# New particle formation leads to enhanced cloud condensation nuclei concentrations in Antarctic Peninsula

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## 14 Abstract

Few studies have investigated the impact of new particle formation (NPF) on cloud condensation 15 nuclei (CCN) in remote Antarctica, and none has elucidated the relationship between NPF and CCN 16 production. To address that knowledge gap, we continuously measured the number size distribution of 17 2.5–300 nm particles and CCN number concentrations at King Sejong Station in the Antarctic Peninsula 18 from January 1 to December 31, 2018. Ninety-seven new particle formation (NPF) events were detected 19 throughout the year. Clear annual and seasonal patterns of NPF were observed: high concentration and 20 frequency of nucleation-mode particles in summer (December-February: 53 NPF cases) and undetected 21 nucleation-mode particles in winter (June-August: no NPF cases). We estimated the spatial scale of NPF 22 by multiplying the time during which a distinct nucleation mode can be observed at the sampling site by 23 the locally measured wind speed. The estimated median spatial scale of NPF around Antarctic peninsula 24 was found to be approximately 155 km, indicating the large-scale of NPF events. Air back-trajectory 25 analysis revealed that 80 cases of NPF events were associated with air masses originating over the ocean, 26 27 followed by sea-ice (12 cases), multiple (3 cases), and land (2 cases) regions. We present and discuss three major NPF categories: (1) marine NPF (2) sea-ice NPF, and (3) multiple NPF. Satellite-estimates 28 for sea surface dimethylsulfoniopropionate (DMSP; a precursor of gaseous dimethyl sulfide) data showed 29

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that the production of oceanic biogenic precursors could be a key component in marine NPF events, 30 31 whereas halogen compounds released from ice-covered areas could contribute to sea-ice NPF events. 32 Terrestrial sources (wild life colonies, vegetation, and meltwater ponds) from Antarctica could affect aerosol production in multiple air masses. Out of 97 observed NPF events, 83 cases were characterized 33 34 by the simultaneous increase in the CCN concentration by 2-270% (median 44%) in the following 1 to 36 hours (median 8 hours) after NPF events. Overall, Antarctic NPF events were found to be a significant 35 source of particles with different physical characteristics and related to biogenic sources in and around 36 the Antarctic Peninsula, which subsequently grew to cloud condensation nuclei. 37

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## 39 1. Introduction

40 Antarctic peninsula is warming more rapidly than Earth's global mean rate (Chen et al., 2009; Vaughan et al., 2003), leading to shrinking sea-ice coverage and consequent sea-level rise (Pritchard et 41 al., 2009). In the Antarctic region, ambient aerosols play a crucial role in governing radiative transfer, 42 43 directly by the scattering and absorption of solar radiation and indirectly by acting as cloud condensation nuclei (CCN) (IPCC, 2013). The magnitude of the radiative forcing caused by the interactions between 44 aerosols and CCN remains highly uncertain due to a poor understanding of pristine natural aerosols 45 (Carslaw et al., 2013). To reduce this uncertainty, the physicochemical properties of aerosol particles (e.g., 46 number concentrations, size distributions, chemical compositions, and hygroscopicity) have been studied 47 at several Antarctic stations including King Sejong Station (Kim et al., 2019), Aboa (Asmi et al., 2010; 48 Virkkula et al., 2006), Dome C (Järvinen et al., 2013), Halley (Lachlan-Cope et al., 2020; O'Dowd et al., 49 1997), Kohen (Weller et al., 2018), McMurdo (Giordano et al., 2018; Liu et al., 2018), Neumayer (Teinilä 50 51 et al., 2014; Weller et al., 2015), Princess Elisabeth (Herenz et al., 2019) and Syowa (Hara et al., 2011; Ito, 1993). Furthermore, open ocean and coastal Antarctic expeditions such as SIPEXII (Sea Ice Physics 52 and Ecosystems eXperiment, 2012; Humphries et al., 2015; Humphries et al., 2016), PEGASO (Plankton-53 54 derived Emissions of trace Gases and Aerosols in the Southern Ocean, 2015; Dall'Osto et al., 2017; Decesari et al., 2020; Fossum et al., 2018), ACE-SPACE (Antarctic Circumnavigation Expedition - Study 55

of Preindustrial-like Aerosol Climate Effects, 2017; Schmale et al., 2019; Walton and Thomas, 2018), 56 PCAN (Polar Cell Aerosol Nucleation, 2017; Simmons et al., 2021); PI-ICE (Polar atmosphere-ice-ocean 57 Interactions: Impact on Climate and Ecology, 2019; Brean et al., 2021; Dall'Osto et al., 2022) studies on 58 the influences of marine aerosols on climate and ecology. Overall, aerosol particle number concentrations 59 follow a clear annual trend, being much higher in austral summer than in other seasons (Järvinen et al., 60 2013; Kerminen et al., 2018; Weller et al., 2011). For instance, Kim et al. (2017) found that summertime 61 concentrations in the Antarctic Peninsula were ~20 times higher than in winter. This pattern can be largely 62 explained by new particle formation (NPF) events. 63

64 Precursor gases for NPF in this region can originate from the ocean, sea-ice, meltwater ponds, terrestrial animal colonies, anthropogenic activity and continental ecosystem. Oceanic emissions of 65 66 dimethyl sulfide (DMS) represent the largest natural sulfur source in the Antarctic atmosphere (Simó, 2001), and its photooxidation is a key process contributing to NPF (Giordano et al., 2017; Jang et al., 67 2019 and 2022). For instance, in situ (Saiz-Lopez et al., 2007) and satellite (Schönhardt et al., 2008) 68 69 measurements have shown Antarctica to be an iodine emission hotspot, particularly from the sea-ice in 70 the Weddell Sea during spring (Atkinson et al., 2012). Indeed, Sipilä et al. (2016) measured iodic acid (HIO<sub>3</sub>) in Antarctica and found that the Antarctic oceanic regions may be strong sources of molecular 71 72 iodine, which is then converted to  $HIO_3$  in gas-phase reactions. Dall'Osto et al. (2017) reported that 73 microbiota in sea-ice were associated with atmospheric organic nitrogen formation in the Southern Ocean 74 near Antarctica. According to Kyrö et al. (2013), the precursor vapors responsible for NPF and subsequent growth could originate from the cyanobacteria, which are abundant in Antarctic meltwater ponds. In 75 76 addition, continental Antarctica is a habitat for various types of seabirds and penguins, with guano species 77 acting as a crucial source of ammonia and organic compounds and may contribute to NPF in coastal Antarctic areas (Schmale et al., 2013; Weber et al., 1998; Zhu et al., 2011). At continental South Pole NPF 78 event are commonly associated with the local anthropogenic pollution during calm weather conditions 79 80 (Park et al., 2004). In addition, the biomass burning aerosol from South American continental outflow has been observed at Troll Research Station (Fiebig et al., 2009). During the daytime, higher radiation 81

enhances photo-active emissions from land ecosystems (mosses, grasses, and lichens) of the Antarctic
Peninsula and can lead to NPF and aerosol growth (Decesari et al., 2020; Quéléver et al., 2022; Schmale
et al., 2013). However, land sources are rather unlikely due to a small footprint of emerging land and the
associated short overpass over the sparse vegetation.

In recent years, long-term records of aerosol size distribution have become an important aspect of 86 investigations into the sources and dynamical processes of NPF. The majority of Antarctic field studies 87 have focused on the annual and spatial patterns of the number size distribution of particles > 10 nm (Belosi 88 et al., 2012; Järvinen et al., 2013; Kim et al., 2019; Kyrö et al., 2013; Lachlan-Cope et al., 2020). Although 89 90 NPF events are typically characterized by a rapid increase in the number concentration of cluster from 1– 3 nm (Kulmala et al., 2004), datasets for these types of aerosol size distribution remain rare. To date, 91 92 number size distribution of particles > 3 nm has been reported by Asmi et al. (2010) at Aboa during from December 29, 2006 to January 29, 2007; by Pant at al. (2011) at Maitri from January 1 to February 28, 93 2015; by Weller et al. (2015) at Neumayer from January 20 to March 26, 2012; by Jokinen et al. (2018) 94 95 at Aboa from November 2014 to February 2015; by Weller et al. (2018) at Kohnen during January 2015 96 and 2016; by Quéléver et al. (2022) at Marambio during the austral summer between January 15 and February 25, 2018; and by Brean et al. (2021) during the PI-ICE cruise from January 25 to February 4, 97 2019. However, all of these measurements were made during the Antarctic summer due to restricted 98 99 access and, therefore, limited information on seasonal cycles.

100 Newly formed particles can grow into larger sizes that act as CCN, becoming relevant for cloud formation (O'Dowd, 2002; Williamson et al., 2019). In a highly pristine atmosphere such as Antarctica, 101 102 where CCN concentration is extremely low (Kim et al., 2017), NPF may be a significant phenomenon 103 controlling the CCN budget (Kyrö et al., 2013). For instance, Herenz et al. (2019) showed that an elevated 104  $CN_{2.5}$  (total number concentration of particles > 2.5 nm) during NPF events was accompanied by an 105 increase in CCN concentrations at Princess Elisabeth during austral summer (December to February, 106 2013–2016). Ship-based observations during the ACE-SPACE found that the fraction of particle serving as CCN was higher near the coast of Antarctica compared to open ocean, resulting from multiple 107

108 processing cycles of dissipating and condensing clouds and/or the higher availability condensable gases 109 originating from marine microbial activity (Schmale et al., 2019). In addition, seasonal variability in 110 CN<sub>2.5-10</sub> (number concentration of particles within the 2.5 nm and 10 nm range and attributed to NPF) 111 and CCN concentrations at King Sejong Station from March 2009 to December 2016 were investigated 112 by Kim et al. (2019), who concluded that CCN concentrations during NPF events increased by ~11% 113 compared to the background concentration. However, to date, only one study (Kim et al., 2019) has 114 reported the contribution of NPF to CCN in the Antarctic Peninsula, and that study did not consider 115 aerosol number size distribution.

116 In this study, we continuously recorded the number size distribution of 2.5–300 nm particles and 117 CCN number concentrations at King Sejong Station in the Antarctic Peninsula from January 1, 2018, to 118 December 31, 2018. Our primary goals were to (1) characterize the seasonal variation and occurrence of 119 NPF events from the perspective of aerosol physical properties (total number concentration, number size 120 distribution, formation and growth rates, and condensation sink); (2) improve our understanding of the 121 major sources (including open ocean, sea-ice, and land) and processes influencing NPF and particle 122 growth; and (3) estimate the contribution of atmospheric NPF to CCN activity in this pristine environment. To our knowledge, this is the first study to present direct evidence of CCN production associated with 123 124 NPF and growth events in the Antarctic Peninsula, using simultaneous measurements of particle number 125 size distributions (down to 3 nm) and CCN properties for a full year.

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#### 127 **2. Experimental methods**

## 128 **2.1. Sampling site and instrumentation**

Continuous measurements of the physical properties of aerosol particles were conducted from January 2018 to December 2018 at King Sejong Station in the Antarctic Peninsula (62.22° S, 58.78° W). Full details of the sampling site and measurement setup are given in Kim et al. (2017). In brief, a cylindrical stainless inlet (0.1 m diameter and 5.2 m length; total flow rate of the sampled air was 150 L min<sup>-1</sup>) was placed on the observatory roof following Global Atmosphere Watch aerosol measurement

guidelines and recommendations. Two condensation particle counters (TSI model 3776 CPC and TSI 134 135 model 3772 CPC) were used to measure the total number concentration of particles larger than 2.5 136 (corresponding data  $CN_{2.5}$ ) and 10 nm (corresponding data  $CN_{10}$ ) every 1 s, respectively. The aerosol sample flow rates of TSI model 3776 CPC and TSI model 3772 CPC were 1.5 and 1.0 L min<sup>-1</sup>, respectively. 137 A nano-scanning mobility particle sizer (nano-SMPS) consisting of a nano-differential mobility analyzer 138 139 (nano-DMA) (TSI model 3085, USA) and an ultrafine condensation particle counter (TSI model 3776, 140 USA) was used to measure the number size distribution of particles from 2.5-64 nm every 3 minutes. The aerosol flow rate was 1.5 L min<sup>-1</sup> and the sheath flow rate was 15 L min<sup>-1</sup> inside the nano-DMA. 141

The particle number size distribution (from 10-300 nm every 3 min) was measured with a standard-142 SMPS consisting of a long DMA (TSI model 3081, USA) and a CPC (TSI model 3772, USA). The aerosol 143 flow rate was 1.0 L min<sup>-1</sup>, and the sheath flow rate was 10 L min<sup>-1</sup> inside the long DMA. To obtain the 144 number size distribution of particles from 2.5–300 nm, the nano-SMPS and standard-SMPS were merged. 145 146 For particle diameters 2.5–20 nm, nano-SMPS data were chosen because this was optimized to operate 147 with a smaller particle diameter. In the nano-DMA, the aerosol residence time can be reduced by shortening the inlet transport passage (5.0 cm) and increasing the inlet flow (up to 16.5 L min<sup>-1</sup>) (< 10 nm) 148 149 (Chen et al., 1998). Hence, the number size distribution data from both nano-SMPS and standard-SMPS 150 were merged at a diameter of 20 nm. Furthermore, three-point median filter and five point moving average 151 were performed on merging the number size distribution data to remove nano-SMPS noise, as suggested 152 by Kulmala et al. (2012).

The black carbon (BC) concentration was measured using an aethalometer (AE22, Magee Scientific Co., USA) every 5 min to examine long-range polluted aerosol transport from other continents and to assess the influence of local pollution from the station. The flow rate through a sharp-cut 2.5 μm cyclone (BGI, Inc., USA) was set to 5 L min<sup>-1</sup>. The CCN counter (CCNC: CCN-100, Droplet Measurement Technologies, USA) measured CCN number concentrations at five different supersaturation levels of 0.2% 0.4%, 0.6%, 0.8%, and 1% every 30 minute. The total flow rate in the CCN counter was 0.5 L min<sup>-1</sup>. The sample and sheath flow rates of the CCN counter were 0.05 and 0.45 L min<sup>-1</sup>, respectively. In addition, basic meteorological parameters (temperature, pressure, relative humidity (RH), wind speed, wind
direction, and solar radiation intensity) were measured using an automatic weather station (Vaisala
HMP45).

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#### 164 **2.2. Data evaluation**

165 As the observatory is located ~400 m southwest of the main station buildings and several kilometers 166 away from other research stations, measurement data were impacted by local emissions from station 167 activities (e.g., power generators and incineration) or anthropogenic pollutions near the observatory (e.g., 168 plumes from other research station about several kilometers, vessels providing research station supply, 169 and commercial cruise vessels). To obtain an unperturbed aerosol population of pristine Antarctic 170 environment, contaminated measurements were removed manually based on wind direction, wind speed, BC concentration, and total particle number concentration. The following data elimination procedure was 171 applied: (1) the measurements taken within wind sector of  $355^{\circ}$  and  $55^{\circ}$  were discarded as directly 172 impacted by local pollution sources; (2) relative wind speed below 2.0 m s<sup>-1</sup>, as stagnant conditions would 173 have facilitated contaminated particle propagation to the measurement location; (3) equivalent BC mass 174 concentrations exceeding 50 ng m<sup>-3</sup>, because elevated BC concentration unambiguously pointed at 175 176 polluted particles; and (4) a sharp increase in the total number concentration over the entire particle 177 diameter range in a short time scale of less than an hour, as such abrupt peaks and spikes are related to 178 potential contamination or instrumental malfunctions. For instance, CPC and SMPS data were removed for time periods when particle number concentrations suddenly increased to more than twice the 179 180 background values.

Based on a four-year (2016-2019) BC dataset, six types of Antarctic Peninsula air-pollution levels were identified (Grigas et al., 2017): (1) pristine air with BC concentrations  $< 15 \text{ ng m}^{-3}$ , (2) clean air with BC levels 15–50 ng m<sup>-3</sup>, (3) slightly polluted air with BC levels 50–100 ng m<sup>-3</sup>, (4) moderately polluted air with BC levels 100–300 ng m<sup>-3</sup>, (5) polluted air with BC levels 300–1000 ng m<sup>-3</sup>, and (6) extremely polluted air with BC concentrations > 1000 ng m<sup>-3</sup> (Figure 1). Previously, BC data were used as indicators

for local contamination in Antarctica when BC concentration level exceeded 50 ng m<sup>-3</sup> (Herenz et al., 186 2019) or 100 ng m<sup>-3</sup> (Jang et al., 2018; Kim et al., 2017; Kim et al., 2019; Weller et al., 2011; Weller et 187 188 al., 2015). Hara et al. (2019) measured BC concentration at Syowa station Antarctica from February 2005 until December 2016. They found that the daily median BC concentrations were below the detection limit 189 (0.2 ng m<sup>-3</sup>) to 63.8 ng m<sup>-3</sup> at Syowa Station (median, 1.8 ng m<sup>-3</sup>; mean, 2.7 ng m<sup>-3</sup> during the measurement 190 191 period). During the ACE-SPACE expedition, BC concentration reach its background levels of 19.2 ng m<sup>-</sup> <sup>3</sup> (Schmale et al., 2019). Arctic shipborne-observations measured BC concentration throughout the Arctic 192 193 Ocean and Pacific Ocean during the summer of 2017, all pointing to pristine clean marine air masses with BC values of approximately  $20 \pm 10$  ng m<sup>-3</sup>. (Park et al., 2020). 194

Of the total time period assessed, pristine air conditions represented 30% (mean value of BC:  $6 \pm 6$ ng m<sup>-3</sup>), clean for 44% (mean value of BC:  $30 \pm 10$  ng m<sup>-3</sup>), lightly polluted 19% (mean value of BC:  $69 \pm 14$  ng m<sup>-3</sup>), moderately polluted 6% (mean value of BC:  $150 \pm 47$  ng m<sup>-3</sup>), polluted 1% (mean value of BC:  $499 \pm 174$  ng m<sup>-3</sup>), and extremely polluted less than 1% (mean value of BC:  $1537 \pm 595$  ng m<sup>-3</sup>). Together, pristine and clean air conditions accounted for ~74% of the time with the remaining 26% (BC > 50 ng m<sup>-3</sup>) removed prior to data analysis.

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### 202 **2.3. Definition of NPF and growth events**

NPF events were visually identified by the particle number size distribution based on the protocol 203 204 described by Dal Maso et al. (2005) and Kulmala et al. (2012). Here, these were defined when a distinct new mode of particles (initially < 25 nm), appearing in the particle number size distribution at nucleation-205 206 mode size (3–25 nm), prevailed for more than an hour. Using these criteria, the particle size distribution 207 data showed that in some cases, there was only a short burst of nucleation-mode particles without clearly 208 discernible particle growth, whereas in other cases, particle formation with subsequent particle growth 209 lasted for several hours, representing a regional-scale phenomenon (Ström et al., 2009). This enabled us 210 to determine the particle growth rate (GR), which is not possible during short bursts of nucleation-mode particles. 211

The particle growth and formation rates along with the condensation sink were calculated from the 212 213 measured particle number size distribution. The GR was determined using the maximum concentration 214 and mode-fitting methods (Dal Maso et al., 2005; Yli-Juuti et al., 2009). GR was calculated by a linear fit 215 through the geometric mean diameter of the nucleation-mode particles as a function of time during NPF. 216 The formation rate (FR) of nucleation-mode particles  $(J_{3-25})$  was calculated by taking into account the 217 time evolution of the particle number concentration in this size range and particle losses due to the 218 coagulation sink and condensational growth out of the size range (Kulmala et al., 2012). The surface area 219 of particles available for the condensation of gaseous molecules can be characterized by a condensation 220 sink (CS), which determines how rapidly vapor molecules condense onto pre-existing particles (Collins 221 et al., 2017; Dal Maso et al., 2002).

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### 223 **2.4. Backward trajectory analysis and potential source regions**

224 Air mass back trajectories were obtained using the Hybrid Single-Particle Lagrangian Integrated 225 Trajectory (HYSPLIT) model to investigate their relationships with the physical characteristics of aerosol 226 particles (Draxler and Hess, 1998). The 2 days air mass back trajectories (48 hours) were determined at 227 hourly intervals and combined with satellite-derived geographical information to estimate the transport 228 history of the air masses arriving at the observation site (Jang et al., 2022 and Park et al., 2021). The 229 potential origins of the aerosols were divided into three categories based on the retention time of the 2 230 days back trajectories over three major domains: ocean (including the Weddell and Bellingshausen Seas), sea-ice, and land (including the Antarctic Peninsula). Daily geographical information on ocean, sea-ice, 231 232 and land area was obtained from the Sea Ice Index (25 km resolution) provided by the National Snow and 233 Ice Data Center (NSIDC). The sea-ice zone was defined as the area with a sea-ice coverage >15% (Stroeve 234 et al., 2016). Air masses that passed over the Weddell and Bellingshausen Sea-regions were categorized 235 as originating from the ocean (i.e. > 50% retention over the ocean region). The air masses that frequently 236 advected over the sea-ice region were categorized as originated over the sea-ice (i.e. > 50% retention over 237 the sea-ice domain). Air masses that traveled through the Antarctic Peninsula were categorized as

originating from the land (i.e. > 50% retention over the land). Finally, the air masses which passed over the ocean, sea-ice, and land regions simultaneously were categorized as originating from the multiple regions (i.e., 20–40 % retention over each ocean, sea-ice, and land domain).

To evaluate the influence of oceanic biological characteristics on NPF properties, the phytoplankton 241 242 biomass of the ocean domains was estimated by calculating their chlorophyll concentration from the 243 Moderate Resolution Imaging Spectroradiometer on the Aqua (MODIS-Aqua) satellite at 4 km resolution 244 during the entire study period. Phytoplankton produces dimethylsulfoniopropionate (DMSP, a precursor 245 of gaseous DMS) and other organic vapors all of which are potential precursors to new particle formation. 246 Thus, the spatiotemporal distribution of sea-surface DMSP could be an indicator of contemporary DMS 247 emissions. The total DMSP concentration on the sea-surface was estimated using the algorithm developed 248 by Galí et al. (2015). The algorithm for the total DMSP concentration was based on the satellite-derived 249 chlorophyll concentration and photosynthetic radiation exposure. To calculate the air mass exposures to 250 ocean chlorophyll and DMSP (Jang et al., 2019), hourly back trajectory position was combined with 251 satellite-derived chlorophyll concentration and total DMSP concentration, providing a good measure for 252 quantitatively investigating the biological exposure history of sampled air over the several days before its arrival at the observation site (Park et al., 2018 and 2021). However, satellites are not typically used to 253 254 directly measure the biomass of sea ice algae because satellite sensors cannot penetrate through thick 255 layers of sea ice to directly measure the biomass of algae beneath it (Lee et al., 2015; Lange et al., 2017). 256 Thus, calculated chlorophyll exposures (i.e., satellite-estimates of biological activity) cannot account for 257 the biological activities thriving within and beneath of the sea ice.

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## 259 **3. Results and discussion**

## 260 **3.1. General features and annual cycle**

We investigated the overall seasonality of particle number size distributions focusing on NPF events. In addition, local meteorological parameters (e.g., temperature, RH, wind speed, wind direction, pressure, and solar radiation) and air mass back trajectories were used to support the interpretation of the seasonal trends of the particle number size distribution and the dynamics of NPF events observed at the station.

265 **3.1.1. Particle number concentrations and size distributions** 

266 Figure 2 shows a time series of the one-hour average total particle number concentration and size-267 segregated particle number concentrations over the entire measurement period conforming to pristine (BC < 15 ng m<sup>-3</sup>) and clean (BC: 15–50 ng m<sup>-3</sup>) conditions. In addition, monthly medians for total number 268 269 concentration of particles, size-segregated particles number concentration, CCN number concentration at supersaturation of 0.4%, and meteorological parameters are included in Table 1. The CN<sub>2.5</sub> and CN<sub>10</sub> 270 ranged from 60 to 3982 cm<sup>-3</sup> and 30 to 3304 cm<sup>-3</sup>, respectively. The annual median number concentrations 271 of particles for the nucleation mode (N<sub>NUC</sub>; 2.5–25 nm in diameter), Aitken mode (N<sub>AIT</sub>; 25–100 nm in 272 diameter), and accumulation mode (NACC; 100-300 nm in diameter) were 46.8 cm<sup>-3</sup>, 53.5 cm<sup>-3</sup>, and 21.7 273 274 cm<sup>-3</sup>, respectively. The highest median N<sub>NUC</sub>, N<sub>AIT</sub>, and N<sub>ACC</sub> values were recorded in December (193.5 cm<sup>-3</sup>), December (227.6 cm<sup>-3</sup>), and January (83.8 cm<sup>-3</sup>), respectively (Table 1). The lowest N<sub>NUC</sub>, N<sub>AIT</sub>, 275 and N<sub>ACC</sub> values were recorded during austral winter in June – 12.2 cm<sup>-3</sup>, 12.5 cm<sup>-3</sup> and 9.2 cm<sup>-3</sup>, 276 277 respectively. Overall, clear annual and seasonal patterns of particle number concentrations in all size 278 classes were observed: high concentrations in summer (December-February) and low concentrations in 279 winter (June-August), similar to those observed at Marambio Station in the Antarctic Peninsula (Asmi et 280 al, 2018), at coastal Neumaver Station (Weller et al., 2011), at Concordia Station Dome C (Järvinen et al., 281 2013), and at Troll Station (Fiebig et al., 2014). Furthermore, the hourly average CN<sub>10</sub> value was positively correlated with the hourly average  $N_{NUC}$  (R = 0.88; not shown), implying that the summer 282 maximum of total particle number concentrations was largely influenced by newly formed particles in the 283 284 Antarctic atmosphere.

#### **3.1.2. Influence of meteorological parameters on NPF events**

The meteorological parameters after data filtering (BC < 50 ng m<sup>-3</sup> indicating pristine and clean conditions) were characterized by a solar radiation range of 0–919 W m<sup>-2</sup> (median 10.7 W m<sup>-2</sup>), a temperature range of -20–6 °C (median -1.2 °C), an RH range of 52–98 % (median 88 %), a pressure range of 950–1022 hPa (median 988 hPa), a wind speed range of 0.3–21 m s<sup>-1</sup> (median 7.4 m s<sup>-1</sup>), and

wind direction range of 3–357° (median 296°) (Figure S1). To understand impacts on the particle number 290 291 size distributions, we determined the relationships between the size-segregated particle number concentrations and meteorological parameters (Figure S2). CN10, NNUC, NAIT, and NACC were positively 292 293 correlated with both solar radiation intensity and temperature. In particular, N<sub>NUC</sub> had the highest 294 correlation with solar radiation intensity (R = 0.39) of any meteorological condition, suggesting that solar 295 radiation is one of the most important factors influencing NPF events, as it can drive photochemical 296 reactions leading to the production and further reaction of precursor gases. In contrast, there was a weak 297 anticorrelation between RH and N<sub>NUC</sub>, supporting the view that NPF occurs preferentially at low RH 298 (Dada et al., 2017; Hamed et al., 2011; Jeong et al., 2010; Laaksonen et al., 2008). Field observations 299 have reported that during NPF events, RH was negatively related to the number concentration of freshly 300 formed particles (Jeong et al., 2004; Lachlan-Cope et al., 2020; Weber et al., 1997) because of the 301 enhanced coagulation from scavenging effect of sub-3 nm nanoparticles at high RH and the diminished 302 solar radiation at high RH. Previously, some NPF events were associated with high wind speeds at various Antarctic stations, such as Neumayer (Weller et al., 2015) and Aboa (Asmi et al., 2010; Virkkula et al., 303 304 2007). These studies found an enhanced particle number concentration < 10 nm during stormy weather and suggested ion production by frictional processes in fast-moving snow and ice crystals, followed by 305 306 subsequent ion-mediated nucleation during strong winds. However, in our study, wind speed was not 307 correlated with N<sub>NUC</sub> (R = -0.18), N<sub>AIT</sub> (R = -0.04), or N<sub>ACC</sub> (R = -0.05), as recently suggested by Liu et 308 al. (2018). Our results indicated that wind speed did not affect NPF events. A possible explanation for the 309 wind speed independence is that an increase in wind speed contributes to the increase of cluster size ion 310 number concentrations by friction processes (Virkkula et al., 2007), but it was also accompanied by 311 cloudy conditions. In summary, the elevated N<sub>NUC</sub> values (i.e., indicator of NPF events) at King Sejong 312 Station were more likely to be accompanied by high solar radiation, high temperature, and low RH, 313 regardless of wind speed. Recent studies concluded that Antarctic NPF occurred under combined high 314 solar radiation, high temperature and low RH conditions, similar to previous study measured at the 315 Marambio Antarctic research station (Quéléver et al., 2022). Quéléver et al. (2022) found all NPF events

were observed during the daytime with high solar radiation (clear-sky conditions), mostly with above-

317 freezing temperature and with low RH.

## 318 **3.1.3. Characteristics of NPF events**

NPF events in this study were identified based on the size distribution data measured using the 319 standard-SMPS (Figure 2e) and nano-SMPS (Figure 2f). During the pristine and clean periods 320 321 (comprising of 355 observation days and 169166 size distribution spectra for the standard-SMPS, and of 322 349 observation days and 165259 size distribution spectra for nano-SMPS), NPF events were frequently observed at King Sejong Station, as shown by the size distribution data (Figure 2f). 97 events (26% of 323 observation days) with elevated N<sub>NUC</sub> were observed when taking place in pristine (BC < 15 ng m<sup>-3</sup>) and 324 clean (BC: 15–50 ng m<sup>-3</sup>) conditions. Median value of BC concentrations during NPF events was 21.0 ng 325 326 m<sup>-3</sup>, similar to that of whole measurement periods after data filtering (median BC value: 18.8 ng m<sup>-3</sup>) 327 (section 2.2). This indicated that NPF events are independent of occasional increases of BC during clean 328 periods. The NPF events were classified into: (1) burst event and (2) nucleation with growth event 329 according to the classification by Dal Maso et al. (2005) as seen in Figure S3. The burst events and 330 nucleation with growth events were observed on 1 January 2018 and 16 December 2018, respectively. NPF events were more frequently observed in summer (~55%) than in any other season (Figure 4), with 331 332 the highest frequency in January (22%) and December (22%) followed by spring (September–November, 333 34%) and autumn (March-May, 11%). Similar results were reported by Järvinen et al. (2013) based on 334 observations from Dome C and Kim et al. (2019) based on observations from King Sejong Station. 335 Although Järvinen et al. (2013) reported winter events that occurred in the absence of sunlight, we did 336 not detect NPF events during austral winter from May through to August.

In order to investigate the seasonal characteristics of NPF event, we compared mean size distributions of aerosol particles for summer, spring (transition period of the melting ocean), and autumn (transition period of refreezing of the ocean) (Figure S4). Trimodal distributions were presented in all seasons excepting winter when nucleation mode or particle formation was not observed. For instance, a trimodal distribution was seen at 7 nm, 30 nm, 122 nm during summer months. The number concentration

of nucleation and Aitken modes were higher than the accumulation modes, indicating that NPF event 342 343 regulates the aerosol processes in Antarctic peninsula. The largest mode at 126 nm or 103 nm may be due 344 to a combination of primary (produced by bubble-bursting process) and secondary (produced by gas-to-345 particle conversion process) aerosol components. Results are broadly in line with previous results published from the Arctic and Antarctic regions. A ship-borne field campaign over Arctic Ocean found a 346 347 trimodal distribution at 18 nm, 53 nm and 150 nm for open-ocean marine Arctic NPF event (Park et al., 348 2020). Lachlan-Cop et al. (2020) presented k-mean cluster analysis of particle size distribution measured 349 at Halley, Antarctica, showing a nucleation peak at 15 nm for "nucleation" ultrafine category and a 350 nucleation peak at 27 nm for "bursting" ultrafine category.

351 Air mass back trajectories were calculated at hourly intervals to investigate possible source regions 352 for the observed NPF events. Figure 2g shows the residence times of air masses over the three domains. 353 Based on 2-days air mass transport history analysis, air masses allocated to ocean, sea-ice and land 354 account for 83, 12 and 5%, respectively, during the study period. 97 cases were identified as NPF events, 355 80 of which were observed when the air mass originated over the ocean domain (Figure 4). 12 NPF events 356 were observed in air masses originating over the sea-ice domain, while the remaining 5 events were 357 associated with multi-regional origin (3 cases) and land origin (2 cases). Multi-regional origin indicated 358 air masses simultaneously influenced by all three domains. Median BC concentration for ocean, sea-ice, and multiple air masses found to be 23.8 ng m<sup>-3</sup>, 12.7 ng m<sup>-3</sup>, 9.8 ng m<sup>-3</sup>, respectively, (Figure S5), 359 360 indicating pristine clean air masses with minimum influence from anthropogenic pollutions during each NPF event case. Our results indicated that NPF events were more common in air masses originating over 361 362 the ocean and sea-ice compared to those originating from the land. Precursors released by both ocean and 363 sea-ice could play an important role in the formation of new particles in the Antarctic atmosphere.

### 364 **3.1.4. Spatial extension of regional nucleation event**

Many previous studies have reported that key steps of the nucleation process (e.g., cluster stabilization) occur in the size range ~2 nm, in line with recent direct observations of atmospheric molecular clusters (Kerminen et al., 2018; Kulmala et al., 2013). However, during NPF events, we did

not observe particle formation starting directly from the lower end of the particle size spectrum (2.5 nm), 368 369 showing that the formation of freshly nucleated particles could not have actually taken place at the site. 370 Indeed, the initial diameter of particles that arrived to the measurement site during the NPF ranged from 371 4 nm to 16 nm (Figure 3a). Median values of NPF event duration (Figure 3b) and growth rate (Figure 3c) were 4.0 hour and 0.83 nm hr<sup>-1</sup>, respectively. We assumed that they were transported from elsewhere or 372 373 produced aloft, and detected the appearance of an already grown mode. Consistent with these studies, 374 NPF events can be a regional-scale phenomenon extending over spatial scales of tens to hundreds of kilometers in several regions, such as the remote marine boundary layer (Zheng et al., 2021), Canadian 375 376 high Arctic (Eureka, Nunavut, on Ellesmere Island in the Canadian Arctic Archipelago) (Tremblay et al., 377 2019), and Arctic ship-based observations.

378 Assuming the region is characterized by homogenous meteorological conditions, we estimated the 379 spatial scale of NPF by multiplying the time during which a distinct nucleation mode can be observed at 380 the sampling site by the locally measured wind speed (Birmili et al., 2003; Crippa and Pryor, 2013). As 381 shown in Figure 3d, the spatial extend of NPF event associated with substantial particle growth can be 382 16-816 km (median value: 155 km), indicating the large-scale NPF events. Weller et al. (2015) measured size distributions at the coastal Antarctic station Neumayer during two summer campaigns (from 20 383 384 January to 26 March 2012 and 1 February to 30 April 2014). They found that the spatial extend of NPF 385 event was estimated to be around  $170 \pm 85$  km, taking into account the prevailing wind velocity (around  $8 \pm 4 \text{ m s}^{-1}$ ) and the confined NPF duration (around 6h). 386

387

## 388 **3.2. Case studies**

389 This section presents a detailed overview of the ocean, sea ice, and multi-regional NPF events.

390 **3.2.1. Marine NPF event** 

A striking series of NPF events took place over seven days (Figure 6), starting at approximately 00:00 on December 9, 2018. Events starting at midnight can likely indicate their formation few hours earlier during afternoon sunlight, because the events are observed with an already grown nucleation mode. 394 Time series of meteorological parameters, air mass origins, oceanic biological activity (estimated by 395 chlorophyll and DMSP exposures), particle size distribution (measured by nano-SMPS and standard-396 SMPS), and CCN concentrations are shown in Figure 6. During this time, the prevailing northerly winds (median 307 °) were stable at 7.7 m s<sup>-1</sup>. Air temperature varied from -1.5 to 2.1 °C (median 0.5 °C) and 397 398 RH varied from 75–97% (median 89%). There were no data for solar radiation during these events. Air 399 masses predominantly traveled over the Antarctic Ocean (46.9, 0.7, and 0.4 h over ocean, land, and sea 400 ice, respectively) and could be categorized as originating from the Antarctic Ocean. Specifically, the air 401 mass originated mainly from Bellingshausen Sea (Figure 5a). During this event, the median total DMSP and chlorophyll exposures in the surface sea were 18 nmol L<sup>-1</sup> and 0.26 mg m<sup>-3</sup>, respectively. 402

Between 00:00 and 20:00 on December 9,  $N_{NUC}$  increased from 196 to 688 cm<sup>-3</sup>. At the same time, CCN concentrations at 0.4 % supersaturation gradually increased from 138 (00:00 on December 0) to 326 cm<sup>-3</sup> (12:00 on December 11), an increase of 135%. In addition, elevated  $N_{NUC}$  occurred at 00:00 on December 13, ranging from 118–522 cm<sup>-3</sup>. CCN number concentration at 0.4% supersaturation began to increase at this time (95 cm<sup>-3</sup>) and reached its maximum at 18:00 (503 cm<sup>-3</sup>), with a concentration increase of 431%.

409

## 410 **3.2.2. Sea-ice NPF event**

411 The NPF event with subsequent particle growth were detected from around 19:00 on January 13, 412 2018, to around 08:00 on January 14, 2018 (Figure 7). Air temperature and RH during the event were 0.1 °C and 85%, respectively, while solar radiation decreased from 131.7 to 0.2 W m<sup>-2</sup>. Winds were mild 413 and stable (1.9-5.7 m s<sup>-1</sup>), with a prevailing northwesterly (262-350°) direction and air masses 414 415 predominantly coming from sea-ice. The average retention times of the 2 d back trajectories traveling 416 over ocean, sea-ice, and land were 20.0, 20.9, and 7.1 h, respectively, indicating sea-ice-influenced air 417 masses (Figure 5b). During the NPF event, both total DMSP and chlorophyll exposure values are stable, with median exposures of 13.3 nmol  $L^{-1}$  and 0.2 mg m<sup>-3</sup>, respectively. 418

419 During the event,  $CN_{2.5}$  and  $CN_{10}$  increased to 5669 and 5097 cm<sup>-3</sup>, respectively. Furthermore, the

median N<sub>NUC</sub>, N<sub>AIT</sub>, and N<sub>ACC</sub> values were 508, 376, and 66 cm<sup>-3</sup>, respectively. Elevated CCN concentrations at 0.2 and 0.4 % supersaturations were not observed, whereas CCN concentrations at 0.6, 0.8, and 1.0 % supersaturations slightly increased during the event. For instance, CCN concentration at 0.8 % supersaturation was 517 cm<sup>-3</sup> at 20:00 on January 13, then increased to 688 cm<sup>-3</sup>, until 23:00 on January 13. The CCN concentration at 0.6, 0.8, and 1.0% supersaturations increased by 11%, 33%, and 58%, respectively.

426

## 427 **3.2.3. Multiple NPF event**

An intensive NPF event occurred from November 16 to November 17, 2018 (Figure 8). Air 428 temperature during the event ranged from -2.5 to -0.1 °C (median -1.3 °C). RH ranged from 70–95% 429 430 (median 79%), slightly lower than that for the marine and sea-ice NPF events described above. During the NPF event observed from 20:00 on November 16 to 02:00 on November 17, solar radiation decreased 431 from 30 to 0 W m<sup>-2</sup>. This suggests that the actual formation and growth occurred during daylight hours 432 433 upwind from measurement location, but very slow growth continued over the Antarctic Peninsula allowing the detection of observed grown mode at ~ 7 nm after the sunset. Wind speed ranged from 4.3– 434 9.5 m s<sup>-1</sup> with a constant direction from the southwest (median 239 °). Air mass back trajectories showed 435 436 multiple origins before reaching the station, passing over ocean (25.7 h, 53% of residence time), sea-ice (12.4 h, 26% of residence time), and land (10.0 h, 21% of residence time) (Figure 5c). During the event, 437 the median total DMSP and chlorophyll exposures in the sea surface were 6.0 nmol L<sup>-1</sup> and 0.2 mg m<sup>-3</sup>, 438 439 respectively.

440 At the start of the event (17:00 on November 16), N<sub>NUC</sub>, N<sub>AIT</sub>, and N<sub>ACC</sub> were 687, 83, and 13 cm<sup>-3</sup>, 441 respectively. The particle number concentration of the nucleation mode sharply increased to 1610 cm<sup>-3</sup> at 442 the NPF time, and its peak concentration occurred 7 h after the start of the event (00:00 on November 17), 443 indicating spatial extent of the formation region. The peak concentration of Aitken mode particles 444 successively appeared 14 h after the start of the event (07:00 on November 17) and 22 h respectively for 445 accumulation mode particles (15:00 on November 17). The values in the Aitken and accumulation mode ranges were 448 and 92 cm<sup>-3</sup>, respectively. We also observed a gradual increase in CCN concentration for
23 h. CCN concentration at 0.4 % supersaturation increased from 78 (17:00 on November 16) to 272 cm<sup>-3</sup>
(15:00 on November 17). This NPF event may have been a source of CCN, which enhanced CCN
concentrations by 248%.

450

### 451 **3.3. Influence of air mass origin on the NPF event**

## 452 **3.3.1. Parameters related to NPF**

453 Our results show that NPF and its growth events had largely different features depending on air 454 mass origin (Figure S6). Although only 3 cases of multi-regional NPF events occurred during the pristine 455 and clean periods (not included in Figure 9), the most intense NPF event was observed with multi-regional source region. Here, we compared N<sub>NUC</sub>, FR GR, and CS, for the ocean and sea-ice air masses (Figure 9 456 457 a-d). The FR, GR, and CS values agreed well with those reported in previous studies at other Antarctic sites (Järvinen et al., 2013; Kim et al., 2019; Kyrö et al., 2013; Weller et al., 2015), but significantly lower 458 459 than the values reported by the Quéléver et al. (2022), who showed the average FR and GR were 0.686 cm<sup>-3</sup> s<sup>-1</sup> and 4.2 nm h<sup>-1</sup>, respectively. The median N<sub>NUC</sub> and FR values for the ocean air mass (N<sub>NUC</sub>: 220 460 cm<sup>-3</sup> and FR:  $1.2 \times 10^{-2}$  cm<sup>-3</sup> s<sup>-1</sup>) were 1.6 and 3.0 times lower than those of sea-ice air mass (N<sub>NUC</sub>: 343 461 cm<sup>-3</sup> and FR: 3.6  $\times$  10<sup>-2</sup> cm<sup>-3</sup> s<sup>-1</sup>), respectively. This implies that marine NPF events are frequent, but 462 463 weak in terms of N<sub>NUC</sub> and FR values. Unlike N<sub>NUC</sub> and FR, there were no marked differences between 464 the median values of GR and CS by air mass category. The median particle GR values for ocean, sea-ice, and multiple air masses were 0.8, 0.7, and 0.9 nm h<sup>-1</sup>, respectively. In comparison, Jokinen et al. (2018) 465 reported that GR values ranged from 0.3 to 1.3 nm h<sup>-1</sup> at Aboa, and Brean et al. (2021) showed GR of 0.4 466 to 0.6 nm h<sup>-1</sup> measured during the PI-ICE cruise. To examine the effects of oceanic biological activity on 467 468 NPF properties, we examined solar radiation intensity, chlorophyll exposure and DMSP exposure for the 469 two air mass (or source region) categories (Figure 9 e-g). There was no difference in the median value in 470 solar radiation, while the median values for chlorophyll exposure and DMSP exposure were higher in air

471 masses originating from the ocean than in air masses originating from the sea ice. The median chlorophyll 472 exposure in ocean-influenced air masses ( $0.2 \text{ mg m}^{-3}$ ) was roughly twice that of the sea-ice-influenced air 473 mass ( $0.1 \text{ mg m}^{-3}$ ). Total DMSP exposure for the ocean-influenced air mass was ~2.7 times that of the 474 sea-ice air mass.

475

## 476 **3.3.2. Potential sources facilitating new particle formation**

477 The chlorophyll exposure and DMSP exposure during marine NPF events were higher than those during sea ice NPF events, suggesting a large chance to carry biologically-derived organic compounds 478 479 from the open ocean areas to the observation site. DMSP, a metabolite of oceanic phytoplankton, is partly 480 converted into gaseous DMS through enzymatic cleavage (Simó, 2001), which is the largest natural sulfur 481 source in the atmosphere (Barnes et al., 2006). Hence, the photooxidation products of biogenic DMS in the Antarctic atmosphere (e.g., Sulfuric acid and Methane sulfonic acid) could be a major contributor to 482 483 NPF and its growth when the air mass originates from the ocean. Jang at al. (2019) reported that NPF 484 events were more frequent in air masses originating from the Bellingshausen Sea than the Weddell Sea 485 during the biologically productive austral summer, and it is likely that the taxonomic composition of phytoplankton can be related to the formation of new particles in the Antarctic Ocean. Biogenic DMS 486 487 was found to be a precursor of NPF in coastal Antarctica (Yu and Luo, 2010).

488 The air mass exposure to chlorophyll and DMSP for sea-ice NPF events were 1.8 and 2.7 times 489 lower than those of marine NPF events. This could be explained by volatile iodine compounds released from ice-covered areas in Antarctica (Jokinen et al; 2018; Saiz-Lopez et al., 2007; Sipilä et al., 2016); 490 491 however, iodine compounds were not measured during our study period. Previously, iodine compounds 492 were found in large concentrations in and above the sea-ice of the Weddell Sea in Antarctica during spring 493 and summer (Atkinson et al., 2012). Roscoe et al. (2015) also confirmed that iodine compounds may 494 contribute to the secondary production of a significant number of particles measured at Halley and 495 Neumayer on the Antarctic coast.

496

In our study, sea-ice NPF events occurred frequently in January (middle of austral summer) and

497 September (early austral spring) (Figure 4). We compared the JR, GR, and CS values for the sea-ice NPF 498 cases observed between January and September (Figure S7) because of their notable differences in ice 499 coverage. In Antarctica, the minimum ice coverage is observed in February and the maximum in 500 September (Parkinson and Cavalieri, 2012). Our results showed that JR, GR, and CS values were much 501 higher in January than in September, indicating different NPF processes. The January events occurred 502 under low ice-coverage conditions, similar to previous studies from polar areas such as Svalbard 503 (Dall'Osto et al., 2017) and Greenland (Dall'Osto et al., 2018). Both studies showed that NPF events are 504 related to biogenic precursors released by open water and melting sea-ice regions, particularly during the 505 summer. In contrast, the September events occurred under high ice-coverage conditions. The monthly 506 median values of solar radiation showed that solar radiation intensity was very low from May to August 507 and then started to increase from September (Table 1). During the September events, median solar radiation intensity was found to be 63 W m<sup>-2</sup>. It is therefore possible that elevated sea-ice concentrations 508 509 under sufficient solar radiation around Antarctica lead to an increase in the concentration of halogen 510 species, resulting in the production of newly formed particles.

The most intensive NPF event was observed in multiple air masses, although the oceanic biological 511 activity was lower than that in the oceanic air mass. This indicated that terrestrial sources from continental 512 513 Antarctica, in addition to both DMS (mainly from the ocean) and iodine (mainly from sea-ice), may have 514 contributed to NPF. First, previous studies have reported that precursors emitted from seabird colonies 515 are linked to NPF (Schmale et al., 2013; Weber et al., 1998). The Chottaebawi area in the southwestern part of King George Island (around 2 km away from our observation site) is an important penguin colony 516 517 in the Antarctic region (Lee et al., 2009), while the cape area near King Sejong Station is abundantly 518 populated by flying seabirds such as skua. Given the proximity and abundance of seabird colonies at King 519 Seiong Station, seabird colony emissions are the likely sources of precursor gases to NPF (e.g., ammonia 520 and amine) (Quéléver et al., 2022). In fact, 2 NPF events (4 February 2018 for marine air mass origin and 521 18 February 2018 for multiple air mass origin) were observed when winds were seen to originate from the south sector where strong emission from the penguin colonies (southeast sector of 106–140°). Figure 522

523 S8 showed the contour plots of the size distributions and wind roses during those days. Although we did 524 not directly measure the precursor gases such as ammonia and amine that can trigger the NPF, we can 525 speculate that the fauna on the land or at the shore such as penguin and seabird colonies could not be 526 excluded as the potential source of NPF events locally although highly productive and ice melting 527 Weddell sea is coinciding with southeast direction too. Previous studies reported that precursor gases for 528 NPF (e.g., ammonia) can originate from the decomposition of excreta from seabirds and penguins 529 (Lachlan-Cope et al., 2020; Legrand et al., 1998; Liu et al., 2018; Schmale et al., 2013). More recently, 530 Quéléver et al. (2022) proposed that nitrogen-containing species could be land-sourced (e.g., from a high 531 penguin population during the summertime) or marine-sourced (e.g., from the biological activity of 532 plankton in the ocean and melting sea ice). The ammonia from seabird-colony guano is a key factor 533 contributing to bursts of newly formed particles, which are observed in the summertime Arctic (Croft et 534 al., 2016). Second, biogenic emissions from vegetation in the Antarctic Peninsula, mainly composed of 535 relatively small and sparse patches of lichens and mosses (Miranda et al., 2020), could be associated with 536 NPF and growth. Kim et al. (2006) studied plant communities on the Barton Peninsula around King 537 Sejong Station in the maritime Antarctic and concluded that ~47% of the investigated area was covered by vegetation although generally sparse. Finally, biogenic precursors from meltwater ponds in continental 538 539 Antarctica have also been suggested (Kyrö et al., 2013) as a possible source of aerosol production (Weller 540 et al., 2018). Overall, our data suggest that complex interconnected ecosystems across ocean, sea ice, and 541 land can lead to an enhancement in Antarctic NPF.

#### 542 **3.3.3. NPF as a source of CCN**

For a given SS of 0.4%, the median CCN number concentrations were rather similar 184, 144, and 178 cm<sup>-3</sup> for ocean, sea ice, and multiple air masses, respectively (Figure 9h). The values are in line with previous studies published from the Antarctic regions. Humphries et al. (2023) reported CCN concentrations nearby East Antarctic observations from Macquarie Island and Kennaook / Cape Grim as well as recent ship voyages of the RSV Aurora *Australis* and the RV *Investigator* in the region. The median CCN value at a SS of 0.5% was in the ranges of 88–145 cm<sup>-3</sup> at Macquarie Island, 57–158 cm<sup>-3</sup> at

Kennaook / Cape Grim, and 40–230 cm<sup>-3</sup> during the voyages (No voyage data exist for the winter months), 549 550 respectively. The PCAN project exhibited that a median particle number concentration larger than 3 nm of 354 cm<sup>-3</sup> was observed from the voyage and median CCN at 0.55 % supersaturation were 167 cm<sup>-3</sup>, 551 552 implying approximately half the particles measured as CN<sub>3</sub> could be activated as CCN (Simmons et al., 2021). Recently, several ship-based measurements over the Southern Ocean found significantly increased 553 554 MSA concentrations in air masses originating close to the Antarctic coastline, alongside enhancements in 555 CCN concentration (Humphries et al., 2021). Of the 83 NPF events, CCN concentrations increased by 2-556 268% (median 44%) following 1 to 36 hours (median 8 hours) after NPF events. The median increase in 557 CCN concentrations was 44 %, 34 %, and 107 % for ocean, sea ice, and multiple air masses, respectively. 558 NPF can be an important source of CCN in Antarctica, and the highest CCN enhancement was observed 559 when air masses passed through multiple regions, followed by ocean and sea-ice regions. Our results 560 provide the first direct evidence of CCN production resulting from an NPF event in the Antarctic 561 atmosphere, based on simultaneous measurements of particle number size distribution (e.g., diameter 562 ranges of 2.5–300 nm) and CCN number concentrations in real time throughout the year.

563 We also compared CCN activity and critical diameter for the three selected periods (Figure 9i and j). The median values of CCN activity, i.e. the ratio of the number concentration of particles that activated 564 565 to become CCN at a given supersaturation to the total number concentration or particles larger than 10 nm (CN<sub>10</sub>), were similar (about 5%) in three different air masses. The critical diameter ( $D_c$ ), the diameter 566 567 at which the integration of aerosol size distribution from the largest particle diameter to the lowest one 568 matches the measured CCN concentration, was determined using the measured aerosol size distribution,  $CN_{10}$ , and CCN concentrations (Furutani et al., 2018). The median  $D_c$  value at 0.4% supersaturation was 569 570 estimated to be 41 nm, 32 nm, and 37 nm for ocean, sea ice, and multiple air masses, respectively. These 571 results agreed well with those reported in previous studies that determined  $D_c$  at the Finnish Antarctic 572 Research Station, Aboa (Kyrö et al., 2013), a clean subarctic background site (Komppula et al., 2005), 573 and over remote Southern Ocean around Antarctica (Fossum et al., 2018; Fossum et al., 2020). For instance, Kyrö et al. (2013) found the smallest  $D_c$  at 48 nm. The median  $D_c$ , as suggested by Komppula 574

et al. (2005), varied from 50–128 nm (average 80 nm). The  $D_c$  value for maritime polar and marine modified continental Antarctic air masses were 71 and 59 nm, respectively (Fossum et al., 2020).

577 A time series of daily mean CCN concentrations at five different supersaturation ratio of 0.2, 0.4, 0.6, 0.8, and 1 was illustrated in Figure S10. To understand the contribution of growing particles on the 578 579 CCN concentrations during NPF event, we determined the increase in CCN concentration during growth 580 periods (i.e., growth to smaller than 40 nm particles and growth to larger than 40 nm particles) compared 581 to baseline values under different supersaturation conditions (Figure 10), according to the method 582 suggested by Chang et al. (2022). When particle growth was smaller than to 40 nm (growth  $\leq$  40 nm), the mean CCN concentrations increased by 59–178 cm<sup>-3</sup> for a SS of 0.2 %–1.0 %, representing a 172–217 % 583 584 increase compared to the values during baseline conditions. When particle growth was larger than to 40 nm (growth > 40 nm), the mean CCN concentrations increased by 57–227 cm<sup>-3</sup> for a SS of 0.2 %–1.0 %, 585 representing a 170–249 % increase compared to baseline values. Our results indicate that particles formed 586 587 from NPF events can lead to the significantly enhanced CCN concentration in Antarctic Peninsula, and 588 this effect is more pronounced if we consider particle growth larger than 40 nm, consistent with ship-589 based observations (Chang et al., 2022) and aircraft-based observation (Willis et al., 2016) in the Canadian 590 Arctic during summer.

591

#### 592 **4. Conclusions**

593 We measured the number size distribution of 2.5–300 nm particles and CCN number concentrations at King Sejong Station in the Antarctic Peninsula continuously from January 1 to December 31, 2018. 594 The annual median values of N<sub>NUC</sub>, N<sub>AIT</sub>, and N<sub>ACC</sub> were 46.8 cm<sup>-3</sup>, 53.5 cm<sup>-3</sup>, and 21.7 cm<sup>-3</sup>, respectively. 595 596 Overall, clear annual and seasonal patterns of particle number concentrations in all size classes were 597 observed (high concentrations in summer and low concentrations in winter). Furthermore, the monthly 598 CN<sub>10</sub> value was positively correlated with the monthly N<sub>NUC</sub>, implying that summer maximum particle 599 concentrations could be largely influenced by newly formed particles in the Antarctic atmosphere. Among 600 meteorological parameters, the elevated N<sub>NUC</sub> values (i.e., indicators of NPF events) were more likely to

be accompanied by high solar radiation, high temperature, and low RH, regardless of wind speed.

602 NPF events were identified based on size distribution data measured using two SMPSs. During the 603 pristine and clean periods, 97 events (26% of observation days) with elevated N<sub>NUC</sub> were observed. NPF 604 events occurred more frequently in summer than in any other season. Based on air mass back-trajectory 605 analysis, we distinguished three different types of NPF events: marine (80 cases), sea ice (12 cases), and multiple (3 cases). Marine NPF events were frequent and weak (N<sub>NUC</sub>, 220 cm<sup>-3</sup>; FR =  $1.2 \times 10^{-2}$  cm<sup>-3</sup> s<sup>-</sup> 606 <sup>1</sup>; GR = 0.8 nm hr<sup>-1</sup>) and occurred when the air mass exposure to oceanic phytoplankton was high 607 (chlorophyll, 0.2 mg m<sup>-3</sup>; DMSP, 18 nmol L<sup>-1</sup>). The photooxidation of biogenic DMS in the Antarctic 608 609 atmosphere could be a major contributor to marine NPF events. In contrast, sea-ice NPF events (N<sub>NUC</sub>, 343 cm<sup>-3</sup>; FR, 3.6  $\times$  10<sup>-2</sup> cm<sup>-3</sup> s<sup>-1</sup>; GR, 0.7 nm h<sup>-1</sup>) were observed when the air mass exposure to oceanic 610 phytoplankton was relatively low (chlorophyll, 0.1 mg m<sup>-3</sup>; DMSP, 7 nmol L<sup>-1</sup>), which may be due to 611 volatile iodine compounds released from ice-covered areas. Strong NPF events (N<sub>NUC</sub>, 516 cm<sup>-3</sup>; FR, 3.2 612  $\times$  10<sup>-2</sup> cm<sup>-3</sup> s<sup>-1</sup>; GR, 0.9 nm hr<sup>-1</sup>) were associated with multiple air masses, indicating complex 613 614 interconnected ecosystems leading to an enhancement in Antarctic NPF.

615 To investigate the connection between newly formed particles and CCN production, we compared 616 CCN properties for the three air mass categories. The median CCN number concentrations at a given SS of 0.4% were 184, 144, and 178 cm<sup>-3</sup> for ocean, sea ice, and multiple air masses, respectively. Of the 83 617 618 events, an increase in CCN concentrations after the NPF events was detected, ranging from 2-268 % 619 (median 44 %). The median increase in CCN concentrations was 44 %, 34 %, and 107 % for ocean, sea ice, and multiple air masses, respectively. NPF events led to increased CCN concentrations at King Sejong 620 Station. The median value of  $D_c$  at a supersaturation of 0.4% was estimated to be 41 nm, 32 nm, and 37 621 622 nm for ocean, sea ice, and multiple air masses, respectively. This study is the first to report CCN 623 production resulting from Antarctic NPF events in the Antarctic Peninsula. However, further detailed 624 measurements of the chemical properties of aerosol particles and precursor gases (e.g., ammonia) during 625 NPF events are required to better understand the contribution of these compounds to the formation and growth of aerosol particles and to explore their impacts on CCN formation in the remote Antarctic 626

627	environment.
628	
629	Data availability
630	The data analyzed in this publication will be readily provided upon request to the corresponding author
631	(yjyoon@kopri.re.kr).
632	
633	Author contributions
634	JP and YJY designed the study. JP, HK, YG, EJ, K-TP, SP, and YJY analyzed data. JP wrote the
635	manuscript. CHJ, DC, and CO'D all commented on and discussed the manuscript.
636	
637	Competing interests
638	The authors declare that they have no conflict of interest.
639	
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Table 1. Monthly median for total particle number concentration > 10 nm (CN<sub>10</sub>), particle number concentrations of the nucleation mode (N<sub>NUC</sub>), Aitken mode (N<sub>AIT</sub>), accumulation mode (N<sub>ACC</sub>), CCN number concentration at supersaturation of 0.4% (CCN<sub>0.4%</sub>), and meteorological parameters such as solar radiation, temperature, RH, pressure, wind speed, and wind direction for 2018, after data filtering (BC < 50 ng m<sup>-3</sup> indicating pristine and clean conditions), measured at King Sejong Station in the Antarctic Peninsula from January 1 to December 31, 2018.

	CN <sub>10</sub> (cm <sup>-3</sup> )	N <sub>NUC</sub> <sup>a</sup> (cm <sup>-3</sup> )	N <sub>AIT</sub> <sup>a</sup> (cm <sup>-3</sup> )	NACC <sup>a</sup> (cm <sup>-3</sup> )	CCN <sub>0.4%</sub> (cm <sup>-3</sup> )	Solar radiation (W m <sup>-2</sup> )	Temp. (°C)	RH (%)	Pressure (hPa)	Wind Speed (m s <sup>-1</sup> )	Wind direction (°)
January	506.2	101.1	188.7	83.8	235.2	129.2	1.1	88.6	986.0	5.78	315.8
February	594.3	111.3	200.0	69.9	229.8	103.5	1.8	90.8	987.2	7.72	319.9
March	357.3	86.0	112.4	42.1	138.7	58.0	1.1	88.4	981.8	8.21	342.3
April	184.1	49.9	39.1	17.5	58.6	26.2	-0.7	87.1	988.2	7.88	350.0
May	106.7	25.1	23.8	14.2	51.1	7.3	-2.3	81.8	990.1	7.34	277.7
June	75.9	12.2	12.5	9.2	35.4	3.4	-4.1	88.4	995.9	7.21	339.8
July	84.3	28.2	16.8	11.6	39.1	5.5	-2.9	86.5	992.2	9.08	300.8
August	109.8	39.3	19.6	14.8	52.1	21.8	-3.3	85.9	986.2	8.57	327.8
September	266.4	123.8	51.3	20.9	79.3	65.6	-3.6	86.5	992.6	9.52	313.2
October	287.0	88.9	62.0	26.9	105.3	122.1	-2.1	84.6	994.4	6.50	290.8
November	498.2	79.3	136.8	46.1	150.3	143.3	-0.6	89.3	980.0	7.59	307.9
December	511.9	193.5	227.6	67.7	189.1	136.5	0.4	87.2	980.4	6.72	302.7

 $^{a}N_{NUC}$ ,  $N_{AIT}$ , and  $N_{ACC}$  represent the particle number concentrations in the nucleation mode (2.5–25 nm), Aitken mode (25–100 nm), and accumulation mode (100–300 nm).



Figure 1. Frequency of occurrence of BC mass concentration for six types of Antarctic Peninsula air-pollution levels classified from four-year BC data.



Figure 2. Time series of 1 h averages for (a)  $CN_{2.5}$  and  $CN_{10}$ , (b) nucleation mode ( $N_{NUC}$ ; 2.5–25 nm), (c) Aitken mode ( $N_{AIT}$ ; 25–100 nm), and (d) accumulation mode ( $N_{ACC}$ ; 100–300 nm); contour plots of the size distributions measured using (e) standard and (f) nano-SMPS; and (g) residence time of air masses passing over ocean, sea ice, and land. CN 2.5 data are only available from January to March due to the instrumental malfunctions.



Figure 3. Frequency of (a) initial diameter of particles, (b) duration time, (c) growth rate, and (d) extension for the NPF event. Two NPF cases were excluded when the wind speed was higher than 10 ms<sup>-1</sup>.



Figure 4. Seasonal variations in the number of NPF days by air mass origin.



Figure 5. Air mass back trajectories for arrival at 50 m for the three case study NPF events: (a) marine, (b) sea ice, and (c) multiple.



Figure 6. Marine NPF event observed from December 9–15, 2018. (a–b) meteorological variables, (c) the residence time of air masses that passed over the ocean, sea ice and land areas, (d) total DMSP and chlorophyll exposures, (e–f) number size distribution with the standard-SMPS and nano-SMPS, and (g) CCN number concentration. The x-axis represents local time.



Figure 7. Sea ice NPF event observed from January 13–14, 2018. (a–b) meteorological variables, (c) the residence time of air masses that passed over the ocean, sea ice and land areas, (d) total DMSP and chlorophyll exposures, (e–f) number size distribution with the standard-SMPS and nano-SMPS, and (g) CCN number concentration. The x-axis represents local time.



Figure 8. Multiple NPF event observed from November 16–17, 2018. (a–b) meteorological variables, (c) the residence time of air masses that passed over the ocean, sea ice and land areas, (d) total DMSP and chlorophyll exposures, (e–f) number size distribution with the standard-SMPS and nano-SMPS, and (g) CCN number concentration. The x-axis represents local time.



Figure 9. Box plots for (a) number concentration of nucleation-mode particles (N<sub>NUC</sub>), (b) formation rate (FR), (c) growth rate (GR), (d) condensation sink (CS), (e) solar radiation, (f) chlorophyll exposure, (g) DMSP exposure, (h) CCN number concentration, (i) CCN activity, and (j) critical diameter ( $D_c$ ) for ocean, sea ice, and multiple air masses. Upper/lower box limits and solid lines indicate the 75th/25th percentiles and median, respectively.



Figure 10. The increase in CCN concentration during growth to larger than 40 nm particles (green) and smaller than 40 nm (blue) times compared with background times at five different supersaturations.