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Arctic ship-based evidence of new particle formation events in the Chukchi and East Siberian Seas

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HIGHLIGHTS

- Particle size distributions were collected during an Arctic ocean cruise.
- Elevated concentrations of aerosols (N < 100 up to 3,000 cm^{-3}) were found.
- New particle formation events associated with open ocean and sea ice regions.

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ABSTRACT

Arctic aerosol-climate interactions are controlled by multiple factors including sources, processes and removal mechanisms of particles. The Arctic is mostly ocean, surrounded by mostly land, and our understanding of Arctic aerosol processes is incomplete due to scarce measurements carried out in sea ice regions. In particular, it is currently not known if these particular regions are sources of aerosols of primary or secondary origin. We present new results from ship-based measurements illustrating that marine new particle production and growth events occur in open ocean and melting sea ice regions in the Chukchi and East Siberian Seas. We report two new particle formation events during which a recently formed nucleation mode (<15 nm diameter) is detected and is observed to slowly grow into an Aitken mode (0.1–3.8 nm h^{-1}). Our results suggest that new particle formation occurs in the marine boundary layer contributing to the Arctic aerosol population in the study region for the first time studied and herein reported.

1. Introduction

In order to understand the climate system and to estimate the impact of humans on climate change, it is imperative to apportion the natural versus the anthropogenic component of the total aerosol (Hamilton et al., 2014). The continuous decrease of the Arctic sea ice extent - caused by the variability of the warming climate - amplifies the control that the ocean has on the atmospheric composition. Key factors in aerosol-climate interaction include: sources, chemical transformations and mechanisms that remove aerosols (Carslaw et al., 2013; Abbatt et al., 2019). One of the largest remaining uncertainties in climate change is the impact of aerosol particles on the formation and microphysical properties of clouds (Carslaw et al., 2013). The aerosol population making cloud condensation nuclei (CCN) depends upon the ambient aerosol particle size distribution (PSD) and the associated chemical composition (Farmer et al., 2015). In the Arctic, two main seasonal patterns dominate the cycle of the aerosol concentration, size and composition. In the winter and spring, accumulation mode aerosols dominate (Tunved et al., 2013; Lange et al., 2018). In contrast, the number size distribution during summer is dominated by nucleation and Aitken mode particles; natural boundary layer local Arctic aerosol sources dominate the summer population relative to long range

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continental transport (Leaitch et al., 2013; Heintzenberg et al., 2015; Dall'Osto et al., 2017a). Air pollution in the Arctic caused by local emission sources is also a challenge that is important but often overlooked (Schmale et al., 2018). Current knowledge on the composition and sources of summer aerosols is insufficient.

Furthermore, we still know too little on aerosol lifetime and removal near mid latitude regions, as well as during transport (Willis et al., 2018; Abbatt et al., 2019). To tackle this problem, an increasing number of studies are being carried out, and a consensus seems to be emerging that marine and snow or ice-related sources are the main candidates for production of biogenic precursors forming new particles (Willis et al., 2018; Abbatt et al., 2019). However, the relative importance of primary wind-driven particle production at the ocean surface over secondary (gas-to-particle conversion) production to Arctic marine cloud formation remains unclear. Primary sea spray related to marine polymeric gels produced by phytoplankton and sea-ice algae biological secretions have been reported in the polar atmosphere (Leck and Bigg, 2005; Orellana et al., 2011), although more recent evidence suggests that in situ formation of new aerosol particles via secondary processes from emissions of biogenic volatile species may dominate the aerosol population in the Arctic (Fu et al., 2013; Tunved et al., 2013; Heintzenberg et al., 2015; Burkart et al., 2017a,b; Mungall et al., 2017). These secondary processes are expected to increase in the future, given that the summer-ice coverage is decreasing due to Arctic warming. Indeed, air mass trajectory analysis has linked frequent nucleation events to biogenic precursors released by open water and melting sea ice regions, especially during the summer season (Dall'Osto et al., 2017a,b; 2018 a).

It is important to mention that ultrafine particles have been measured previously and extensively in different locations throughout the Arctic including Alert, Canada (Leaitch et al., 2013), Ny Alesund and Zeppelin on Svalbard islands (Engvall et al., 2008; Ström et al., 2003; Tunved et al., 2013), Tiksi, Russia (Asmi et al., 2016), Eureka and Alert on Ellesmere Island in Nunavut, Canada (Tremblay et al., 2019), Utqiaġvik (Barrow, Polissar et al., 2001; Freud et al., 2017; Kolesar et al., 2017), and Station Nord, Greenland (Nguyen et al., 2016). All the studies show a strong shift to smaller particles during the summer months relative to winter. Recently, an emerging multi-year set of observed aerosol number size distributions (10–500 nm) from five sites around the Arctic Ocean (Alert, Villum Research Station – Station Nord, Zeppelin, Tiksi and Utqiaġvik) was assembled and analysed (Freud et al., 2017).

Three different sites (Zeppelin research station and the nearby Gruvebadet Observatory in the Svalbard archipelago; and Villum Research Station at Station Nord, 600 km west-northwest of Zeppelin, at the tip of northeastern Greenland) across a more specific area in the Arctic North Atlantic sector were also recently compared (Dall'Osto et al., 2019). It was argued that there is no single site in the Arctic that can be considered as fully representative for the entire Arctic region. Furthermore, despite extensive studies into Arctic aerosol sources and various hypothesized species involved, very few studies have reported ship-borne Arctic measurements suggesting that such events occur only under particular or exceptional conditions. Despite the "NETwork on Climate and Aerosols: addressing key uncertainties in Remote Canadian Environments" (NETCARE) (Abbatt et al., 2019) and the Arctic Summer Cloud Ocean Study (ASCOS) (Tjernström et al., 2014) programmes - carried out with the Swedish ice breaker Oden and the Canadian Coast Guard Ship ice breaker Amundsen - there are large areas of the Arctic ocean that remain unexplored where aerosol measurements have never been made to date. Motivated by the need to further understand sources of Arctic aerosols - especially in open ocean areas - we present open ocean ship-borne measurements of aerosol size distributions obtained on board of the RV Araon (KOPRI polar research vessel) in the Chukchi and East Siberian Seas (CESS), Pacific Arctic Ocean.

The East Siberian Sea is a marginal sea in the Arctic Ocean located between the Arctic Cape to the north and the coast of Siberia to the south. The Chukchi Sea is bounded on the west by the Long Strait, and in the east by Utqiaġvik, Alaska, beyond which lies the Beaufort Sea. This study analyses and probes an open ocean aerosol size distribution dataset for occurrence of open ocean events as opposed to events detected near coastal areas. Atmospheric aerosols over the Northwest Pacific Ocean, the Bering Sea, and the Arctic Ocean polar regions have attracted considerable attention for their effects on climate change. Previous measurements around the CESS include Total Suspended Particles (TSP) collected to study the impacts of Siberian biomass burning on organic aerosols (Ding et al., 2013), the impacts of secondary organic aerosols over oceans via oxidation of isoprene and monoterpenes (Hu et al., 2013), the seasonal variations of biogenic secondary organic aerosol tracers in ambient aerosols from Alaska (Haque et al., 2016), the spatial distribution of Methanesulphonic Acid in the Arctic aerosol collected during the Chinese Arctic Research Expedition (Ye et al., 2015). Bulk TSP measurements were also reported over Chukchi Sea and Bering Sea (Zhu et al., 2004). By means of High Resolution Time of Flight Aerosol Mass Spec-trometer (AMS), Choi et al. (2017) reported PM_1 aerosol concentrations and composition in the North Pacific marine boundary layer; the MSA/sulphate ratio showed a sharp gradient approach to clean marine condition. As regards of size resolved particle number concentrations, a number of measurements in the coastal site of Utqiaġvik (Barrow) exists (Polissar et al., 2001; Freud et al., 2017). Recently, a similar numbers of Arctic particle growth events were influenced by marine (46%) and Prudhoe Bay air masses (33%) (Kolesar et al., 2017). Measurements of aerosol size distributions and aerosol composition were also recently taken during the summers of 2015 and 2016 at coastal measurement sites Eureka and Alert on Ellesmere Island in Nunavut, Canada (Tremblay et al., 2019). Kim et al. (2015) previously reported ship-borne measurements of ambient aerosols were conducted over the Arctic Ocean and the Pacific Ocean. Time series of $N_{>2.5nm}$ and $N_{2.5–10nm}$ were reported, concentrations as high as 4 10^5 (particle cm^{-3}) were reported but not clear association with new particle formation was shown. Furthermore, number size distributions in the range 30–600 nm were reported, not allowing obtaining any information on new particle formation events. Results from open ocean icebreaker expedition measurements of physical-chemical characteristics of atmospheric aerosol in areas of the Arctic and Far East seas showed concentrations of aerosol particles with diameters from 0.3 to 20 μm (Sakerin et al., 2015), and from 0.5 to 10 μm (Tian et al., 2019). Ship-borne observations of normalized mass distributions of the refractory black carbon (rBC) component of ambient aerosol particles over the Arctic Ocean, Bering Sea, and North Pacific Ocean were reported by Taketani et al. (2016). Atmospheric black carbon along a cruise path through the Arctic Ocean during the Fifth Chinese Arctic Research Expedition was also reported (Xing et al., 2014).

Our main objective is to characterize ultrafine particle size distributions in the CESS area - an atmospheric environment that can provide a background concentration baseline to compare against future measurements likely affected by a warming world.

2. Experimental measurements

2.1. Study area

Aerosol sampling was conducted onboard the IBRV Araon from 4 August to 11 September 2017 divided in two legs: 4 August to 22 August (first leg) and 30 August to 11 September (second leg). In addition to the onboard sampling, a sea ice field survey was conducted in the CESS during the first leg. The ship track is shown in Fig. 1. Whilst the first leg aimed at studying sea ice field studies, the second leg was mainly focusing on geological studies near shallow waters near the coast of Alaska. In this study, we focus on measurements taken in the open ocean areas close to marginal sea ice regions, showing real time data collected between 8 and 22 August 2017 (Fig. 1, pink line, cruise track).

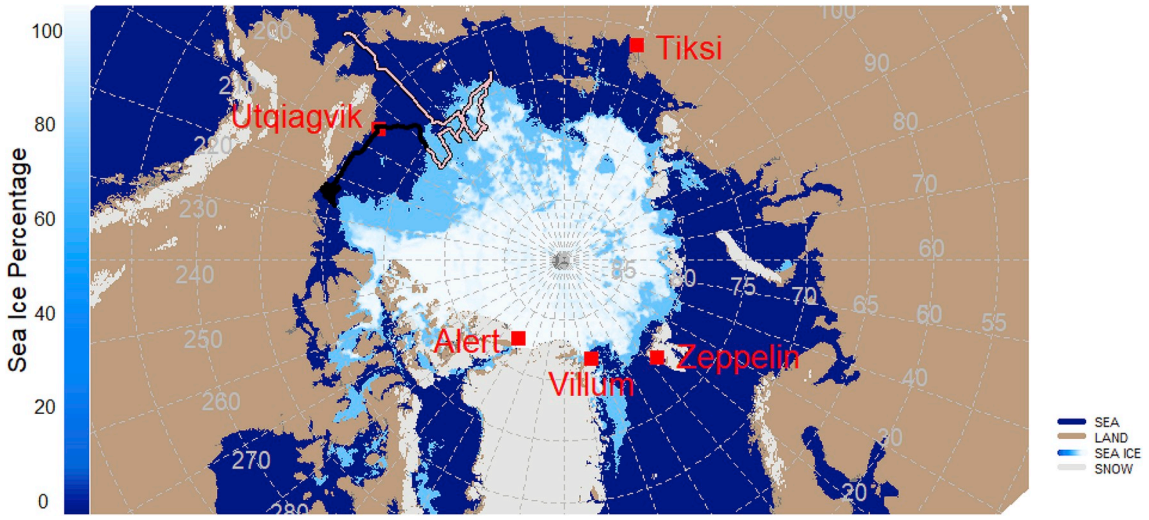


Fig. 1. A physical map of the Arctic region, with the five measurement sites marked (as described in Freud et al., 2017 and Dall'Osto et al., 2019). Sea ice maps (sea ice in light blue and white scale) for the period August 2017 (plotted is the map of 15 th August 2017). Land borders are marked in brown. Snow in dark white, land in brown. RV Araon ship track is shown as rose line (first leg, this study), and shown the rest of the RV cruise (leg 2) on black line, ending in Utqiaġvik (Alaska, USA). Please note that the Gruvebadet Observatory (GRU) in the Svalbard archipelago is at the bottom of the hill of the Zeppelin research station (Dall'Osto et al., 2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2.2. Instruments used

The size distribution of ambient aerosols in the size range 5–60 nm was measured with a nano scanning mobility particle sizer (nano-SMPS) (Differential mobility analyzer (DMA): TSI 3085, CPC: TSI 3776), and in the size range from 8 to 290 nm, the size distribution was measured with a SMPS (DMA: TSI 3081, CPC: TSI 3772). In the nano SMPS, the aerosol and sheath flow rates were 1.5 lpm and 15 lpm, respectively; for the SMPS, the aerosol and sheath flow rates were 1.0 lpm and 10 lpm, respectively. Black Carbon measurements were made by an aethalometer (AE22, Magee Scientific Co., USA), data were collected at 5 min interval, and used to remove atmospheric data contaminated by local ship emissions including diesel generators, cooking emissions, local operation on the ship. Inlet for SMPS was a 1 m long 1/4 inch stainless pipe connecting the SMPS to ambient air via a window looking at the bow of the ship, air was dry with a dryer before entering the SMPS system. All data points with BC higher than 20 ng m^{-3} were removed from the analysis, concentrations used as pristine environments in previous pristine marine environments (Dall'Osto et al., 2011, 2012). Visual inspection of all SMPS data was carried out, removing data points associated to local contamination (e.g. high black carbon concentration, short time spikes of elevated (>5,000 cm^{-3} particle number concentrations). Balloon-borne radiosonde (Vaisala RS-41) observations were made 4 times a day (00, 06,12 and 18 UTCs) to have meteorological vertical profiles of temperature, relative humidity and horizontal winds. Short measurements (1 h) of ultrafine particle number concentrations by means of the diffusion size classifier (DiSC, about N > 10–30 nm, Fierz et al., 2011 - inlet line 1 m 1/4 inch conductive tubing) during a number of helicopter flights (5) during our cruise were also carried out, providing vertical particle number concentrations during part of our field study. Such helicopter flights (carried out mainly during the period 12-15th August 2017) were not made during the NPF events herein described, and the DiSC aerosol particle concentration data are described here only in a qualitative manner.

2.3. Air mass back trajectories analysis

Using the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT), two day back trajectories arriving at the ship (400 m) were calculated at hourly resolution. The length of the back-trajectory calculation was chosen as a balance between the typical lifetime of the aerosols in the Arctic troposphere in the summer for the particles, and the increasing uncertainty in the calculation the further back in time it goes (Tunved et al., 2013). These were calculated based on meteorological files selected from the NCEP/NCAR Reanalysis Project which is a joint project between the National Centers for Environmental Prediction (NCEP, formerly "NMC") and the National Center for Atmospheric Research (NCAR). The REANALSYS meta files consist of 2.5° latitude-longitude global 144x73 grid of points covering from 90N–90S, 0E-357.5E from 1/1/1948 - present with output every 6 h.

For each of the positions along each of the trajectories, the surface information was logged into a file off daily maps indicating whether the air mass was passing over land, sea, sea ice or snow. Surface coverage maps (4 km and 24 km resolution) were produced by National Ice Center, 2008. Similarly, a sea ice concentration was selected for each step along the trajectories using daily 12.5 km resolution ice concentration maps collected by the Special Sensor Microwave Imager (SSM/I) (Ezraty et al., 2007, ftp:/ifremer/cersat/products/gridded/psi-concentration/data/arctic/daily/).

2.4. K-means cluster and PMF PSD analysis

After removing local ship contamination, a relatively small data set of 193 hourly aerosol size distributions were cluster analysed (Beddows et al., 2009, 2014, Dall'Osto et al., 2018 b). Four clusters were selected to best represent the data, giving a Dunn Index of 0.016 and Silhouette Width of 0.36. However, a statistically higher number of clusters would be preferred. A Dunn-Index of 0.018 and 0.033 was observed at 15 and 16 clusters respectively with Silhouette Width of at 0.37 and 0.42. This indicated that as the number of clusters was increased from 4 to 15–16 clusters, the separation of the clusters increased and the similarity of the elements within each cluster also increased. In this case, due to the small data set, the view was taken that there was a tendency for the data to be 'over-clustered' and that a smaller number of clusters gave a better description of the environment. With this in mind, the cluster result presented is simply a means with which to present the data by splitting the data into 4 rather than presenting it as 4 natural clusters within the data.

3. Results

3.1. SMPS overview and clustering results

SMPS data were averaged in hourly bins, and local ship emissions were removed from the current analysis. For the first leg of the RV Araon herein presented, the data coverage was 193 h (47% of the time). Fig. 2 shows the average size distributions for the entire period of study, superimposed with the monthly average concentrations obtained at three stations (Gruvebadet (GRU), Zeppelin (ZEP) and Villum Research Station at Station Nord (VRS)) and recently discussed in Dall'Osto et al. (2019). Data from GRU, ZEP and VRS were simultaneously collected for three whole years (2013–2015), in Fig. 2 the average for the month of August is reported (as shown also in Fig. 2h in Dall'Osto et al., 2019). Whilst the sites in the Svalbard islands (GRU, ZEP, green and blue lines, Fig. 2) show similar aerosol size distributions peaking at about 41 nm, the VRS site (North East Greenland) shows about 35% lower particle number concentrations, peaking at a smaller mode (31 nm, grey line, Fig. 2). The aerosol size distribution found for this study detected on board the RV Araon shows a strong Aitken mode at 30±5 nm (red line, Fig. 2). However, the mode is reduced in particle number concentrations by about 50% relative to the VRS monitoring site, suggesting Greenland coastal sources may be responsible for this difference. In other words, the size distributions collected in the open ocean (this study) is broadly similar with the one characterizing the VRS site, with a peak at about 33±5 nm nm. This is reasonable, as broadly out of the three fixed monitoring sites, VRS is the one more confined in sea ice regions, hence the greater similarity to the sea ice regions analysed in this work. As discussed in Tunved et al. (2013) and Dall'Osto et al. (2019), there is a shift from about 20 to 30 nm (June) to about 40–50 nm (August), due to a number of factors including higher nucleating gas and precursor concentrations and reduced condensation sink dominating the summer months. It is interesting to note that the particle size distributions in the accumulation mode (>100 nm, particularly >200 nm) are fairly similar aerosol modes among the four study areas (although double in particle number concentrations), the reasons may be multiple and are unknown at this stage.

In order to more fully elucidate the processes affecting aerosol size distributions, we used a statistical tool - K-means clustering - to reduce the complexity of this open ocean SMPS dataset. Four K-means clusters were obtained, the temporal abundance did not vary much among them (18–31%). The average size distributions are shown in Fig. 3, and it is likely that a number of individual sources and processes contribute to the overall shape of the size distributions. However, the name of the four clusters is kept the same as that used in previous work; additional information can be found elsewhere (Dall'Osto et al., 2017a, b, 2018 a, b; 2019). Briefly, Cluster 1 (Fig. 3, green line) shows an average number size distribution with an ultrafine mode peaking at about 25 nm. This

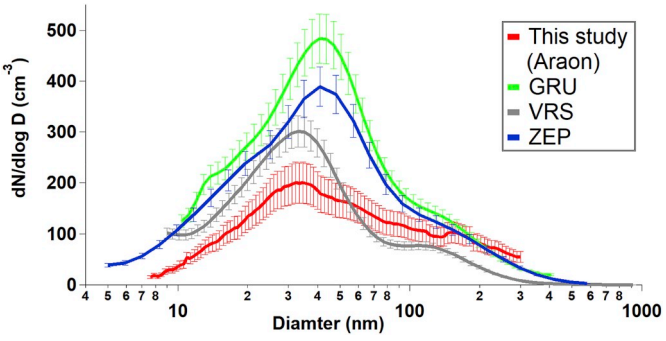


Fig. 2. Monthly average size distributions taken at the three sampling sites for the period August 2017 from three monitoring sites (GRU, ZEP, VRS) described in details in Fig. 2h in Dall'Osto et al. (2019), and average size distribution for this study (7–23 August 2017, RV Araon).

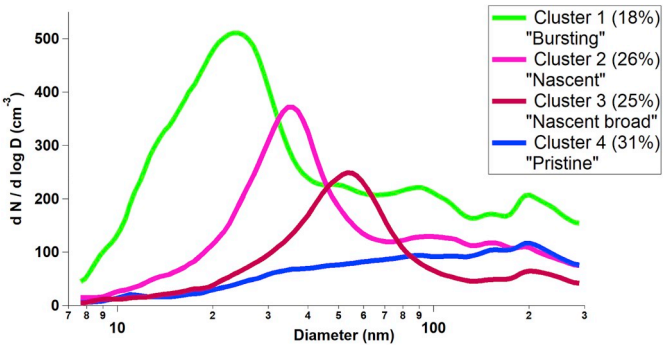


Fig. 3. Average number aerosol size distributions for each of the K-means groups. Aerosol size distributions are for: cluster 1 (green), cluster 2 (violet), cluster 3 (dark violet) and cluster 4 (blue). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Arctic size distribution was previously referred as the "Bursting" category: an aerosol population that begin to exist if it is developing to larger sizes. Multiple origins may contribute to this cluster shape, including NPF with limited growth, open ocean nucleation events, possibly also an Arctic ultrafine primary origin, although recent studies show that ultrafine aerosols mainly of secondary origin dominate the Arctic aerosol population (Willis et al., 2018; Abbatt et al., 2019). Clusters 2 and 3 (Fig. 2, dark and bright magenta) can be attributed to two Aitken categories ("Nascent" and "Nascent broad"), showing similar particle size distributions (peaking at 35 nm and 55 nm, respectively) and contributing altogether to 51% of the sampled aerosol (26% and 25%, respectively). The name nascent was previously discussed (Dall'Osto et al., 2017 a,b; 2018 a,b), emerging from different primary and secondary aerosol processes linked to emissions of local and regional marine origin. Finally, Cluster 4 is characterized by very low particle number concentrations (<100 particles cm^{-3}, previously categorized as "Pristine" ultrafine, with three minor modes at 90 nm, 150 nm and 200 nm (Fig. 3, blue line), contributing 31% of the hourly aerosol size distributions characterized.

In summary, the K-means categorization was in line with previous studies carried out at fixed monitoring stations across the Arctic (Fig. 2, Freud et al., 2017; Dall'Osto et al., 2019). An important conclusion from this study is that about 18% of the time where SMPS measurements were taken, a nucleation mode with a mean diameter of 23 nm was detected, implying NPF events can occur in open ocean and sea ice regions. This is the first of this type of study in the Chunkchi and East Siberian seas carried out on board of an icebreaker vessel. It is also worth noting that - as briefly mentioned earlier - two sets of studies using research vessels cruising the Arctic seas detected nucleation mode particles. The first was carried out during the Arctic Summer Cloud Ocean Study (ASCOS, Tjernström et al., 2014), and was mainly attributed to a marine primary biological source of particles from the surface microlayer in open-water leads, also reported in previous ASCOS studies. Recently, during the "NETwork on Climate and Aerosols: addressing key uncertainties in Remote Canadian Environments" (NETCARE) (Abbatt et al., 2019), a number of NPF events were detected, although these were close to the Arctic Canadian Archipelagos and likely affected by coastal sources including bird colonies (Croft et al., 2016). In the next section we report two case studies of two NPF events detected in the sea ice marginal zone of the Chukchi and East Siberian seas.

3.2. Open ocean NPF event case studies

3.2.1. First NPF event

The first new particle nucleation event was detected on the 9th August 2017. Fig. 4 a-b shows that air mass back trajectories were travelling most of the time over open waters and sea ice regions (83%

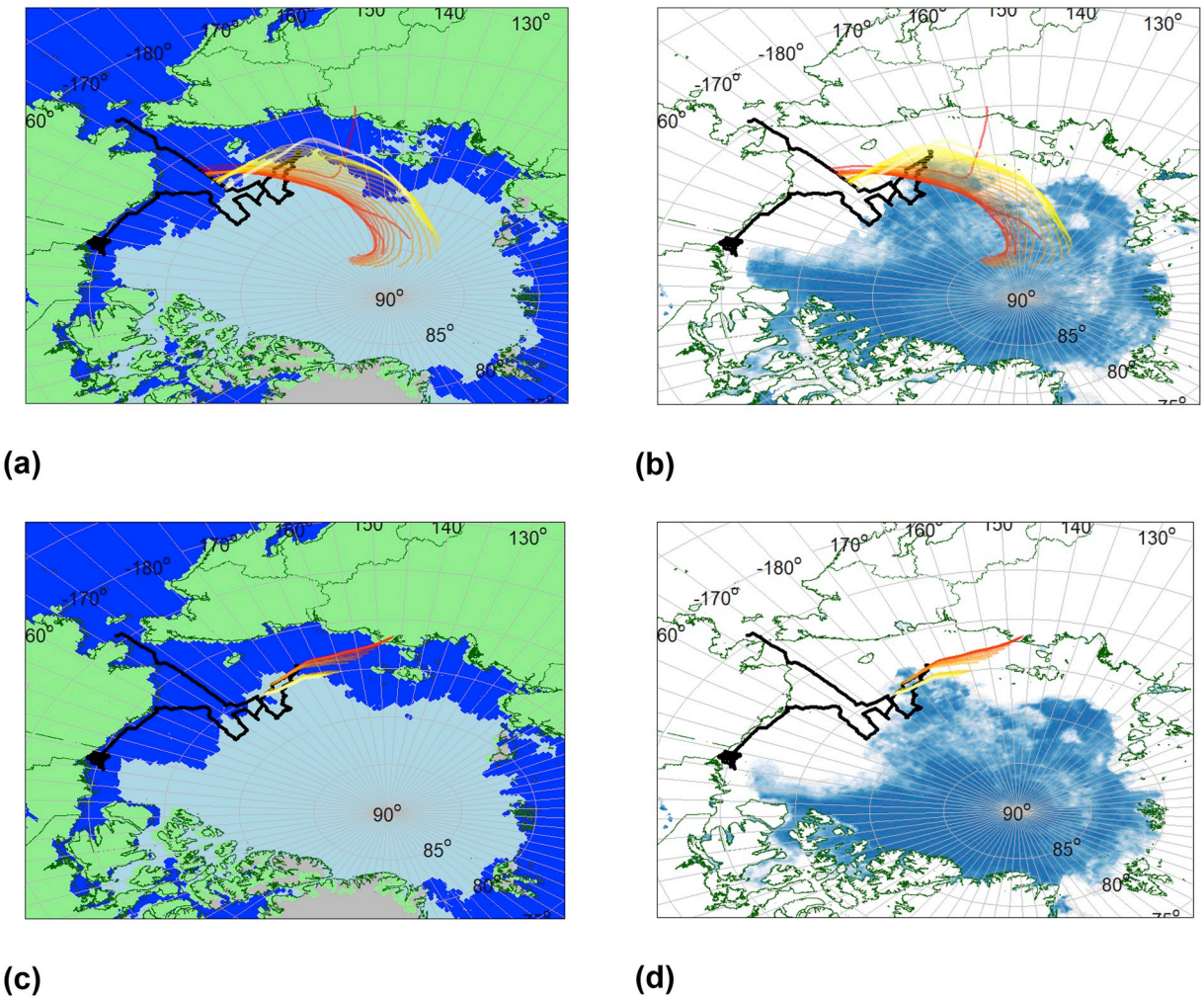


Fig. 4. a-d. Backward trajectories during two open ocean nucleation periods detected on 9th (a, b) and 20th August (c, d) 2017. Fig. 4 a and c show different regions as land (green), open ocean (bark blue), sea ice (bright blue) and snow (grey). Fig. 4 b and d shows sea ice distributions (0–100% where 0% is white and 100% is blue). Maps for the days of 9th August and 20th August 2017 are taken from the NOAA/NESDIS Interactive Multisensor Snow and Ice Mapping System (IMS) and plotted in Fig. 4 a-b and 4 c-d, respectively.

and 17%, respectively) for the 48 h previous to reaching the location of the RV Araon. Fig. 5a shows the temporal trends detected with the nano-SMPS (5–60 nm) and SMPS (8–290 nm), as well as meteorological parameters and air mass back trajectory history. A nucleation mode peaking at about 17 nm is seen appearing at about 9am on the 9th August, slowly growing till about midnight at 35 nm, resulting in a growth rate of 1.2 nm per hour. Following that, a much slower growth is noticed till about 6pm the following day (10th August), reaching a stable mode at about 39 nm (0.22 nm h^{-1} growth rate).

Vertical profiles of meteorological variables were obtained by balloon profiling every 6 h during the RV Araon cruise. Fig. 6a shows that the first NPF event was characterized by dry air mass (Fig. 6a, about 85% RH), from cold regions (Fig. 6b, temperature), with strong constant winds coming from the North West sector (260-300°) at about 8 m s^{-1} (Fig. 6 c, d). Previous studies at various locations showed that NPF events take often place in concomitance with dry air masses, supporting the current study (Laaksonen et al., 2009; Hamed et al., 2011).

3.2.2. Second NPF event

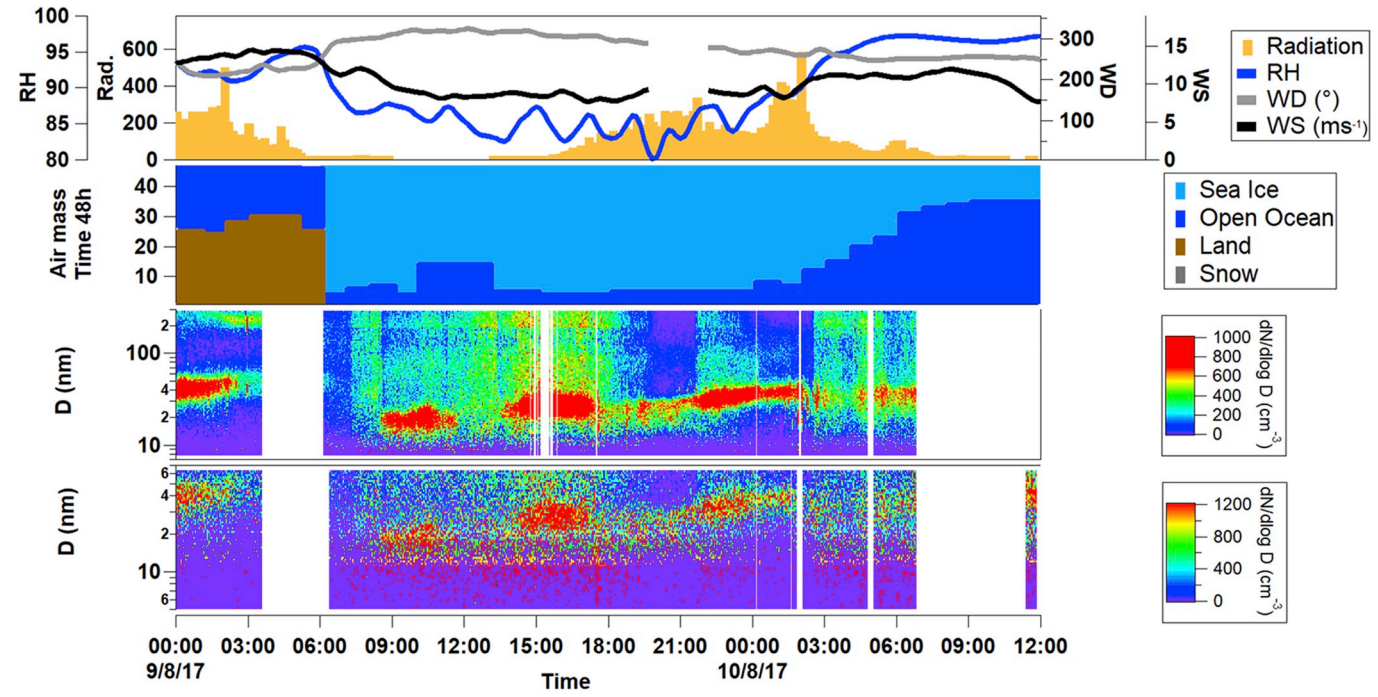
The second nucleation event was detected on the 20th of August 2017. Air mass back trajectories (Fig. 4c and d) showed a possibly coastal origin about 48th before reaching the RV Araon, therefore crossing regions mainly of sea ice (90% of the time) and open ocean waters (the remaining 10% of the time). Fig. 5b shows a nucleation mode at about 21 nm appearing at around 2am, and growing till about 8 am at a size of about 48 nm, resulting growth rate of 3.8 nm h^{-1}. This is much higher than the average growth rate reported for previous Arctic studies, of about 0.1–0.7 nm h^{-1} (Willis et al., 2018). The mode is seen for an additional 3 h, until about 11am, without further growth. Fig. 7 shows vertical profiles for the period of the detected event: drier air masses (Fig. 7a), colder temperature (Fig. 7b), low wind speeds from the North West region (Fig. 7c and d).

4. Discussion

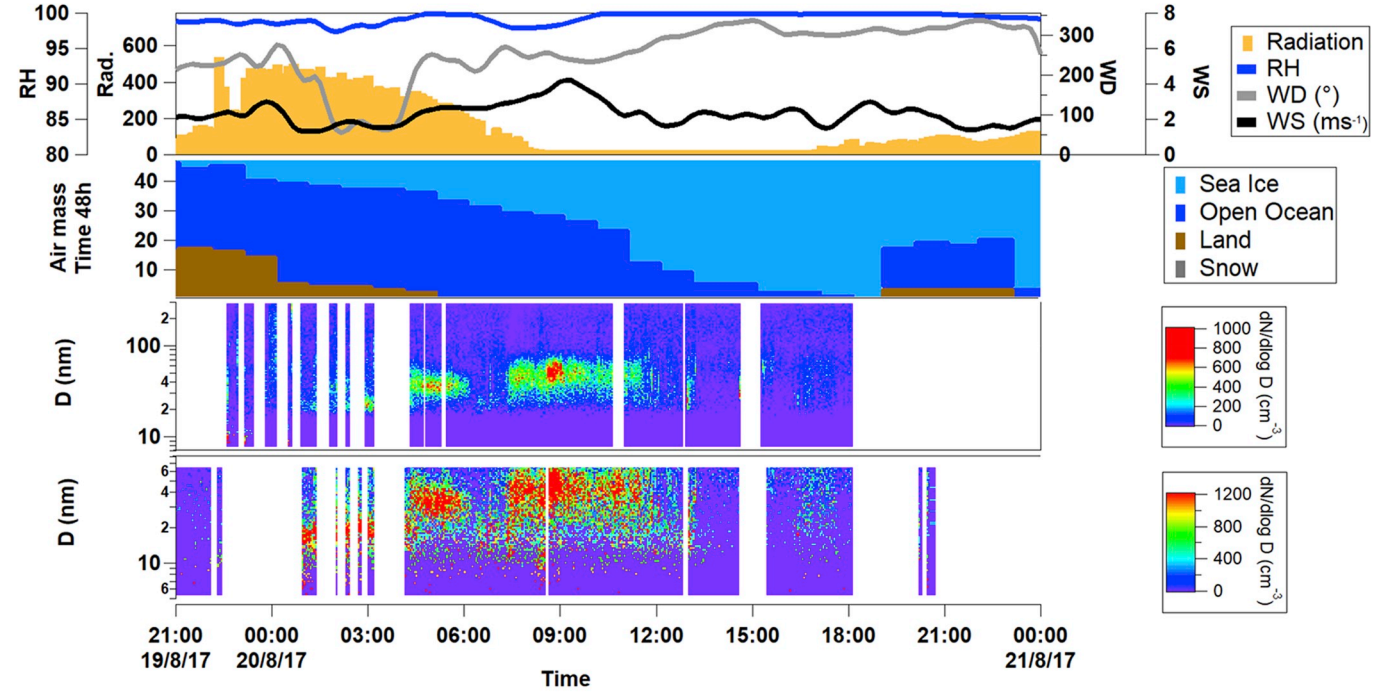
4.1. Detection of open ocean NPF events in sea ice marginal zones

There is increasing evidence showing that secondary gas-to-particle aerosol formation drives the aerosol population in the Arctic (Willis et al., 2018). However, most of the available measurements have been taken in a number of fixed monitoring station in Arctic coastal areas (Freud et al., 2017; Dall'Osto et al., 2019), where seabird colonies (Croft et al., 2016) and intertidal zones (Allan et al., 2015; Sipilä et al., 2016) may also contribute to aerosol sources. However, emissions of precursor gases associated with biological communities on or near sea ice margins may dominate (Dall'Osto et al., 2017a; b; Levasseur, 2013).

The summer sea ice extent has been retreating dramatically over the past decades; some studies report the possibility that the Arctic may be



(a)



(b)

Fig. 5. a-b Number-size distributions with nano-SMPS and SMPS (bottom) and meteorological variables (top) temporal trends (UTC time) for two new particle formation events detected on (a) 9th August 2017 and (b) 20th August 2017. During both NPF events, air mass back trajectories travelling over sea ice regions were mostly (>95% of the time) travelling over open pack ice (regions with sea ice concentration higher than 15% and lower than 80% within the consolidated ice region), with an only very minor part (<5% of the time) travelling over consolidated pack ice (regions with pack ice concentration higher than 80%).

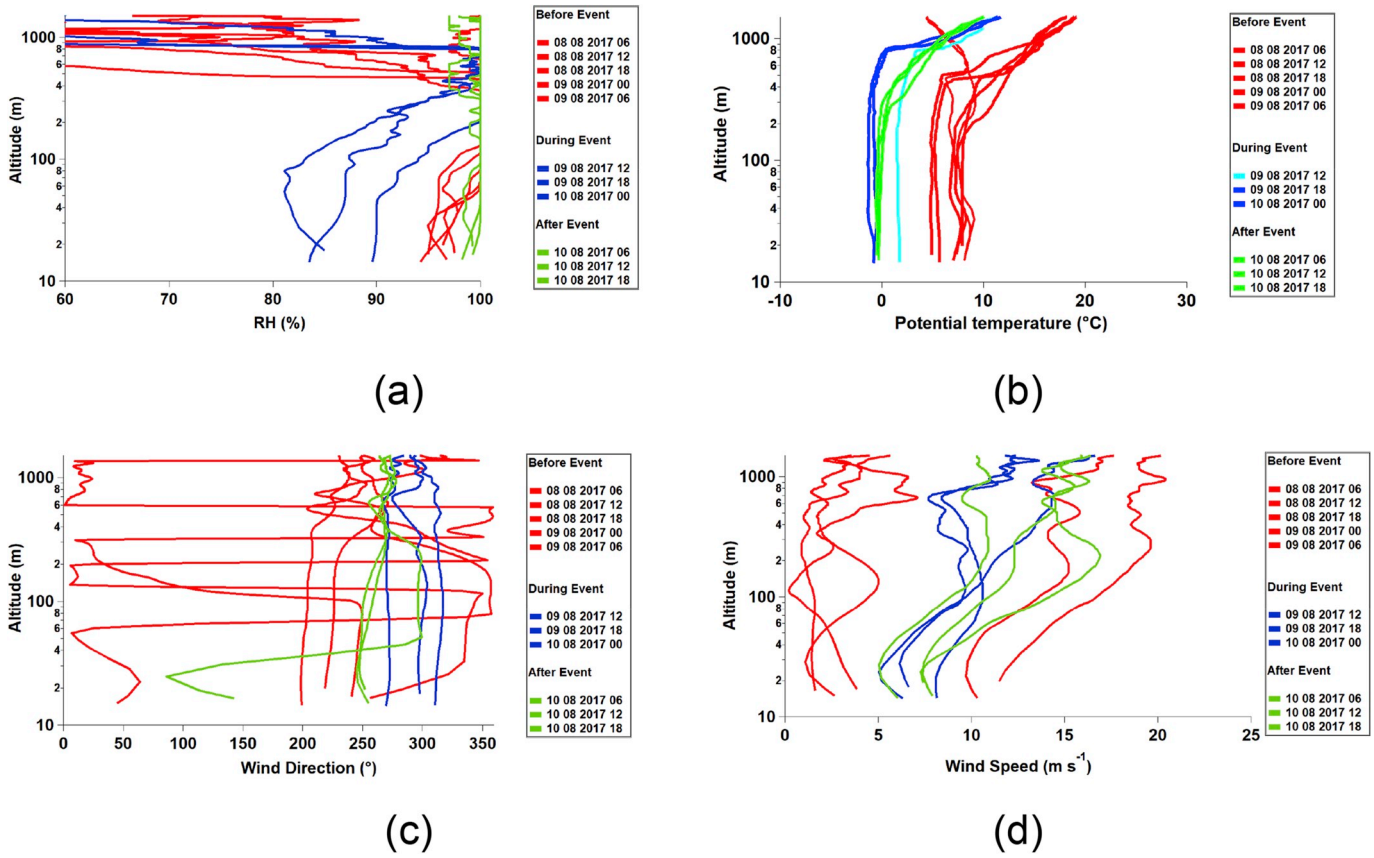


Fig. 6. Meteorological profiles of (a) Relative Humidity, (b) Temperature, (c) Wind Direction and (d) Wind Speed for the first nucleation event (9th August 2017, UTC time).

ice free by the end our XXI century (Boé et al., 2009; Serreze and Barry, 2011). Recently, a 7 year record (2010–2016) of aerosol size distributions taken at the VRS station in North Greenland were analysed, it was shown that NPF events occur annually 9% of the time, peaking at 39% during summer time (Dall'Osto et al., 2018 a). Such events are suggested to have originated in the boundary layer in contact with open water and melting sea ice regions, related to marine biological activity. The results from 2010 to 2016 at the VRS station strongly support previous similar studies carried out in the 2000–2010 period at the Zeppelin mountain station (Dall'Osto et al., 2017 a).

Our open ocean measurements strongly support the concept that the marginal sea ice zone is a source of ultrafine aerosols, and particles can grow to Aitken modes of about 50 nm in diameter. The Chukchi and East Siberian Seas (CESS) are among the most rapidly changing areas in the Arctic Ocean, where the annual sea ice retreat is beginning earlier and primary production is rapidly increasing (Arrigo and van Dijken, 2011; Lee et al., 2019). The marine production of secondary aerosol precursors is particularly sensitive to summer sea ice and melt ponds present in spring and summer (Gabric et al., 2017). The shrinking of the sea ice area in the Arctic will result in more open water available for gas exchange, which in turn may increase the background particle number concentrations (Dall'Osto et al., 2017 a).

4.2. Boundary layer origin

Aerosol observations made on the Arctic surface provide important insights into aerosol sources and processes. Nevertheless, surface-based observation may not be representative of the overall composition of the aerosols in the vertical aerosol profile. For example, some seasonal airborne observations have shown that aerosol sulphate may differ aloft compared to that measured near the surface (Klonecki et al., 2003; Scheuer et al., 2003). Most of the studies reporting in situ NPF in the Arctic associate such aerosols with natural emissions of volatile species that are oxidized within the Arctic boundary layer. In this study, we detected an aerosol nucleation mode during 18% of the time in our measurements recorded in the planetary boundary layer (Cluster 1, Bursting, Fig. 1). It is important to remember that the Arctic aerosol vertical distribution is governed by complex interactions between air mass origin (local and long-distance transport), aerosol sources (marine and anthropogenic), aerosol processes and cloud processing.

A number of observations have shown that new particle events can take place at high altitude, including in the free troposphere (Wiedensohler et al., 1996). However, recent vertical profiles taken in the last decade have revealed that nucleation events in the boundary layer near sea ice and open water regions may be a more plausible and much common dominating source. For example, Willis et al. (2016, 2017, 2018) reported aircraft-based measurements of High Arctic springtime aerosol showing evidence for vertically varying sources, transport and composition. Burkart et al. (2017a) reported summertime observation of elevated levels of ultrafine particles in the high Arctic marine boundary layer. Overall - by studying ultrafine particle number concentrations between 5 and 20 nm in diameter - it was concluded that NPF occurs frequently in the Canadian high Arctic marine boundary layer. Values of few thousand particles cm^{-3} were often observed especially just above ocean and clouds. By contrast, particle number concentrations in the free troposphere were much lower.

We did measure ultrafine particle number concentrations by means of minidisc ($N > 10$–30 nm) during a number of helicopter flights during our cruise. It was found that particle number concentrations at about 100–400 m were always about 30–45% lower than that measured at the sea surface, suggesting the ultrafine aerosol population of this study has likely a marine boundary layer origin. Unfortunately, none of these

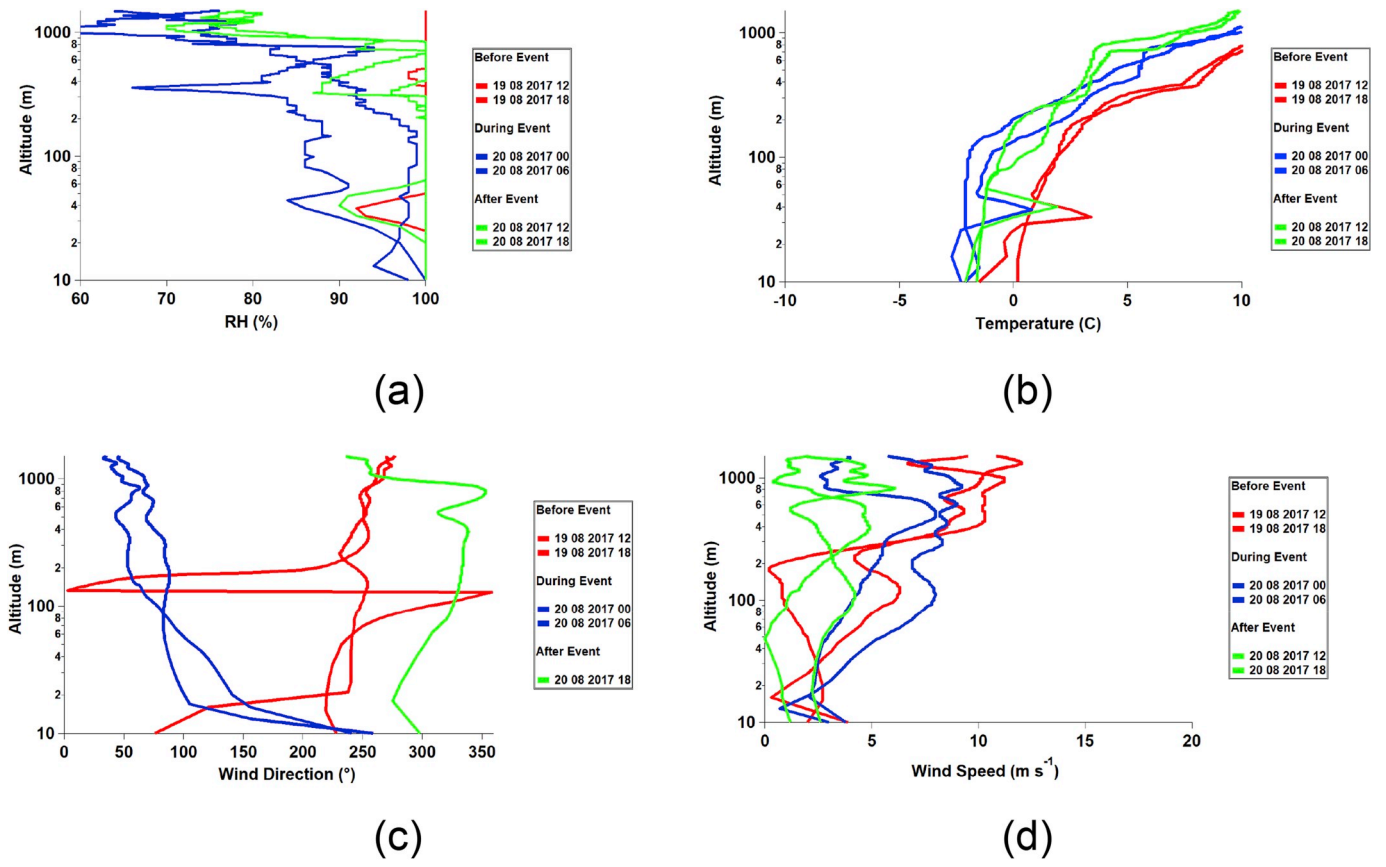


Fig. 7. Meteorological profiles of (a) Relative Humidity, (b) Temperature, (c) Wind Direction and (d) Wind Speed for the second nucleation event (20th August 2017, UTC time).

flights (12-15th August 2017) were done on the two nucleation days studied here. Our study suggests the dominant source of grown nucleation mode dominating the aerosol does not arise by mixing from aloft but most likely from marine sources in pelagic and sympagic regions. The origin of the nucleating particles (D < 10 nm) detected remains unknown at this stage; we only managed to detect NPF events already underway, which grew over time.

4.3. Chemical composition

The chemical composition of the nucleation range particles herein detected is not known because we did not deploy any instruments able to measure it (Junninen et al., 2010). Future work should be carried out in the study area in order to evaluate the chemical composition of the compounds involved in aerosol nucleation and growth. It is very likely that multiple chemical vapours from both biotic and abiotic processes may be responsible for the detected NPF events. It should be mentioned briefly that the current NPF is particularly challenging in the Arctic sea due to the very low concentrations of particles encountered (often < 100 cm^{-3}). Measurements of aerosol chemical composition at the VSR monitoring site identified that methanesulfonic acid (MSA) and molecular iodine (I_2) may be involved in the NPF mechanisms. The source of MSA has a well known biological origin in the ocean and sea ice (Lana et al., 2011; Levasseur, 2013; Becagli et al., 2016). By contrast, iodine may be associated with air masses over snow on land and over sea ice, suggesting both abiotic and biotic sources. In other words, iodine may originate in photochemical inorganic reactions in the snowpack, even on land (Raso et al., 2017), and also from marine algae from intertidal zones (Allan et al., 2015; Sippila et al, 2016; Dall'Osto et al., 2018 c). Recently, it was argued that Arctic marine secondary organic aerosol contributes significantly to summertime particle size distributions in the Canadian Arctic Archipelago (Willis et al., 2017; Collins et al., 2017; Croft et al., 2019). However, it was stressed that this organic aerosol is not typical biogenic secondary organic aerosols; instead has features with long hydrocarbon chains, implying a fatty-acid-type precursor. A possible source may be marine oxygenated volatile organic compounds (Mungall et al., 2017), possibly related to heterogeneous air-sea reactions of biogenic organic matter enriched in the sea surface microlayer (Wurl et al., 2017). Organic-rich particles contributed significantly to Arctic boundary layer aerosol mass, and correlations were found between such particles and elevated cloud condensation nuclei concentrations (Willis et al., 2016a,b, 2017).

Some consideration should be also made regarding anthropogenic pollution on Arctic coasts. The reduced sea ice in the Arctic sea is making development of oil and gas extraction and shipping, and generally human activities, to increase. Gunsch et al. (2017) reported the contribution of transported Prudhoe Bay oil field emissions to the aerosol population in Utqiaġvik, Alaska. It was found that increased smaller aerosol modes and higher total particle number were observed in air masses perturbed by Prudhoe Bay human activities in comparison to cleaner Arctic Ocean air masses. Our studies in open ocean regions of the Chukchi and East Siberian Seas show that biogenic sources - and not anthropogenic - are responsible for the detected NPF events.

Given the different bioregions characterising the Arctic environment, it is likely that a number of gaseous precursors of different chemical composition contribute to secondary aerosol formation, and further studies across different Arctic regions are strongly needed.

5. Conclusions

NPF events in the Arctic are being observed frequently (Willis et al., 2018; Abbatt et al., 2019), our Arctic ship-based measurements also

provide strong evidence of secondary aerosol formation in the Chukchi and East Siberian Seas. To our knowledge, these are the first studies of this type in this unstudied Arctic open ocean region, adding to recent work in different study areas (Park et al., 2019a, 2019b). Future ship-borne measurements in the study area should aim to elucidate the chemical composition of the NPF events, in order to better represent the exchanges between sea ice/ocean/snowpack and the atmosphere in Earth system models. It is also important to determine the balance between primary aerosols (e.g. sea spray, organic micro gels (Orellana et al., 2011),) and secondary aerosols in the Arctic; previous studies show these two sources may occur simultaneously (Collins et al., 2017; Köllner et al., 2017). This may be happening particularly in late spring and autumn, when biogenic gas precursors may be coexisting with sea spray generation from strong winds. Our work helps to understand the aerosol sources and spatial distribution within the Arctic sea, especially in poorly studied open waters.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.117232.

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