughly 12% of all events. The top panel shows that the related trajectories not only stayed below 500 m through most of the last five days before arrival but close to the surface until they started rising to the station level about 24 h before arrival. The peak of the sum of event occurrences now coincided with the main MSA peak in May, (see center panel in Fig. 9). For DGR-events the May-maximum was particularly strong whereas the PCT-predominantly occurred in May and June and MEV-events remained clustered around the later part of summer, possibly coupled to SST and DMSPt.

A number of environmental parameters indicated substantial deviations from their respective reference values during the months with most frequent occurrence of this sub-population of NPF-events. Strongest deviations were noted for precipitation that was elevated above reference levels two to five days before trajectory arrival, most prominently for DGR-events in May, (by a factor of six 36 h before trajectory arrival). Strong positive deviations in aerosol-chemical parameters only occurred with Na^+ in PCT and MEV-events, indicating relatively high wind speeds near sea surface in the related air masses. MSA was elevated up to 50% above reference levels only during MEV events. Elevated levels of DMSPt were noted with all three types of NPF-formation, most prominently for DGR-events 12 to 36 hours before which DMSPt was raised by factors up to 1.7 relative to reference levels.

The bottom panel of Fig. 9 gives average trajectory positions in 12 h steps for the months May through September. The circles around the steps comprise 95% of all trajectories. During all months the trajectories stayed in the ice-free and marginal ice zone between Fram Strait and Eastern Svalbard as illustrated by average July ice cover for the ten study years, (for average monthly ice covers cf. Fig. 8). In particular during the earliest and latest months of May and September the trajectories swing farthest south over the open water south of Svalbard. We note that the complementary sub-population of results with trajectories remaining above station level did not yield results that differed strongly from those for the whole population of back trajectories.

As a last step in the discussion of seasonal variations in new particle formation a model is formulated that describes the average sum of NPF-events, (TNPF), as a function of three parameters, two of which are directly measurable at the site. With the linear combination of the solar flux, (SFL, Wm^{-2}), average sea surface temperature under back trajectories 36 to 48 hours before their arrival at the site, (T48, °C), and condensation sink, (CS, 10^5 s^{-1}):

$$TNPF = 0.57 \cdot SFL + 15.4 \cdot T48 - 0.69 \cdot CS$$

TNPF as shown in Fig. 10, can be described within an average deviation of 5% taken over the major months with new particle formation, April - October. Any other of the sea surface parameters describes TNPF less well.

4.3 Diurnal variability

Average hourly occurrence of the three types of NPF-events is plotted in Fig. 11, (top). The three approaches yield rather similar diel variations. From their minimum during the night and early morning hours they reach their maximum occurrence between 12 and 16 h UTC in the afternoon. One might expect the differences between the NPF-types to be due to the requirement of the three types of NPF formation being mutually exclusive. However, this constraint does not exclude that they occur at the same time of day, only that they occur at the same time on the same day.

Over the continents new particle formation and growth events of the classical "Banana-type" usually exhibit an increase in measurable precursors such as sulfuric acid shortly after sunrise followed by the detection of increased numbers of nanometer-sized particles between one and two hours later (Kulmala et al., 2004), who deduce a connection to photochemically produced condensable vapors from this daily pattern. In the Svalbard region the sun is up all day between mid-April and the end of August. Consequently we would expect the photochemical production of condensable vapors to have a smaller diurnal amplitude than at lower latitudes, which in turn should even out the diurnal pattern of NPF-events to some degree. Despite the relatively small daily variations in solar elevations the solar flux on Mt. Zeppelin varied on average by more than a factor of five during the sunlit days (see curve SFL in Fig. 11, bottom). The daily maximum of SFL between 12 and 15 UTC coincides well with the average diel change in N25 and NPF-occurrence. As expected in particle growth due to condensable vapors after initial nucleation the daily maximum in N10 precedes that of N25 by a few hours.

The other process controlling the development of newly formed small particles is the diurnal development of the planetary boundary layer, (Kulmala et al., 2004). We have no data on the daily variation in boundary layer structure over or near the measurement site. The ceilometer data yield the only high-resolution information with some connection to the structure of the planetary boundary layer. During the summer months these data show a consistent daily variation with a jump in most frequent hourly cloud base by about 100 m from about 1570 m after 09 UTC with rather stable values following until 16 UTC, after which cloud base decreases again to values comparable to the early morning hours. The hourly medians of the vertical displacement parameter DZ, (see Fig. 11, bottom), provide a clearer diurnal variation. While being negative throughout the day, i.e. indicating subsiding air during the last hour before arrival at Mt. Zeppelin, DZ indicates the weakest subsidence in early afternoon. We interpret diurnal variation in cloud base and DZ as indicative of local clearing and convection during the day that may be conducive to photochemical processes and mixing in the boundary layer, both of which would be enhancing new particle formation.

5. Conclusions

Three different types of events of new particle formation, (NPF), were identified through objective search algorithms formulated for the present study. The first and simplest algorithm utilizes short-term increases in particle concentrations below 25 nm, (PCT-events). The second one builds on the growth of the sub-50 nm diameter-median, (DGR-events), and is most closely related to the classical "banana-type" of events, (Kulmala et al., 2004) involving the presence of photochemically generated DMS oxidation precursors. The third and most complex, so-called multiple-size approach to identifying NPF-events builds on the hypothesis of Leck and Bigg (2010), suggesting the concurrent production of polymer gel particles at several sizes below about 60 nm, (MEV-events).

With these algorithms NPF-events were identified in a ten-year record of hourly number-size distributions taken at the research station on Mt. Zeppelin, Spitsbergen. As a first and

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general conclusion we can state that NPF-events are a summer phenomenon and not related to Arctic haze, which is a late winter-to-early spring event. The seasonal distribution of the available information on cloudiness does not suggest any direct connection with NPF-formation. The MODIS derived cloud fraction generally is very high (70 – 90%) and rather evenly distributed over the Svalbard region during the months with high frequencies of NPF-events. As already reported in Tunved et al. (2013) NPF-events appear to be somewhat sensitive to the available data on precipitation derived from the trajectory model, in particular when constrained to cases with back trajectories staying below 500 m. In this subpopulation of NPF-events DGR-events show the strongest change in precipitation parameters in connection with new particle formation.

The seasonal distribution of solar flux suggests some photochemical control that may affect marine biological processes generating particle precursors and/or atmospheric photochemical processes that generate condensable vapors from precursor gases. Whereas the seasonal distribution of the biogenic MSA follows that of the solar flux it peaks before the maxima in NPF-occurrence. For PCT-events, and more distinctly so for DGR-events, this one-month delay disappears in the subpopulation with back trajectories staying below 500 m. MEV-events, however, maintain their peak occurrence later in summer and early fall.

With the limited information on particle size, composition, particle precursors, and environmental conditions no definitive statements can be made about the processes leading to the formation of new particles in the Svalbard region. A host of findings, however, point to varying and rather complex marine biological source processes. The potential source regions for all types of new particle formation appear to be restricted to the marginal ice and open water areas between Northeastern Greenland and Eastern Svalbard. During earliest and latest months with high numbers of NPF-events the back trajectories reach farther south into the open waters of the North Atlantic. Depending on conditions yet to be clarified new particle formation may become visible as short bursts of particles around 20 nm, (PCT-events), longer events involving condensation growth, (DGR-events), or extended events with elevated concentrations of particles at several sizes below 100 nm, (MEV-events). The seasonal distribution of NPF-events peaks later than that of MSA and, DGR and in particular of MEV-events reach into late summer and early fall with much open, warm, and DMSPt-rich waters around Svalbard, promoting the production of *Phaeocystis pouchetii* together with polymer gels. Consequently, a simple model to describe the seasonal distribution of the total number of NPF-events can be based on solar flux, and sea surface temperature, representing environmental conditions for marine biological activity, and condensation sink, controlling the balance between new particle nucleation and their condensational growth. Based on the sparse knowledge about the seasonal cycle of gel-forming marine microorganisms and their controlling factors we hypothesize that the seasonal distribution of DGR and more so MEV-events reflect the seasonal cycle of the gel-forming phytoplankton.

Despite the rather small diel changes expected during the summer Arctic there is a significant diurnal variation in aerosol and environmental parameters. Diurnal distributions of particle numbers below ten, (N10), and below 25 nm, (N25) follow that of the solar flux rather closely with a maximum between 14 and 16 UTC with the maximum of N10 occurring a few hours before that of N25. This delay in maxima may be caused by a slow particle growth due to photochemically produced condensable vapors. With a peak around noon MEV-events show the earliest daily peak occurrence with PCT and DGR-events peaking between 15 and 17 h, more closely to the maximum solar flux. Considering the diurnal variation in vertical trajectory displacement, (DZ), the early daily maximum in MEV-occurrence may be simply controlled by boundary layer dynamics.

With the large database of ten years of aerosol data on Mt. Zeppelin enriched by environmental atmospheric and marine data occurrences, pathways and potential source areas of different types of new particle formation in the Svalbard region were elucidated by the present study. More process related information about new particle formation would require dedicated mechanistic experiments with more detailed information on particle precursors, ultrafine particles, and boundary layer mixing processes. DGR and MEV-types of new particle formation seem to be more closely related to near-surface processes. Thus, a low-level site such as the reopened Station Nord, (Nguyen et al., 2016), would be more suitable for related mechanistic experiments. Station Nord has the additional advantage of being close to the potential source regions of DGR and MEV-events identified by the present study.

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Parameter	TR (h)	Explanation
C25	1 min	25% percentile of cloud base from AWI ceilometer (m)
CF12, 24, 36, 48, 5D	24	Average MODIS cloud fraction during the last 12h, 24h, 36, 48h, and days 3-5 before trajectory arrival
D50	1	Number-median diameter of particles < 50 nm diameter
CS	1	Condensation sink (s^{-1})
DZ	1	Vertical trajectory displacement ($m\ h^{-1}$) during the last hour before arrival
MSA	≥1 day	Methane sulfonate ($nmolm^{-3}$)
N10	1	Number concentration of particles up to 10 nm (>2010, cm^{-3})
N20	1	Number concentration between 10 and 20 nm (cm^{-3})
N25	1	Number concentration of particles up to 25 nm (cm^{-3})
N40	1	Number concentration between 20 and 40 nm (cm^{-3})
N60	1	Number concentration between 40 and 60 nm (cm^{-3})
N100	1	Number concentration between 60 and 100 nm (cm^{-3})
N300	1	Number concentration between 100 and 300 nm (cm^{-3})
Na	≥1 day	Sodium concentrations ($nmolm^{-3}$)
NCO	1	Number concentration of particles > 300 nm (cm^{-3})
$nssSO_4^{2-}$	≥1 day	Non-sea salt sulfate concentrations ($nmolm^{-3}$)
NTO	1	Number concentration of particles ≥ 10 nm, cm^{-3})
OC12, 24, 36, 48, 5D	120	Average MODIS DMSPt (nmol) during the last 12h, 24h, 36, 48h, and days 3-5 before trajectory arrival
OW12, 24, 36, 48, 5D	24	Average open water (%) during the last 12h, 24h, 36, 48h, and days 3-5 before trajectory arrival
PRE	1	Trajectory precipitation (mm) at arrival
RH	1	Trajectory relative humidity (%) at arrival
SFL	1	Solar flux at trajectory arrival (Wm^{-2})
SP12, 24, 36, 48, 5D	1	Accumulated precipitation (mm) during the last 12h, 24h, 36, 48h, and days 3-5 before trajectory arrival
T12, 24, 36, 48, 5D	24	Average sea surface temperature (C) during the last 12h, 24h, 36, 48h, and days 3-5 before trajectory arrival
TEM	1	Trajectory temperature (C) at arrival
WDR	1	Trajectory wind direction (°) during the last hour before arrival
WSP	1	Trajectory wind speed ($m\ sec^{-1}$) during the last hour before arrival

Table 1 Aerosol, atmospheric, and ocean parameters utilized in the present study. DMSPt = Total dimethylsulfoniopropionate in surface ocean waters. TR = temporal resolution in which the respective data were available.

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Approach	**Acronym**	**Criteria and thresholds**	**Total number of events**	**Number of unique events**
Percentiles	PCT	N25 >93%-percentile	4143	240
Diameter-growth	DGR	D50-Growth >1.5	1199	235
Multi-size growth	MEV	Multi-growth >1.6	1191	266
Sum			6533	741

Table 2 Total and unique number of events of new particle formation identified by the three approaches to identify NPF-events.

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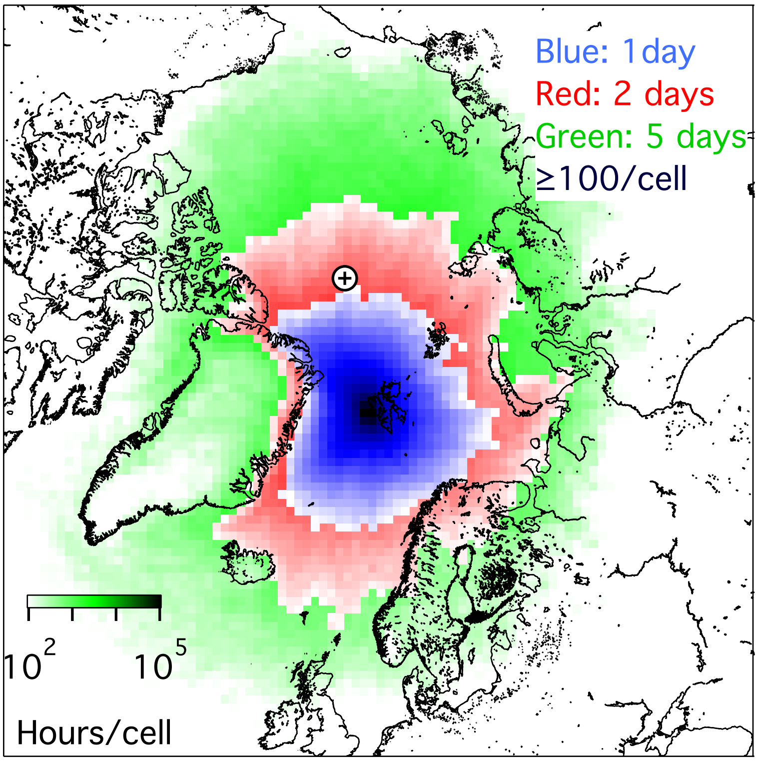
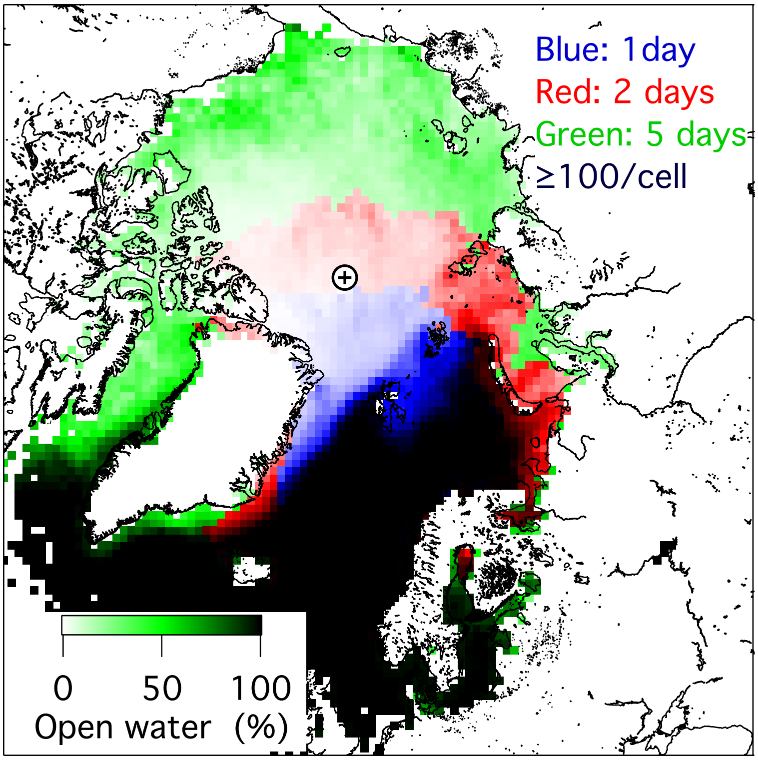


1213

1214 Fig. 1 Map of the regional distribution of 5-day (green), 2-day (red), and 1-day (blue) hourly back trajectories to Mt. Zeppelin during the months March through October of the years 2006 - 2015. Black symbol: North Pole. The colored areas are covered with at least 100 trajectory hours per geocell and the color saturation corresponds to the number of trajectory hours per grid cell on a log-scale.

1219



1220

1221 Fig. 2 Map of the regional distribution of open water under 87648 5-day (green), 2-day
1222 (red), and 1-day (blue) hourly back trajectories to Mt. Zeppelin during the during the
1223 months March through October of the years 2006-2015. Black symbol: North Pole.
1224 The areas are covered with at least 100 trajectory hours concurrent with data values
1225 per geocell.

1226

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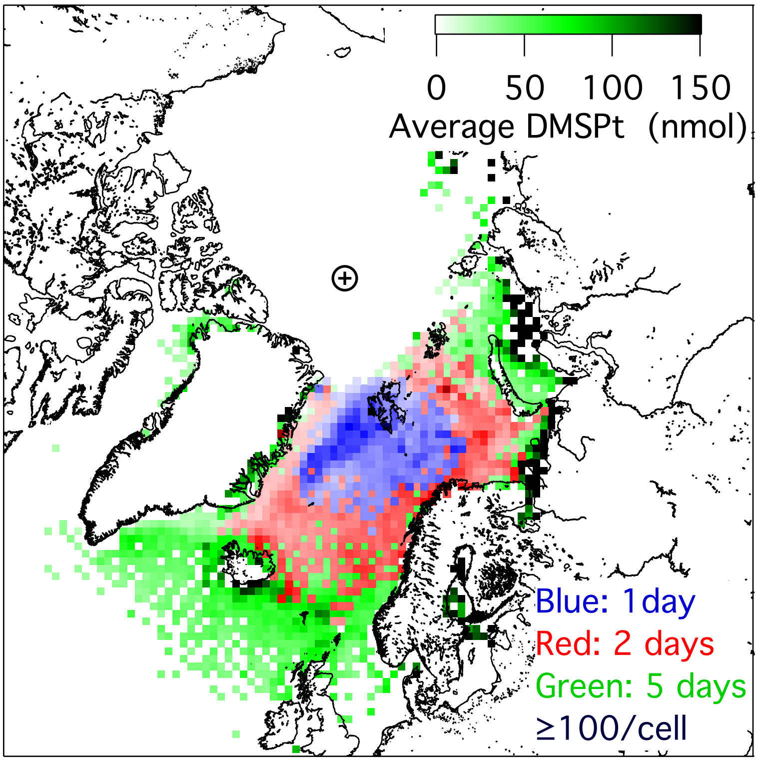

1227

1228 Fig. 3 Map of the regional distribution of DMSPt along 87648 5-day (green), 2-day (red),
1229 and 1-day (blue) hourly back trajectories to Mt. Zeppelin during the during the
1230 months March through October of the years 2006-2015. Black symbol: North Pole.
1231 The relative color scale holds for all colors. The areas are covered with at least 100
1232 trajectory hours with data values per geocell.

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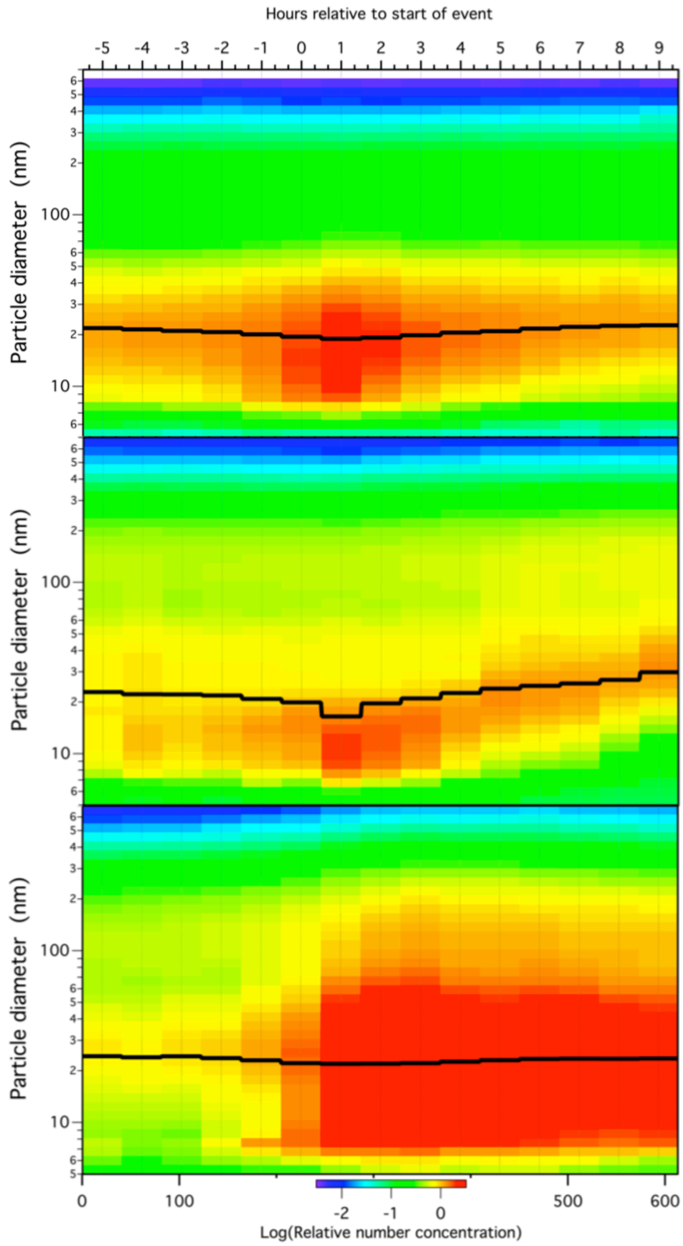

Fig.4 Average temporal development of the relative number size distribution before and during NPF-events identified by the three approaches. The black curve gives the median sub-50-nm particle diameter D50 during the events. Top: PCT-events; center: DGR-events; bottom: MEV-events.

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Atmospheric Chemistry and Physics
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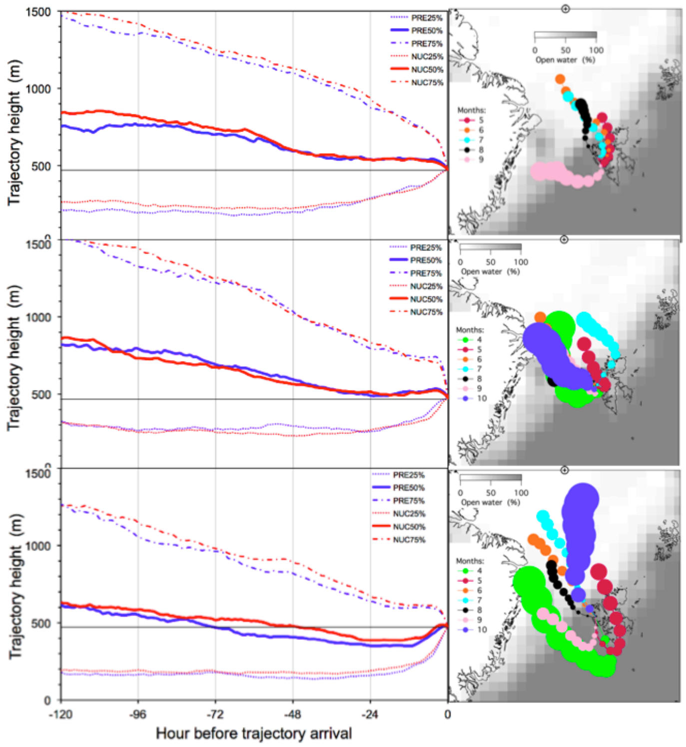

Fig. 5 Left panels: Median back trajectory height profiles (m) during the six pre-event hours (full line in blue, PRENUC) and during the nine DGR-event hours (full line in red, NUC). 25% and 75% percentiles are shown as dotted, and dash-dotted lines, respectively. Top: PCT-events; center: DGR-events; bottom: MEV-events. The thin horizontal line marks the station level.

Right panels: Average monthly trajectory positions in 12 h steps for the months April though October. Only months with at least 10 NPF-events are shown. The circles comprise 95% of all trajectories at any trajectory step. The underlying grey-scale map indicates July ice cover averaged over the years 2006 – 2015

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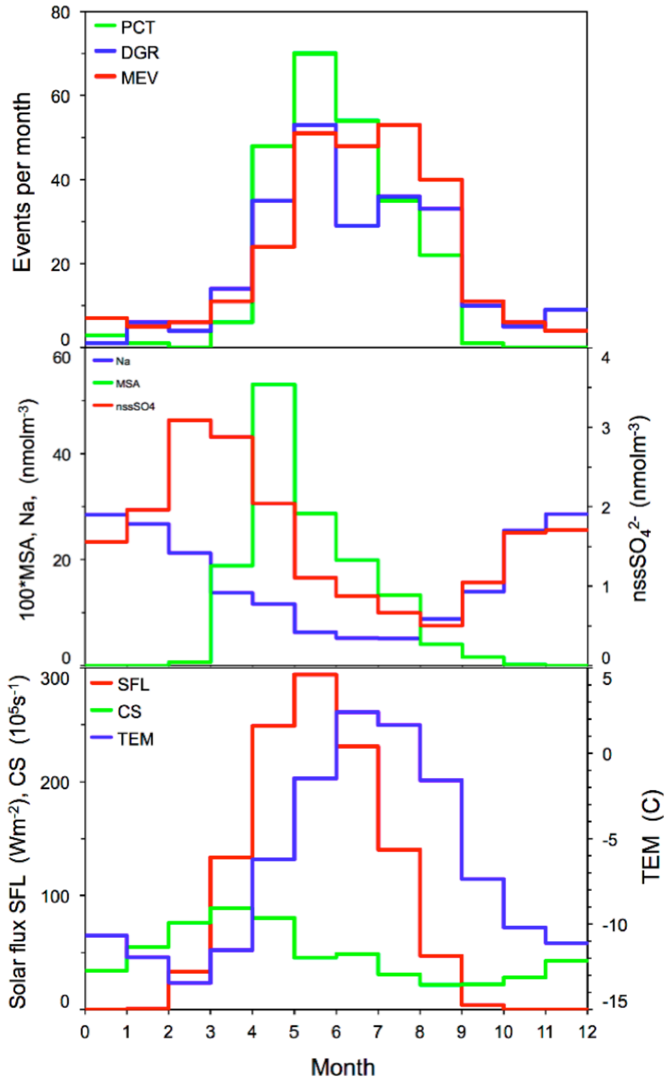

Fig. 6 Top: Monthly numbers of new particle formation events according to the three approaches. PCT: Upper percentile of N25; DGR: Diameter growth; MEV: Multiple size events.

Center: Average seasonal distribution of particle composition in nmolm^{-3}. Na = sodium, nssSO4 = nssSO_4^{2-}, MSA = Methane sulfonate times 100.

Bottom: Monthly average solar flux (SFL, red, Wm^{-2}), and temperature (TEM, blue °C), and condensation sink, (CS, $10^5 s^{-1}$), at Mt. Zeppelin, Spitsbergen.

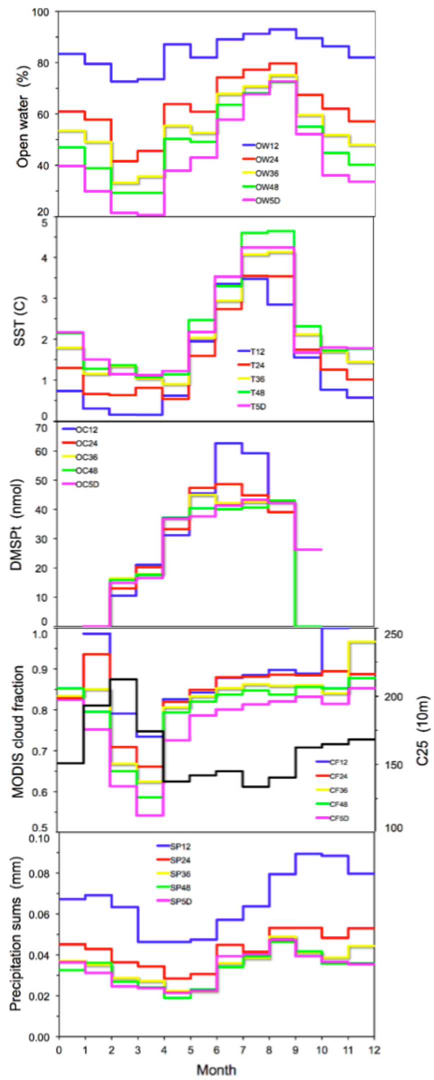

Fig. 7 Monthly averages of environmental parameters averaged along back trajectories to Mt. Zeppelin. From top to bottom: OW12-5D: Open water in % during last 12, 24, 36, and 48h, and days 3-5 before trajectory arrival at Mt. Zeppelin. T12-5D: Same for sea surface temperature in °C. OC12-5D: Same for DMSPt in nmol in surface waters. CF12-5D: Same for MODIS cloud fraction. SP12-5D: Same for precipitation sums in mm. C25 = 25%-percentile of cloud base in decameter.

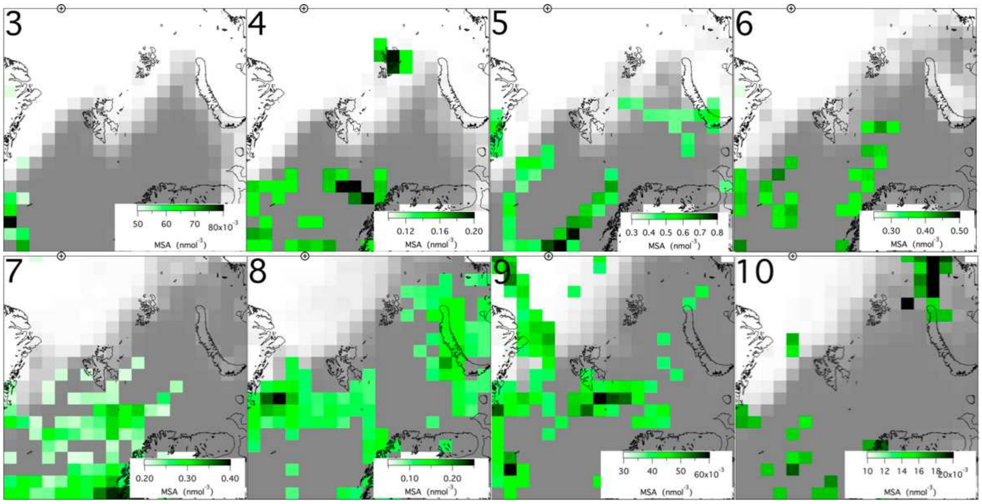

Fig. 8 Average monthly distribution of methane sulfonic acid, (MSA, nmolm^{-3}), during the months March – October of the years 2006 – 2015, constructed from MSA-concentrations measured on Mt. Zeppelin, which were extrapolated along 5-day back trajectories. Average open water percentages during the respective months are indicated as white (0% open water) to dark grey (100% open water) areas. The position of the North Pole is marked as cross in circle on the upper border of the maps.

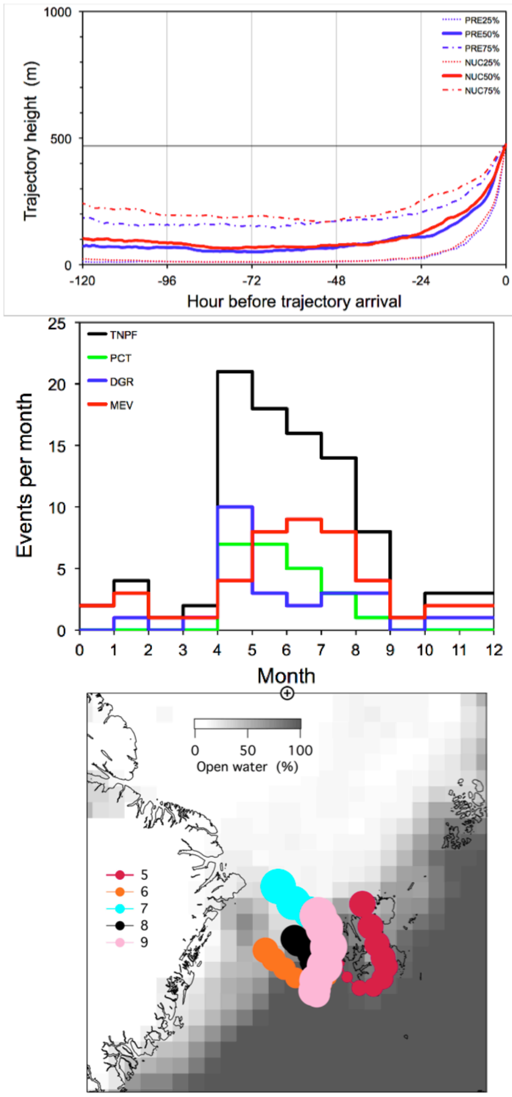

Fig. 9 Characteristics of the subpopulation of 93 NPF-events with back trajectories that stayed below 500 m for five days before arrival. Top: Statistics of related vertical trajectory coordinates as in Fig. 5. Center: Average monthly occurrence of PCT, DGR, and MEV-events. Bottom: Related average monthly trajectory positions in 12 h steps for the months May through September. The circles comprise 95% of all trajectories at any trajectory step. The underlying grey-scale map indicates July ice cover averaged over 2006 – 2015.

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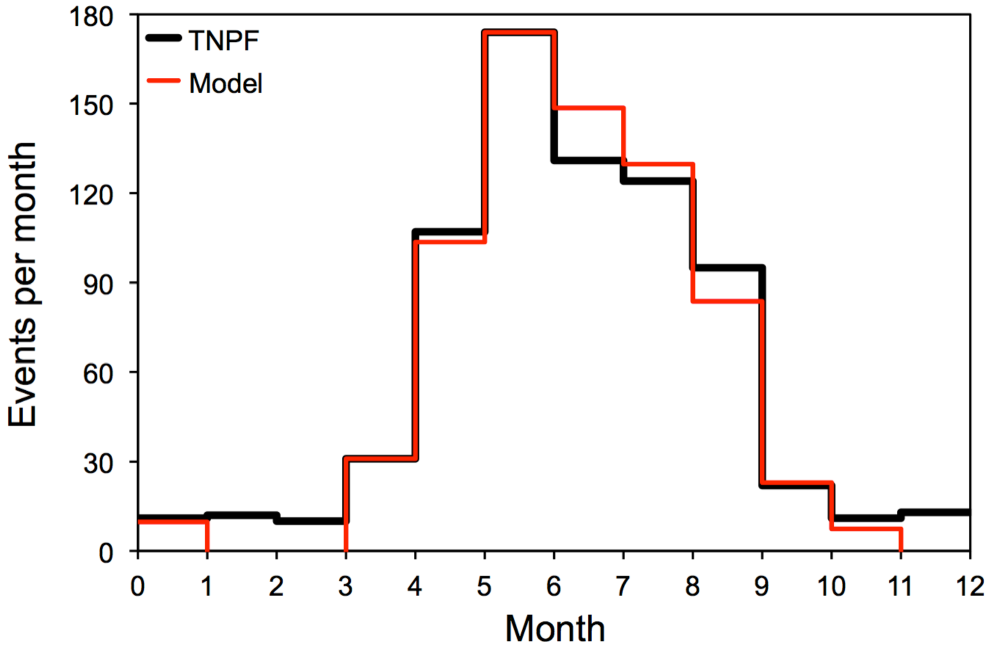
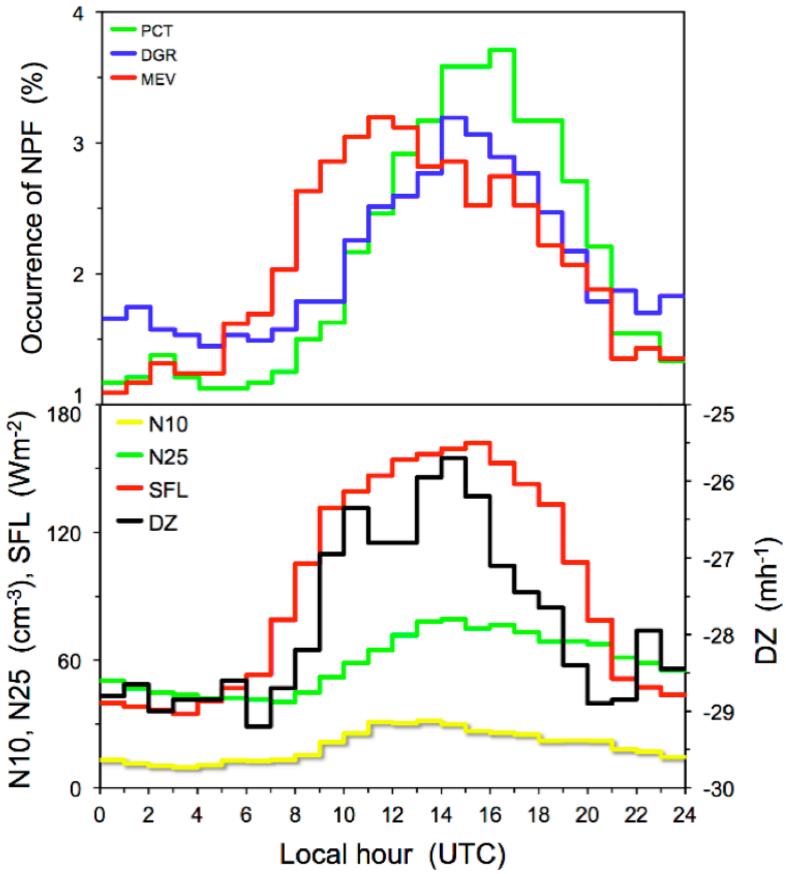


Fig. 10 Average monthly sums of NPF-events due to the three types of new particle formation, (TNPF, black). Red: Three-parameter model to describe TNPF.



1291

1292 Fig. 11 Top: Relative average diurnal occurrence of the three types NPF-events. PCT: Upper percentile of N25; DGR: Diameter growth; MEV: Multiple size events.
1293
1294 Bottom: Average diurnal variation of the HYSPLIT-modeled solar flux (SFL, Wm^{-2}),
1295 the integral particle concentrations N10, and N25 in cm^{-3}, and of the vertical
1296 displacement parameter (DZ, mh^{-1}). N10 is based on data of the years 2011 – 2015
1297 whereas the other parameters are based on data of the years 2006 – 2015.