

Interactive comment on “Shipborne observations reveal contrasting Arctic marine, Arctic terrestrial and Pacific marine aerosol properties” by Jiyeon Park et al.

Anonymous Referee #2

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This manuscript describes approximately one month of measurements of aerosol physical properties and CCN concentrations taken aboard ship in the Pacific region of the Arctic, and Northern Pacific Ocean. This work demonstrates that terrestrial inputs in coastal areas can be an important driver of the particle size distribution in Arctic regions, and can drive strong new particle formation relative to open ocean areas of the Pacific. This is an important and interesting conclusion that has not been made in the past. Also, notably, this work presents measurements of particle size distributions starting at 3nm, which has not been done in many Arctic regions. This manuscript is well written, and presents the main results clearly. I support publication in ACP once the following issues can be addressed.

We sincerely appreciate all valuable comments and suggestions, which helped us to greatly improve the quality of the manuscript. We corrected the manuscript point by point accordingly.

Major comments:

(1) I am somewhat surprised that the authors have not demonstrated agreement between the two SMPS systems in their overlapping size range (i.e., 10-80nm). This is a crucially important size range for understanding particle growth and adequate quality control of these measurements should be addressed. How does N10-80 measured by the two instrument compare across the measurement period? Were any size calibrations of the two instruments made in the field? A clearer description of how the two measurements of particle size were aggregated over the overlapping range to produce Figure 7 is needed. To obtain the number size distribution in the size range from 7 nm to 300 nm as shown in Figure 7, we used nano SMPS data from 7 nm to 64 nm and standard SMPS data from 64 nm to 300 nm. We confirmed that nano SMPS and standard SMPS data agreed within ~8% in their overlapping size range (10 – 64 nm) (Figure S4). In fact, Watson et al. (2011) compared four types of scanning mobility particle sizers (i.e., TSI nano SMPS, TSI standard SMPS, Grimm SMPS, MSP WPS). They also demonstrated that hourly average particle concentrations in the 10 – 84 nm size range measured with TSI nano SMPS were well correlated with those measured with TSI standard SMPS. To clarify this issue, Figure S4 and description were added as given below.

Page 13, Line 334: “To obtain the number size distribution in the size range from 7 nm to 300 nm as shown in Fig. 7, we used nano SMPS data from 7 nm to 64 nm and standard SMPS data from 64 nm to 300 nm. The nano SMPS and standard SMPS data agreed within ~8.8% in their overlapping size range (10 – 64 nm) (Fig. S4), similar to a previous study (Watson et al., 2011).”

(2) How does the condensation sink and average growth rate compare (1) across the different types of air masses sampled, and (2) to previous Arctic measurements? Was the condensation sink significantly higher in the Pacific air masses? Figure 7 would suggest that is the case. I suggest that the authors make full use of their unique data set by calculating the above two quantities wherever possible, and including them in relevant figures. Statistics of these quantities could be included in Table 1. This would facilitate improved comparison with other studies in similar coastal Arctic regions (e.g., Collins et al., ACP, 2017 <https://doi.org/10.5194/acp-17-13119-2017> and Burkart et al., GRL, 2017 doi: 10.1002/2017gl075671). In particular, Collins et al 2017 provides a detailed comparison of the condensation sink in their

observations with that sampled globally (their Figure 8). Placing the author's results in this broader context would strengthen the paper significantly.

Response: We calculated growth rate and condensation sink for Arctic marine, Arctic terrestrial and Pacific marine air masses. The resulting values are given in Table 1 and compared with other studies in Arctic regions as given below.

Page 17, Line 422: "Table 1 shows particle growth rate (GR) and condensation sink (CS) for Arctic marine, Arctic terrestrial and Pacific marine air masses. The GR was calculated by fitting a linear regression to the peak diameter of the aerosol size distribution for the nucleation-mode between 4 and 20 nm against time during the NPF cases (Dal Maso et al., 2005; Pierce et al., 2014). The GR observed during the Arctic marine and Arctic terrestrial air masses were the $0.4 \pm 0.3 \text{ nm h}^{-1}$ and $0.8 \pm 0.2 \text{ nm h}^{-1}$, respectively, which was similar to the values previously observed from other Arctic regions. A shipboard expedition conducted during the summers of 2014 and 2016 throughout the Canadian Arctic, indicated that the GR varied widely from 0.2 to 15.3 nm h^{-1} (Collins et al., 2017). The GR observed at Summit, Greenland was $0.2 \pm 0.1 \text{ nm h}^{-1}$ (range of 0.09 to 0.3 nm h^{-1}) (Ziemba et al., 2010). Similarly, in Utqiagvik (Barrow), Alaska, the GR was 1.0 nm h^{-1} in air mass influenced by Beaufort Sea, whereas the value was 11.1 nm h^{-1} in air mass influenced by Prudhoe Bay (i.e., oil field area) (Kolesar et al., 2017)."

Page 17, Line 439: "The CS is a key parameter assessing the NPF and growth and determines how rapidly gaseous molecules condense onto pre-existing particles. The CS was calculated, following Dal Maso et al. (2002) and Collines et al. (2017). The resulting CS values are given in Table 1. The CS observed during the Arctic marine and Arctic terrestrial air masses were $0.5 \pm 0.4 \text{ nm h}^{-1}$ and $0.9 \pm 0.5 \text{ nm h}^{-1}$, respectively. The CS in this study is on the low end of the values observed during the summer in Arctic marine boundary layer (shipborne expeditions) (Collins et al., 2017), Utqiagvik, Alaska (Kolesar et al., 2017), and Ny-Ålesund, Svalbard (Giamarelou et al., 2016). In case when air mass passed over the Pacific Ocean, the CS was 2 or 4 times higher than those of Arctic air masses. It seems that such higher CS for Pacific marine air masses lowered the concentration of condensable vapors, thereby resulting in the non-event days in Pacific marine air masses."

(3) What other environmental variables were different between the three air mass source regions? e.g., wind speeds, cloud cover?

Response: Following the reviewer's comments, environmental variables such as wind speeds and wind direction for the three air masses were also shown in Table 1.

(4) It appears as though multiple modes were present in some cases (e.g., during 9/13-9/17), and based on Figure 7 appear in average size distributions. As discussed in Burkart et al., GRL, 2017 (doi: 10.1002/2017gl075671), comparing the growth rates of different particles modes can provide significant insight into the physical chemical properties of the condensing/partitioning species. I do not suggest that the authors undertake a detailed modelling exercise as in Burkart et al, rather that the authors calculate growth rates for the different modes and discuss what these quantities might mean in the context of the Burkart et al analysis.

Response: To clarify this issue, we determined the growth rate of the distinct modes based on Burkart et al analysis. Figure S6 and a few sentences were newly added as follows.

Page 17, Line 432: "Particularly, simultaneous growth of multiple modes was present in some cases (9/13/2017– 9/21/2017). We calculated the GR of the distinct modes, as shown in Fig. S6. The results showed that growth of the larger mode (e.g., preexisting mode) was faster than the smaller mode (e.g., nucleation mode), consistent with ship-based aerosol measurements in the summertime Arctic by Burkart et al. (2017b). They proposed that growth was largely via condensation of semi-volatile organic material, because lower volatile organics could lead to faster growth of the smaller mode."

Minor comments:

L67-68: I agree with this statement in general; however, the authors miss two very relevant studies of ship-based coastal Arctic measurements: Collins et al., ACP, 2017 <https://doi.org/10.5194/acp-17-13119-2017> and Burkart et al., GRL, 2017 doi: 10.1002/2017gl075671

Response: As suggested by the reviewer, we added the two references (Collins et al., 2017 and Burkart et al., 2017).

L69-82: This paragraph focuses on studies that have linked NPF events to increases in measured CCN; however, the authors focus not only on studies from Arctic regions and seem to focus on global observations as the expense of being exhaustive in discussing all of the very few relevant Arctic studies. The two studies mentioned in the comment above should be included here, and are more relevant to the authors' discussion than Kalivitis 2015 and Rose 2017. Burkart et al., ACP, 2017 (doi: 10.5194/acp-17-5515-2017) also connect NPF with CCN in coastal Arctic environments.

Response: These references (Collins et al., 2017 and Burkart et al., 2017) were addressed in the manuscript (Line 67, Line 70, Line 75 and Line 77), and the sentence was modified as given below.

Page 3, Line 75: "Several examples of increase in the CCN concentration after a few hours from the beginning of NPF events were presented by Burkart et al. (2017b) in the summer marine Arctic during the 2014 NETCARE Amundsen ice breaker campaign, by Kim et al (2019) at King Sejong Station in the Antarctic Peninsula, by Pierce et al. (2012) in a forested mountain valley in western Canada, and by Willis et al. (2016) in an Arctic aircraft campaign in Nunavut, Canada."

L123-126: Time resolution of the aethelometer?

Response: Raw time resolution of the aethelometer is 5 min. We added this information in the experimental section.

Page 5, Line 123: "The BC concentration was measured using an aethalometer (AE22, Magee Scientific Co., USA) with a 5-min time resolution to assess the influence of anthropogenic sources (such as local pollution and ship emissions)."

L141: Can the authors be more quantitative in describing "particularly high (spike) and varied dramatically in a short time"?

Response: The statement was modified to make quantitative description:

Page 6, Line 143: "The data collected when total aerosol number concentrations were higher than 8000 cm⁻³ were removed. In addition, the CPC and SMPS data were removed for the time periods when total aerosol number concentrations suddenly increased more than two times higher than the background values."

L235: Tremblay et al, ACP, 2019 (<https://doi.org/10.5194/acp-19-5589-2019>) demonstrated regional growth events taking place over the Northern Canadian Arctic Archipelago, and their results are relevant to this discussion.

Response: We agreed with the reviewer's comments. We added the reference in the manuscript (Page 10 and Line 240) and the description was rewritten as given below.

Page 10, Line 243: "Tremblay et al (2019) also concluded that particle nucleation events occurred over spatial scales of at least 500 km during the summertime in the Canadian High Arctic."

L262-263: Could a satellite-based measurement of cloud fraction (e.g., from Aqua MODIS that is

openly available) be used to assess any impact of cloudiness on the observations of NPF?

Response: To classify this issue, we compared weekly averaged liquid cloud fraction from Aqua/MODIS retrieval data for each period (week 1: 8/29/2017– 9/5/2017, week 2: 9/6/2017– 9/13/2017, week 3: 9/14/2017– 9/21/2017, and week 4: week 4 (9/22/2017– 9/29/2017)). Then, Figure S5 was added and the sentence (including references) was rewritten as given below.

Page 11, Line 270: “In addition, cloudiness which usually affects the real solar radiation reaching the surface was compared based on Moderate Resolution Imaging Spectroradiometer (MODIS) cloud fraction retrievals (Fig. S5). The data showed that cloud fraction was significantly high during the entire sampling periods, in general agreement with some other studies over the western Arctic region (e.g., Dong et al., 2010; Collines et al., 2017). In detail, the cloud fraction was relatively low for week 1 (8/29/2017– 9/5/2017; Fig. S5a) and week 3 (9/14/2017– 9/21/2017; Fig. S5c) when NPF event and growth was frequently observed (Fig. 3). This suggests that solar radiation at the surface, which is affected both by the cloud cover and SZA, may have influenced aerosol concentration and NPF observed here.”

L422-435: Burkart et al., ACP, 2017 (doi: 10.5194/acp-17-5515-2017) also estimated the CCN activation diameter to be approximately 80nm in a coastal Arctic environment.

Response: We added the reference in the manuscript as given below.

Page 19, Line 478: “For instance, the D_c of 80 nm at 0.6 % SS was observed during the aircraft measurement in July 2014 in the high Arctic marine boundary layer of Resolute Bay, Nunavut, Canada (Burkart et al., 2017a).”

Table 1: Do $N_{\text{StandardSMPS}}$ and N_{nanoSMPS} include the overlapping size range? I suggest that the authors be explicit about the size range here, rather than referring to the instrument. Presenting mean concentrations in specific size ranges from aggregated size distribution data may be more useful (e.g., as shown in figure 2)

Response: As suggested by the reviewer, we presented mean concentrations of nucleation mode (3 – 20 nm) (N_{NUC}), Aitken mode (20 – 100 nm) (N_{AIT}), accumulation mode (100 – 300 nm) (N_{ACC}), and coarse mode (> 300 nm from OPS) (N_{OPS}) particles for the three selected periods in Table 1.

Figure 7: Specify which SMPS is used in the size range from 10-80nm

Response: This is similar to a comment above. Again, we used nano SMPS data from 7 nm to 64 nm and standard SMPS data from 64 nm to 300 nm. We confirmed that the nano SMPS and standard SMPS data agreed within ~8.8% in their overlapping size range (10 – 64 nm), as shown in Fig S4.

Specific comments:

L61: Ice and snow-covered regions

Response: It was corrected.

L264: change 'trends' to 'tends'

Response: It was corrected.

L286: missing 'were'

Response: It was corrected.

Newly added references

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Shipborne observations reveal contrasting Arctic marine, Arctic terrestrial and Pacific marine aerosol properties

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Abstract

There are few shipborne observations addressing the factors influencing the relationships of the formation and growth of aerosol particles with cloud condensation nuclei (CCN) in remote marine environments. In this study, the physical properties of aerosol particles throughout the Arctic Ocean and Pacific Ocean were measured aboard the Korean ice breaker R/V Araon during the summer of 2017 for 25 days. A number of New Particle Formation (NPF) events and growth were frequently observed in both Arctic terrestrial and Arctic marine air masses. By striking contrast, NPF events were not detected in Pacific marine air masses. Three major aerosol categories are therefore discussed: (1) Arctic marine (aerosol number concentration $CN_{2.5}$: $413 \pm 442 \text{ cm}^{-3}$), (2) Arctic terrestrial ($CN_{2.5}$: $1622 \pm 1450 \text{ cm}^{-3}$) and (3) Pacific marine ($CN_{2.5}$: $397 \pm 185 \text{ cm}^{-3}$), following air mass back trajectory analysis. A major conclusion of this study is that not only that the Arctic Ocean is a major source of secondary aerosol formation relative to the Pacific Ocean; but also that open ocean sympagic and terrestrial influenced coastal ecosystems both contribute to shape aerosol size distributions. We suggest that terrestrial ecosystems - including river outflows and tundra - strongly affects aerosol emissions in the Arctic coastal areas, possibly more than anthropogenic Arctic emissions. The increased river discharge, tundra emissions and melting sea ice should be considered in future Arctic atmospheric composition and climate simulations. The average CCN concentrations at a supersaturation ratios of 0.4% were $35 \pm 40 \text{ cm}^{-3}$, $71 \pm 47 \text{ cm}^{-3}$, and $204 \pm 87 \text{ cm}^{-3}$ for Arctic marine, Arctic terrestrial, and Pacific marine aerosol

30 categories, respectively. Our results aim to help to evaluate how anthropogenic and natural atmospheric
31 sources and processes affect the aerosol composition and cloud properties.

32

33 **1. Introduction**

34 The climate change experienced in the Arctic is more rapid than that occurring at mid-latitudes in a
35 phenomenon known as Arctic amplification (ACIA, 2005). In the warming Arctic, the extent and
36 thickness of sea-ice have dramatically decreased over the past few decades (Stroeve et al., 2012). It has
37 been estimated that the Arctic may seasonally become sea ice-free Arctic in the next 30 years (Wang
38 and Overland, 2009). Aerosol particles in the atmosphere are a major driver of the Arctic climate (IPCC,
39 2013), as they directly affect the climate through scattering and absorbing solar radiation (Stier et al.,
40 2007), and indirectly by modifying the formation, properties, and lifetimes of clouds (Twomey, 1974).
41 These direct and indirect effects are the leading uncertainty in current climate predictions. New particle
42 formation (NPF), a predominant source of atmospheric particles, occurs through the formation of
43 nanometer-sized molecular clusters (~ 1 nm) (i.e., nucleation) and their subsequent growth into aerosol
44 particles of a few nanometers ($\sim 1 - 10$ nm) and larger ($\sim >10$ nm) (Kulmala et al., 2004; Zhang et al.,
45 2012). NPF can significantly increase the number of aerosol particles in the atmosphere. During
46 summer, the Arctic is more isolated from anthropogenic influences (Arctic Haze) and experiences
47 comparatively pristine background aerosol conditions (Heintzenberg et al., 2015; Law and Stohl, 2007).
48 As the number concentrations of particles in the Arctic during summer are very low (of an order of $\sim 10^2$
49 cm^{-3}) (Merikanto et al., 2009), the physicochemical properties of aerosol particles in the Arctic
50 atmosphere is highly sensitive to NPF.

51 NPF events have been frequently observed within a wide range of environmental conditions at
52 various Arctic locations, such as Zeppelin (Tunved et al., 2013; Croft et al., 2016; Heintzenberg et al.,
53 2017), Tiksi (Asmi et al., 2016), Alert (Croft et al., 2016), Station Nord (Nguyen et al., 2016), and
54 Barrow (Kolesar et al., 2017), and from limited ship-based observations (Chang et al., 2011; Kim et al.,
55 2015; Heintzenberg et al., 2015). The formation and growth of particles in the Arctic atmosphere are

56 strongly influenced by marine, coastal, marginal ice, and/or anthropogenic sources. Oceanic dimethyl
57 sulfide (DMS) and other volatile organic precursors (such as isoprene, monoterpenes, and amines) play
58 important roles in the formation and growth of new particles in the Arctic (Leaitch et al., 2013; Willis et
59 al., 2016; Park et al., 2017; Abbatt et al., 2019; Mungall et al., 2016). In addition, iodine oxides
60 significantly contribute to NPF in marine and coastal Arctic environments owing to emissions from
61 marine microalgae at low tide or snowpack photochemistry in [ice and snow-covered regions](#) (Allan et
62 al., 2015; O'Dowd et al., 2002; Raso et al., 2017). Biogenic gaseous precursors released by the melting
63 Arctic sea-ice margins have also been associated with NPF (Dall'Osto et al., 2017; Willis et al., 2018).
64 Recent studies in Alaska have indicated that the formation and growth of particles are influenced by
65 emissions from oil and gas extraction activities in Prudhoe Bay (Gunsch et al., 2017; Kolesar et al.,
66 2017). Although several observations have been made in the Arctic under different environmental
67 conditions ([Burkart et al., 2017b](#); [Collins et al., 2017](#)), there are few detailed size distribution analyses
68 of particle formation and growth events within the Arctic marine environment.

69 Several studies have attempted to investigate the impacts of NPF on the concentrations of cloud
70 condensation nuclei (CCN) (Willis et al., 2016; [Burkart et al., 2017b](#); [Collins et al., 2017](#)). Model-based
71 studies have predicted that a large fraction of CCN (up to 78% of CCN at 0.2 % supersaturation) in the
72 global atmosphere results from atmospheric NPF and growth (Merikanto et al., 2009; Westervelt et al.,
73 2014; Spracklen et al., 2008). Field observations have also observed substantial increases in the
74 concentrations of CCN due to atmospheric nucleation in various environments (Pierce et al., 2012;
75 Kalivitis et al., 2015; [Burkart et al., 2017b](#); [Collins et al., 2017](#); Kim et al., 2019). [Several examples of
76 increase in the CCN concentration after a few hours from the beginning of NPF events were presented
77 by Burkart et al. \(2017b\) in the summer marine Arctic during the 2014 NETCARE Amundsen ice
78 breaker campaign, by Kim et al \(2019\) at King Sejong Station in the Antarctic Peninsula, by Pierce et al.
79 \(2012\) in a forested mountain valley in western Canada, and by Willis et al. \(2016\) in an Arctic aircraft
80 campaign in Nunavut, Canada.](#) However, due to the infrequency of aerosol measurements collected
81 onboard ice breakers, very few studies have measured the simultaneous aerosol size distribution and

82 CCN concentrations over the Arctic Ocean.

83 In this study, the physical characteristics of aerosol particles over the Arctic and Pacific Oceans were
84 investigated between [August 31 and September 24, 2017](#), using aerosol particle monitoring instruments
85 installed on the Korean ice breaker R/V Araon. Data of the aerosol size distribution, the concentrations
86 of the total aerosol number (CN), black carbon (BC), and CCN were continuously collected using
87 various aerosol instruments. The main aims of this study were to (1) investigate the frequency and
88 characteristics of NPF and particle growth over the Arctic and Pacific Oceans, (2) determine the major
89 sources that are associated with NPF based on backward air mass trajectory analysis, and (3) explore the
90 potential contribution of NPF to the CCN concentrations in the remote marine environment.

91 92 **2. Experimental methods**

93 **2.1. Study area and ship tracks**

94 Ambient atmospheric aerosol measurements were collected over the Arctic and Pacific Oceans
95 onboard the ice breaker R/V Araon, operated by the Korea Polar Research Institute (KOPRI), Korea.
96 The ship's track is presented in Fig. 1. The cruises covered two main areas: the Arctic Ocean (including
97 both Beaufort and Chukchi Seas) and the remote Northwest Pacific Ocean. The ship departed from
98 Barrow, USA, on August 28, 2017, crossed the Beaufort (August 29-September 13, 2017) and Chukchi
99 Seas (September 15, 2017), and reached Nome, USA, on September 16, 2017. The Beaufort Sea
100 extends across the northern coasts of Alaska and the Northwest Territories of Canada. After completing
101 the Arctic survey, the ship departed from Nome, USA, on September 18, 2017, crossed the Bering Sea,
102 Sea of Okhotsk, and East Sea, and reached Busan, Korea, on September 28, 2017.

103 104 **2.2. Atmospheric aerosol measurements**

105 The aerosol sampling inlet was placed on the front deck of the ship (13 m above sea level), ahead of
106 the ship's engines to avoid any influences from the emissions of the ship's exhaust. In addition, kitchen
107 ventilation systems were connected by a plastic cylindrical pipe (~15 m length) and moved back on the

108 deck (far away from the sampling inlet) to minimize the potential effects of cooking emissions on the
109 atmospheric measurements during the sampling periods. Aerosols were sampled through a stainless
110 steel tube (inner diameter of 1/4 in, and length of ~1 m), which was connected to the various
111 instruments by electrically conductive tubing to minimize particle losses in the sampling line.

112 The physical properties of the aerosols were measured with various aerosol instruments, including
113 two condensation particle counters (TSI 3776 CPC and TSI 3772 CPC), two scanning mobility particle
114 sizers (SMPS), an optical particle sizer (OPS), an aethalometer, and a cloud condensation nuclei counter
115 (CCNC). The TSI 3776 CPC and TSI 3772 CPC measured the total number concentrations of particles
116 larger than 2.5 and 10 nm every 1 sec, respectively. The aerosol sample flow rates of TSI 3776 CPC and
117 TSI 3772 CPC were 1.5 and 1.0 lpm, respectively. The number size distributions of the particles were
118 measured using the nano SMPS every 3 min (Differential mobility analyzer (DMA): TSI 3085, CPC:
119 TSI 3776), covering a size range of 3 to 63.8 nm, and the standard SMPS (DMA: TSI 3081, CPC: TSI
120 3772) every 3 min, covering a size range of 10 to 300 nm. The aerosol and sheath flow rates of the
121 nano-SMPS were 1.5 and 15 lpm, respectively; and those of the standard SMPS were 1.0 and 10 lpm,
122 respectively. An OPS (TSI 3330) was used to determine the size distribution of particles in the range of
123 100 nm to 10 μm diameter with a sample flow rate of 1.0 lpm every 3 minutes. The BC concentration
124 was measured using an aethalometer (AE22, Magee Scientific Co., USA) with a 5-min time resolution
125 to assess the influence of anthropogenic sources (such as local pollution and ship emissions). The
126 instrument uses the absorption of light at a wavelength of 880 nm by the ambient aerosols collected on a
127 quartz filter tape to determine the BC concentration. The flow rate through a sharp-cut 2.5 μm cyclone
128 (BGI, Inc., USA) was set to 5 lpm and the integration time was 5 min. The Droplet Measurement
129 Technologies CCN counter (DMT CCN-100) was operated to measure the CCN number concentrations.
130 The total flow rate in the CCN counter was 0.5 lpm, and the counter was operated at five different
131 supersaturation ratios (SS) (0.2, 0.4, 0.6, 0.8, and 1.0 %) every 30 min. The sample and sheath flow
132 rates of the CCN counter were 0.05 and 0.45 lpm, respectively.

133

134 **2.3. Identification of ship exhaust**

135 To obtain a data set that reflects background aerosol loading, measurement data affected by the
136 exhaust emissions of the ship's engine should be excluded prior to further data analysis. For this,
137 aerosol data were filtered based on the BC concentration, wind direction, wind speed, and total particle
138 number concentration. The data with the following properties were discarded: (1) BC concentrations
139 exceeding 100 ng m^{-3} , (2) relative wind direction against the ship's heading between 110° and 260° , as
140 this originates directly from the ship's exhaust, (3) relative wind speed lower than 2 m sec^{-1} as air
141 masses under a calm environment could become contaminated due to local turbulence, and (4) the total
142 particle number concentrations were particularly high (spike) and varied dramatically in a short time.
143 Ship plumes were clearly observed in the data collected during the campaign. **The data collected when**
144 **total aerosol number concentrations were higher than 8000 cm^{-3} were removed. In addition, the CPC**
145 **and SMPS data were removed for the time periods when total aerosol number concentrations suddenly**
146 **increased more than two times higher than the background values.** Typically, the ship exhaust differs
147 from the NPF events as the enhanced number concentration during the NPF events lasted for at least an
148 hour with a low BC concentration (Ehn et al., 2010).

149 150 **2.4. Backward air mass trajectory and satellite observations**

151 The backward air mass trajectories were analyzed using version 4 of the Hybrid Single-Particle
152 Lagrangian Integrated Trajectory (HYSPLIT) model (<http://ready.arl.noaa.gov/>) to examine their
153 relationships with the physical characteristics of aerosol particles. The 2-day air mass back trajectories
154 (48 h) were determined at hourly intervals from the ship's position at an arrival height of 50 m to
155 estimate the transport history of the air masses arriving at the observation site (Park et al., 2018). The
156 potential origins of the aerosols were divided into three categories based on the retention time of the 2-
157 day back trajectories over the three major domains: Arctic Ocean (including the Beaufort and Chukchi
158 Seas, and sea-ice region), Pacific Ocean (including the Bering Sea and Sea of Okhotsk) and land
159 (including Alaska and the eastern part of Siberia) (Fig. 1). The phytoplankton biomass was obtained by

160 calculating the chlorophyll-*a* concentration from the level-3 product of Aqua Moderate Resolution
161 Imaging Spectroradiometer at a 4 km resolution (Fig. S1). Geographical information over the ocean,
162 land and sea-ice was obtained from the sea-ice index, which was provided by the National snow and Ice
163 Data Center (NSIDC) (Fig. S2). Note that the sea-ice extent was defined as the area having an ice
164 concentration of $\geq 15\%$ (Pang et al., 2018). Air masses that intensively passed over the Beaufort and
165 Chukchi Seas and sea-ice region were categorized as Arctic Ocean originated air masses (i.e., $> 50\%$
166 retention over the ocean $> 65^\circ\text{N}$ and sea-ice region). Air masses that intensively passed over Northern
167 Alaska and the eastern Siberia were potentially affected by the Arctic tundra and categorized as land
168 originated air masses (i.e., $> 50\%$ retention over the land domain). Finally, air masses that traveled
169 through the Bering Sea and Sea of Okhotsk were categorized as air masses originated from Pacific
170 Ocean domain (i.e., $> 50\%$ retention over the ocean domain $< 65^\circ\text{N}$).

171 172 **2.5. Oceanic measurements**

173 To examine the influence of oceanic conditions on NPF and growth, seawater samples were collected
174 from sea surface at a depth of ~ 1 m by Niskin bottles. The sampling locations and methods have been
175 described previously in more detailed (Park et al., 2019). In brief, concentrations of dissolved organic
176 carbon (DOC) were measured with a Shimadzu TOC-V high-temperature combustion total organic
177 carbon analyzer. To identify the source and composition of DOC in surface seawater, three-dimensional
178 excitation-emission matrixes (EEMs) were scanned using a fluorescence spectrometer (Varian, USA).
179 The excitation wavelength range was between 250 and 500 nm, and emission between 280 and 600 nm.
180 In this study, the four major fluorescent components were classified into 4 groups; terrestrial humic
181 substances peak (A) (EX: 260 nm, EM: 380–460 nm), the terrestrial fulvic substances peak (C) (EX:
182 350 nm, EM: 420–480 nm), the marine fulvic substances peak (M) (EX: 312 nm, EM: 380–420 nm),
183 and the proteinaceous peak (T) (EX: 275 nm, EM: 340 nm) (Coble, 2007).

184 185 **3. Results and discussion**

186 3.1. Overall particle number concentrations

187 Fig. 2a presents a time series of the 1 hour average total particle number concentration (CN)
188 measured using TSI 3776 CPC and TSI 3772 CPC throughout the sampling periods. The number
189 concentration of particles larger than 2.5 nm (CN_{2.5}) or 10 nm (CN₁₀) in the Arctic and Pacific marine
190 environments had a range of approximately three orders of magnitude ($\sim 10^1 - 10^3 \text{ cm}^{-3}$). In most cases,
191 the CN_{2.5} and CN₁₀ concentrations were less than $\sim 2000 \text{ cm}^{-3}$, with averages of 505 ± 280 and $492 \pm$
192 264 cm^{-3} , respectively, which were in agreement with those reported in previous studies conducted at
193 other Arctic stations (Asmi et al., 2016; Burkart et al., 2017a; Freud et al., 2017) and remote marine
194 regions (O'Dowd et al., 2014; Sellegri et al., 2006; Kim et al., 2019; Jang et al., 2019; Yum et al., 1998;
195 Hudson and Yum, 2002). For example, four years of observational data from the Arctic Climate
196 Observatory in Tiksi, Russia, showed that the monthly median CN concentration ranged from $\sim 184 \text{ cm}^{-3}$
197 in November to $\sim 724 \text{ cm}^{-3}$ in July (Asmi et al., 2016). Furthermore, Sellegri et al. (2006) reported CN
198 concentrations under clean marine sector conditions at Mace Head of a few hundreds of cm^{-3} (e.g., ~ 200
199 cm^{-3} in January and $\sim 450 \text{ cm}^{-3}$ in June). Elevated CN_{2.5} and CN₁₀ concentrations were concentrated over
200 the period from September 13 to 20, when the ship sailed over Chukchi and Bering Seas. During this
201 period, CN_{2.5} and CN₁₀ concentrations exceeding $\sim 2000 \text{ cm}^{-3}$ were frequently observed. The peak
202 concentrations of aerosol particles were notable, as the CN_{2.5} and CN₁₀ concentrations exceeded ~ 6016
203 and $\sim 5750 \text{ cm}^{-3}$, respectively.

204 To elucidate further details of the variations in CN_{2.5} and CN₁₀, the particle size distributions
205 measured with the nano SMPS, standard SMPS, and OPS were divided into four size groups: nucleation
206 (3 – 20 nm), Aitken (20 – 100 nm), accumulation (100 – 300 nm), and coarse ($> 300 \text{ nm}$ from OPS), as
207 shown in Fig. 2b–e. The average number concentrations of the nucleation-mode (N_{NUC}), Aitken-mode
208 (N_{AIT}), accumulation-mode (N_{ACC}), and coarse-mode (N_{OPS}) particles were 169 ± 142 , 201 ± 131 , $40 \pm$
209 17 , and $4 \pm 2 \text{ cm}^{-3}$, respectively. The temporal variations in N_{NUC} and N_{AIT} exhibited a distinct pattern,
210 compared to that of N_{ACC} and N_{OPS}. Overall, N_{NUC} and N_{AIT} concentrations larger than $\sim 1000 \text{ cm}^{-3}$ were
211 observed from September 13 to 20 (e.g. the ship sailed over Chukchi and Bering Seas), whereas

212 relatively high concentrations of N_{ACC} and N_{OPS} were observed from September 21 to 23 (e.g., the ship
213 sailed over Sea of Okhotsk). As shown in Fig. 2b, sudden bursts of nucleation-mode particles occurred
214 frequently, as indicated by a sudden increase in the N_{NUC} concentration rising from tens to several
215 thousands of cm^{-3} . Whenever the $CN_{2.5}$ concentration exceeded $\sim 2000\ cm^{-3}$, the N_{NUC} concentration
216 exceeded $\sim 600\ cm^{-3}$ (except for the results observed in the evening of September 18). In addition, the
217 $CN_{2.5}$ concentration was strongly correlated with the N_{NUC} concentration ($r^2 = 0.69$) (Fig. S3),
218 suggesting that the high CN concentration was mainly derived from nucleation-mode particles.
219 Instances of elevated N_{NUC} occurred along the northern coast of Alaska (September 13 – 14, 2017),
220 throughout the Chukchi Sea (September 15, 2017), near the Nome and Eastern Siberia (September 16 –
221 18, 2017), and throughout the Bering Sea (September 19 – 20, 2017). During the cruises, the satellite-
222 derived chlorophyll-*a* concentration data indicated strong biological activity over the Chukchi and
223 Bering Seas, as shown in Fig. S1. Thus, the high occurrence of nucleation-mode particles may be
224 related to multiple processes that influence the formation of secondary aerosols (e.g., oceanic biological
225 activities, regional anthropogenic emissions on land (Alaska or eastern Siberia), and terrestrial sources
226 in the tundra ecosystems of Alaska).

227

228 **3.2. Case studies**

229 As mentioned in Section 3.1, significant increases in N_{NUC} were frequently observed during the
230 cruise (Fig. 2 b). Typically, N_{NUC} is used to indicate the presence of newly formed particles produced by
231 gas-to-particle conversion (i.e., secondary aerosol formation) (Asmi et al., 2016; Burkart et al., 2017a).
232 Here, an NPF event was defined as a sharp increase in the N_{NUC} with elevated $CN_{2.5}$ that lasted for at
233 least one hour. Fig. 3 presents contour plots of the size distributions measured using nano SMPS and
234 standard SMPS. This strong NPF and growth event occurred over the Chukchi and Bering Seas, which
235 border the western and northern sides of Alaska, suggesting that there may be a substantial source of
236 precursors in this region. Bursts of the smallest particles at the lowest detectable sizes ($\sim 2.5\ nm$) were
237 not observed; however, we hypothesize that, during the NPF event, particle formation occurred

238 elsewhere and that subsequent horizontal extension caused the particles to reach the sampling site.
239 Previously, NPF events have been identified on the regional scale in several locations around the world
240 (Kerminen et al., 2018; Németh and Salma, 2014; Tremblay et al., 2019; Vana et al., 2004; Väänänen et
241 al., 2013). For instances, Németh and Salma (2014) found that a nucleating air mass in regional NPF
242 events may originate horizontally as far as several hundreds of kilometers (~400 or 700 km) away from
243 the sampling site. Tremblay et al (2019) also concluded that particle nucleation events occurred over
244 spatial scales of at least 500 km during the summertime in the Canadian High Arctic. In this section,
245 case studies are discussed, including (i) marine Arctic NPF event, (ii) terrestrial Arctic NPF event, and
246 (iii) pacific marine aerosol categories. During these temporal periods, the influences of the origins and
247 pathways of air masses on the characteristics of particle formation and growth were investigated.

248 249 **3.2.1. Open ocean marine Arctic NPF event case study**

250 The marine Arctic NPF event was observed on September 3, 2017, and time series plots of the
251 particle size distribution and air mass origins are presented in Fig. 4. N_{NUC} increased from 77 cm^{-3} to
252 757 cm^{-3} , while N_{AIT} varied little. The elevated number concentration of nucleation-mode particles
253 lasted for over five hours and then disappeared. Geometric mean diameter (GMD) varied from 14.6 to
254 18.2 nm with an average of 16.3 nm, indicating that particle growth hardly occurred. The GMD is
255 defined as the particle diameter at which the cumulative probability becomes 50% for the fitted log-
256 normal distribution (Hinds, 1999). During the day, air masses traveled over the Arctic Ocean (explicitly,
257 47.6, 0 and 0.4 h over the Arctic Ocean, Pacific Ocean and land domain, respectively), and have been
258 categorized as Arctic Ocean originated air masses. As shown in Fig. S1, the satellite-derived
259 chlorophyll-*a* concentration indicated a relatively high level of biological activity in the ocean during
260 the time period focused upon in this study. It was noteworthy that the monthly mean chlorophyll
261 concentration in the Beaufort and Chukchi Seas ($2.24 \pm 3.44 \text{ mg m}^{-3}$; 65°N – 74°N and 170°E – 120°W)
262 was approximately 3-fold greater than that estimated in the Pacific Ocean including the Bering Sea and
263 the Sea of Okhotsk ($0.83 \pm 1.30 \text{ mg m}^{-3}$; 40°N – 65°N and 145°E – 168°W) (Fig. S1). Moreover, the

264 marginal ice zone is commonly associated with intense algae blooms during the melting season,
265 therefore, significant emissions of biogenic trace gases such as DMS have been detected in the sea-ice
266 edge (Levasseur, 2013; Oziel et al., 2017). Accordingly, as our measurements were collected over the
267 Arctic Ocean onboard the ice breaker, marine biogenic sources could be considered as an important
268 factor inducing NPF events.

269 Fig. 4d shows cosine of the solar zenith angle ($\cos(\text{SZA})$) data that can be used as a proxy for solar
270 energy reaching the ground surface. In addition, cloudiness which usually affects the real solar radiation
271 reaching the surface was compared based on Moderate Resolution Imaging Spectroradiometer
272 (MODIS) cloud fraction retrievals (Fig. S5). The data showed that cloud fraction was significantly high
273 during the entire sampling periods, in general agreement with some other studies over the western
274 Arctic region (e.g., Dong et al., 2010; Collines et al., 2017). In detail, the cloud fraction was relatively
275 low for week 1 (8/29/2017– 9/5/2017; Fig. S5a) and week 3 (9/14/2017– 9/21/2017; Fig. S5c) when
276 NPF event and growth was frequently observed (Fig. 3). This suggests that solar radiation at the surface,
277 which is affected both by the cloud cover and SZA, may have influenced aerosol concentration and NPF
278 observed here. As illustrated in Fig 4, the NPF event occurred when the sun was below the horizon (i.e.,
279 Arctic nighttime nucleation). Typically, nucleation tends to take place preferably with high solar
280 irradiation during the daytime (Kulmala et al., 2004). In several locations, however, also nighttime
281 nucleation has been observed at Tumbarumba in Australian (Sunı et al., 2008), at Värriö measurement
282 station in Finnish Lapland (Vehkamäki et al., 2004), and at a subarctic site in northern Sweden (~14 km
283 east of Abisko) (Svenningsson et al., 2008). The possible explanation for nighttime events is that the
284 actual formation and growth occurred even during daylight, but very slow growth in the Arctic and
285 marine atmosphere allowed to detect the particles (~ 8 nm) only after sunset (Vehkamäki et al., 2004).
286 Previous study reported that 32% of strong nighttime nucleation events (2.5 times as frequent as
287 daytime nucleation event) were appeared in the presence of a very efficient ion source such as the
288 strong radon efflux from the Tumbarumba soil (Sunı et al., 2008). Due to their rarity, the major
289 mechanisms for nocturnal aerosol production are still unclear and require more study.

290

291 **3.2.2. Open ocean terrestrial Arctic NPF event case study**

292 The terrestrial Arctic NPF event was observed during September 13–14 2017. As shown in Fig. 5,
293 significant strong NPF events occurred frequently during this period. The number concentration of total
294 particles increased considerably, as a $CN_{2.5}$ value exceeding $\sim 6016 \text{ cm}^{-3}$ was observed during this event.
295 In addition, the average concentrations of N_{NUC} and N_{AIT} during the terrestrial Arctic NPF were $931 \pm$
296 222 and $1127 \pm 380 \text{ cm}^{-3}$, respectively. This indicates that high $CN_{2.5}$ concentration mainly contributed
297 by nucleation and Aitken-mode particles (45 and 54% of the size distribution for nucleation-mode and
298 Aitken-mode particles, respectively). GMD increased from 13.9 to 33.3 nm, indicating that the
299 nucleation-mode particles subsequently increased in size. The formation and growth of aerosol particles
300 were observed during the daytime (Fig. 5d), suggesting that photochemistry is involved. [During this](#)
301 [period, air masses were heavily influenced by northern Alaska.](#) The average retention times of the 2-day
302 back trajectories arriving at the ship position over the northern Alaska, Arctic Ocean and Pacific Ocean
303 were 40.8, 7.2 and 0 h, respectively (Fig. 5e). It can be seen that the photochemical reactions of
304 precursor gases (e.g., volatile organic compounds (VOCs) such as isoprene, monoterpenes, and
305 sesquiterpenes) emitted by terrestrial ecosystems in Alaska were associated with new particle formation
306 and growth (Schollert et al., 2014; TAPE et al., 2006; Kolesar et al., 2017; Ström et al., 2003).

307

308 **3.2.3. Pacific marine aerosol case study**

309 A typical aerosol scenario for Pacific marine air masses was observed on September 21–22, 2017,
310 when the air masses passed over mainly the Pacific Ocean (including the Bering Sea and Sea of
311 Okhotsk) (explicitly, 0, 47.9 and 0.1 h over the Arctic Ocean, Pacific Ocean and land domain,
312 respectively) (Fig. 1a). As shown in Fig. 6, the aerosol number concentrations exhibited a bimodal size
313 distribution, peaking at size ranges of 30 – 80 nm (Aitken mode) and 100 – 300 nm (accumulation
314 mode), respectively. In contrast, the concentrations of nucleation-mode particles were very low. For
315 example, the concentration of N_{NUC} ranged from 1 to 38 cm^{-3} with an average of $8 \pm 4 \text{ cm}^{-3}$. We also

316 observed CN_{2.5} values at the background level of $\sim 460 \pm 70 \text{ cm}^{-3}$, which are consistent with the
317 measurements collected at a coastal Antarctic station during summer ($\sim 600 \text{ cm}^{-3}$) (Kim et al., 2017) and
318 from flight-based measurements over the Arctic Ocean ($\sim 300 \text{ cm}^{-3}$) (Burkart et al., 2017a).

319

320 **3.3. Overview of aerosol properties according to different air mass back trajectories**

321 Air masses comprising marine Pacific along with marine and terrestrial Arctic air masses were
322 encountered during the campaign. In the section 3.2, two case studies of NPF events (Fig. 4 and Fig. 5)
323 were found in the Arctic atmosphere. As stressed in Willis et al., (2018), NPF and growth is frequently
324 observed in the boundary layer in the both Arctic open ocean and coastal regions. These events seem to
325 occur more frequently than lower-latitude marine boundary layers (Quinn and Bates, 2011); there are
326 multiple reasons including summer 24-h high solar radiation, low condensation sink, low temperature
327 and low mixing of surface emissions, as recently reviewed in Abbatt et al. (2019). Our study also
328 confirmed that any NPF was not detected during the Pacific transect.

329 In this section, we present an overall meteorological air mass summary of the open ocean field study,
330 categorizing it into three synoptic period types: Pacific marine, Arctic marine and Arctic terrestrial.
331 These classifications do not represent specific air mass back trajectories analysis, but they can mainly
332 represent air masses that have been travelled over these three distinct geographical regions (section 2.4).
333 Average size distributions for the three selected periods in the different air masses are shown in Fig. 7.
334 To obtain the number size distribution in the size range from 7 nm to 300 nm as shown in Fig. 7, we
335 used nano SMPS data from 7 nm to 64 nm and standard SMPS data from 64 nm to 300 nm. The nano
336 SMPS and standard SMPS data agreed within $\sim 8.8\%$ in their overlapping size range (10 – 64 nm) (Fig.
337 S4), similar to a previous study (Watson et al., 2011). In addition, a summary of total number
338 concentrations of particles for these periods is included in Table 1.

339

340 - *Arctic Marine*. A trimodal distribution was seen at $18 \pm 3 \text{ nm}$, $53 \pm 6 \text{ nm}$ and $150 \pm 6 \text{ nm}$. The first
341 mode is due to NPF arriving from open pack sea ice and open ocean Arctic regions, as discussed in

342 Section 3.2.1 where a case study is presented. The Aitken mode (~ 53 nm) is remarkably similar to the
343 Pacific Ocean aerosol size distribution and to previous studies detected in the Arctic regions (Tunved et
344 al., 2013; Freud et al., 2017; Dall'Osto et al., 2019). The largest mode at ~ 150 nm may be due to a
345 combination of primary and secondary aerosol components.

346
347 - *Arctic terrestrial*. A bimodal distribution is seen with two main modes at 24 ± 3 nm and 151 ± 3 nm,
348 respectively. The nucleation and Aitken modes are much higher than the accumulation mode, suggesting
349 that NPF governs the aerosol processes in this coastal region at this time of the year.

350
351 - *Pacific marine*. The Pacific Ocean aerosol size distributions showed a trimodal size distribution at 56
352 ± 3 nm, 130 ± 3 nm and 220 ± 6 nm. The lowest peak at ~ 56 nm (i.e., Aitken mode) is likely a
353 combination of primary and secondary marine aerosol components, whereas the largest peak at ~ 220
354 nm might be caused by cloud processing and aged aerosols. The mode at ~ 130 nm could originate from
355 primary sea spray aerosols in the Pacific atmosphere (Quinn et al., 2015). When the distribution is fitted
356 with log-normal modes, the inter-modal minimum is calculated to be ~ 120 nm - often known as Hoppel
357 minimum as a signature of cloud processing (Hoppel et al., 1994) - although, it is difficult to draw a
358 firm conclusion due to the overlap with the third mode at ~ 130 nm.

359
360 This study shows that aerosol originating from higher and lower marine latitudes – although both
361 being treated as marine air masses - have very different features, as pointed out in several previous
362 studies (Dall'Osto et al., 2010; Frossard et al., 2014). A key conclusion of this study is that we also need
363 to separate different bioregions in the Arctic, especially given the current results showing very different
364 aerosol size distributions in the Arctic study areas (Fig. 7; Arctic marine and Arctic terrestrial). The
365 reasons for the much higher aerosol concentrations near the coast of Alaska relative to the open ocean
366 sympagic and pelagic regions may be multiple. We discuss at least two major sources may contribute to
367 the high aerosol concentrations recorded.

368 The first source of aerosols in the [late summer](#) terrestrial Arctic air masses may be due to

369 anthropogenic sources. Due to sea ice retreat and better technologies, the Arctic is now easily accessible
370 to human activities, including oil and gas extraction (Law and Stohl, 2007; Peters et al., 2011). These
371 Arctic oil fields can emit the large amounts of aerosols, and with on-going Arctic development, such
372 local combustion emissions may increase in the future, possibly affecting local air quality (Gunsch et al.,
373 2017; Schmale et al., 2018a). In fact, some NPF events were reported within the North Slope of Alaska
374 (e.g., Prudhoe Bay oil fields) during August and September 2016 at Oliktok Point Alaska. This
375 observation was suggested to be linked with oil fields emissions (Kolesar et al., 2017). However, our
376 measurements were conducted in the open ocean, quite far from any land oil field local emissions. BC
377 data were collected as shown in Fig. 8; they revealed very high standard deviations due to high
378 detection limit of the instrument used relative to the concentrations detected. However, no remarkable
379 differences can be seen, all pointing to pristine clean marine air masses with BC values of
380 approximately $20 \pm 10 \text{ ng m}^{-3}$. The two Arctic categories (Marine and Terrestrial) shows similar BC
381 values, whereas higher values can be seen for the Pacific marine aerosol category, probably due to
382 contamination from nearby Asian high pollutant sources.

383 The second source of aerosol in the [late summer](#) terrestrial Arctic air masses may be due to terrestrial
384 natural sources. We believe that this may be a much more probable reason. The Arctic Ocean is
385 submerged under areas of relatively shallow water known as a shelf sea for ~50% of its area. It is a
386 relatively small ocean, characterized by pronounced riverine influence and a complex hydrography. Up
387 to 11% of the entire global river discharge ends up in the Arctic Ocean (Shiklomanov et al., 2000),
388 which is only 1% of the global ocean volume. The discharge of freshwater is increasing (Peterson et al.,
389 2002), impacting coastal salinity and carbon cycle. Indeed, this continental runoff is a major source of
390 freshwater, nutrients and terrigenous material to the Arctic Ocean (Benner et al., 2005; Fichot et al.,
391 2013; Massicotte et al., 2017). The warming climate in the region is causing permafrost degradation,
392 alterations to regional hydrology and shifting amounts and composition of dissolved organic matter
393 (DOM) transported by streams and rivers (Mann et al., 2016; Chen et al., 2017). Overall, there is a
394 considerable spatial and temporal heterogeneity in the distribution of the DOC in the Arctic, owing to

395 strong biological and physicochemical processes. It is important to remember that sea ice formation and
396 melting also affects the concentrations and distributions of DOC, although its impact is still difficult to
397 resolve (Fichot et al., 2013; Shen et al., 2012).

398 In a recent paper (Park et al., 2019), we suggested that the large amount of freshwater from river
399 runoff may have a substantial impact on primary aerosol production mechanisms, possibly affecting the
400 cloud radiative forcing. We showed that the Arctic riverine organic matter can be directly emitted from
401 surface seawater into the atmosphere via bubble bursting (Park et al., 2019). The high amount of DOC
402 populating the sea-surface microlayer (SML) in the Arctic waters - including UV absorbing humic
403 substances - can also produce VOCs (Ciuraru et al., 2015; Fu et al., 2015), which are known precursors
404 of secondary organic aerosols. Recently, Mungall et al. (2017) reported that the marine microlayer in the
405 Canadian Arctic Archipelago is a source of oxidized VOCs (OVOCs), which could be an important
406 source of biogenic secondary organic aerosol (Croft et al., 2019). Previous studies also reported
407 fluorescent water-soluble organic aerosols in the High Arctic atmosphere (Fu et al., 2015). It is worth
408 noting that terrestrial VOCs from tundra and lakes at elevated concentrations were reported (Potosnak et
409 al., 2013; Lindwall et al., 2016; Steinke et al., 2018).

410 Fig. 9 shows DOC concentrations from water samples taken in the areas where the NPF marine and
411 terrestrial case studies (Section 3.2.1 and 3.2.2) were detected. It is clear that as much as twice higher
412 concentrations are seen for the coastal marine areas, relative to the open ocean marine regions. The
413 origin of this organic matter can be obtained by the FDOM analysis. Fig. 9 (bottom) shows specific
414 peaks attributed to different chemical features. The ratio of terrestrial humic substances (peak A) was
415 3.5 for the terrestrial/marine samples. By striking contrast, marine fulvic substances (peak M) and
416 proteinaceous (peak T) had a ratio of 0.45 and 0.27, respectively, showing two very distinct chemical
417 compounds. This suggests that coastal oceanic water enriched in river organic material as well as fresh
418 water tundra and lake may be a source of VOC (both from biotic and abiotic emission processes) that
419 may be responsible for the high secondary aerosols detected near these areas.

420

3.4. Particle growth rates and condensation sink

Table 1 shows particle growth rate (GR) and condensation sink (CS) for Arctic marine, Arctic terrestrial and Pacific marine air masses. The GR was calculated by fitting a linear regression to the peak diameter of the aerosol size distribution for the nucleation-mode between 4 and 20 nm against time during the NPF cases (Dal Maso et al., 2005; Pierce et al., 2014). The GR observed during the Arctic marine and Arctic terrestrial air masses were the $0.4 \pm 0.3 \text{ nm h}^{-1}$ and $0.8 \pm 0.2 \text{ nm h}^{-1}$, respectively, which was similar to the values previously observed from other Arctic regions. A shipboard expedition conducted during the summers of 2014 and 2016 throughout the Canadian Arctic, indicated that the GR varied widely from 0.2 to 15.3 nm h^{-1} (Collins et al., 2017). The GR observed at Summit, Greenland was $0.2 \pm 0.1 \text{ nm h}^{-1}$ (range of 0.09 to 0.3 nm h^{-1}) (Ziemba et al., 2010). Similarly, in Utqiagivik (Barrow), Alaska, the GR was 1.0 nm h^{-1} in air mass influenced by Beaufort Sea, whereas the value was 11.1 nm h^{-1} in air mass influenced by Prudhoe Bay (i.e., oil field area) (Kolesar et al., 2017). Particularly, simultaneous growth of multiple modes was present in some cases (9/13/2017– 9/21/2017). We calculated the GR of the distinct modes, as shown in Fig. S6. The results showed that growth of the larger mode (e.g., preexisting mode) was faster than the smaller mode (e.g., nucleation mode), consistent with ship-based aerosol measurements in the summertime Arctic by Burkart et al. (2017b). They proposed that growth was largely via condensation of semi-volatile organic material, because lower volatile organics could lead to faster growth of the smaller mode.

The CS is a key parameter assessing the NPF and growth and determines how rapidly gaseous molecules condense onto pre-existing particles. The CS was calculated, following Dal Maso et al. (2002) and Collines et al. (2017). The resulting CS values are given in Table 1. The CS observed during the Arctic marine and Arctic terrestrial air masses were $0.5 \pm 0.4 \text{ nm h}^{-1}$ and $0.9 \pm 0.5 \text{ nm h}^{-1}$, respectively. The CS in this study is on the low end of the values observed during the summer in Arctic marine boundary layer (shipborne expeditions) (Collins et al., 2017), Utqiagivik, Alaska (Kolesar et al., 2017), and Ny-Ålesund, Svalbard (Giamarelou et al., 2016). In case when air mass passed over the Pacific Ocean, the CS was 2 or 4 times higher than those of Arctic air masses. It seems that such higher

447 CS for Pacific marine air masses lowered the concentration of condensable vapors, thereby resulting in
448 the non-event days in Pacific marine air masses.

449 3.5. Impact on CCN number concentrations

450 Fig. 10a illustrates the CCN concentrations for the three selected periods under different
451 supersaturation conditions. For a given SS of 0.4%, CCN concentrations for Arctic marine, Arctic
452 terrestrial and Pacific marine air masses were $35 \pm 40 \text{ cm}^{-3}$, $71 \pm 47 \text{ cm}^{-3}$, and $204 \pm 87 \text{ cm}^{-3}$,
453 respectively. Higher concentrations of CCN were observed when the air mass originated from the
454 Pacific marine for a SS of 0.2%–1.0 %. This may have occurred due to the differences in the CCN
455 sources between the Arctic and Pacific Oceans. It was noted that that accumulation and coarse-mode
456 particles, which are predominant over the Pacific Ocean (Fig. 7), can easily act as CCN. Our results
457 agreed well with values reported in previous studies that measured CCN at a ground-based Arctic
458 station (Jung et al., 2018), but was slightly higher than those measured from high-Arctic expeditions
459 (Leck et al., 2002; Martin et al., 2011; Mauritsen et al., 2011). For example, Jung et al. (2018) reported
460 seasonal variations in the CCN concentration over seven years (2007 –2013) at the Zeppelin station, and
461 found that the monthly mean CCN concentrations ranged from 17 cm^{-3} in October 2007 to 198 cm^{-3} in
462 March 2008 at a SS value of 0.4%. However, Mauritsen et al. (2011) observed CCN concentrations
463 lower than $\sim 100 \text{ cm}^{-3}$ at five different supersaturations (SS = 0.10%, 0.15%, 0.20%, 0.41%, and 0.73%),
464 with median values ranging from 15 to 50 cm^{-3} , in four High Arctic expeditions during the Arctic
465 Summer Cloud Ocean Study. Such values were also in line with the long term measurement at an Arctic
466 station in Barrow, which indicated that the median CCN concentrations at 0.2% SS was smaller than
467 100 cm^{-3} (Schmale et al., 2018b).

468 We also compared CCN activity and critical diameter for the three selected periods, as shown in Fig.
469 10b and c. The CCN activity is defined as the ratio of the number concentration of particles that
470 activated to become CCN at a given supersaturation to the total number concentration of particles larger
471 than 2.5nm ($\text{CN}_{2.5}$). The CCN activity followed a similar pattern as the CCN concentration.
472 Furthermore, the critical diameter (D_c) (i.e., the diameter at which the integration of aerosol size

473 distribution from the largest particle diameter to the lower ones matches with the measured CCN
474 concentration) was estimated using the measured aerosol size distribution, $CN_{2.5}$, and CCN
475 concentrations with a time resolution of 1 h, as described by Furutani et al., (2014). The D_c at a SS of
476 0.4% was found to be 103 ± 43 nm, 83 ± 18 nm, and 136 ± 67 nm for Arctic marine, Arctic terrestrial,
477 and Pacific marine periods, respectively. These values are comparable to previous studies obtained in
478 the Arctic and subarctic regions. For instance, the D_c of 80 nm at 0.6 % SS was observed during the
479 aircraft measurement in July 2014 in the high Arctic marine boundary layer of Resolute Bay, Nunavut,
480 Canada (Burkart et al., 2017a). Jaatinen et al. (2014) reported that the D_c value of 98 ± 16 nm (SS =
481 0.4%) from the subarctic area of Finland (Pallas-Sodankylä Global Atmospheric Watch station). Anttila
482 et al. (2012) also showed that a D_c value was in the range of 90 to 120 nm at a SS of 0.4% during the
483 same field campaign as reported in Jaatinen et al. (2014). For a maximum SS between 0.18 and 0.26%,
484 D_c varied between 110 and 140 nm at the same measurement sites.

485 486 **4. Summary and conclusions**

487 This study presents the physical properties of aerosol particles measured aboard the R/V Araon ice-
488 breaker during 2017 throughout the Arctic and Pacific Oceans. The $CN_{2.5}$ value commonly ranged
489 between 13 and 2000 cm^{-3} with an average of 505 ± 280 cm^{-3} . An elevated $CN_{2.5}$ concentration reaching
490 ~ 6016 cm^{-3} was observed from 13 September to 20 September. The temporal variations in the $CN_{2.5}$
491 concentration followed a similar pattern to those of N_{NUA} and N_{AIT} . We also found that the $CN_{2.5}$
492 concentration was strongly correlated with N_{NUA} ($r^2 = 0.69$), suggesting that CN was mainly derived
493 from nucleation-mode particles.

494 NPF events caused by gas-to-particle conversion frequently occurred over the Arctic Ocean.
495 Overall, two major NPF sources (i.e., Arctic marine and Arctic terrestrial) were identified based on the
496 backward air mass trajectory analysis. NPF events were associated with Arctic marine air masses,
497 indicating the impact of marine biogenic emissions from the Arctic Ocean. Strong NPF events with
498 particle growth were associated with Arctic terrestrial air masses, which may be due to the biogenic

499 precursor gases emitted by terrestrial ecosystems including river discharge and Alaskan tundra in the
500 Arctic coastal areas. In contrast, relatively larger particles with broad Aitken and accumulation-mode
501 peaks were observed over the Pacific Ocean. Our study confirmed that any NPF was not detected during
502 the Pacific transect. We also compared the average CCN concentrations for each of the cases. Our data
503 showed that the impact of aerosols on CCN concentrations ($SS = 0.4\%$) was significant: $35 \pm 40 \text{ cm}^{-3}$,
504 $71 \pm 47 \text{ cm}^{-3}$, and $204 \pm 87 \text{ cm}^{-3}$ for Arctic marine, Arctic terrestrial, and Pacific marine periods,
505 respectively. Our interpreted data showed that river outflows and tundra strongly influence Arctic
506 aerosol properties. Further detailed measurements of the chemical characteristics of marine aerosols are
507 required to provide more direct evidence for the contribution of biogenic precursors to the NPF and
508 CCN in the remote Arctic atmosphere.

509 Arctic areas are currently experiencing drastic climate change, with air temperatures increasing at
510 twice the rate of the global average. This warming is causing clear changes, such as the increases in
511 biogenic emissions from tundra vegetation and changes in vegetation cover (Faubert et al., 2010;
512 Peñuelas and Staudt, 2010; Potosnak et al., 2013; Lindwall et al., 2016). Lindwall et al. (2016) observed
513 a 280% increase in VOC emissions relative to the ambient level in response to a 4 °C increase in the
514 summer temperature of the Subarctic. Increases in VOC emissions from river discharge and tundra
515 vegetation in the Arctic are critical factors that induce NPF and particle growth events, which may
516 impact the CCN concentrations during the Arctic summer.

517 518 **Data availability**

519 The data analyzed in this publication will be readily provided upon request to the corresponding author
520 (yjyoon@kopri.re.kr).

521 522 **Author contributions**

523 JP, YJY designed the study, JP, MD'O, KP, YG, HJK, EJ, KTP, MP, SSY, JJ, and BYL analyzed data.
524 JP, MD'O, KTP and YJY prepared the manuscript with contributions from all co-authors.

525 526 **Competing interests**

527 The authors declare that they have no conflict of interest.

528

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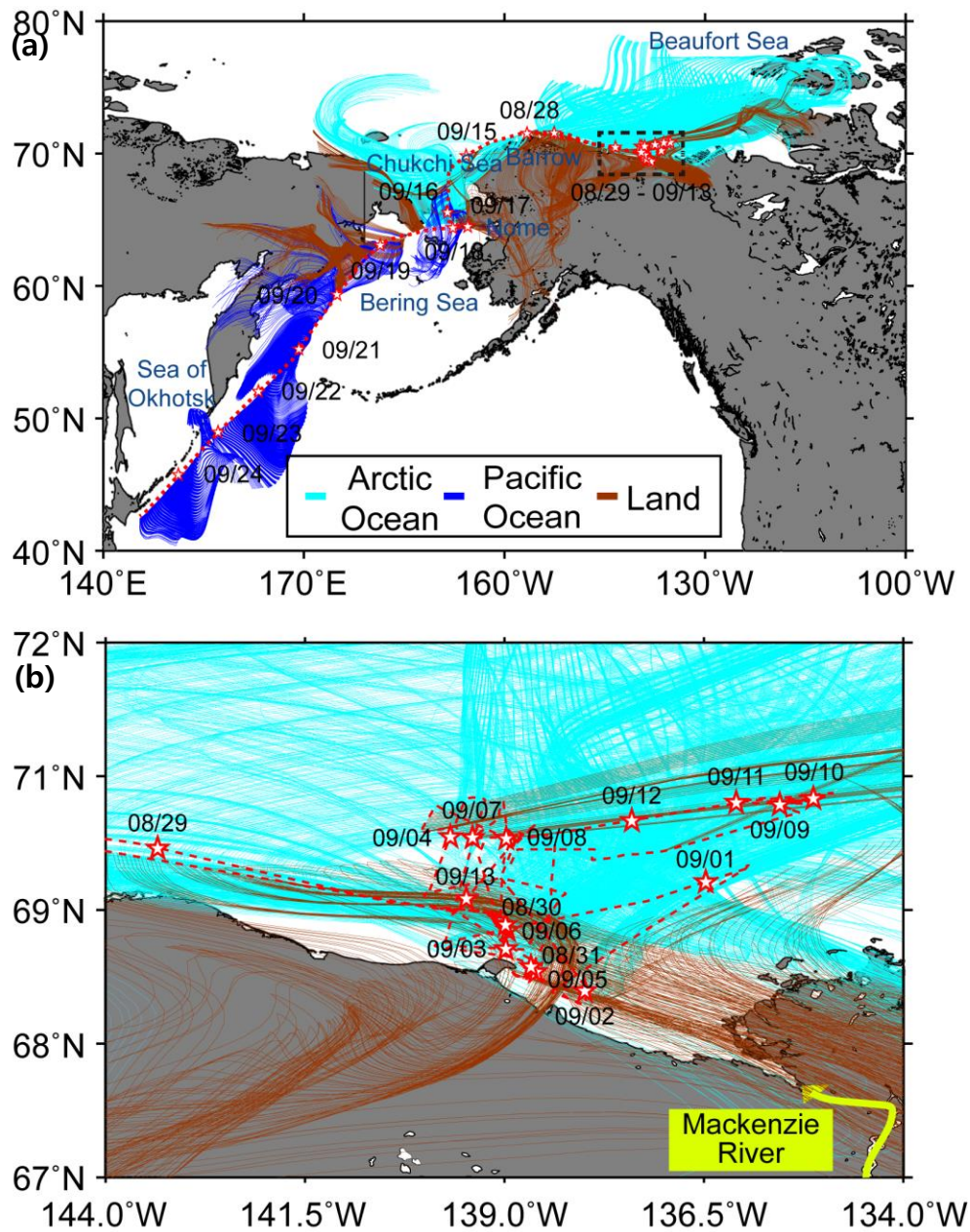
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890 Table 1. A summary of meteorology, total number concentrations of particles (measured with TSI 3776
 891 CPC, TSI 3772 CPC, Standard SMPS, nano SMPS), growth rate (GR), and condensation sink (CS) for
 892 the three selected periods. The $CN_{2.5}$ and CN_{10} represents the total number concentration of particles
 893 larger than 2.5 nm and 10 nm, respectively. The N_{NUC} , N_{AIT} , N_{ACC} , and N_{OPS} represents total aerosol
 894 nucleation-mode (3 – 20 nm), Aitken-mode (20 – 100 nm), accumulation-mode (100 – 300 nm), and
 895 coarse-mode (> 300 nm from OPS) number concentrations.

	Arctic Marine	Arctic Terrestrial	Pacific Ocean
Periods	9/02/2017– 9/05/2017, 9/10/2017– 9/12/2017	9/13/2017– 9/17/2017	9/21/2017– 9/23/2017
Wind speed ($m s^{-1}$)	6.1 ± 6.0	8.7 ± 5.7	8.4 ± 4.3
Wind direction ($^{\circ}$)	352.3 ± 38.7	344.7 ± 28.1	338.3 ± 23.0
$CN_{2.5}$ (cm^{-3})	413 ± 442	1622 ± 1450	397 ± 185
CN_{10} (cm^{-3})	414 ± 452	1396 ± 1279	384 ± 86
$CN_{2.5-10}$ (cm^{-3})	62 ± 130	263 ± 318	35 ± 195
N_{NUC} (cm^{-3})	118 ± 198	350 ± 393	46 ± 103
N_{AIT} (cm^{-3})	108 ± 132	405 ± 425	116 ± 93
N_{ACC} (cm^{-3})	19 ± 14	33 ± 20	95 ± 30
N_{OPS} (cm^{-3})	2 ± 2	3 ± 2	11 ± 6
GR ($nm h^{-1}$)	0.4 ± 0.3	0.8 ± 0.2	-
CS (h^{-1})	0.5 ± 0.4	0.9 ± 0.5	2.1 ± 0.7

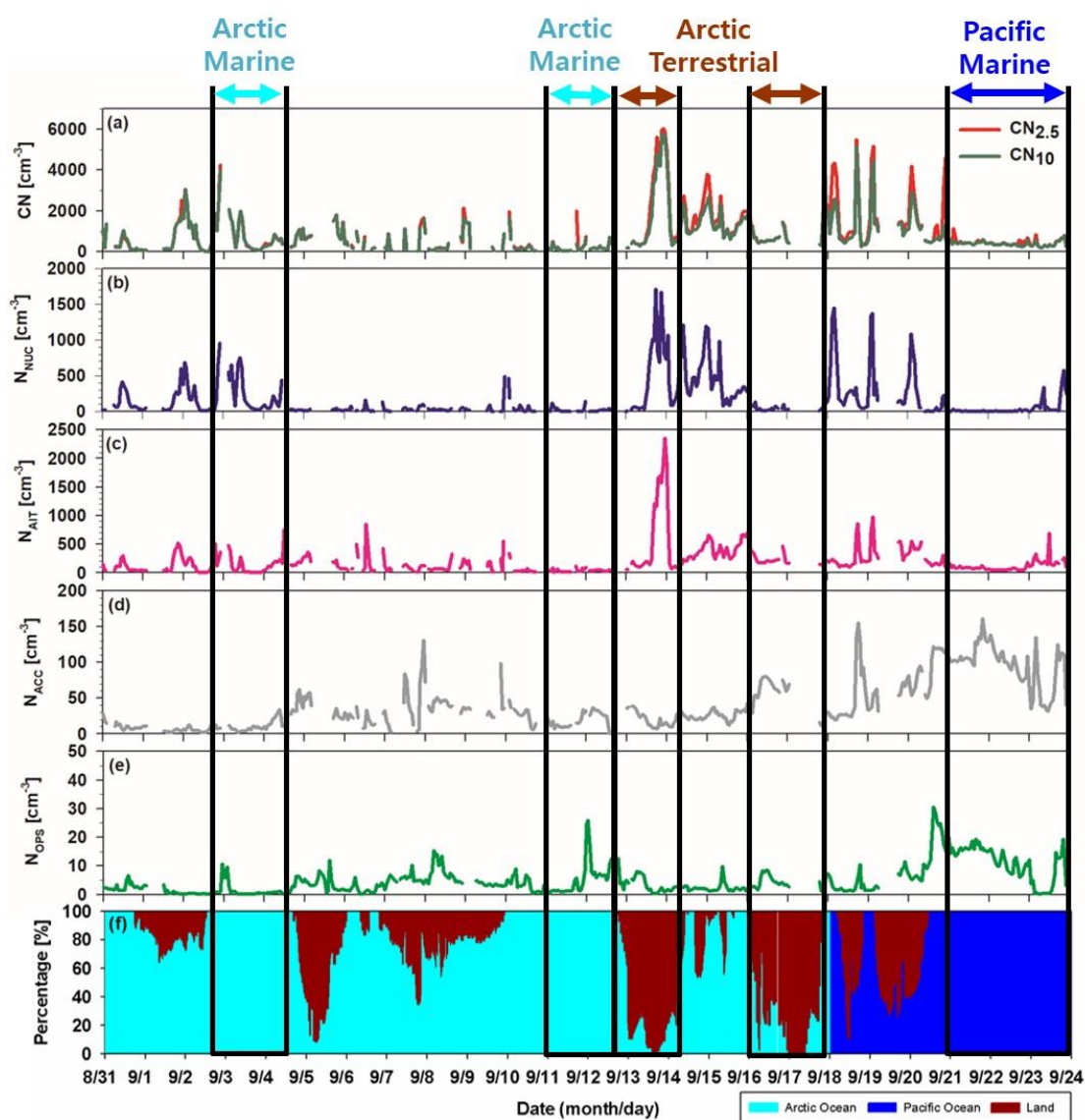
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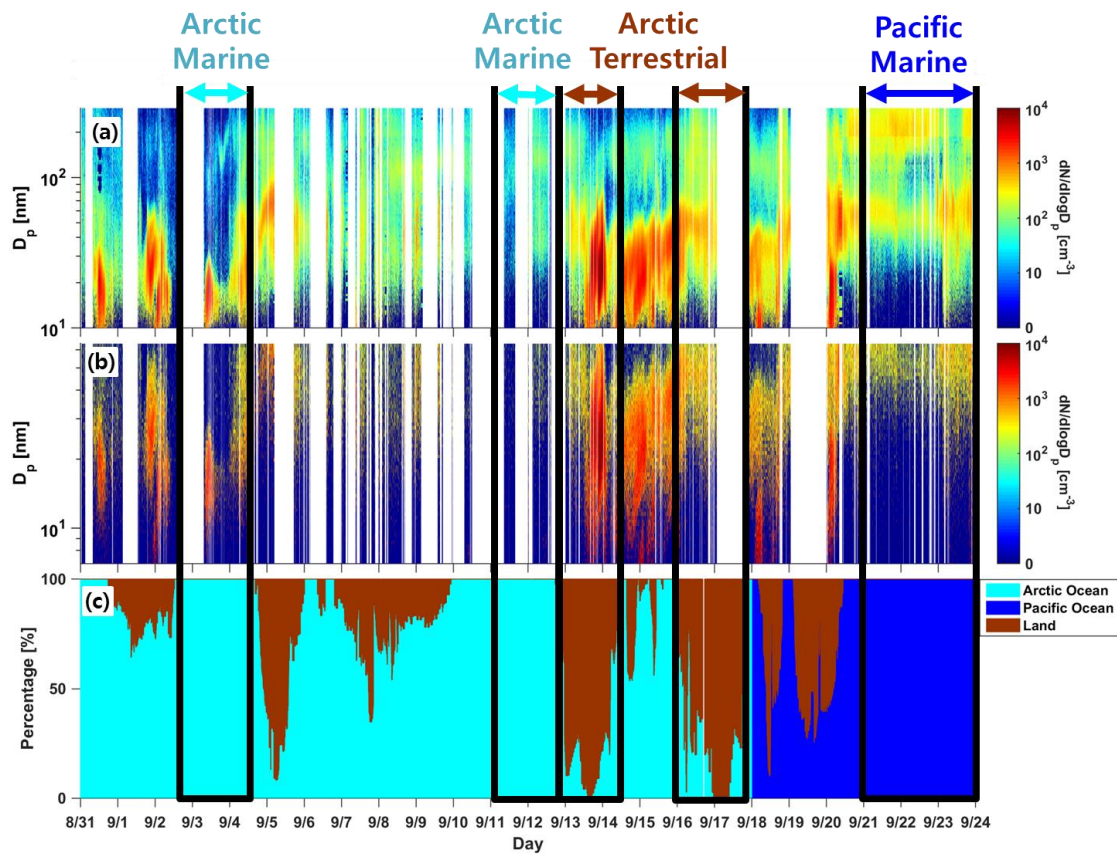
899 Figure 1. Ship tracks across (a) the Arctic (8/28/2017–9/18/2017) and Pacific Oceans
 900 (9/18/2017–9/25/2017) and (b) zoom into the dotted black square region in Fig. 1a. A dotted red line
 901 including star symbols represents ship tracks during the entire cruise. The star symbols represent the
 902 daily ship location at midnight. Light blue, blue and brown lines denote the 2-day air mass trajectories
 903 categorized into three main domains such as Arctic Ocean, Pacific Ocean, and land, respectively.



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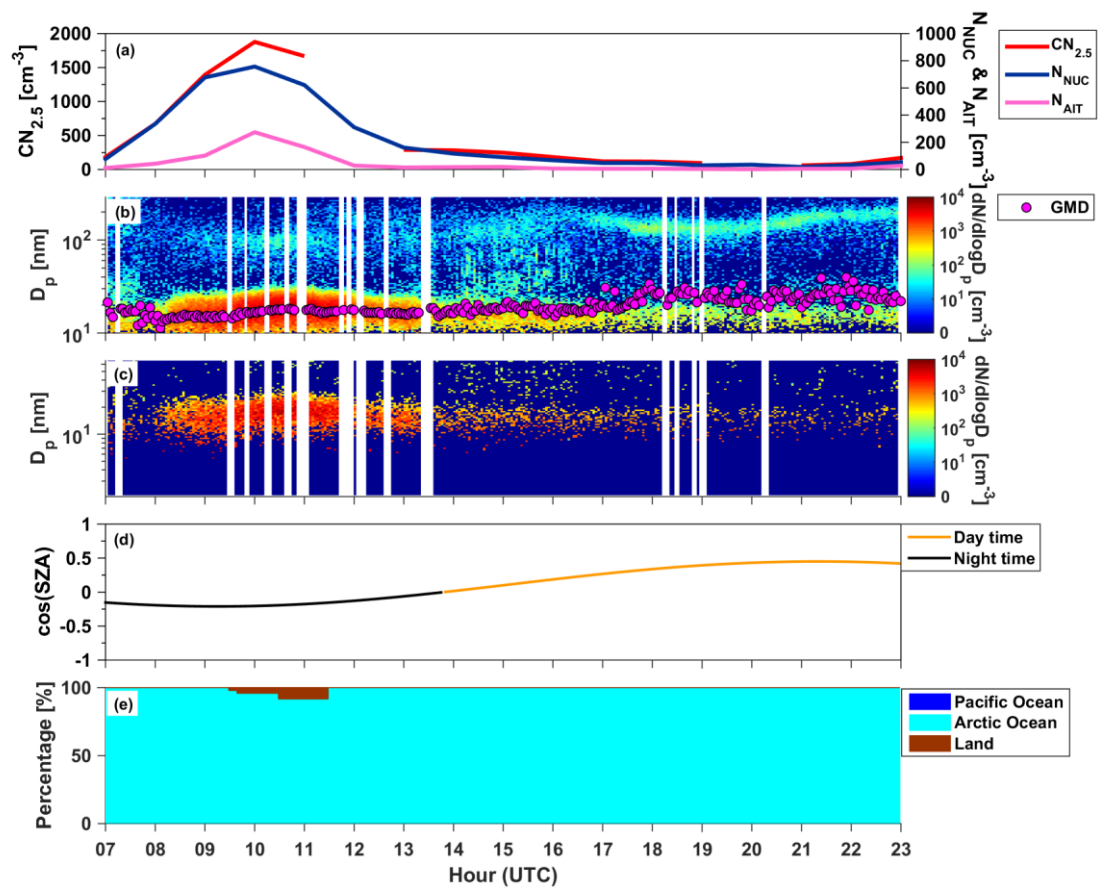
906 Figure 2. Time series of the 1 hour average (a) total aerosol ($CN_{2.5}$ and CN_{10}), (b) nucleation-mode (3 –
 907 20 nm) (N_{NUC}), (c) Aitken-mode (20 – 100 nm) (N_{AIT}), (d) accumulation-mode (100 – 300 nm) (N_{ACC}),
 908 (e) coarse-mode (> 300 nm from OPS) (N_{OPS}) number concentrations, and (f) the residence time of air
 909 masses that passed over the Arctic Ocean, Pacific Ocean, and land. The $CN_{2.5}$ and CN_{10} represent total
 910 number concentration of particles larger than 2.5 and 10 nm, respectively.

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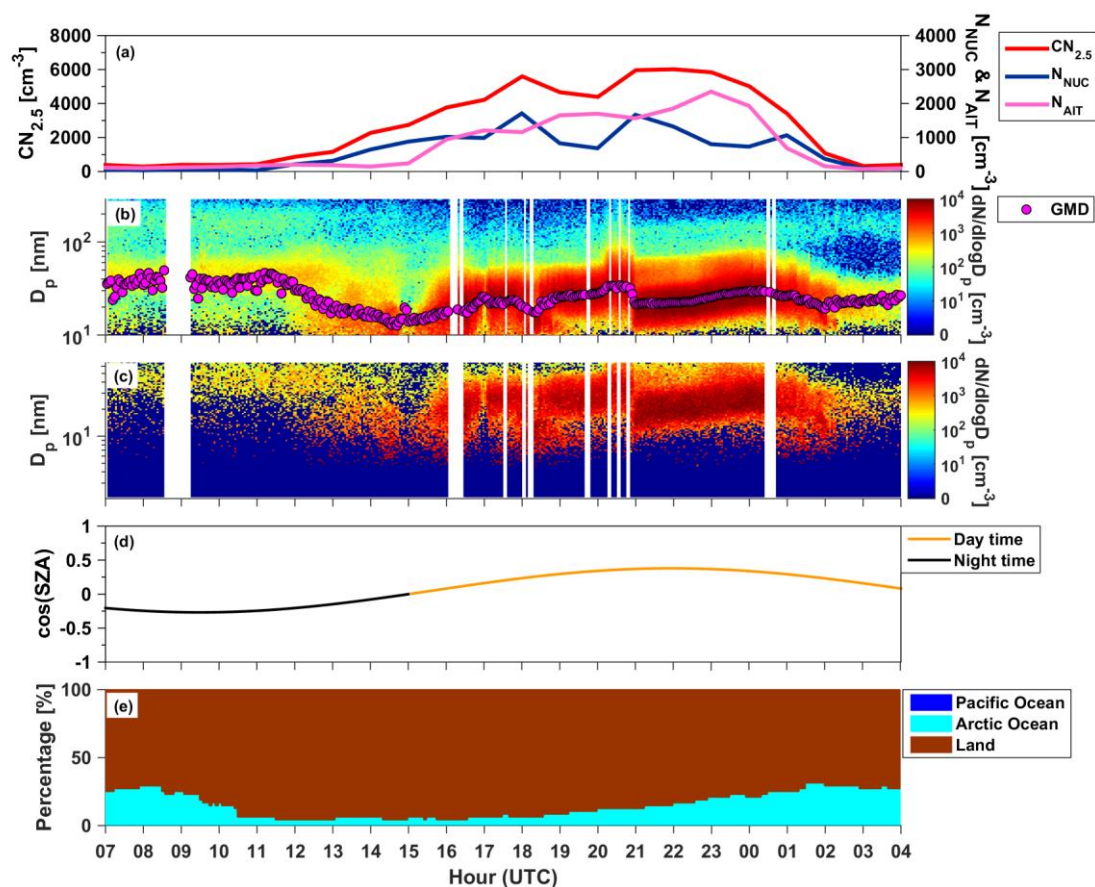
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914 Figure 3. Contour plots of the size distributions measured using (a) standard and (b) nano SMPS and (c)
 915 the residence time of air masses that passed over the Arctic Ocean, Pacific Ocean, and land throughout
 916 the sampling periods.



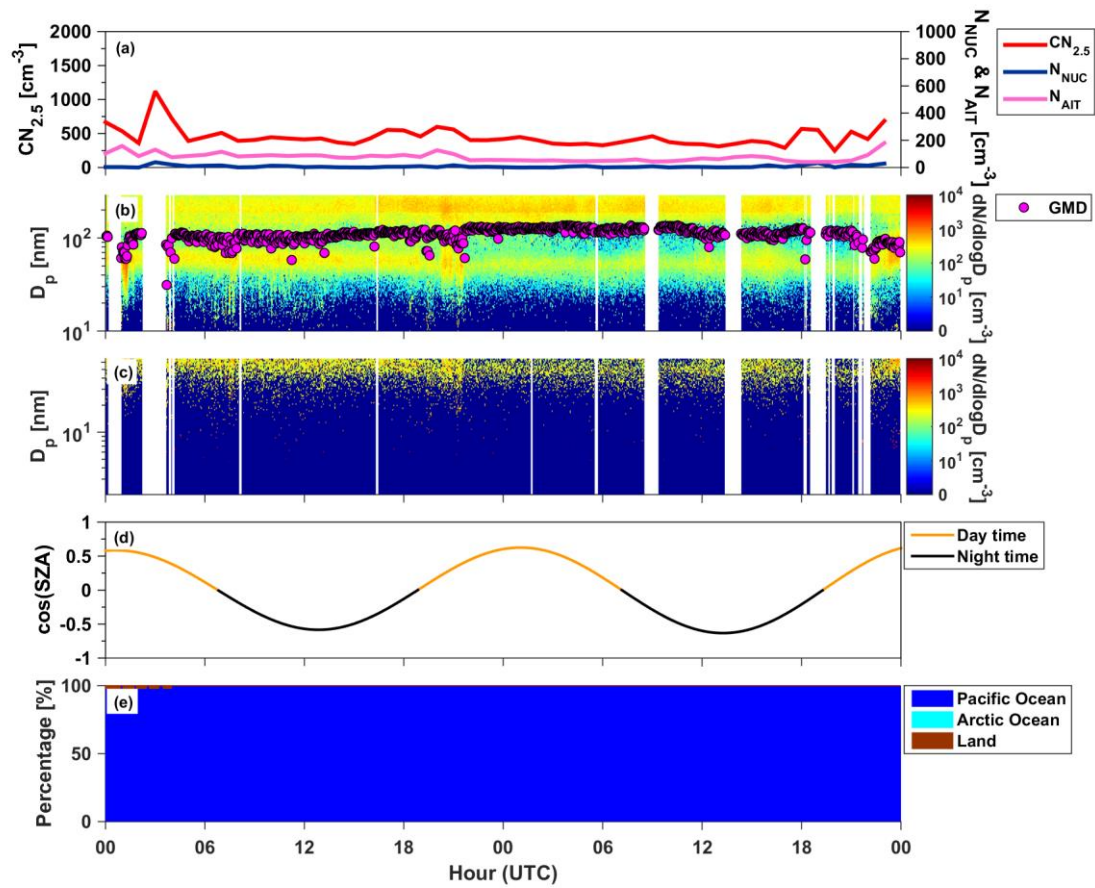
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919 Figure 4. Example of a case-I event observed on 3 September 2017. From top to bottom, the parameters
 920 are: (a) the total number concentration of particles smaller than 2.5 nm, nucleation-mode particles, and
 921 Aitken-mode particles; (b) a time series of the standard SMPS size distribution and GMD; (c) a time
 922 series of the nano SMPS size; (d) Cosine values of solar zenith angle; (e) the residence time of air
 923 masses that passed over the ocean, land, and sea-ice areas.



925

926 Figure 5. Example of a case II event that was observed on September 13–14, 2017. From top to bottom,
 927 the parameters are: (a) the total number concentration of particles smaller than 2.5 nm, nucleation-mode
 928 particles, and Aitken-mode particles; (b) a time series of the standard SMPS size distribution and GMD;
 929 (c) a time series of the nano SMPS size; (d) cosine values of solar zenith angle; (e) the residence time of
 930 air masses that passed over the ocean, land, and sea-ice areas.



932

933 Figure 6. Example of a case III event that was observed on September 21–22 2017. From top to bottom,
 934 the parameters are: (a) the total number concentration of particles smaller than 2.5 nm, nucleation-mode
 935 particles, and Aitken-mode particles; (b) a time series of the standard SMPS size distribution and GMD;
 936 (c) a time series of the nano SMPS size; (d) cosine values of solar zenith angle; (e) the residence time of
 937 air masses that passed over the ocean, land, and sea-ice areas.

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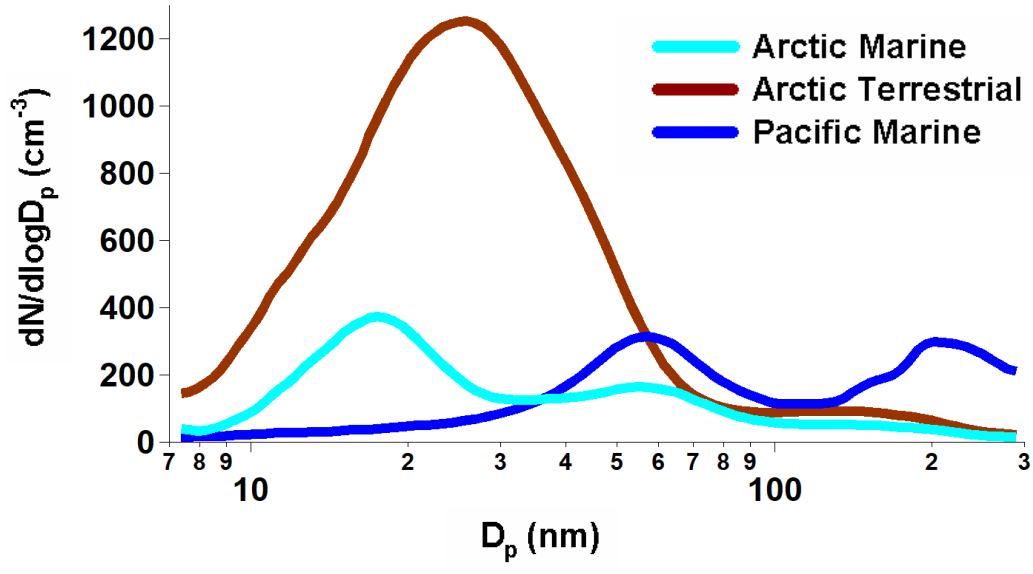
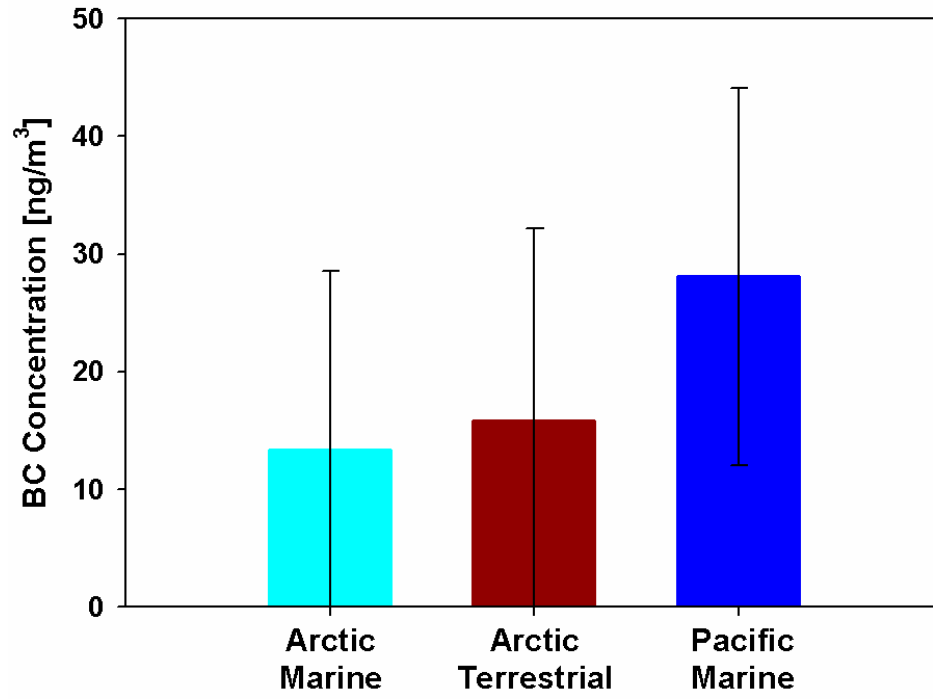
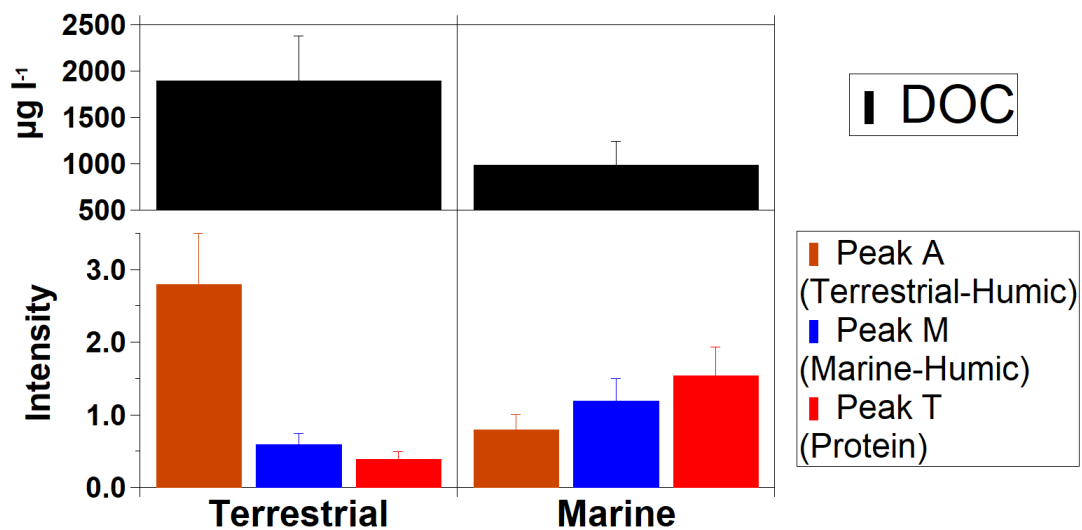


Figure 7. Average size distributions of aerosol particles for Arctic marine, Arctic terrestrial and Pacific marine air masses



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953 Figure 8. Average mass concentrations of black carbon for each air mass.



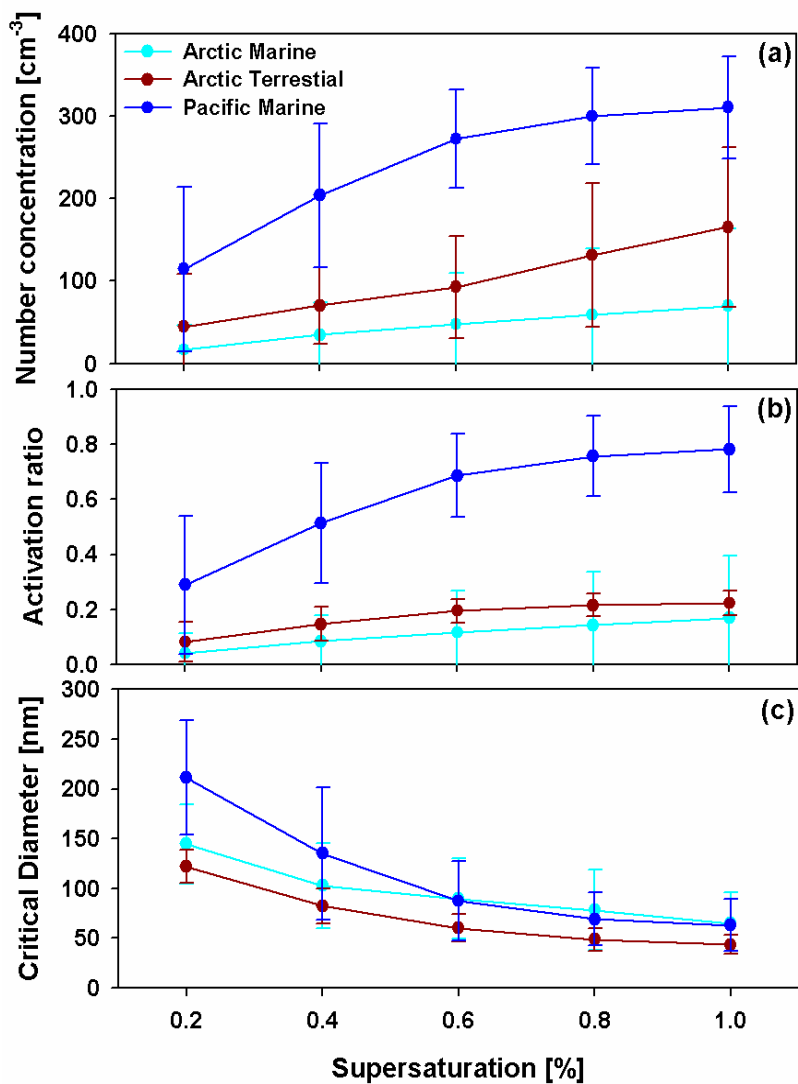
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956 Figure 9. Average DOC concentrations for surface seawater samples collected during this cruise,
 957 simultaneously during the atmospheric measurements herein reported. Peak A, M, and T represent
 958 terrestrial-humic substances, marine-fulvic substances, and protein, respectively.

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964 Figure 10. Comparisons of (a) CCN number concentrations, (b) CCN activity, and (c) critical diameter
 965 for Arctic marine, Arctic terrestrial and Pacific marine air masses under different supersaturation
 966 conditions. The error bars represent a standard deviation.

968 **Shipborne observations reveal contrasting Arctic marine, Arctic**
969 **terrestrial and Pacific marine aerosol properties**

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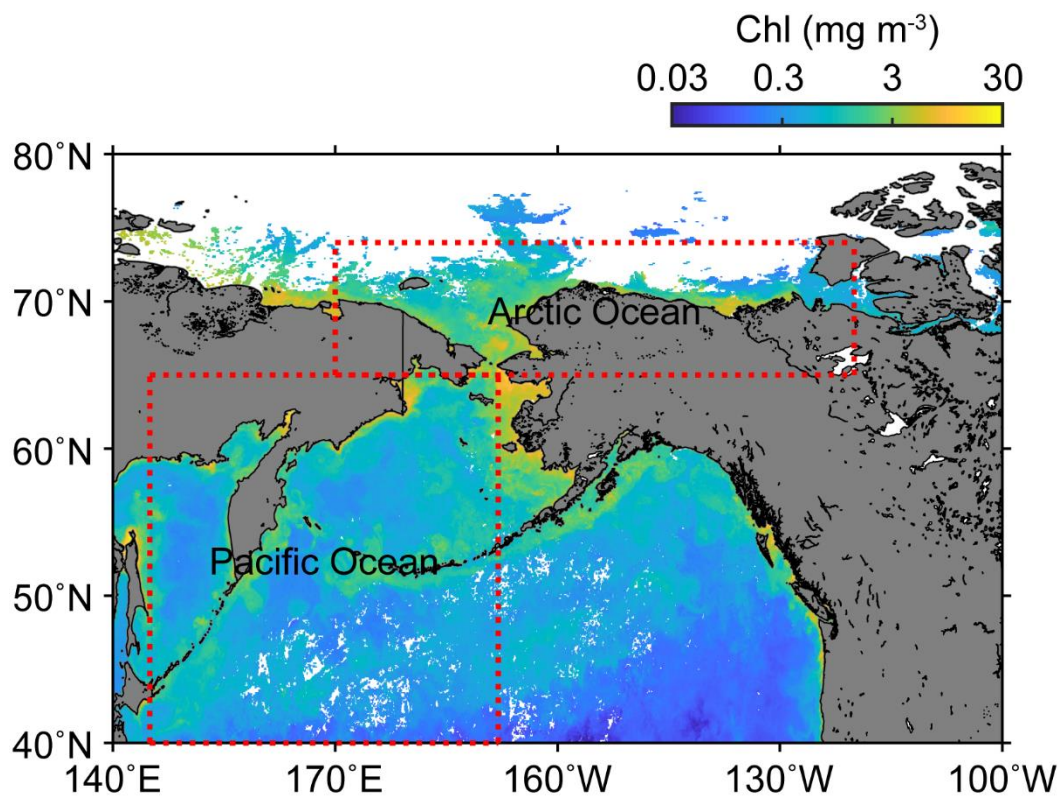
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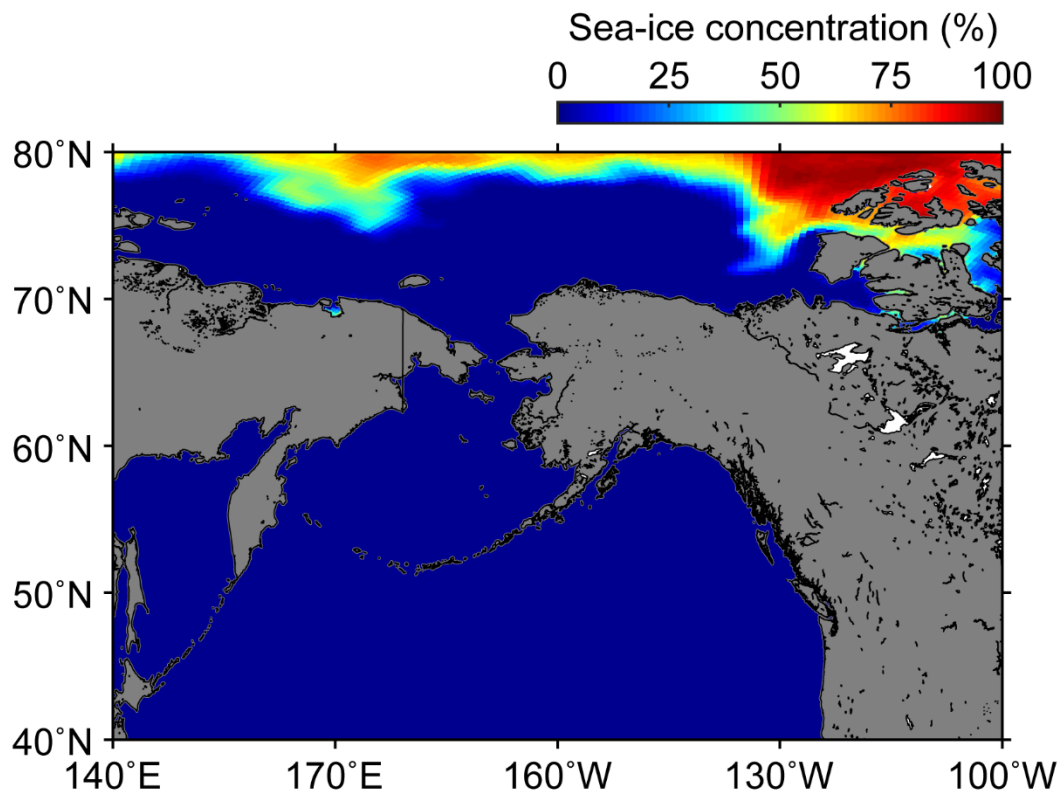
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982 Figure S1. The monthly mean Chlorophyll-*a* concentration in September 2017 (an index of
983 phytoplankton biomass over the ocean) obtained from Satellite data (Aqua Moderate Resolution
984 Imaging Spectroradiometer). The dotted red lines indicate the domains for Arctic Ocean (65°N–74°N
985 and 170°E–120°W) and Pacific Ocean (40°N–65°N and 145°E–168°W).

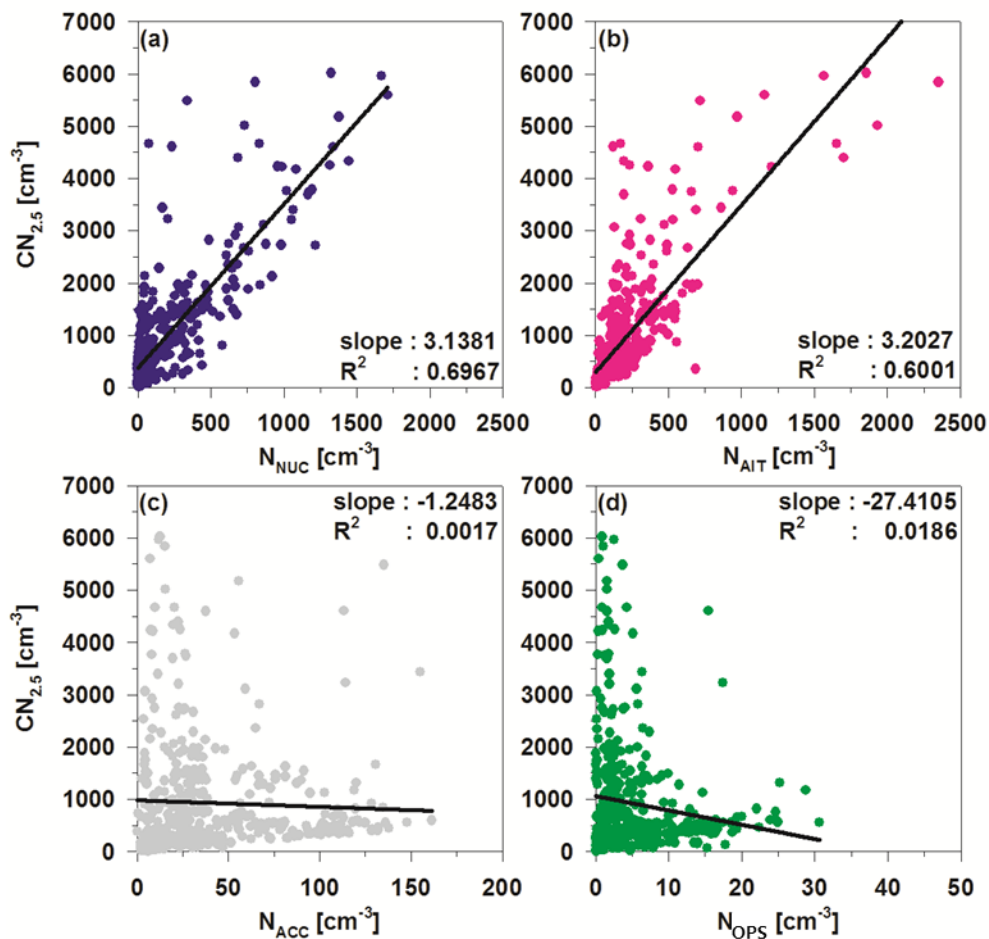
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989 Figure S2. The monthly mean sea-ice concentration in September 2017 obtained from the Sea Ice Index
990 (SII) provided by the National Snow and Ice Data Center, (<https://doi.org/10.7265/N5K072F8>).

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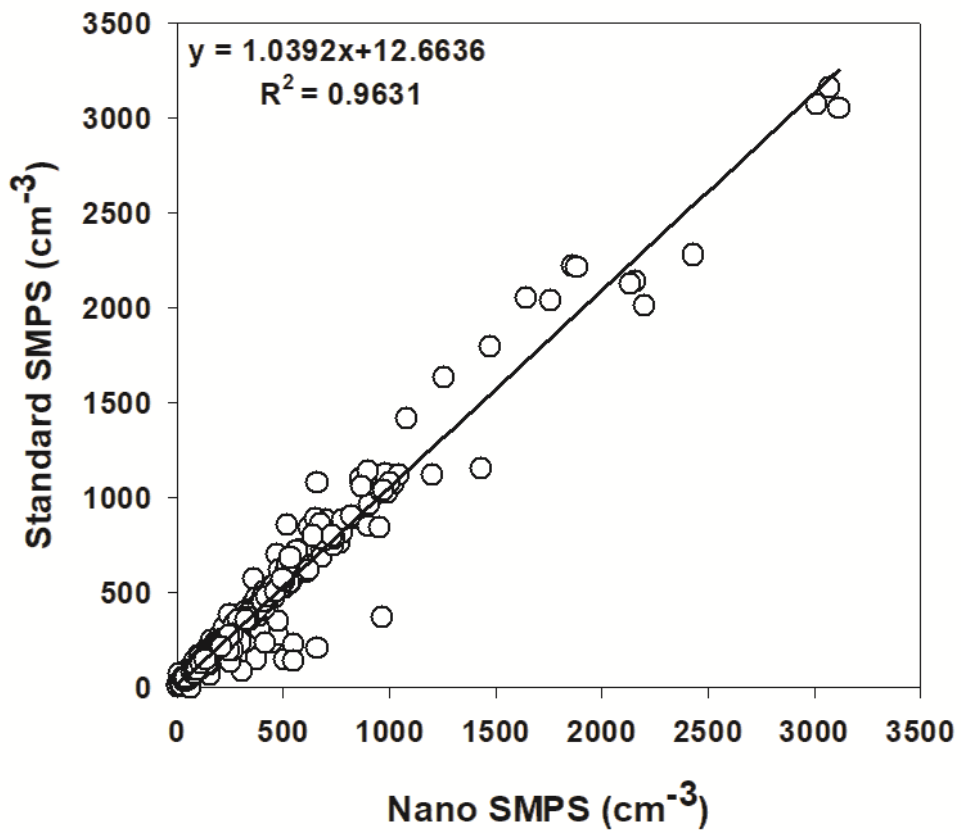
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993 Figure S3. Correlations between (a) $CN_{2.5}$ and N_{NUC} , (b) $CN_{2.5}$ and N_{AIT} , (c) $CN_{2.5}$ and N_{ACC} , and (d)
 994 $CN_{2.5}$ and N_{OPS} during the entire sampling periods

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999 [Figure S4. Comparisons of hourly average particle number concentrations in the 10 – 64 nm size range](#)
1000 [measured with nano SMPS and standard SMPS](#)

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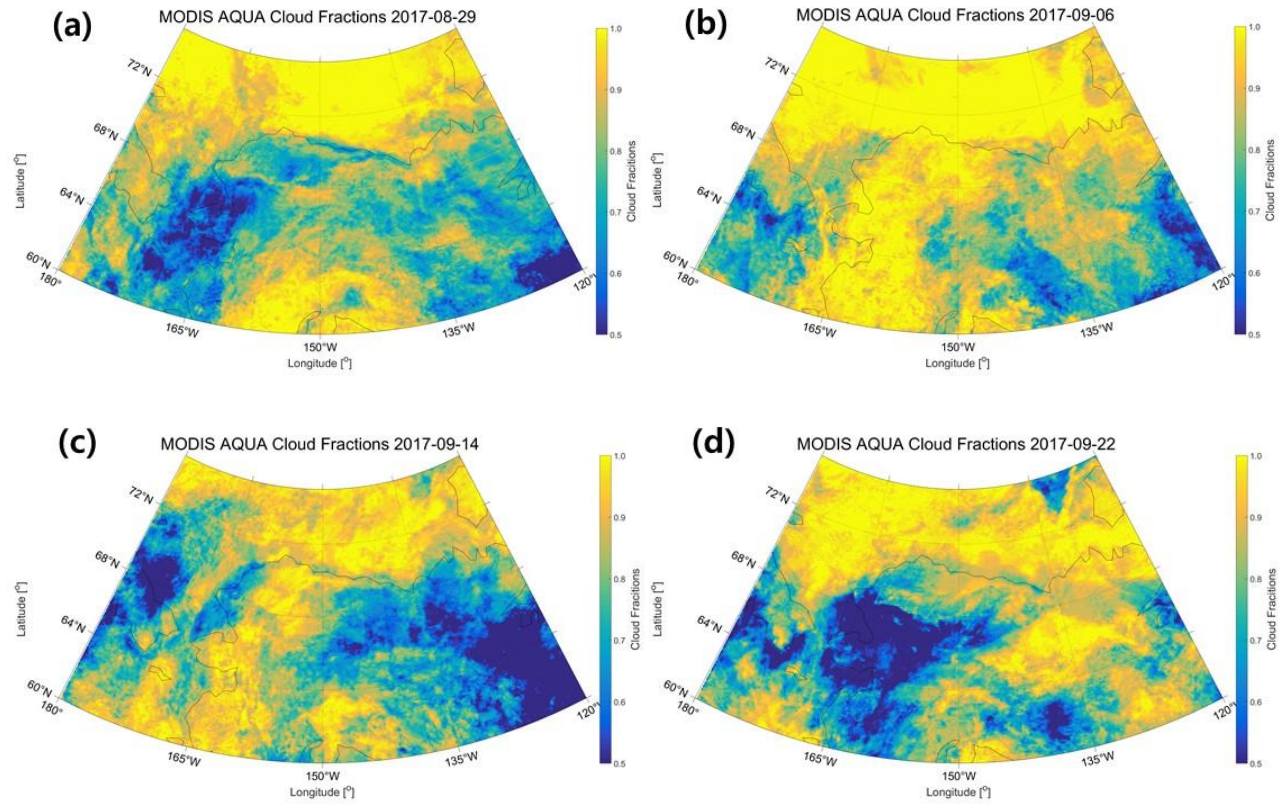
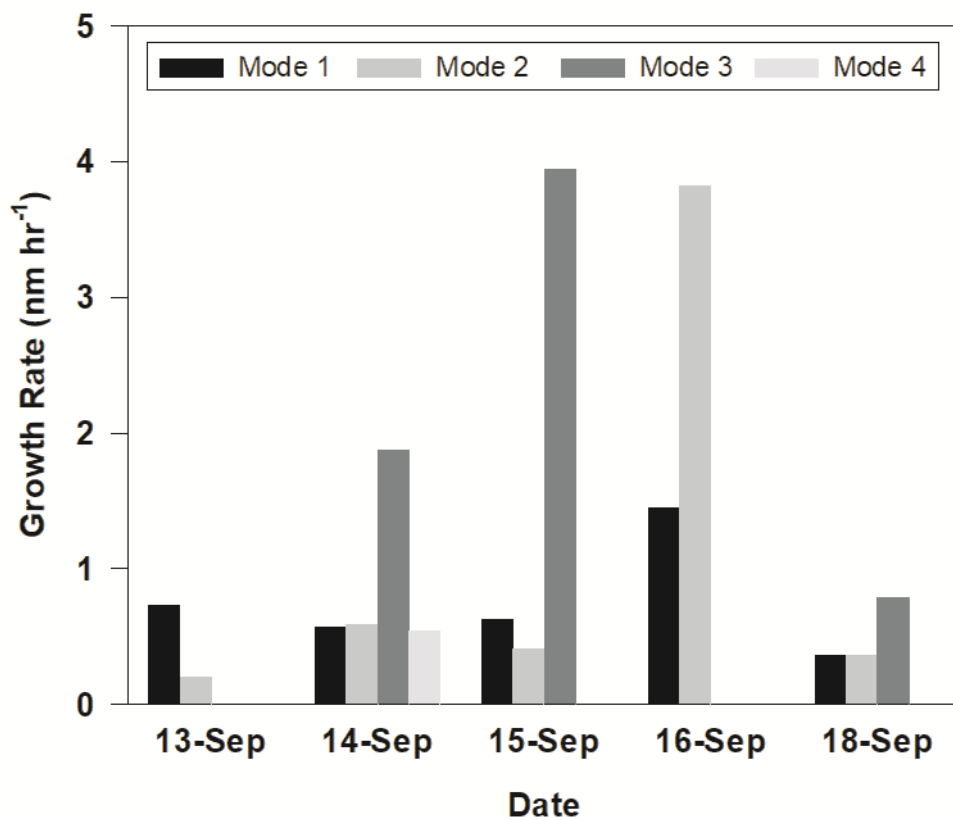


Figure S5. Weekly averaged liquid cloud fraction from Aqua/MODIS retrieval data for (a) week 1(8/29/2017– 9/5/2017), (b) week 2 (9/6/2017– 9/13/2017), week 3 (9/14/2017– 9/21/2017), and week 4 (9/22/2017– 9/29/2017). Data were obtained from <https://neo.sci.gsfc.nasa.gov/>.



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1011 [Figure S6. Growth rate for the different mode particles. Mode 1 is the smallest mode and Mode 2 the](#)
1012 [next smallest, etc.](#)

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