

# 1 Drivers governing the seasonality of new particle formation in

## 2 the Arctic

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## 45 Abstract

46 New particle formation (NPF) is the phenomenon wherein gaseous precursors form critical clusters of barely a  
47 few nanometres in diameter, after which, under favourable conditions these particles can grow to  
48 climate-relevant sizes. Here we present measurements from 2022 to 2024 of particle and ion number size  
49 distributions from the Zeppelin Observatory (ZEP), an Arctic research station situated on the western edge of  
50 Svalbard atop a mountain. NPF events begin in April and continue occurring into November. The events at the  
51 start of the NPF season (i.e. April/May) are considerably stronger (i.e. a larger production of nucleation mode  
52 particles) than compared with other months of the year. The peaks in NPF strength coincide with peaks in the  
53 accumulated solar insolation experienced by arriving air masses. During the summer period NPF events occur  
54 on 20-40% of days each month, however, there is a consistent decline starting in June. We show that the  
55 combined influence of solar insolation and the surface area of pre-existing aerosols (i.e. condensation sink, CS)  
56 are strong predictors for the likelihood of NPF. We develop a simplified predictive model which matches the  
57 frequency of NPF events identified via the classification schemes used in this study; we show that the ratio of  
58 solar insolation over CS corresponds well to the frequency of NPF events ( $R^2$  of 0.78). We show that NPF events  
59 occur during the polar night (i.e. when the Sun does not pass above the horizon), and that these events are linked  
60 to high altitude air masses. Furthermore, we detail the likely geographic origins of nucleation mode particles as  
61 measured at ZEP. We show that NPF events are considerably more likely to originate from marine regions  
62 towards the west of Svalbard, particularly the Greenland Sea, which is the marine region with the greatest  
63 likelihood of originating air masses linked to an NPF day. We also show that NPF events lead to an increase in  
64 the number of Aitken mode particles, indicating that a significant proportion of the Aitken mode particles  
65 originate from NPF. Of the measured NPF events, 37% of them exhibited nucleation mode particles that grew  
66 beyond 25nm, a diameter representing the minimum activation diameter for particles to act as cloud  
67 condensation nuclei. Overall, we present a concise picture of the lifecycle of nucleation mode particles in the  
68 Arctic, including the effect wet scavenging has in reducing the condensation sink, which in turn promotes NPF  
69 events to occur.

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## 72 1. Introduction

73 New particle formation (NPF) describes the production of secondary aerosol particles from precursor gases to  
74 clusters of molecules a few nanometres in diameter. Newly formed particles belong to the nucleation mode (i.e.  
75 typically <10nm) and may grow through the continued coagulation and condensation of vapours into Aitken  
76 mode particles (i.e. tens of nanometres in diameter). The phenomenon of NPF has been observed globally  
77 (Kerminen et al., 2018), however, in each region the chemical characteristics and controlling source and sink  
78 processes leading to NPF may vary.

79 Arctic aerosol seasonality is governed by a combination of atmospheric general circulation, meteorology, and  
80 several source and sink processes including the direct emission of local primary particles, NPF, the long-range  
81 transport of anthropogenic emissions from lower latitudes, and the removal of particles through wet scavenging  
82 (Garrett et al., 2011; Schmale et al., 2022; Tunved et al., 2013; Willis et al., 2018). One distinctive feature of the  
83 annual cycle is the Arctic haze in late winter and early spring, which is characterised by a sustained increase in  
84 the number of accumulation mode aerosol (i.e. particles typically around 100nm) (e.g. Shaw (1995)). Reduced  
85 wet scavenging and increased transport efficiency from anthropogenic source regions at lower latitudes during  
86 this part of the year give rise to this phenomenon (Garrett et al., 2011). The Arctic summertime, by contrast, is  
87 less influenced by long-range transported pollutants, and the increased precipitation effectively reduces the once  
88 pronounced accumulation mode. Moreover, increased photochemistry and biological activity (e.g. phytoplankton  
89 blooms) provide a source of precursor gases encouraging NPF and the growth of newly formed particles  
90 (Tunved, Ström and Krejci, 2013; Croft et al., 2016; Price et al., 2023); as a result, the smaller Aitken and  
91 ultrafine aerosol particles increase in number.

92 Changes in aerosol size, abundance and chemical composition can perturb the radiative balance either indirectly  
93 by acting as cloud condensation nuclei (CCN), altering cloud formation, brightness, and lifetime or directly  
94 through their interaction with both short- and long-wave radiation (Li et al., 2022). The Arctic region can at times  
95 be CCN-limited, which means that small changes in CCN concentrations could potentially impact cloud  
96 properties via cloud-mediated indirect aerosol effects, which in this and similar environments could result in  
97 surface warming due to enhanced long-wave cloud forcing (Garrett et al., 2004; Garrett and Zhao, 2006;  
98 Mauritsen et al., 2011). Thus, local-regional scale production of aerosols during the summertime and changes to  
99 this production mechanism may have significant consequences for the Arctic (Kecorius et al., 2019). Still  
100 however, the impact of newly formed particles on Arctic cloud properties remains poorly quantified.

101 Depending on certain environmental factors, for example the rate of particle nucleation, the production rate and  
102 volatility of condensable vapours, and the condensation and coagulation sinks, to mention a few, these newly  
103 formed particles can reach climate-relevant sizes; In the Arctic, studies have suggested that particles as small as  
104 25nm can participate in cloud formation by acting as CCN (Leaitch et al., 2016; Karlsson et al., 2020, 2021;  
105 Pöhlker et al., 2021; Gramlich et al., 2023; Motos et al., 2023) thus affecting cloud properties and climate (i.e.,  
106 by influencing the radiative balance). Motos et al. (2023) predicted the smallest particle activation diameters in  
107 the summertime to be 20nm, with a modal average of 60 nm, but suggested that the dominant summertime  
108 Aitken mode originating from NPF may stay in the interstitial (inactivated) phase. Other studies have indicated  
109 that a substantial fraction of CCN in the Arctic are produced via NPF (Gordon et al., 2017; Merikanto et al.,  
110 2009).

111 Air ions are an additional focus area of aerosol effects, as they can play a role in nucleation (Hirsikko et al.,  
112 2011; Kirkby et al., 2011) and can influence aerosol particles through their formation and growth mechanism.  
113 Both positive and negative ions contribute to the production of newly formed particles in the Arctic, via  
114 ion-induced nucleation (see Beck et al., 2021 for negative ions and Jokinen et al. (2018) for positive ions) The  
115 overall contribution of ions to NPF is disputed as quantifying their impact on nucleation rates in ambient  
116 conditions remains difficult. However, studies suggest that the dynamics of sub-2nm clusters is dominated by  
117 neutral clusters (Hirsikko et al., 2011; Kulmala et al., 2013) . Ions can help stabilise clusters which form initially  
118 (Yu and Turco, 2001) , with most particles formed from ion-induced nucleation reaching neutrality after growing  
119 beyond 2.5nm (Wagner et al., 2017).

120 Observations of ultrafine particles (~3-20nm) in the Arctic (onboard the Swedish icebreaker Oden, 70°N-85°N)  
121 were reported by Covert et al. (1996) and Wiedensohler et al. (1996). At Zeppelin Observatory (ZEP), Ström et  
122 al. (2003) and Tunved et al. (2013) presented measurements of small (>10nm) aerosol particles showing an  
123 increased concentration and frequency of formation events during the Arctic summer. Tunved et al. (2013)  
124 suggested that the summertime, coinciding with increased photochemical production of precursor gases, and the  
125 removal of larger pre-existing aerosol, generates favourable conditions in which NPF can occur. Other Arctic  
126 sites have been the focus of research into NPF, including Villum Research Station (hereafter Villum) (see  
127 Nguyen et al. (2016)) and Summit (see Ziemba et al. (2010)), both on Greenland, and Utqiagvik, Alaska  
128 (Kolesar et al., 2017a). Other studies, e.g. Brean et al. (2023) present NPF data from several locations, including  
129 Alert, Villum, Tiksi, ZEP, Gruvebadet (Ny-Ålesund), and Utqiagvik; a commonality shared by all Arctic sites  
130 was the occurrence of NPF events during the summer, and also that events coincided with air masses generally

131 influenced by marine regions, including at Utqiagvik; and Alert whereby a correlation was observed between  
 132 biogenic methane sulphate (MSA-) and summertime particle number concentrations (Leaitch et al., 2013;  
 133 Quinn et al., 2002). Beck et al. (2021) through a comparison of measurements from both Ny-Ålesund and  
 134 Villum, showed that different nucleation mechanisms can also occur at different times of the year at these two  
 135 locations; at Villum iodic acid (IA,  $\text{HIO}_3$ ) was found to be the primary driver of NPF events during springtime,  
 136 and sulphuric acid (SA,  $\text{H}_2\text{SO}_4$ ) -ammonium (AM,  $\text{NH}_4^+$ ) driven events were shown to occur in the summer. At  
 137 Ny-Ålesund, NPF was driven by SA-AM during the springtime and highly oxygenated molecules (HOMs)  
 138 during the summer. There is no evidence that anthropogenic precursors contribute to NPF at ZEP (Schmale and  
 139 Baccarini, 2021). Instead, Dimethyl Sulphide (DMS), produced by phytoplankton, can serve as a precursor gas  
 140 contributing to NPF. Marine regions with strong chlorophyll a concentrations and DMS production capacity  
 141 have been linked to increased concentrations of nanoparticles, which suggests that marine biogenic sources are  
 142 an important source of nucleating and condensing material (Lee et al., 2020). Observed DMS concentrations  
 143 vary with air mass origin, season and residence time over different marine source regions. For example, it has  
 144 been shown that air masses exposed more to the Greenland Sea, compared with the Barents Sea bring about  
 145 higher DMS concentrations (Park et al., 2018). In the Arctic, there are low concentrations of nucleating agents as  
 146 opposed to continental rural and urban locations (Karl et al., 2012; Pirjola et al., 2000). However, in spring and  
 147 summer increased solar radiation, a lower total surface area of pre-existing particles (i.e. reduced condensation  
 148 sink, CS), and coinciding phytoplankton blooms are shown to give rise to precursor gas concentrations sufficient  
 149 to sustain ion-induced nucleation, and growth of NPF-induced particles beyond 20nm (Beck et al., 2021).

150 Recent modelling and global aerosol-climate coupling studies both laboratory and field-based have increased our  
 151 understanding of biogenic sulphur and iodine pathways. For sulphur pathways, Hoffmann et al. (2016) showed  
 152 that the aqueous-phase (i.e. cloud and aerosol water chemistry), and not just the gas-phase, play an important  
 153 role in the oxidation products of DMS, demonstrating the complexity of atmospheric sulphur chemistry in part  
 154 due to the multiple oxidation pathways, which in turn are not represented well in climate models. Beyond  
 155 sulphur-derived precursors, Finkenzeller et al. (2023) shed light on the formation mechanism for iodic acid  
 156 ( $\text{HIO}_3$ ) under atmospheric conditions and helps to corroborate how iodine can be produced and therefore act as a  
 157 strong nucleating vapour in polar regions. Iodine-assisted NPF is also directly supported by the experimental  
 158 findings shown in Baccarini et al. (2020). When iodine chemistry is included iodine acts as a missing source of  
 159 low-volatility vapours, and improves the modeled Aitken mode representation (Xavier et al., 2024).

160 In this study, we present two and a half years' worth of measurements of particle and ion number size  
161 distributions, including three summer periods (in total data covers the period April 2022 – October 2024).  
162 Continuous measurements of sub-3nm up to ~850nm particles were performed at the ZEP, Svalbard, providing  
163 unique information of the initial stages of nucleation and the subsequent growth. NPF events were identified,  
164 formation rates were calculated, and subsequent growth was analysed. The seasonality and coinciding  
165 environmental parameters encouraging NPF events and subsequent growth have been explored in detail.

## 166 2. Materials and methods

### 167 2.1 Measurement site

168 Zeppelin observatory (ZEP) (78.90°N, 11.88°E, 474 m a.s.l.) is situated on the ridge of Mount Zeppelin, 2 km  
169 south from Ny-Ålesund research village on the western edge of the Norwegian Svalbard archipelago. ZEP  
170 represents regional background conditions in the Atlantic sector of the Arctic, as it is largely unaffected by local  
171 air pollution due to its location well above Ny-Ålesund and the prevailing wind patterns driven by  
172 Kongsfjorden's geography. ZEP is one of the most developed atmospheric observational sites in the Arctic where  
173 a broad set of parameters has been measured for decades. It is part of numerous regional and global monitoring  
174 networks, including the Global Atmosphere Watch (WMO/GAW), Integrated Carbon Observation System  
175 (ICOS), The Aerosol, Clouds, and Trace Gases Research Infrastructure (ACTRIS) and the co-operative  
176 programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe (EMEP).  
177 For a detailed description of the observatory, local meteorology and climatology, history and observational  
178 program see Platt et al. (2022).

### 179 2.2 Instrumentation

180 For this study, an array of instruments has been used to measure aerosol and ion size distributions, between 0.8  
181 and 850 nm including a Neutral Cluster and Air Ion Spectrometer (NAIS5, Airel Ltd. Tartu, Estonia), a  
182 Nanoparticle Scanning Mobility Particle Sizer (Nano SMPS, TSI), and a Differential Mobility Particle Sizer  
183 (DMPS) system. Particle and ion number size distributions and concentrations form the core data used in this  
184 study. Several other supporting data sets were used for data analysis and interpretation and are described below.  
185 This study covers the period from 2022-04-17 to 2024-10-04.

#### 186 2.2.1 Differential Mobility Particle Sizer (DMPS) system

187 Particle number size distributions, from 5 to 850nm, were measured using a custom-made twin-DMPS system  
188 with a closed loop sheath circulation and composed of two Hauke-type Differential Mobility Analysers (DMA).

DMPS-1, with a short (5.3 cm) DMA, measured aerosol size distribution from 5 to 57 nm using sample-to-sheath air ratio close to 1:10. DMPS-2, with medium length DMA (28 cm), measured size distribution from 20 to 850 nm using sample-to-sheath air ratio close to 1:5. The aerosol particles were counted using condensational particle counters (CPCs) at a frequency of 1 Hz with the two CPCs. Particles > 3 nm were measured using the Ultrafine CPC (UCPC) TSI model 3776 (cut-off of 2.5nm) coupled to the DMPS-1. Particles > 10 nm were measured with the CPC TSI model 3010 (cut-off of 10nm) coupled to the DMPS-2. The two DMAs were run in a stepwise mode, providing two particle number size distribution cycles approximately every 27 minutes. The overlap between the size ranges measured by both DMPSs was used to check consistency and to merge both size distributions into one. Additional information can be found in Karlsson et al. (2021). The DMPS measurements were corrected for diffusional losses using the Particle Loss Calculator (PLC) according to Von Der Weiden et al. (2009) (see Fig. S1 in the supplement for details).

### 2.2.2 Neutral Cluster and Air Ion Spectrometer (NAIS)

NAIS was utilised to measure concentrations of ions (charged particles and cluster ions) of both polarities between 0.8 and 40 nm, and particles between 2.5 and 40 nm. The NAIS is an aerosol mobility spectrometer; when the sample is left unmodified it detects naturally-charged ions and particles, and when the corona charger is used to charge the particle population it measures all particles including the uncharged (Manninen et al., 2011; Mirme et al., 2007; Mirme and Mirme, 2013). It consists of two mobility analyser columns, with a total of 25 electrometers per column. Its lower size thresholds and high temporal resolution make it highly suitable for observing the early stages of NPF. The NAIS provides very accurate size information, regardless of the choice of the inversion algorithm used (in this case, the 25-channel V14.1 inverter algorithm) (Wagner et al., 2016). The lowest detection limit for the NAIS in particle mode is set based on the diameter of the corona charger ions, and the inability of the electrical filter to remove all naturally charged particles (Manninen et al., 2011), however, there is a broad consensus that measurements of concentrations of particles below 2.5nm are inaccurate.

The NAIS at ZEP was installed with a very short metallic inlet, less than 1m long, to minimise diffusional losses. The inlet pointed slightly downward, and a metallic rain shield was attached at the end, to ensure that rain did not enter into the instrument. The NAIS inlet was heated to prevent ice blocking the inlet, and also to limit water condensing inside the tubing. The NAIS was serviced and cleaned once a year. Additional aspects of the NAIS observations, including the apparent overestimation of particle number concentrations when compared to DMPS measurements (see also Kangasluoma et al., 2020) and the loss of negative cluster ions can be found in section S2.6 of the supplementary material.

### 219 2.2.3 Complementary observations

220 A Nanoparticle Scanning Mobility Particle Sizer (Nano SMPS, TSI) comprising of a nano-differential mobility  
221 analyser (nano-DMA, TSI 3085, USA) and an ultrafine CPC (TSI 3776, USA), provided size distributions of  
222 nanoparticles (3-60nm). See Lee et al. (2020) for further details. Atmospheric Dimethyl Sulphide (DMS) was  
223 measured using an analytical system in which DMS is trapped and eluted, before being quantified using gas  
224 chromatography equipped with a pulsed-flame photometric detector. The detection limit is approximately 1.5  
225 pptv given air samples of around 6 litres. For more details, see Jang et al. (2016) and Park et al. (2018).  
226 Meteorological parameters including visibility, relative humidity, wind speed and direction, ambient temperature  
227 and pressure were also further utilised throughout the study.

### 228 2.3 Data Analysis of aerosol measurements

229 For processing of the NAIS data the *nais-processor* was used (<https://github.com/jlpl/nais-processor>). The  
230 *nais-processor* Python package corrected for diffusional losses in the inlet (Gormley and Kennedy, 1948),  
231 applied an ion mode calibration (Wagner et al., 2016), converted ion mobility to particle diameter, remapped the  
232 distribution to a new size grid, and adjusted the data to standard conditions (273.15 K, 101325 Pa).

233 Further information on the removal of artefacts can be found in Sect. S2 of the supplementary material. In  
234 addition, a comparison between the particle concentrations measured by NAIS and the DMPS-system can be  
235 found in Sect. S2.5; we report a significant overestimation of particle number concentrations as measured by the  
236 NAIS in comparison to the DMPS-system, particularly at the smallest diameters (see Fig. S9-S11). It should be  
237 noted that the particle number size distributions presented in this study represent that of dry aerosol particle  
238 sizes. The measured particle size distributions for both the NAIS and DMPS were not corrected for hygroscopic  
239 growth, and thus do not reflect ambient conditions.

240 When assessing the qualitative and quantitative nature of NPF events, it is important to distinguish between on  
241 the one hand the production of newly formed particles (~2-4nm) and on the other hand subsequent particle  
242 growth. To qualify as an event, not only does formation of ultrafine particles need to be present, but also the  
243 subsequent growth of the particles. Growth can last for several hours and in some cases multiple days, with the  
244 latter timescale suggesting that the NPF events occur on a regional scale (Kecorius et al., 2019; Ström et al.,  
245 2009). In this study, we distinguish between (1) *on-site formation*, defined as occasions when the concentration  
246 of 2-4nm particles clearly rises above the background level; and (2) *formation and subsequent growth*, defined as

a combination of *on-site formation* followed by subsequent growth. The latter is identified by visual inspection of the behaviour of the growing nucleation mode during the time it is present until when it disappears.

### 2.3.1 NPF Classification

New particle formation events were classified using schemes described by Dal Maso et al. (2005) and Aliaga et al. (2023). The classification schemes are used complementary. The classification by Dal Maso et al. (2005) focuses more on formation and growth, and is perhaps more intuitive to understand; however, it struggles with a large number of undefined cases. On the other hand, the nano-ranking analysis developed by Aliaga et al. (2023) can be used to explore the NPF intensity and focuses on the formation of nucleation mode particles as opposed to the subsequent growth. The Aliaga-method classifies NPF events into 3 groups based on the overall ranking of NPF intensity, avoiding the problem with unidentified event days (see Fig. S16 for the groups). The two classification methods can be compared with one another (see Fig. S14). We used the method for the intensity parameter and the diameter range that showed the best comparison with the Dal Maso et al. (2005) i.e. the daily maximum for the total number concentration of 2.8 nm and 5 nm particles ( $\Delta N_{2.8-5 \text{ nm}}$ ).

#### *Dal Maso classification system*

In the Dal Maso et al. (2005) scheme an NPF event is defined if there exists a nucleation mode (3-25nm) which prevails for several hours and shows signs of growth; these events are further subdivided into three separate classes, namely, Class Ia and Ib, and Class II. Class I and Class II events are separated on the basis of whether the growth and formation rates, of the well-behaved size distribution, can be determined with good confidence. Class I is further divided into Class Ia and Class Ib depending on whether there are pre-existing particles obscuring the newly formed mode (Dal Maso et al., 2005). Undefined events are days where there are sporadic occurrences of nucleation-mode particles, and this classification is used to separate clear events from clear non-events. For examples of the different classifications and their respective averages see Fig. S12 and S13 respectively in the supplement.

In this study, undefined days include days where the occurrence of nucleation mode particles (3-25nm) is linked to windblown snow and situations when ZEP is enveloped by clouds. For more details regarding these sampling conditions see Sect. S2.1 and S2.2 in the supplement for in-cloud-sampling and windblown snow events respectively. As a result, days classified as undefined are ones which experience bursts of nucleation mode particles, either from windblown snow (see Fig. S4), in-cloud conditions (see Fig. S2), from non-growing NPF or a combination of all three. There is a definite increase in the concentration of particles with diameters around

5nm when the NAIS measures in conditions of low visibility and this is not observed in other instruments (see Fig. S3). For windblown snow events, particles with diameters less than 20nm observed considerable increases in concentrations when the wind speed passed roughly 10ms<sup>-1</sup> (see Fig. S5). Non-events are days that display no nucleation mode (3-25nm). It is important to note that this scheme assumes that particle formation occurs over a geographically wide area more or less simultaneously. Identification and classification were performed using daily surface plots of particle number size distribution based solely on NAIS data, that is, 2.5-40nm.

It should be noted that there is a degree of subjectivity to this classification approach, as to what is considered a certain class. The addition of the NAIS, in combination with the DMPS-system, made it easier to detect events using surface plots, especially when the growing mode struggled to surpass ~6nm. However, we were not able to identify any NPF events with the NAIS that we could not identify with the DMPS-system, as close inspection of the number concentration of the lowest size bins of the DMPS-system was sufficient to recognise NPF events.

### *Nanoparticle Ranking Analysis*

The Nanoparticle Ranking Analysis method developed by Aliaga et al. (2023) provides a more continuous method to characterise atmospheric NPF, allowing users to gauge the strength of NPF. The nanoparticle ranking analysis was utilised using the daily maximum concentration of 2.8-5nm particles ( $\Delta N_{\text{max}, 2.8-5}$ ), from the negative channel of the NAIS, as the intensity parameter (note that no background value was used, unlike in Aliaga et al. (2023), due to the low concentrations measured at ZEP). In this study, periods which experienced windblown snow or in-cloud events were removed before applying the Nanoparticle Ranking method (see Figs. S7 and S8), as opposed to the Dal Maso et al. (2005) approach which classified them as undefined days. Special care was taken to clean the data before utilising the nanoparticle ranking analysis method, however it may still be the case that not all in-cloud and windblown snow events were removed successfully (see Sect. S2.1 and S2.2 in the supplement). Windblown and in-cloud events can influence the daily maximum without necessarily reflecting NPF activity. The strength of NPF activity was split into 3 groups depending on the intensity (i.e.,  $g1$ ,  $g2$ ,  $g3$ ), where  $g3$  is the most intense event, followed by  $g2$  and then  $g1$ ; see Aliaga et al. (2023) for more details (see Fig. S15 & Fig. S16).

### **2.3.2 Growth rates**

The growth rate (GR) describes the rate of change of the modal diameter during an NPF event. For the GR estimation, NAIS and DMPS size distributions were merged at the maximum diameter of the NAIS size range (i.e. 40nm). The NAIS and DMPS data were both interpolated to 15-minute arithmetic means using linear

interpolation. The start and end of each NPF event including their sequential growth (i.e. (2) *formation and subsequent growth*,) were estimated by visual inspection. The growing nucleation mode was isolated, and the GR was estimated for the full duration of the NPF event (see Fig. S17). For each size bin, we applied a Gaussian filter (sigma = 1) and found the time in which the concentration of a particular size bin experienced the highest maximum rate of change. Essentially, the evolution of the nucleation mode was tracked using the timings for each identified peak in the rate of change of the growing nucleation mode diameter. The GR was calculated using a simple linear ordinary least squares (OLS) method.

It should be noted that the aforementioned issues concerning the overestimation of particle number concentrations for the NAIS measurements do not impact the calculation of GR as it only examines the changes to the diameter of the nucleation mode. Very few events grew beyond the limit of the NAIS (i.e. 40nm), hence the impact of a diameter jump from NAIS to DMPS was not considered. We tried to measure the GR of all NPF events (e.g. Class 1a, 1b and II), even though by definition Class II events lack the strength and consistency, making it difficult to calculate GRs (Dal Maso et al., 2005). The GRs for some Class II events were not estimated due to uncertain tracking of the nucleation growth.

### 2.3.3 Particle Formation Rates

The formation rates namely  $J_{3-7}$ ,  $J_{7-25}$ , and  $J_{3-25}$  ( $\text{cm}^{-3}\text{s}^{-1}$ ) were estimated using the following equation:

$$J_{i-j} = \frac{dN_{i-j}}{dt} + \text{CoagS} \times N_{i-j} + \frac{GR}{\Delta d_p} \times N_{i-j}, \quad (1)$$

where  $J_{i-j}$  is the formation rate of particles between diameters  $i-j$  nm, CoagS is the coagulation sink of the pre-existing particle population, GR is the growth rate between  $i-j$  nm,  $\Delta d_p$  is the difference in diameter between  $i$  and  $j$  in nm and  $N_{i-j}$  is the total concentration of particles between  $i$  and  $j$  nm (Fig. S18 represents an example for calculating formation rates for particles between 3-7nm).

The Python GUI, *npf-event-analyzer*, was used to provide additional complementary estimations of the GRs and formation rates for the NPF events (i.e. in addition to the OLS fitting method). When using the *npf-event-analyzer* the max concentration method was used for the estimations of GR and J.

### 2.3.4. Condensation sink

The condensation sink (CS) is defined as the rate at which non-volatile vapours condense onto pre-existing particles (Kulmala et al., 2012). The twin DMPS system (5-850nm) was used to calculate CS, for SA vapour. The method for the determination of CS is described by Dal Maso et al. (2002), and uses the following equation:

$$CS = 2\pi D \int_i^{d_{p,i \max}} d_{p,i} \beta(d_{p,i}) N(d_{p,i}) dd_{p,i}, \quad (2)$$

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333 where  $d_{p,i}$  is the diameter of a particle in size class  $i$ ,  $N_i$  is the particle number concentration in the respective size  
 334 class, and  $D$  is the diffusion coefficient of the condensing vapour (in this case condensing vapours were assumed  
 335 to have  $\text{H}_2\text{SO}_4$  diffusion properties). We used the transition regime correction factor  $\beta$  from Fuchs and Sutugin  
 336 (1970).

### 337 2.3.5 Concentration of condensable vapours and source strength

338 The concentration of vapours ( $C_v$ ) required to sustain the calculated GRs and their respective source rates ( $Q$ )  
 339 were estimated using the following equations, and assuming the condensation of  $\text{H}_2\text{SO}_4$ .

$$C_v = A \times dD_p / dt. \quad (3)$$

340 where  $A$  is the constant  $1.37 \cdot 10^{-7} \text{ h cm}^{-3} \text{ nm}^{-1}$  representative for the molecular properties of SA and  $dD_p / dt$  is  
 341 the rate of change of the nucleation mode i.e. GR from the start of the NPF until it stops growing (see Sect. 3.1.5  
 342 below for further details). See Dal Maso et al. (2005) for more details. The constant  $A$  incorporates the  
 343 probability that sulfuric acid molecules will ‘stick’ to particles upon collision, the change in volume which  
 344 occurs after collision and condensation, and also the frequency of collisions (Fiedler et al., 2005).

## 345 2.4 Airmass analysis

### 346 2.4.1 Transport model

347 Airmass back trajectory analysis was performed using the Hybrid Single-Particle Lagrangian Integrated  
 348 Trajectory model (HYSPLIT V5.2.1) (Draxler et al., 1998; Stein et al., 2015). Ensemble back trajectories were  
 349 initialised every hour starting at the latitude and longitude of ZEP, and at a height of 250m above ground level  
 350 (AGL). The ensemble was generated by offsetting the meteorological grid point by one in the horizontal (i.e.  $dx_f$   
 351  $= 1.0$ ,  $dy_f = 1.0$ ) and by the default offset of 0.01 sigma units in the vertical ( $\sim 250\text{m}$ ), thus generating 27 back  
 352 trajectories for all possible offsets in X, Y and Z. The starting height of 250m ensured that the starting location  
 353 of all the ensemble members was at the surface or above. The back trajectories were calculated for 5 days back  
 354 in time. Global Data Assimilation System (GDAS)  $1^\circ \times 1^\circ$  archive data (<http://ready.arl.noaa.gov/archives.php>,  
 355 last access: 08-08-2024) was used as the meteorological fields. No averaging of the back trajectories was  
 356 performed, reducing them to single back trajectories. HYSPLIT was run from the Desktop using the Python  
 357 package *pysplit* (<https://github.com/mscross/pysplit>).

The accumulated solar flux in the most recent 6 hours prior to arrival was used for the solar insolation term used throughout this study. The solar flux was taken from the original GDAS-derived HYSPLIT output. Furthermore, only endpoints within the mixed-layer were used as this is when air masses are influenced by the surface. The last 6 hours were used, as opposed to a longer duration, to place more significance on solar radiation closer to the receptor; an accumulated solar flux consisting for a longer duration could lead to misleading results.

#### 2.4.2 Chlorophyll a data

Daily mapped chlorophyll a data derived from satellite observations (Aqua MODIS) (last accessed: 2024-10-04, downloaded from <https://oceancolor.gsfc.nasa.gov/13/>) were utilised along with the HYSPLIT output to estimate the chlorophyll a exposure. Exposure of the air masses to chlorophyll a was calculated based on the equation developed by Park et al. (2018), which linked chlorophyll a with DMS measurements on site at ZEP. Chlorophyll a exposure was essentially used as a proxy for DMS emissions. DMS in turn can form H<sub>2</sub>SO<sub>4</sub> via SO<sub>2</sub>, although this was not explicitly addressed by Park et al. (2018). It should be noted that DMS is not only a precursor to H<sub>2</sub>SO<sub>4</sub>, but also MSA. Park et al. (2018) utilised the relationship between DMS and air mass exposure to chlorophyll a to account for the differences in the species-specific phytoplankton population, between the Greenland Sea and the Barents Sea. For more details on the calculation of the chlorophyll exposure see Sect. 12.

#### 2.4.3 Nucleation site estimation

The method utilised here is similar to the NanoMap analysis developed by Kristensson et al. (2014) in which the geographic position of where new particle formation took place was estimated based on the particle number size distribution measurements during NPF events and HYSPLIT back trajectories. In this study, the duration of the nucleation mode was calculated. For every hour of the NPF event, back trajectories were initialised. The length at which the back trajectories were initialised depended on how long the event had lasted. In short,  $i$  hours after the event began, the back trajectory will have been initialised to run  $i$  hours backwards. So, for every hour except the start time the back trajectories were initialised for the following length of time: *back trajectory length* =  $duration - (end\ time - t)$ , where  $t \in (end\ to\ start\ times)$ . Hence, for an NPF event that lasts 7 hours, 6 back trajectory ensembles were calculated for varying lengths from 7 hours to 1 hour long.

#### 2.4.4 Air mass marine regions

The endpoints for every 5-day back trajectory ensemble were assigned to a marine region e.g. Arctic Ocean, Greenland Sea, Barents Sea, Norwegian Sea and North Atlantic Ocean (see Fig. S21 in supplement for defined

regions). Days were assigned to a specific marine region based on the cumulative residence time in the specific sector. A threshold of 0.5 (50%) was used as a qualifier. Days without clear source preference, i.e. the airmasses that did not reach the 50% threshold for either of the pre-defined source regions, were classified as mixed.

### 3. Results & Discussion

#### 3.1 Seasonality and interannual variability

##### 3.1.1 Annual cycle of NPF events

The 848 days of valid NAIS measurements (2022-04-17 to 2024-10-04) are classified according to the Dal Maso et al. (2005) classification system. 157 days are classified as NPF events, either Class Ia events (28), Class Ib (19) or Class II (110). A total of 438 days are classified as Undefined, and the remaining 271 days are considered to be either Non-event days (253) or consisting of too little data (i.e. defined as <12 hours of data per day) (18) (see Fig. 1). Overall, this meant that 19% of days with valid NAIS measurements experienced an NPF event (i.e. Class Ia, Ib or II). The majority of days (52%) are classified as undefined due to the appearance of a non-growing nucleation mode that persisted for some hours. The undefined days are further sub-divided into those containing windblown snow events (19% of all data), in-cloud events (8% of all data) (see Fig. S6 in the supplement). Of the 438 Undefined days, 72 are a result of non-growing bursts (i.e. 17% of the Undefined days).

The monthly frequency of NPF events (i.e. class Ia, Ib or II) shows events first appearing in April (see Fig. 1), reaching a peak, in terms of the occurrences per month, in May to July with approximately 30 - 40% of days experiencing NPF. The daily occurrence decreases during the month of June compared with the preceding and succeeding months, a consistent finding throughout the three years of data. After July, the frequency of events declines towards the end of summer and into autumn, where NPF events cease to be present beyond November. NPF events were observed as late in the year as November, which is interesting given that this is during the polar night when the Sun stays below the horizon (as witnessed by ZEP) and thus ZEP experiences negligible solar radiation. Multiple NPF events were observed during the polar night period (see Sect. 3.1.3 for more details).

April 2022 appeared anomalous perhaps given that the measurements for that month were not an entire month as the measurements started on 17<sup>th</sup> April 2022.

Prior studies, at ZEP, which used slightly different event classification schemes, reported the occurrence of NPF events to be within the range of 18%-23% (Dall'osto et al., 2017; Heintzenberg and Leck, 2012; Lee et al., 2020). Nieminen et al. (2018) reported NPF day frequencies of 14% (Mar-May), 34% (Jun-Aug) and 6.6% (Sep-Nov) and 0% (Dec-Feb). Lee et al. (2020) observed the highest frequency in terms of the formation and growth of

416  $N_{3-25}$  particles in June (>40%), with the NPF frequency in May and July being ~40% and <40%. Dome-C, in  
 417 Antarctica and Alert, on Greenland are two other polar sites with annual NPF frequencies between 10-20%  
 418 (Nieminen et al., 2018), comparable to ZEP.

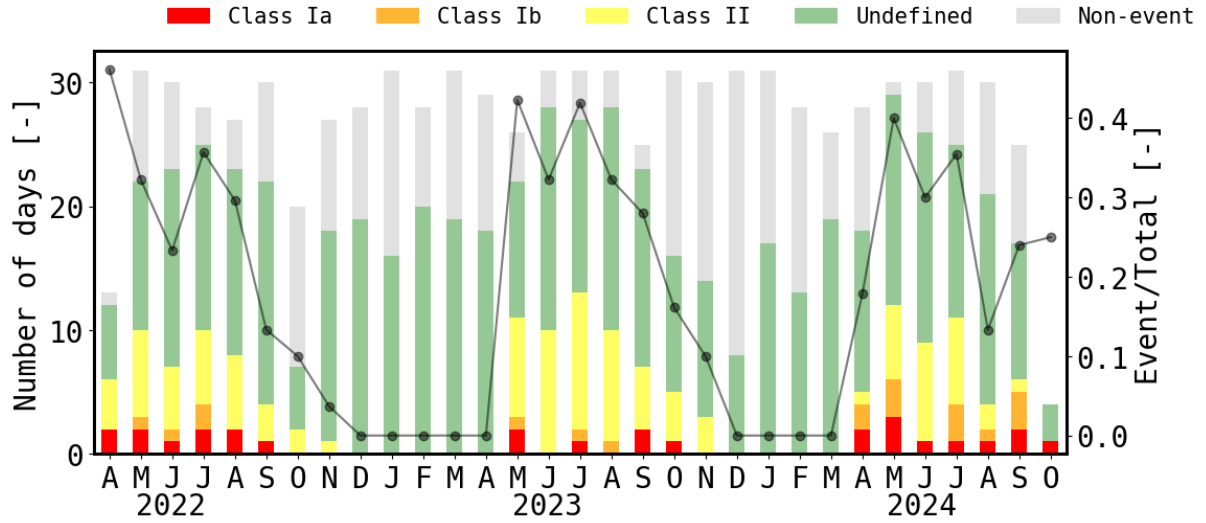
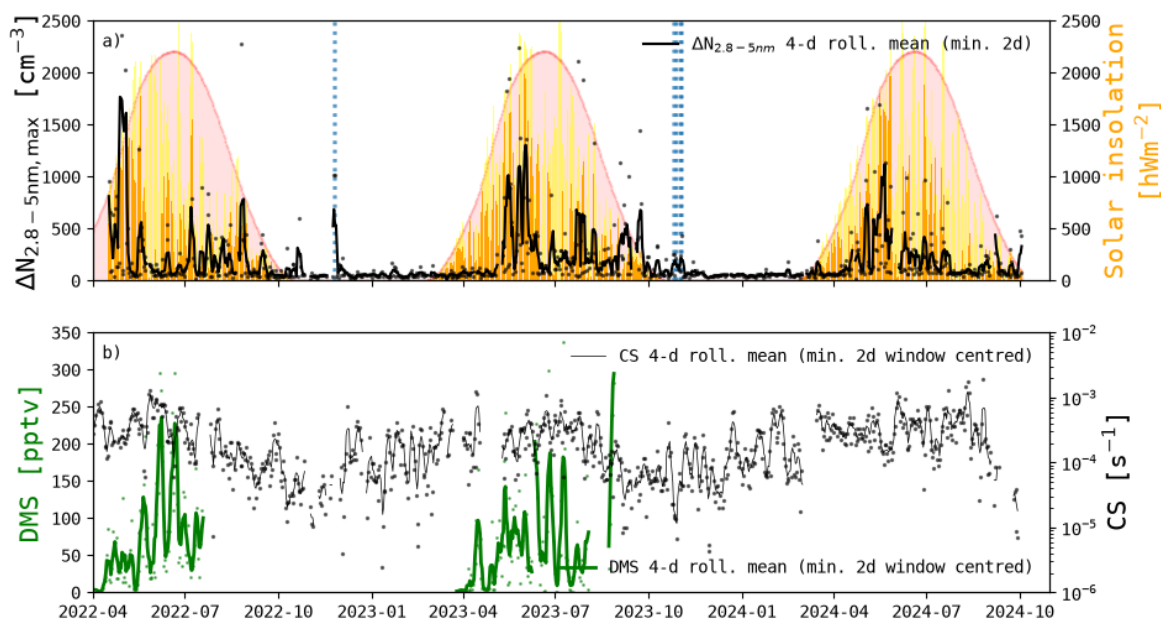


Fig 1: Number of days per month classified as either Class Ia (red), Ib (orange), II (yellow), Undefined (green) and Non-event (grey). The left-hand axis is the number of days classified based on the total number of valid measurement days (i.e. Total). The right-hand side is the fraction of events (i.e. Class Ia, Ib, and II) over the total number of valid measurement days (i.e. Class Ia, Ib, II, Undefined and Non-event). The fraction is given by Event/Total. Almost three years of Neutral cluster and Air Ion Spectrometer (NAIS) particle number size distribution data is classified using the Dal Maso et al. (2005) classification system.

419

### 420 3.1.2 Interannual variability of NPF intensity, solar insolation, DMS and CS

421 The interannual variability of the following main parameters, namely NPF intensity, solar insolation, DMS  
 422 concentration and the CS, are explored in figures 2a-b. A proxy for the intensity of events, as opposed to the  
 423 frequency, was determined by utilising elements of the nanoparticle ranking analysis developed by Aliaga et al.  
 424 (2023). The maximum daily concentration in 2.8 – 5nm particles ( $\Delta N_{max\ 2.8-5}$ ), was used to represent the strength  
 425 of NPF activity (shown to correlate well with calculated J) i.e. the NPF intensity. See Sect. 2.4.1 in the Methods  
 426 for more details concerning the nanoranking method and about the definition of solar insolation.



**Fig. 2:** Time series of (a) the NPF intensity parameter ( $\Delta N_{max,2.8-5}$ ; black dots), defined as the daily maximum particle number concentration between 2.8 and 5 nm. The black line shows the centred 4-day running mean. Orange bars indicate daily solar insolation accumulated over the preceding 6 h for air masses arriving at ZEP within the mixed layer (Sect. 2.4.1); faint yellow bars show the same for all air masses. The red shaded region represents six times the average hourly clear-sky global horizontal irradiance (GHI) at ZEP. (b) Valid DMS measurements (green) and the condensation sink (CS; black dots, daily means) with its centred 4-day running mean (black line). Blue dashed lines indicate polar night periods.

427

428  $\Delta N_{max,2.8-5}$  shows various peaks throughout the annual cycle, however, the one striking feature is that every year  
 429  $\Delta N_{max,2.8-5}$  has its first and highest peak occurring in late spring and the beginning of summer (i.e. end of April in  
 430 2022, beginning of June in 2023, and towards the end of May in 2024) (see the red curve in Fig. 2a)). In terms  
 431 of the annual cycle in NPF event frequency (see Fig. 1), the first peak in NPF event occurrences matches the  
 432 April-June peak in increased  $\Delta N_{max,2.8-5}$  (Fig. 2a)). However, the succeeding peak in NPF event frequency (i.e. in  
 433 July, in Fig. 1) coincides with reduced intensity ( $\Delta N_{max,2.8-5}$ ). The seasonality of the  $\Delta N_{max,2.8-5}$  value follows that of  
 434 the solar insolation, and for the years 2023 and 2024, the maximum NPF strength/intensity (i.e.  $\Delta N_{max,2.8-5}$ )  
 435 coincides with the greatest mean solar insolation; a much better relationship is observed when selecting for air  
 436 masses within the mixed layer as this is when arriving air masses are influenced by the surface (i.e. the orange  
 437 bars in Fig. 2a)). Averaging the intensity parameter and correlating it to the solar insolation calculated based on  
 438 the entire air mass and the parts within the ML shows that selection of air masses within the ML leads to a better  
 439 correlation (see Fig. S27 in supplement). The annual cycle for the calculated CS exhibits a sustained maximum  
 440 in late winter and early spring due to the Arctic Haze (i.e. Feb. - May), and the CS stays high into the summer  
 441 before reaching a minimum in autumn, typically the cleanest part of the year (Tunved et al., 2013) (see Fig. 2b)).  
 442 During the Arctic Haze period, long-range transported air masses from Eurasia bring aged aerosol leading to the

443 higher CS. At the same time this period is characterised by higher sulphate and SO<sub>2</sub> concentrations, mainly of  
 444 anthropogenic origin (Platt et al., 2022), and hence there are NPF events occurring and high increased NPF  
 445 intensity. One additional aspect is that it is clear that increases in  $\Delta N_{max\ 2.8-5}$  occur during periods where there was  
 446 a slight decrease in CS, below that of the seasonal CS average. For example, the peak in  $\Delta N_{max\ 2.8-5}$  in April in  
 447 2022 (see Fig. 2b)), which occurs slightly prior to the maximum solar insolation, can potentially be explained by  
 448 the reduction in the CS during that period.

449 The solar intensity is a controlling factor in the production of SA (i.e. H<sub>2</sub>SO<sub>4</sub>) via the oxidation of SO<sub>2</sub>. In the  
 450 Arctic, DMS acts as a significant source of sulphate, however other sulphur compounds can be oxidised e.g.,  
 451 carbon disulphide and hydrogen sulphide (Pei et al., 2021). The coagulation sink (CoagS) acts as a sink for the  
 452 newly-formed particles, and the CS acts as a sink for nucleating vapours, thus high CoagS and CS can inhibit  
 453 NPF. NPF days with greater intensity (i.e.,  $\Delta N_{max\ 2.8-5}$ ) are skewed towards the end of spring and beginning of  
 454 summer. Similarly, solar insolation is also shifted towards the start of summer, even though the theoretical solar  
 455 maximum has a symmetrical distribution centred around the middle of summer (see red shaded region in Fig.  
 456 2a)). The skewed pattern in both the solar insolation and NPF intensity ( $\Delta N_{max\ 2.8-5}$ ) can perhaps be explained by  
 457 annual changes in cloud cover (see Maturilli and Ebell (2018) for the frequency occurrence of cloudy sky,  
 458 increasing to ~80% during the summer from an April minimum of ~40-60%). Late autumn experiences  
 459 numerous, but relatively weaker, NPF events, with peaks in  $\Delta N_{max\ 2.8-5}$  (~100-500cm<sup>-3</sup>) every year at the end of the  
 460 summer despite the relatively small solar insolation. The transition period between summer and haze (i.e. Oct. –  
 461 Jan.) is typically the cleanest part of the year, and thus exhibits the lowest CS (Tunved et al., 2013). During this  
 462 part of the year the small amount of solar insolation and vapours present are enough to trigger NPF, possibly as a  
 463 result of the low CS.

464 Finally, DMS is the main source of sulphur for clean marine aerosol particles, and constitutes an important  
 465 compound in regards to NPF via its oxidation products. Numerous peaks in DMS are observed during the  
 466 summertime, with a maximum observed in July (see Fig. 2b)). Elevated concentrations of DMS at ZEP coincide  
 467 with air masses which traverse over marine regions containing chlorophyll a (see Fig. S30 – S37 in supplement,  
 468 but also noted by Park et al. (2018b)).

### 469 3.1.3 Events with minimal solar insolation (i.e. Polar night events)

470 Contrary to expectations, during the polar night (i.e. when the sun does not cross the horizon at Ny-Ålesund) a  
 471 number of NPF events are observed. As shown earlier, solar insolation is clearly one of the driving parameters

472 for NPF occurrence and intensity, and Ny-Ålesund in November does not receive any direct solar radiation (see  
 473 Fig. S19 for examples of the polar night events). For the November polar night events, air mass back trajectories  
 474 indicate that air masses have to travel at least half a day backwards before reaching Ny-Ålesund to experience  
 475 direct solar radiation. In total, five types of these events are observed, and are detailed in Table 1.

476 *Table 1: Polar night events observation from 2022-04-17 to 2024-10-04*

| Polar night day | Datetime                                                             | Duration [hrs] | GR [nm hr <sup>-1</sup> ] |
|-----------------|----------------------------------------------------------------------|----------------|---------------------------|
| 1.              | 2022-11-25 - 2022-11-26                                              | 15             | 0.38                      |
| 2.              | 2023-10-25 - 2023-10-26                                              | 23             | 0.41                      |
| 3               | a. 2023-10-28 01:00 – 2023-10-29<br>b. 2023-10-28 12:00 – 2023-10-29 | 36<br>27       | 0.25                      |
| 4.              | 2023-11-01 - 2023-11-02                                              | 28             | 0.32                      |
| 5.              | 2023-11-02 -2023-11-03                                               | 8              | 0.78                      |

477

478 The polar night NPF event starting on the 2023-10-25 appears to be a transported event (TE, see Dada et al.  
 479 (2018) for details on the definition of a transported event) as we do not observe the entire growth of freshly  
 480 nucleated particles; instead the growing nucleation mode begins at 4 nm. The extremely low accumulated solar  
 481 radiation experienced by the arriving air masses during these events means that there should be little available  
 482 nucleating vapours (less oxidation before reaching ZEP). Two other Arctic sites namely Utqiagvik, Alaska and  
 483 Tiksi, Russia, also observe NPF events without the sites experiencing any solar radiation (Asmi et al., 2016;  
 484 Kolesar et al., 2017b). However, in these two cases the suggestion is that anthropogenic emissions of  
 485 semi-volatile gases from nearby oil fields or the build-up of anthropogenic emissions during the haze period can  
 486 explain these events; an interpretation which is not applicable for ZEP.

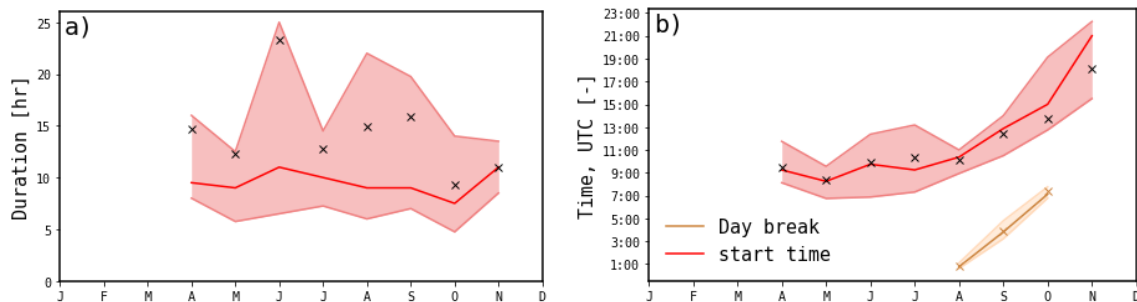
487 The vertical component of the arriving back trajectories associated with these polar night events shows that air  
 488 masses originated from higher altitudes (Figure S17, in the supplement), and that these air masses descend  
 489 before arriving at ZEP. One possible explanation for these types of events could be that precursor gases,  
 490 originally emitted from the surface, are transported to high altitudes where the CS is sufficiently low enough to  
 491 allow them to survive long enough to be nucleated on descent to ZEP. The air masses travel further south and

492 higher in altitude, where the small amount of solar radiation potentially allows for photochemical production of  
 493 nucleating vapours to build up.

#### 494 3.1.4 Start times and duration throughout the year

495 The median duration of NPF events, including their subsequent growth, is approximately  $\sim 10$  hours and seems  
 496 to be fairly consistent across the annual cycle, with a decrease towards the autumn (see Fig. 3a)). Of all events,  
 497 75%  $> 7$  hours, and 90%  $> 4$  hours. During the sunlit period, the starting time of events is typically around 09:00  
 498 (UTC) for the period April to August. From September onwards, the onset of the observed NPF shifts to later in  
 499 the day (Fig. 3b)). For the period August to October, onset of NPF is approximately 8 hours after sunrise at Ny  
 500 Ålesund (see the gradient of the two slopes in Fig. 3b) remains constant but the start time is shifted by 8 hours).  
 501 This could reflect that during the NPF season there is a delay of approximately 8 hours from the minimum solar  
 502 intensity to the onset of NPF, and this could be related to the time it takes for precursor gases to accumulate.

503 Lee et al. (2020) suggested that the duration of NPF at ZEP was approximately 6-7 hours, with the longest  
 504 duration in summer. In addition, NPF start times were 13:00 – 14:00 (local time, which is 11:00 – 12:00 UTC in  
 505 the summer time). The start times observed at ZEP, here in this study, were typically 2 hours earlier, perhaps as a  
 506 result of the slightly lower minimum particle diameter detection and not the same definition of the onset of  
 507 nucleation.

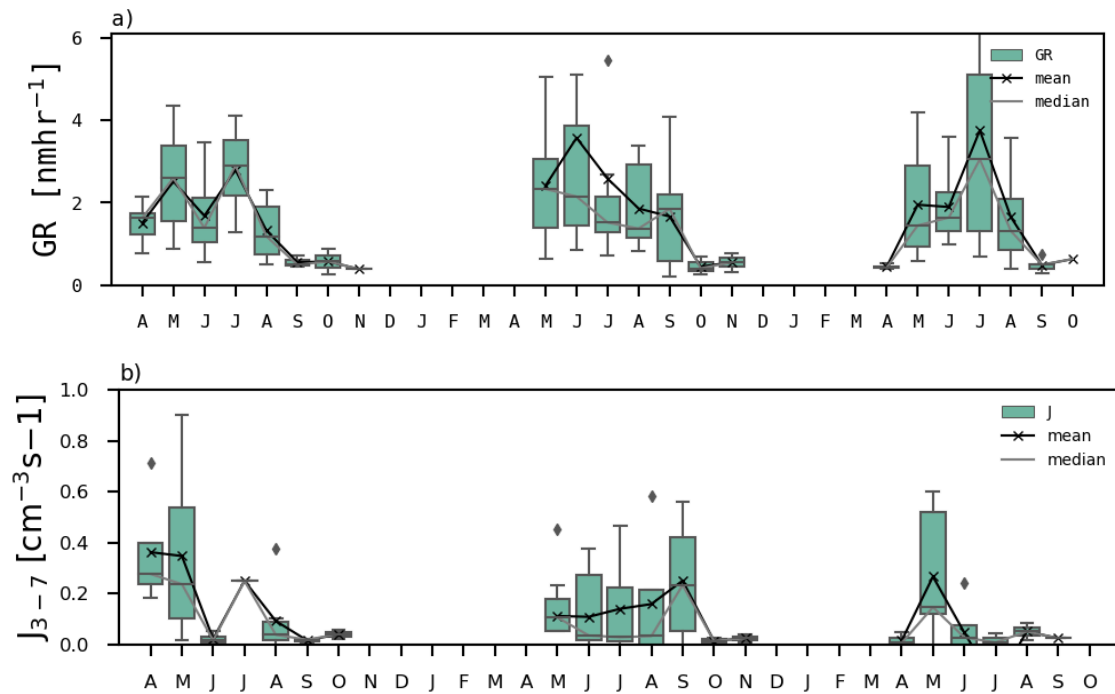


**Fig 3: (a) the duration of the entire growth of newly formed particles from the first measurable size bin i.e. 2.5nm to the largest size bin (b) the average start time of the average new particle formation (NPF) (monthly mean displayed by the cross and median by the red line), along with a shaded region representing the 25<sup>th</sup> and 75<sup>th</sup> percentiles. Along with the estimated time the global horizontal irradiance (GHI) goes beyond zero (i.e. daybreak). The average time of day break for the months April until July is not applicable, since the sun is constantly above the horizon (as experienced by ZEP). Time is given in terms of coordinated universal time (UTC).**

### 3.1.5 Seasonality of Growth Rates and formation rates

The average, mean (median), GRs for all the events is 2.2 (1.5)  $\text{nmhr}^{-1}$ . For class Ia, Ib, and II events the average GRs are 1.4 (1.0), 1.7 (1.2), 2.6 (1.2)  $\text{nmhr}^{-1}$  respectively. The largest GRs occur during July 4.0 (2.1)  $\text{nmhr}^{-1}$ . The polar night events exhibit a significantly reduced GR of 0.43 (0.35)  $\text{nmhr}^{-1}$  (see Fig. 4 a)). Using the *npf-event-analyzer* for the Class Ia events the GRs are 0.9 (1.1), 2.1 (2.6), 4.1 (4.4)  $\text{nmhr}^{-1}$  for 3-7, 3-25 and 7-25 nm diameter ranges.

The GRs presented here agree well with earlier estimates of GRs by Nieminen et al. (2018) reporting a  $\text{GR}_{10-25\text{nm}}$  at ZEP within the range of 1.2 -1.6  $\text{nmhr}^{-1}$ . Kerminen et al. (2018) report a median GR for the Arctic of 2.3 (0.23-4.1, 5<sup>th</sup> and 95<sup>th</sup> percentiles)  $\text{nmhr}^{-1}$ , the site type (i.e. globally) with the smallest GRs.



**Fig 4: a) the calculated growth rates (GRs) b) calculated median formation rates for particles between 3 and 7nm ( $J_{\text{med. } 3-7}$ ) throughout the measurement period are represented by box-whiskers, which display the monthly growth rate averages and include the median and 25<sup>th</sup> - 75<sup>th</sup> percentiles. The green box-whiskers refer to the average GRs/ $J_{3-7}$ s from the start of the NPF until it stops growing. The black line cross represents the mean, and the grey lines join the median values. Diamonds represent outliers.**

517

Formation rates for  $J_{\text{median, } 3-7\text{nm}}$  are  $0.14 \pm 0.23 \text{ cm}^{-3}\text{s}^{-1}$  (mean  $\pm$ std) for all events in which calculations of J and GR could be ascertained (see Fig. 4 b) for the seasonality). Formation rates are generally greater in May with 2023 not exhibiting this maximum in May. The formation rates for  $J_{3-7\text{nm}}$ ,  $J_{3-25\text{nm}}$ ,  $J_{7-25\text{nm}}$  are estimated to be  $0.28 \pm 0.33$ ,  $0.40 \pm 0.35$  and  $0.31 \pm 0.28 \text{ cm}^{-3}\text{s}^{-1}$  (mean  $\pm$ std) for class Ia events (using the max concentration method and *npf-event-analyzer* see Sect. 2.3.3 in the Methods).

Previous studies also estimate  $J$  for various diameters at Ny-Ålesund and ZEP. Beck et al. (2021) reports a  $J_{1.5}$  equal to  $0.27 \text{ cm}^3\text{s}^{-1}$  (Beck et al., 2021). Nieminen et al. (2018) presents the seasonal medians for  $J_{\text{nuc}}$  of nucleation-mode particles (10-25nm) varying from  $0.08 \text{ cm}^3\text{s}^{-1}$  in spring, to  $0.032 \text{ cm}^3\text{s}^{-1}$  in summer and to  $0.0066 \text{ cm}^3\text{s}^{-1}$  in autumn. Lee et al. (2020) gives  $J_{3-7}$ ,  $J_{7-25}$ ,  $J_{3-25}$  values ranging between 0.001-0.54, 0.003-0.5, 0.007-0.61  $\text{cm}^3\text{s}^{-1}$  respectively, and the averages for  $J_{3-7}$ ,  $J_{7-25}$ ,  $J_{3-25}$  are 0.04, 0.09 and 0.12  $\text{cm}^3\text{s}^{-1}$ . Kerminen et al. (2018) report a median Arctic  $J$  (i.e. varying size ranges) of  $0.51 \text{ cm}^3\text{s}^{-1}$ .

In comparison to GRs and  $J$ s at other sites (e.g. Kerminen et al. (2018); Nieminen et al., (2018)), we see that processes within the Arctic, as observed at ZEP, are significantly slower. In urban environments, compared with more remote ones, estimations for  $J$  are significantly larger due to the larger coagulation sink that needs to be overcome (Cai and Jiang, 2017); here, the coagulation sink is much lower than in urban environments.

### 3.1.6 Seasonality of Condensable Vapours

The concentration of condensable vapours ( $C_v$ ) required to sustain the GRs is estimated using equation (3) and the calculated growth rates for the various classes of events (i.e., Class Ia, Ib and II). We show that  $C_v$  increases until July before decreasing during the autumn and into the winter months. It is striking to note that the seasonal variation is similar to that of the sum of the measured concentrations of potential aerosol precursor gases measured in Ny-Ålesund during a 2017 campaign (see Fig. 5). The estimated  $C_v$  is also of a similar magnitude to the combined sum of measured HOMs, sulphuric acid ( $\text{H}_2\text{SO}_4$ ), methanesulphonic acid (MSA) and iodic acid ( $\text{HIO}_3$ ). For Ny-Ålesund, Beck et al. (2021) report increasing IA, SA and MSA concentrations from the start of the sunlight period, and high concentrations prevailing before decreasing around mid-June. HOMs concentrations averaged around  $\sim 1.8 \cdot 10^6 \text{ cm}^{-3}$  during the spring before increasing to  $2.3 \cdot 10^6 \text{ cm}^{-3}$  in June. As a ratio of the total available precursor vapours HOMs account for approximately 0.78 of the entire concentration of precursor vapours after June and prior to June this fraction approximately 0.24 (see Fig. S24). It can therefore be argued that the composition of condensable vapours undergoes a dramatic transformation around the start of June, and the composition of newly formed particles during the initial spring peak is different to that of the subsequent peaks after June (see Fig. 1 & 2). Boyer et al. (2024) measured the concentrations of condensable vapours in the Arctic, however at higher latitudes ( $>80^\circ\text{N}$ ). The estimated  $C_v$  was on average 5.9 times greater than the combined concentrations of the measured precursor gases given in Boyer et al. (2024) (see Fig. S23 in supplement). At higher latitudes ( $>80^\circ\text{N}$ ), there is potentially less photochemistry and biological activity, moreover, the potential sources of precursor gases are more limited (Brean et al., 2023), and may be more

552 influenced by the sea ice cover, further reducing an available and giving rise to the difference between the  
 553 estimated Cv at ZEP and total concentrations during MOSiAC.

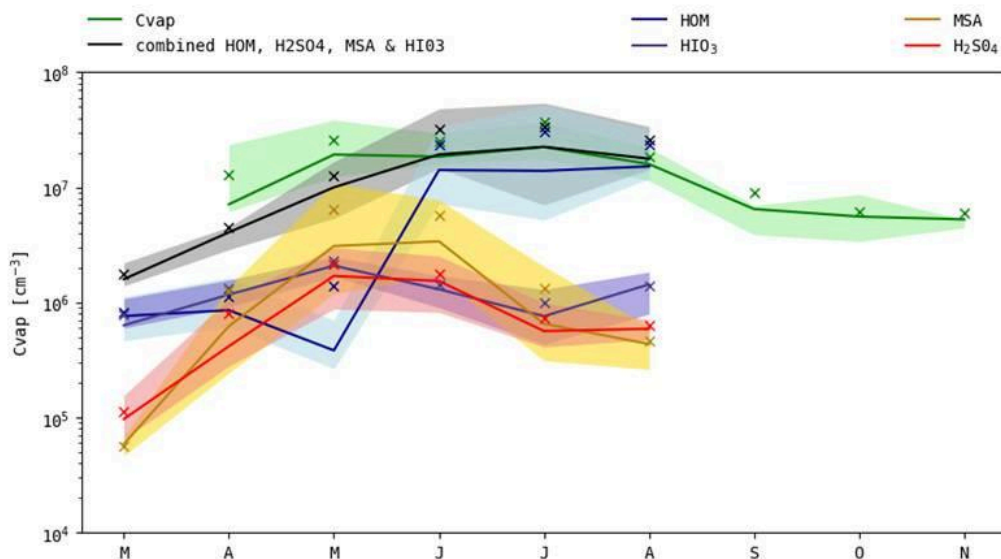


Fig 5: Estimated concentration of condensable vapours (Cv, green) compared with measured concentrations: the monthly means (crosses) and medians (thick line), along with the 25<sup>th</sup> and 75<sup>th</sup> percentiles (shaded region) for both Cv and Ny-Ålesund measurements. Arctic precursor vapours namely sulphuric acid (H<sub>2</sub>SO<sub>4</sub>, red), methanesulphonic acid (MSA, gold) and iodic acid (HIO<sub>3</sub>, purple) and combined (grey) from Beck et al. (2021) are displayed. The estimated Cv measurements cover the years 2022 to 2024, whilst the Svalbard measurements from Beck et al. (2021) cover the period 2017-03-22 to 2017-08-23.

554

### 555 3.2 Positive and negative ions during NPF

556 The polarity of air ions, which aid in nucleation, provides information about the composition of the newly  
 557 formed aerosol particles. Cluster air ions are typically defined between 0.8-1.7nm, whilst intermediate ions range  
 558 from 1.6-7.4nm. Intermediate ions are created in the initial stage of nucleation and are typically formed when  
 559 neutral nanoparticles encounter cluster ions and acquire their charge (Tamm et al., 2014). Negative ions can be  
 560 linked to IA and SA to name a couple of compounds. Unfortunately, due to an instrumental problem related to a  
 561 consistent loss of the negative cluster ions experienced during the measurement campaign (see Sect. S2.6 in the  
 562 supplement) it was not possible to measure the concentration of negative ions less than 2nm.

563 The total number of positive ( $N_{2-4nm}^+$ ) and negative ( $N_{2-4nm}^-$ ) small intermediate ions (2-4nm) within the growing  
 564 mode are calculated for each NPF event. In May, at the start of the NPF season, we observe the smallest fraction  
 565 of positive small intermediate ions compared to the total number of small positive intermediate ions (i.e. the  
 566 mean and median ratio,  $N_{2-4nm}^+ / (N_{2-4nm}^- + N_{2-4nm}^+) \approx 0.2$ ). The low  $N_{2-4nm}^+ / \text{total}$  ratio in May perhaps relates to  
 567 negatively charged molecules dominating the initial stages of ion-induced nucleation (i.e., compared with  
 568 positive ions). Moreover, the low ratio in May could be related to the greater NPF intensity (i.e. larger

production of negative nucleation mode particles in May). However, towards the end of the NPF season (i.e. Sep. - Nov.) there is a shift towards a growing nucleation mode (e.g., 2-4nm) more dominated by positive small intermediate ions, as opposed to negative small intermediate ions (see Fig. 6). The tendency for small intermediate ions (2-4nm) to exhibit a positive charge appears from September onwards, which is also when there begins to be limited solar insolation and the occurrence of polar night events (i.e. late Oct. and Nov.), it is also, and perhaps more importantly, when the composition of condensable vapours changes to being dominated by HOMs (see Sect. 3.1.6.). However, it needs to be noted that there are few events during September and beyond (29 in total). Despite the lack of information about the cluster ions, the shift in the overall charge of the small intermediate ions, from more negative to more positive, potentially hints at a seasonal change in the vapours contributing to the initial formation of critical clusters.

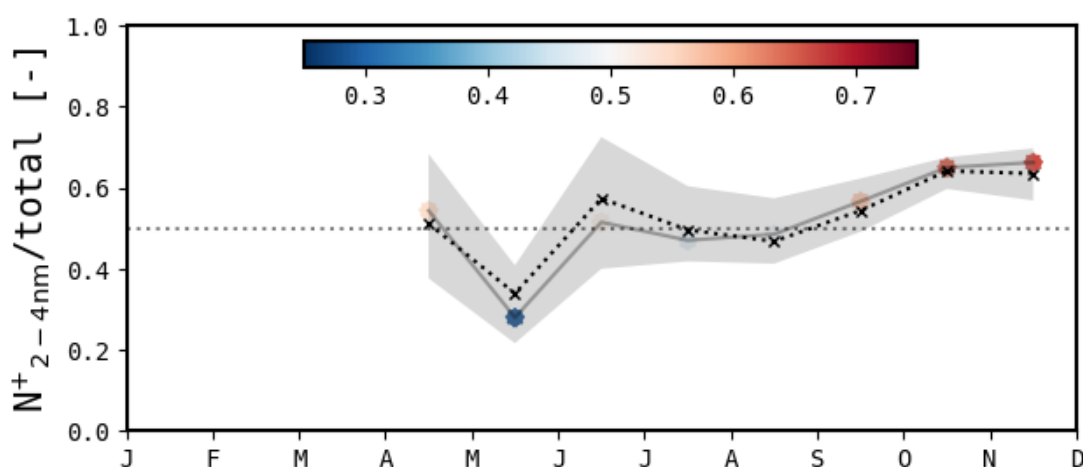


Fig 6: For all new particle formation (NPF) events the monthly ratio of the total number concentration of positively charged small intermediate ions (2-4nm) over the total concentration of both positively and negatively charged small intermediate ions (2-4nm) ( $N_{2-4nm}^+/N_{2-4nm}^- + N_{2-4nm}^+$ ).  $N_{2-4nm}^-$  and  $N_{2-4nm}^+$  includes only ion concentrations within the identified growing mode between 2 and 4nm. The mean monthly  $N_{2-4nm}^+/total$  is presented as a cross, and the median is displayed by coloured circles, blue for an overall negative charge (ratio<0.5) and red for an overall positive charge (ratio>0.5). The grey shading displays the 25<sup>th</sup> – 75<sup>th</sup> percentiles. The dashed black line represents an overall neutral charge of small intermediate ions. It should be noted that there may be some instrumental biases between the two polarities, however, the background concentrations (during no-event periods) display  $N_{2-4nm}^-/N_{2-4nm}^+$  ratios close to unity,  $\sim 1.02$ .

579

As mentioned above, Beck et al. (2021) reports higher concentrations of precursor gases with low proton affinity (e.g., MSA, SA,  $HSO_4^-$ ) during the month of May, while highly oxygenated organic molecules (HOMs) display higher concentrations than SA and MSA during April and from June towards September. Beck et al. (2021) suggest that the events in May were likely initiated by negative ion-induced NPF and relate to sulphuric acid and ammonia nucleation.

Ammonia ( $\text{NH}_4^+$ ) is a possible positively charged cluster ion which could play a role in the shift in polarity (Kirkby et al., 2016), however, it needs to be stressed that further work is needed to ascertain what is the reason for this shift in the polarity. We were unable to compare the overall polarity charges for small intermediate ions with concentrations of  $\text{NH}_4^+$ . However, the shift from more negative to more positive small intermediate ions seems to coincide with the increase in concentrations of HOMs relative to the total concentration of other condensable vapours as also indicated above (see Fig. S24).

### 3.3 Source regions and geo-spatial extent

NPF events are assigned to various marine regions using the method described in Sect. 2.4.5. Air masses that traverse over the Arctic Ocean coincide with the highest number of NPF events (57 in total or 37% of NPF events; see Table 1). 37% of air masses are classified as originating from the Arctic Ocean, the largest marine region in terms of contribution. The air masses that mainly originate over the Greenland Sea result in 29 NPF events and show the highest probability of an NPF event occurring, with 27% of the Greenland Sea air masses being linked to an NPF day. Air masses originating over the Arctic Ocean and Barents Sea have a 19% and 13% likelihood, respectively.

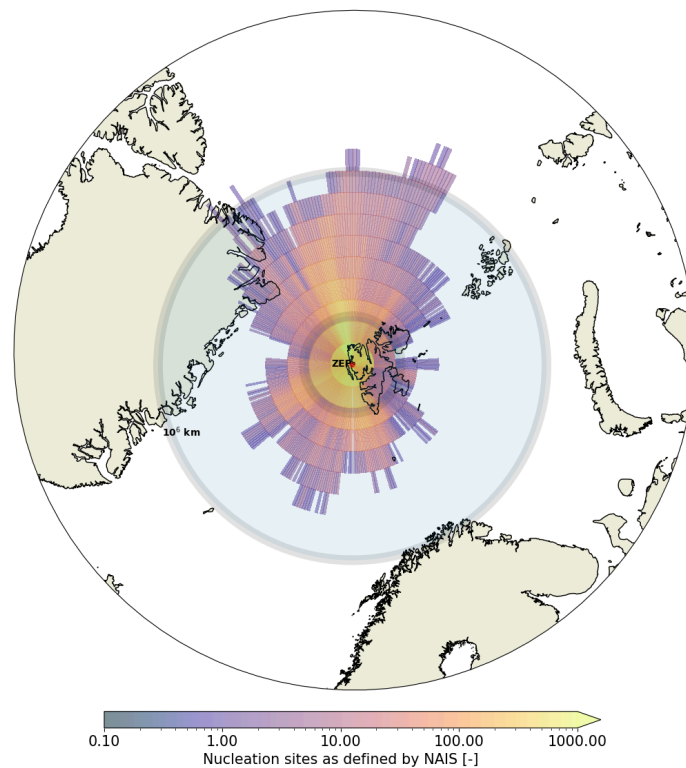
**Table 2: The frequency of occurrence of air masses from different sectors compared with the number of events. Both numbers are given in days, followed by a ratio of the two (i.e. Likelihood). Although not shown in table, back trajectories traversed over the Kara Sea on two occasions. The average CS, solar insolation (both mixed-layer and all), maximum daily increase in 2.8 – 5nm particles ( $\Delta N_{\text{max}, 2.8-5}$ ) and proportion of back trajectory within the mixed layer are given for each marine sector. The averages for these variables are also given during the NPF days and presented in brackets.**

| Air mass      | Region's frequency (days) | Number of NPF events (days) | Likelihood | CS [ $\text{s}^{-1}$ ] | Solar insolation [ $\text{h W m}^{-2}$ ] | $\Delta N_{\text{max}, 2.8-5}$ [ $\text{cm}^{-3}$ ] | Proportion in ML |
|---------------|---------------------------|-----------------------------|------------|------------------------|------------------------------------------|-----------------------------------------------------|------------------|
| Arctic Ocean  | 313                       | 57                          | 0.19       | 0.20 (0.19)            | 185 (378)<br>368 (753)                   | 239 (776)                                           | 0.63 (0.60)      |
| Greenland Sea | 109                       | 29                          | 0.27       | 0.25 (0.25)            | 327 (442)<br>666 (991)                   | 254 (665)                                           | 0.60 (0.54)      |
| Barents Sea   | 69                        | 9                           | 0.13       | 0.36 (0.29)            | 118 (211)<br>597 (1100)                  | 93 (204)                                            | 0.32 (0.28)      |
| Norwegian Sea | 44                        | 7                           | 0.16       | 0.25 (0.43)            | 232 (418)<br>618 (1330)                  | 136 (382)                                           | 0.48 (0.34)      |
| Mixed         | 303                       | 51                          | 0.17       | 0.23                   | 184                                      | 147                                                 | 0.54 (0.50)      |
| Total         | 840                       | 153                         | 0.18       | 0.23                   | 200                                      | 190                                                 | 0.57 (0.53)      |

604

The analysis is expanded, using the method outlined in Sect. 2.4.4, by estimating the location of the formation of newly formed particles. The most common location for nucleation to occur is off the western coast of Svalbard, i.e., in the Greenland Sea and Arctic Ocean (see Fig. 7). Furthermore, the Greenland Sea, south-west of ZEP, as

608 opposed to the Barents Sea, is home to more nucleation sites. It is also clear that the vast majority of nucleation  
609 sites are estimated to occur over the ocean. The estimated nucleation sites are almost all within a  $1 \cdot 10^6$  km radius  
610 of ZEP, and the vast majority are fairly close to ZEP (see Fig. 7), a finding which can also be inferred from the  
611 observation that the majority of events begin growing from 2.5nm. Furthermore, from the air mass back  
612 trajectory analysis, we gather that air masses typically reside between sea level and the altitude of ZEP (i.e.  
613 0-474m, see Fig. S20). There is little difference between the altitudes of the arriving air masses for the NPF  
614 events compared to all days.



**Fig 7: Total number of estimated nucleation sites per grid cell. The unit is based on the number of back trajectories initialised and does not reflect a physical value. Sites of nucleation for class Ia, Ib and II events are estimated by using HYSPLIT to trace the likely origin of particles produced during NPF events, and initialising the back trajectories based on the duration of the NPF event (see Sect. 2.4.4).**

615

616 DMS emissions are closely linked to phytoplankton dynamics and abundance, and typically occur following  
617 phytoplankton biomass maxima (Galí and Simão, 2015). Numerous studies have tried to link chlorophyll a, which  
618 is used as a proxy for phytoplankton biomass, with DMS emissions to the atmosphere (Jang et al., 2019). Park  
619 et al. (2018) note a strong correlation between DMS concentrations measured at ZEP and air masses traversing  
620 over the surrounding ocean with high chlorophyll a. They find that the relationship between DMS concentrations  
621 and air masses likely exposed to high DMS emissions are stronger for air masses traversing over the Greenland  
622 Sea, compared with the Barents Sea. Therefore, suggesting that the Greenland Sea has a DMS production

capacity three times greater than the Barents Sea. By replicating the method detailed in Park et al. (2018), we also find good correlations between chlorophyll a exposure and DMS, however, not for all years, and not for the years related to this study (i.e. 2022 and 2023). In the study by Park et al. (2018) the years 2010, 2014 and 2015 were included, however, 2022 and 2023 did not yield such strong correlations. We were unable to replicate the findings presented in Park et al. (2018) for the years 2022 and 2023 (see Fig. S30-37), however, we could replicate the findings for 2010 (see Fig. S30). We can only speculate about the reasons for the interannual variation between the relationship between chlorophyll a and DMS. We argue that because of the large spatial and temporal interannual variability, possible geo-spatial changes in the DMSP-rich phytoplankton species in the neighbouring seas, a wide range of DMS oxidation timescales depending on photochemistry and available solar insolation (Ghahreman et al., 2019) can all potentially influence this relationship. Furthermore, the influence of various other meteorological and environment factors that affect DMS oxidation and the lifetime of precursor gases, and sea ice and cloud cover could lead to inaccuracies in HYSPLIT outputs and chlorophyll a satellite retrievals. In addition, direct correlations between atmospheric DMS and chlorophyll a exposure are not detected around Iceland, however, a high atmospheric DMS to chlorophyll a exposure coincides with areas containing DMSP-rich phytoplankton species (Lee et al., 2023).

The Greenland Sea is connected to a higher likelihood of NPF events; compared with the Barents Sea there is a distinct difference between both seas in terms of the proportion of air masses coinciding with NPF events, and the number of nucleation sites estimated to arise from each sea (see Fig. 7). The combined effect of a larger number of air masses coming from the west and an increased NPF-likelihood linked to western air masses means that we observe more NPF events off the western edge of Svalbard. This difference could be linked to the finding that the DMS production capacity in the Greenland Sea is estimated to be three times greater than that of the Barents Sea, due to more DMSP-containing phytoplankton species (e.g. *Phaeocystis*) (Park et al., 2018). However, another distinct difference between these two source regions is that air masses, which traversed over the Greenland Sea, experienced more solar insolation and a relatively lower CS average, compared to the air masses coming over the Barents Sea (see Table 1). The combination of both high solar insolation and relatively low CS, could also explain why NPF likelihood is higher for the Greenland Sea as opposed to Barents Sea. One further difference is that the Barents Sea has a much larger sea ice coverage compared with the Greenland Sea, and thus there is potentially less open ocean in contact with the lower atmosphere during the NPF season. The reduced availability of DMS given the greater sea ice coverage could additionally help explain the differences in NPF likelihood of the Greenland and Barents Seas. The Arctic Ocean is another NPF source region (see Fig. 7

and Table 1.). Considering that the Arctic Ocean sector is considerably more covered by sea ice than the NPF precursors are potentially being emitted from sea ice, e.g. iodine and sympagic algae. The Arctic Ocean contributes significantly, in terms of the portion of arriving air masses, between March to May and September to October (see Fig. S22).  $\text{HIO}_3$  and HOMs are both significant contributors prior to May and HOMs increase in concentration after June.

#### 4.1 Predicting NPF likelihood with condensation sink and solar radiation

In Sect. 3.1.2, we show that NPF events and increases in NPF intensity, typically occur during periods of high solar insolation. High solar insolation seems to be a prerequisite for NPF, as it leads to the photochemical production of nucleating compounds. NPF events also seem to require the total surface area of pre-existing aerosol particles (i.e., CS) to decrease below seasonal averages (see Fig. 2). The distribution of solar insolation is statistically significantly different for periods experiencing events as opposed to non-events; or CS it depends on the classification scheme used (as demonstrated using the Mann-Whitney U test, see Fig. S29 a) – b) in supplement). NPF events require the concentration of nucleating vapours to exceed a critical concentration, and as the CS acts as a sink of those highly condensable vapours, low CS values promote the potential for nucleation. In this section, we argue that solar insolation and CS can be used as the two central parameters to determine with high certainty the likelihood that a given day will experience an NPF event. For the definition of the term solar insolation refer to Sect. 2.4.1 in the methods.

The majority of measurements feature the combination of both relatively low solar insolation and low CS ( $<1000 \text{ hW m}^{-2}$  and  $<0.5 \cdot 10^{-3} \text{ s}^{-1}$  see bottom-left of Fig. S25 b) in supplement)), and on very few occasions there are measurements with a combination of high solar insolation ( $>1000 \text{ hW m}^{-2}$ ) and low CS ( $<0.5 \cdot 10^{-3}$ ) (see top-left of Fig. S25 b) ). Nonetheless, we show that when the combination of high solar insolation ( $>1000 \text{ hW m}^{-2}$ ) and relatively low CS ( $<0.5 \cdot 10^{-3} \text{ s}^{-1}$  (i.e. top-left of Fig. S25 a) & b) ) arises, an NPF event that day is likely to occur (as defined by Aliaga et al. (2023), see Fig. 8). We find that periods exhibiting high solar insolation ( $>1000 \text{ hW m}^{-2}$ ) and relatively low CS ( $<0.5 \cdot 10^{-3} \text{ s}^{-1}$ ) are linked to more NPF days. Overall, the combinations of CS and solar insolation which promote NPF are relatively rare, compared to the entire set of measurements. The probability of occurrence is derived from the ratio of the number of measurements for each combination of CS and solar insolation during an NPF day to the number of valid data points for each of these combinations (where both sets of data are composed of hourly measurements) (see Sect. S10.1 in the supplement for more details on the normalisation approach)).

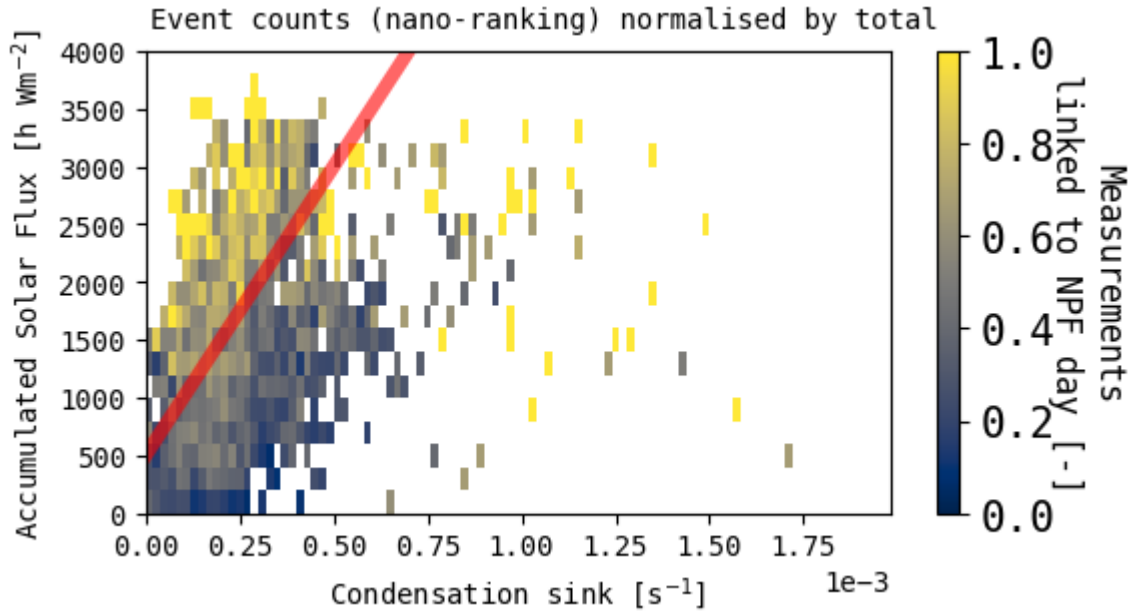


Fig. 8: Grid mesh where the data is binned by accumulated solar insolation and condensation sink (CS) (CS between  $0 - 10^{-3} \text{ s}^{-1}$  every  $2 \cdot 10^{-5} \text{ s}^{-1}$  and solar insolation between  $0 - 4 \cdot 10^4 \text{ h W m}^{-2}$  every  $200 \text{ h W m}^{-2}$ ). Each grid box displays the likelihood, calculated by normalising the data set for the event days (defined using Aliaga et al. (2023)) by the total count for each given bin (i.e. see Fig. S25 a) and b) in supplement)). Likelihood is the fraction of measurement combinations linked to an NPF day. The red line defines the region of interest (ROI) whereby data on the left of it is considered to have the ideal conditions for NPF i.e. high accumulated solar flux and relatively low CS. The equation for the red line is:  $\text{Solar insolation} = 5 \cdot 10^6 \cdot \text{CS} + 500$ . The grids required a count greater than 1.

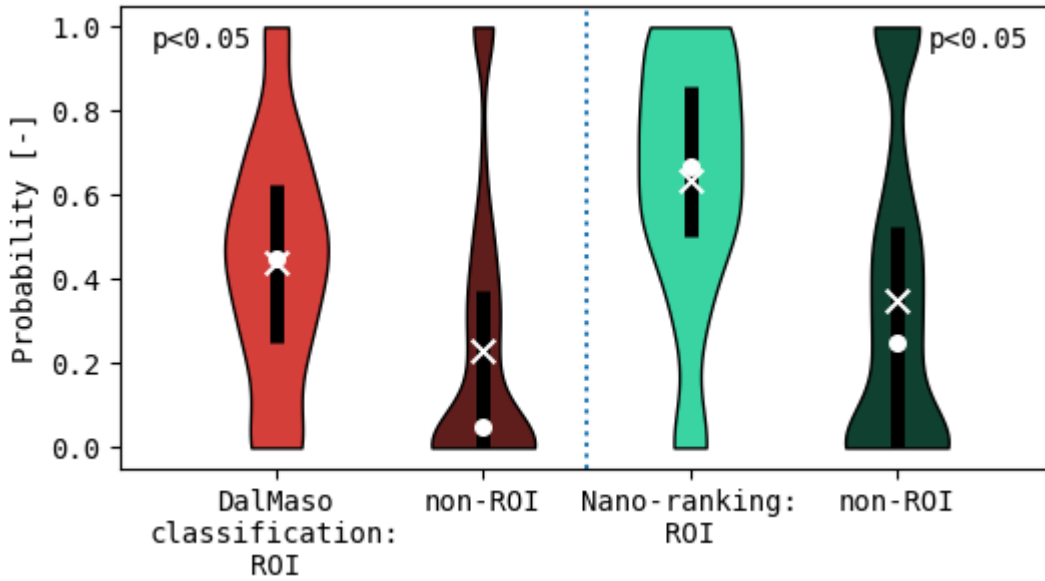
682

683 We define a region of interest (ROI) (represented using the red line, separating the top left-hand corner in figure  
 684 8). The ROI is defined using a simple straight line to separate ideal and non-ideal NPF conditions from each  
 685 other. It should be stressed that it should not be used as an empirical relationship, defining the occurrence of  
 686 NPF, instead it should be used as a guideline to highlight the general conditions most suitable for the promotion  
 687 of NPF. In the ROI, the combination of low CS and plentiful solar insolation provide the most favourable  
 688 conditions for NPF days to occur, and on the contrary, outside of the ROI, NPF days are much less likely to  
 689 occur (i.e. bottom-right of Fig. 8).

690 Moreover, the region outside of the ROI with low solar insolation and low CS might be associated with  
 691 increased cloudiness as the incident radiation is reduced by the presence of clouds, and large accumulation mode  
 692 particles are reduced due to the effect of wet scavenging. The region with low solar insolation and high CS  
 693 could be related to Arctic Haze conditions.

694 The ROI defined in Figure 8 is statistically significantly different from the non-ROI region on the probability  
 695 that an NPF day occurs given such solar insolation and CS measurements occurring; the probabilities which arise  
 696 when normalisation is done via the nanoranking classification scheme, as opposed to the Dal Maso et al. (2005)  
 697 scheme, are more distinct between the ROI and non-ROI regions (see Fig. 9). Here, the statistical significance

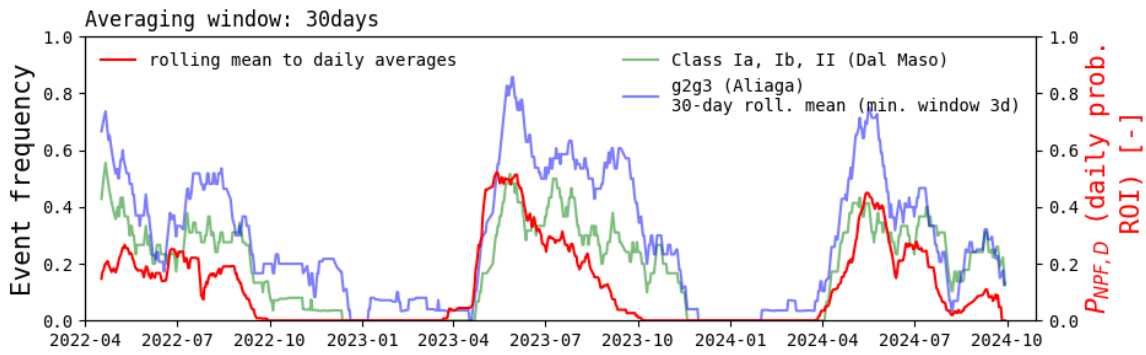
698 between the two regions is conducted using the non-parametric Mann-Whitney U test (where  $H_0$  representing  
699 two identical distributions is rejected). In addition, the ROI and non-ROI represented 23% and 77% of the data  
700 respectively; for the ROI 62.5% of the hourly data were linked to NPF days, as opposed to 23.8% in the non-ROI  
701 (The total number of Nano<sub>events</sub> is 229 (g2=178, g3=51, see Fig.S16 in supplement)).



**Fig 9: Violin plots representing the distribution of the probabilities as defined by the normalisation plot (see Fig. 8). The red violins on the left-hand-side represent the probability distributions related to the Dal Maso et al. (2005) classification and the green violin on the right-hand side represent the probability distributions according to the Nano-ranking approach.**

703 The data points that lie within the ROI and when they occur can be used to try and estimate the likelihood of a  
704 day experiencing an NPF event. A time series representing the daily likelihood of NPF occurrence (i.e.  $P_{NPF, D}$ )  
705 can be generated from the ROI data (see Fig. 10) by labelling hourly data points as either 1 (i.e. within the ROI)  
706 or 0 (i.e. outside of the ROI) and then taking daily averages before applying a rolling average to create the daily  
707 likelihood,  $P_{NPF, D}$ .  $P_{NPF, D}$  generally, matches the frequency of NPF events as defined by Aliaga et al. (2023) or  
708 Dal Maso et al. (2005) (see Fig. 10), however, there are some major discrepancies. Generally, the predicted  
709 frequency of occurrence (i.e., defined by the ROI) is significantly lower than the observations (can also be  
710 observed in the correlation figure Fig. S28 in the supplement). The spring and autumn peaks in both  $P_{NPF, D}$  and  
711 the event frequencies generally line up well in terms of the number of peaks, however, 2023 displays only one  
712 main peak in  $P_{NPF, D}$ .  $P_{NPF, D}$  nicely replicates the distinctive dips in the NPF frequency before July in the years  
713 2022 and 2024. However, in July 2023, the dip in NPF events in the observations is followed by an increase (i.e.  
714 two more sequential peaks in the observations) which is not captured by  $P_{NPF, D}$ . Still, it is striking that the two

parameters alone (CS and solar insolation) are able to serve as predictors for NPF probability, by representing the balance between the production and removal of NPF-precursors. The solar insolation reflects the photochemical production potential (likely via the production of hydroxyl radicals). However, the parameter combination does not account for the reactants required to produce the nucleating vapours (which may be SO<sub>2</sub> (likely via DMS) and/or iodine compounds). Hence, deviations like those observed during July 2023 are expected as the conceptual model is partly incomplete due to a lack of adequate information regarding the original reactants. This said, however, the NPF frequency and likelihood largely seem limited by the balance between photochemical production potential and CS, and this in turn is suggestive of, to some degree, a self-regulating mechanism regarding the number of particles present in the nucleation and Aitken modes. There exist more sophisticated models to predict NPF (Kuang et al., 2010).

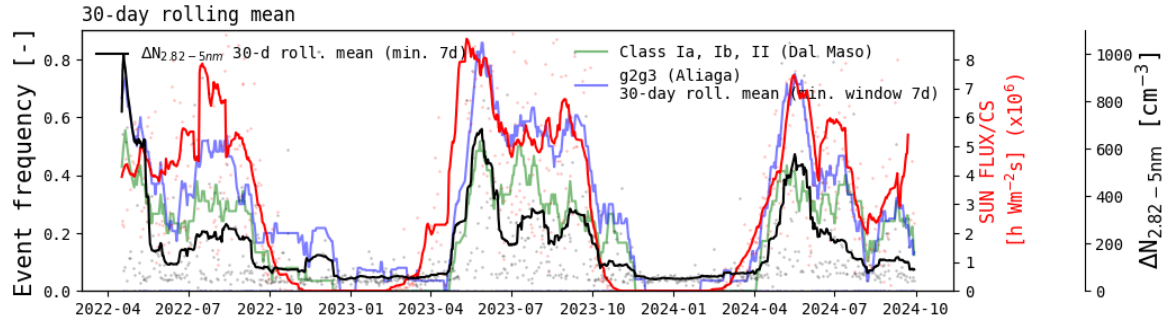


**Fig 10:** The red line is  $P_{NPF, D}$ , the estimated daily likelihood of measurements occurring in the region of interest (ROI). The ROI is based on the relation between CS and 6-hour solar accumulation and is defined in Fig. 8). The green and blue lines represent the rolling averages of the NPF daily event frequencies using the Dal Maso et al. (2005) and Aliaga et al. (2023) classification respectively. The green and blue curves are present here to be able to compare to  $P_{NPF, D}$  (daily prob. ROI).

725

#### 726 4.1.1 Ratio of source and sink: ratio of solar irradiance

727 In section 4.1 we defined a region of interest, whereby measurements within this ROI provide an indication of  
 728 the conditions likely to lead to an NPF day. Observing when these conditions occur offers one approach,  
 729 however, the ability to predict the occurrence of NPF days can be improved by simply using the proxies for a  
 730 source and a sink, and taking their respective ratio (i.e., solar insolation/CS) (see Fig. 11). For a rolling average  
 731 window of 30 days the  $R^2$ , comparing solar insolation/CS and the nano-ranking frequency, is 0.78 (see Fig. S26  
 732 in supplement).



**Fig 11:** The red line is the solar insolation divided by the condensation sink (Solar insolation/CS) with a 30-day rolling mean. The intensity parameter ( $\Delta N_{2.8-5nm}$ ). The green and blue lines represent the rolling averages of the NPF daily event frequencies using the Dal Maso et al. (2005) and Aliaga et al. (2023) classification respectively. The green and blue curves are present to be able to compare to the red line representing a proxy for the balance between sources and sinks.

733

734 Figure 11 demonstrates that by using a simplified parameter i.e., solar insolation/CS, albeit with a large rolling  
 735 averaging window, we achieve a good agreement with both event frequencies (i.e., classified using the Dal Maso  
 736 et al. (2005) or Aliaga et al. (2023) approach). There are some slight discrepancies between the solar  
 737 insolation/CS parameter and the event frequencies, particularly at the start and end of the measurement period;  
 738 the discrepancies could be due to the NPF seasons having started already or not having ended. The relationship  
 739 between Solar insolation/CS and event frequency varies depending on the averaging window utilised, whether  
 740 the ML is selected and also the type of classification used (see Fig. 12). The coefficient of determination ( $R^2$ )  
 741 comparing solar insolation/CS and the daily event frequency (i.e., rolling average applied) increases and begins  
 742 to plateau after a window of 2 weeks is applied (14 days,  $R^2=0.65$ ) (see Fig. 12). Considering that a degree of  
 743 averaging needs to be performed as the daily event frequencies are given as 1s (event) and 0s (non-event), then it  
 744 seems justified that the solar insolation/CS parameter offers a robust predictive capability once a modest amount  
 745 of smoothing is performed.

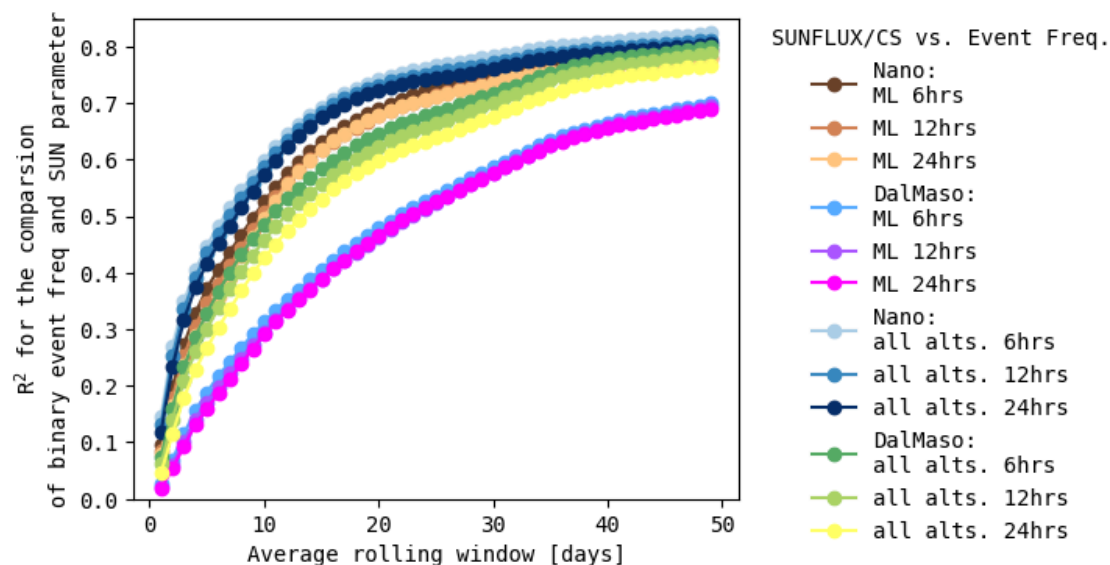
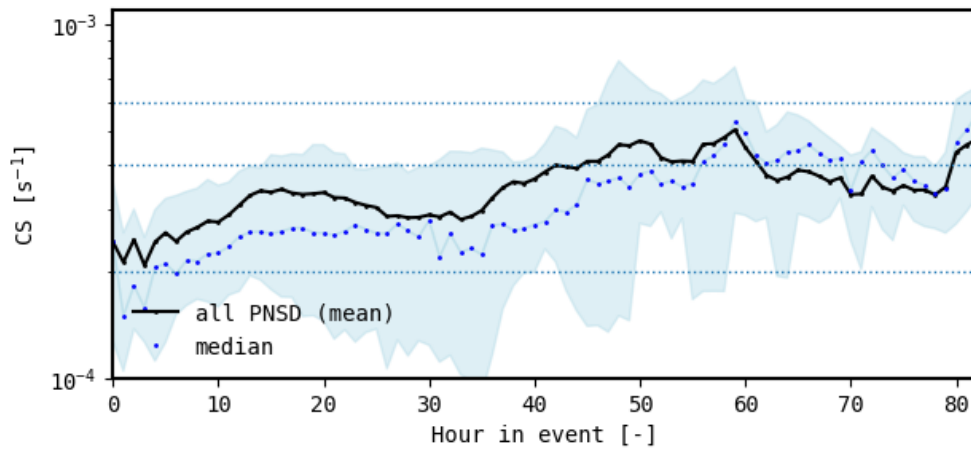


Fig. 12: Correlation between the event frequency (either defined using the Dal Maso or Nano ranking method) and the accumulated solar insolation (SUN parameter) with a rolling average applied. The coefficient of determination ( $R^2$ ) is shown as a function of the rolling window size (number of days). The first 6, 12, and 24 hours of the back trajectory are selected (shown in slightly different shades). The curve labelled ML represents back trajectories that traversed only through the mixed layer, while the curves labelled all alts. represent back trajectories at all altitudes.

746

## 747 4.2 Dynamics driving NPF and growth

748 During NPF events there is an excess availability of condensable precursors, e.g. SA, which contribute to the  
749 growth of newly formed and pre-existing particles, thus increasing CS. The increased CS enhances the removal  
750 of condensable and nucleating vapours. As a result, a typical day with sustained NPF can lead to a relatively  
751 high CS ( $>0.5 \cdot 10^{-3} \text{ s}^{-1}$ ), although the events tend to begin at low CS values ( $\sim 0.1 \cdot 10^{-3} \text{ s}^{-1}$ ) (see Fig. 13). It is  
752 worth noting, that during an event, not only do the nucleation mode particles grow in size, but also all other  
753 existing particles grow as well. Practically, this means that the sink is doubled during the NPF-event.  
754 Consequently, and assuming steady state, this would mean that the source rate of the condensing species must  
755 double to sustain the same concentration of condensing material.

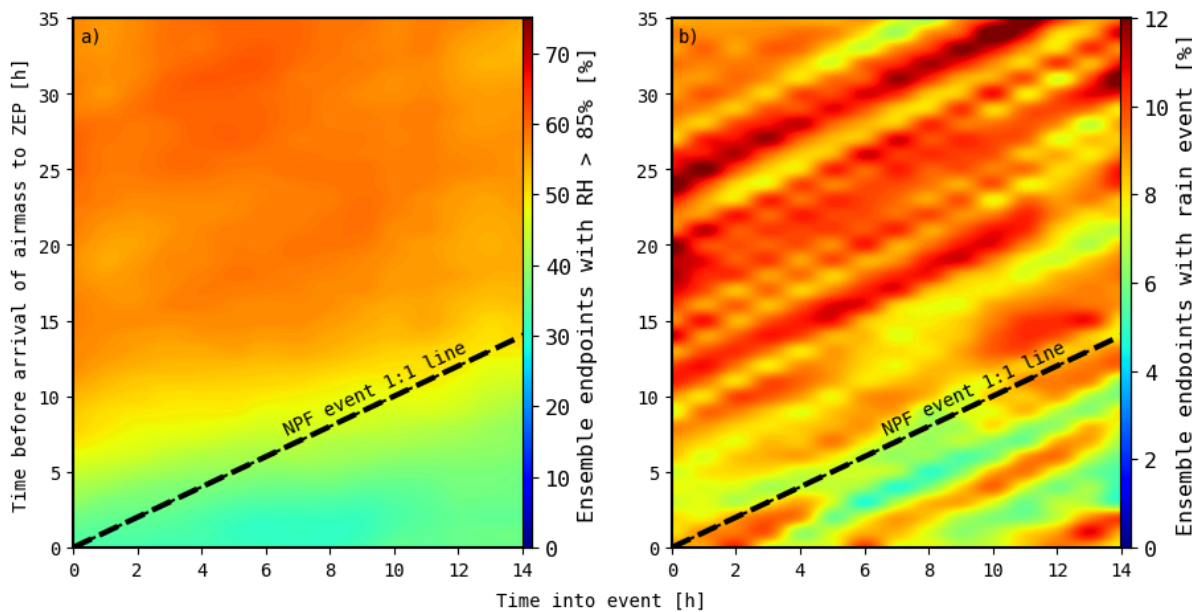


**Fig. 13: The calculated condensation sink (CS) during NPF events. It should be noted that the average NPF typically lasts for 10 hours; however, in some cases events can persist for days until the newly formed mode is removed via wet scavenging or advected away from ZEP. The black line is the mean and the blue is the median. The blue shaded region represents 25<sup>th</sup>-75<sup>th</sup> percentiles. The dashed blue lines display CS values of 0.1, 0.2 and 0.4 • 10<sup>-3</sup> s<sup>-1</sup> and are there to guide the reader.**

756

757 The effect of this additional increase in CS during NPF events can be illustrated by using the ROI. The increase  
 758 in CS shifts the ambient conditions away from the defined ROI ( $>0.5 \cdot 10^{-3} \text{ s}^{-1}$ ) (see Fig. 8), thus potentially  
 759 suppressing any subsequent NPF events. This mechanism will prevent further NPF, if the increase in CS during  
 760 the NPF event is large enough to limit the availability of precursors. This mechanism also helps to explain why  
 761 the combination of low CS and high solar insolation measurements are seemingly rare occurrences, as these  
 762 conditions are soon followed by NPF, and increasing CS.

763 During the Arctic Haze period in late winter and early spring, high concentrations of accumulation mode aerosol  
 764 dominate the contribution to the CS. Reduction in accumulation mode aerosol at the end of the Arctic Haze  
 765 season (i.e., end of May) should result in decreased CS, but ZEP continues to experience relatively high CS. We  
 766 argue that this relatively high sustained CS is the result of particle formation and growth within the Arctic; the  
 767 increasing frequency of NPF events during the summer months increases CS, and maintains the comparatively  
 768 high CS throughout the summer and the most intense part of the NPF season (see Fig. 2b)), additionally Tunved  
 769 et al. (2013)). Towards the late summer and autumn, the solar insolation has decreased substantially, and both  
 770 marine surface water productivity and photochemical production potential are declining. Together with low  
 771 transport efficiency from anthropogenic sources at lower latitudes and efficient wet removal, the aerosol reaches  
 772 a minimum with respect to CS and number during this part of the year.



**Fig 14: Air mass history for a) percentage of back trajectory endpoints within an ensemble with a relative humidity above 85, and b) percentage of endpoints within ensemble which experienced a rainfall event (i.e. rainfall > 0mm). Y-axis is the number of hours backwards before the air masses arrive at ZEP, while the x-axis is the number of hours into the event e.g. if one event started at 6am and another at 9am, their start times would both be 0 hours. The black line is a 1:1 line which represents the start of the NPF event such that the grid points towards the top-left represent endpoints prior to the NPF event, whilst grid points towards the bottom-right represent endpoints during the NPF event. The x-axis has been sliced based on the typical duration of events (most events are <14 hours). This figure consists of the air mass history of Class Ia and Ib NPF events.**

773

774 In the Arctic environment, wet removal is a key factor responsible for the rapid reduction in CS (Garrett et al.,  
 775 2011). For class Ia and Ib NPF events, we can observe that prior to the start of an NPF event there is an  
 776 increased likelihood of cloudy conditions ( $RH > 85\%$ ) and rainfall events (see Figs 14 a-b). Figures 14 a-b  
 777 demonstrate how wet scavenging helps to effectively reduce the CS and provide more suitable conditions for  
 778 NPF events to occur. The changes in the likelihood of cloudy conditions and rainfall prior to and post the start of  
 779 an NPF event is slight, but significantly different, especially considering the duration prior to the event is longer  
 780 and the likelihood of any rainfall should be compounded.

781 Any reduction in CS will shift the system towards the ROI, and as a result the system will rapidly respond to this  
 782 reduction by producing new particles and growing the already existing ones. If suitable gaseous precursors and  
 783 solar insolation are present, NPF will take place and the cycle will be reinitialised. This conceptual model, whilst  
 784 highly simplified, offers substantial predictive value regarding the timing and frequency of NPF events (see Fig.  
 785 10). It does not, however, account for the original source of condensable and nucleating species nor does it  
 786 consider cloud activation and the in-cloud chemistry of Aitken mode particles in non-precipitating clouds.

### 787 4.3 NPF contribution to Aitken and potential CCN

788 Here, we argue that new particle formation is the main contributor to the Aitken mode in the Arctic; through  
789 frequent NPF events and the subsequent growth of nucleation mode particles, NPF activity is able to provide a  
790 significant proportion of the overall concentration of Aitken mode particles. We show in Sect 3.1.2 that at ZEP  
791 the production of nucleation mode particles is largest at the end of spring, however, the production of newly  
792 formed particles persists throughout the summer months and into the autumn (see Fig. 2a)). During NPF events,  
793 including formation and subsequent growth the total number concentration of Aitken mode particles (5-70nm) is  
794 3.6 times the amount compared with pre-event conditions (see Fig. 16). These events are relatively frequent (30  
795 - 40% of summer days experience NPF, (see Fig. 1). Fundamentally, though, it is difficult to use in-situ  
796 measurements at a single location to provide an estimation of the fraction of Aitken mode particles which are  
797 produced via NPF; for the Aitken mode particles at ZEP which are not directly linked to NPF activity, we cannot  
798 rule out the possibility that they are derived from NPF events outside of the vicinity of ZEP and then transported  
799 downwind.

800 At ZEP, NPF and the subsequent growth that follows, results in particles large enough to potentially be activated  
801 and become CCN; on numerous occasions (57 times) we observed newly formed particles able to grow beyond  
802 25nm while in the same regional event (see Fig. 15b), before being scavenged. Here, we use 25nm as the lowest  
803 bound for particles to act as CCN, however, it should be stated that this limit requires strong updrafts and  
804 extremely hygroscopic particles for particles to be activated. We observe that after ~30 hours of growth, we  
805 reach potential CCN-sizes at ZEP (see Fig. 15a)). In addition, on 11, 8 and 3 respective occasions  
806 NPF-produced particles grew beyond 60, 70 and 80nm were observed (see Fig. 15b)). It is important to note that  
807 once produced these particles most likely continue to grow downwind of ZEP by condensation and in-cloud  
808 processing, so a large fraction of the Aitken mode should inevitably reach CCN size in due time, as long as they  
809 are not otherwise scavenged.

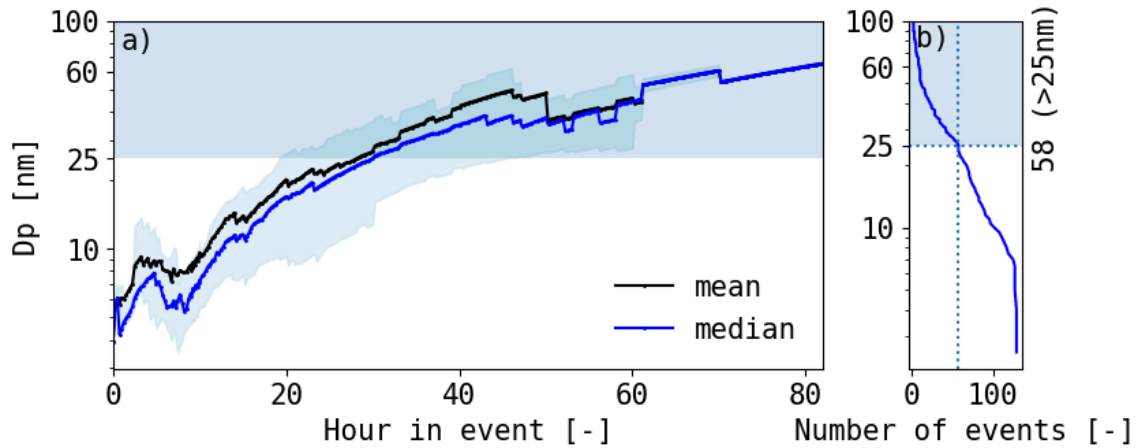


Fig 15: Change in diameter during all class Ia, Ib and II NPF events. The blue shaded region covers 25nm – 60nm. (a) shows the average change in the diameter of the nucleation mode over time. The black and blue lines signify the mean and median diameters respectively. The blue shaded region displays the distance between the 25<sup>th</sup> and 75<sup>th</sup> percentiles respectively (b) displays the number of NPF events against the maximum diameter reached during growth. The horizontal dashed line marks 25nm whilst the vertical dashed line marks the number of NPF events which reached 25nm.

810

811 It is difficult to ascertain the proportion of NPF-derived Aitken mode particles that actually become CCN before  
812 being scavenged or deposited, and estimating an exact fraction is beyond the scope of this study. However,  
813 previous modelling and measurement studies can help to shed some light on the likelihood that NPF events as  
814 measured from ZEP can contribute to the CCN budget. Jung et al. (2018) demonstrate that at supersaturations  
815 (ss) greater than 0.4% there is a considerable number of CCN measured during the summer months at ZEP.  
816 Moreover, the changes in sources of CCN-sized aerosol particles, from mainly anthropogenic to NPF-derived,  
817 during the Haze to summer transition can be seen in the reduction of the geometric mean diameter. For summer,  
818 Jung et al. (2018) suggest a significant number of particles >10nm are activated into cloud droplets, as the  
819 activation ratio during summer is around 0.5 for fairly high ss, i.e. >0.6%. In addition, Karlsson et al. (2021)  
820 show that Aitken mode-sized cloud residuals are fairly common in the summer time (i.e. July onwards); the  
821 transition from accumulation mode-dominated to Aitken mode-dominated aerosol did not lead to a significant  
822 reduction in the concentration of cloud residuals, indicating that particles as small as 20nm can be activated.  
823 Motos et al. (2023) suggest that particles >20nm are activated in the summer, with a modal activation diameter  
824 of 60nm showing that Aitken mode particles act as CCN.

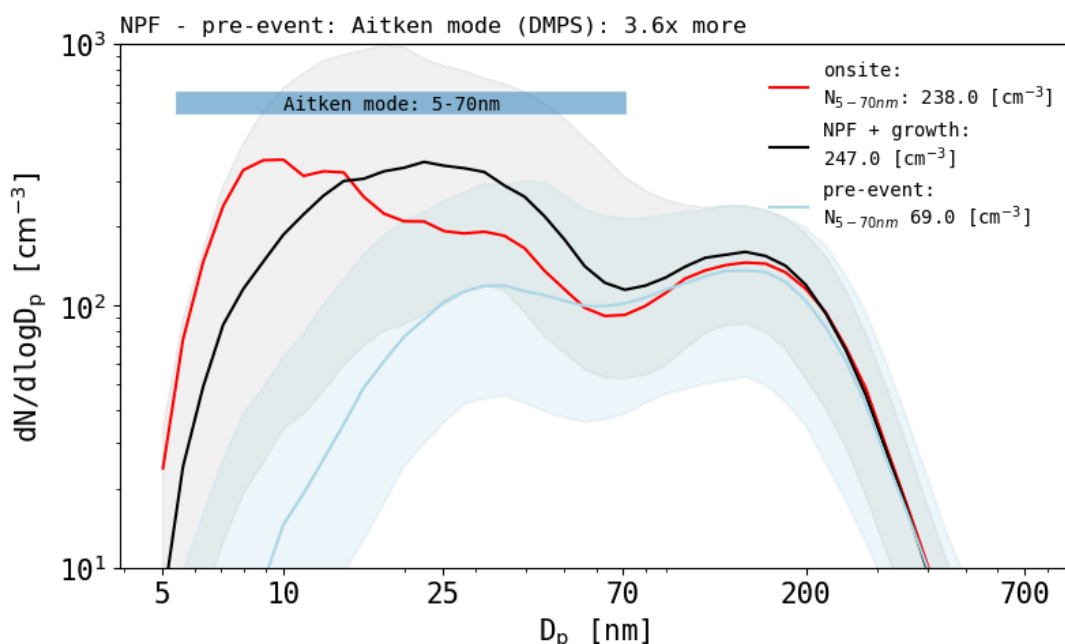


Fig 16: The particle number size distributions (DMPS) for periods before the NPF events (1-2 hours before event started) (pre-event, light blue), during NPF and growth (black) and on-site nucleation (production of 2-4nm particles, red, *on-site formation*).

825

## 826 5. Conclusion

827 In this study, we explore the annual cycle of atmospheric new particle formation (NPF) events from almost  
828 3-years' worth of measurements (2022-2024). In keeping with previous studies, e.g., Tunved et al. (2013) and  
829 Ström et al. (2009), the Arctic summer and more specifically the end of spring and start of summer represent a  
830 peak in NPF intensity as observed at Zeppelin Observatory (ZEP). We find that this peak in the strength of NPF  
831 events coincides roughly with the maximum solar insolation (defined as the last 6 hours of accumulated solar  
832 radiation along HYSPLIT back trajectories); solar insolation allows for the increased availability of precursors  
833 by encouraging oxidation. The NPF frequency during the summer remains fairly consistent, with an estimated  
834 20-30% of days experiencing an NPF event. Towards the end of summer, the decline in the amount of solar  
835 insolation reduces the NPF intensity. In addition, it is also apparent that the condensation sink (CS i.e. linked to  
836 the total surface area of pre-existing aerosol) plays an important role, as the NPF intensity is more pronounced  
837 when there are reductions in CS. For NPF events, the minimum CS was observed at the start of the events and  
838 signifies that a reduced condensation sink is required for events to begin. This study is the first to utilise the  
839 Nano-ranking method in the Arctic. It should be stated that special care needs to be taken in treating wind-blown  
840 events separately from NPF events. Furthermore, this study also highlights potential problems when sampling

841 lines are submerged in dense clouds. It is recommended that relative humidity, visibility and/or web cam footage  
842 is used to help screen for these in-cloud events when sampling at high altitudes.

843 Typically, as the Arctic transitions from the Haze season to summer, cloud coverage and precipitation increase,  
844 which in turn, via wet scavenging, reduces the once predominant accumulation mode. However, the increased  
845 cloud cover, not only reduces the CS, but also reduces the solar insolation. Hence, the transition to summertime  
846 both promotes and discourages NPF. The latter stages of the summer and the autumn provide less solar  
847 insolation; the peak in solar insolation is skewed more towards the start of summer. These competing effects can  
848 explain why we observe such a reduction in NPF intensity as the summer progresses. The dip in frequency and  
849 intensity that occurs around July coincides with a decrease in brightness (solar insolation over potential solar  
850 radiation). It is the interplay between both solar insolation and condensation sink that can potentially give rise to  
851 NPF events, and thus cloudiness will also play a role in NPF activity given interplay with CS and solar radiation.  
852 We can only speculate that a warming climate may appear more similar to the late Arctic summer, being both  
853 warmer and wetter. Some studies indicate that cloud cover in the Arctic could potentially increase (Barton and  
854 Veron, 2012) or has (Eastman and Warren, 2010; Francis et al., 2009; Kay and Gettelman, 2009; Palm et al.,  
855 2010; Vihma et al., 2008) with periods of rapid sea ice loss.

856 One main finding from this study is that we can, with rather good accuracy, predict the occurrence of an NPF  
857 day (Fig. S26 in the supplement) by using CS and solar insolation as the two main predictive parameters.  
858 Periods exhibiting low CS and high solar insolation provide the most ideal conditions for NPF events and the  
859 occurrence of these periods can reproduce the observed frequency of NPF events. It should be noted, though,  
860 that other factors play a role. The sources to the precursors of the condensable vapours, as well as additional  
861 meteorological and environmental parameters such as sea-ice cover, chlorophyll a concentration and the varying  
862 contributions from different source regions all can influence the occurrence and strength of NPF. However, from  
863 this study we argue that both solar insolation and CS can be used to create a rather simple predictive model,  
864 simply by taking the ratio of these two terms. The Arctic perhaps offers a unique environment, where the  
865 relationship between solar insolation and NPF is potentially stronger, due to the typically lower CS and lower  
866 overall solar radiation intensity (Bousiotis et al., 2021).

867 Quantifying the contribution of CCN from NPF on low-level clouds is beyond the scope of this study. However,  
868 we are able to demonstrate that periods of formation and subsequent growth lead to a significant increase in the  
869 proportion of the total number of Aitken mode particles. In combination with this, we show that on many

occasions during NPF events, the nucleation mode particles grow to potential CCN sizes once they are formed (57 NPF events lead to diameters >25nm). With the correct conditions, i.e. a relatively strong updraft, the formation of particles >25nm could lead to droplet formation and in turn alter cloud properties.

Given the high frequency of onsite NPF and the large potential area for formation, it is not unlikely that a majority of Aitken mode particles originate from NPF within or close to the Arctic region. Provided sufficient time for growth up to CCN-sizes, it seems clear that NPF events could constitute a significant source of CCN in the Arctic. For a more detailed picture of the aerosol budget in the Arctic and a more quantitative account of the NPF-contribution to the overall Arctic CCN-population, large scale modelling evaluated against observations and combined with process studies are required.

#### **Code availability**

Code will be made available on GitHub when published.

#### **Data availability**

Data will be on the Bolin Centre database when published (<https://bolin.su.se/data/>).

#### **Author Contributions**

Dominic Heslin-Rees (DHR), Radovan Krejci (RK), and Peter Tunved (PT) worked together to develop the research questions and design the study. DHR performed data analysis and wrote the paper together with assistance from RK and PT. There was good input and advice along the way from Ilona Riipinen (IR), Annica Ekman (AK), Diego Aliaga (DA) and Martina Mazzini (MMi). Janne Lampilahti (JL) developed many of the NAIS specific Python packages and showed DHR how to clean and service the NAIS. Stefania Gilardoni (SG) provided comments. SG, Mikko Sipilä (MS), Mauro Mazzola (MMa) and Roseline Thakur (RT) helped with the Gruvabadet work. Ki-Tae Park (KTP) and Kitack Lee (KT) maintained and operated the Zeppelin DMS analyzer as part of a collaborative effort between KOPRI and POSTECH research team. Kihong Park (KP) and Young Jun Yoon (YJY) provided the Nano-SMPS data.

#### **Acknowledgements**

This study would not be possible without the work done by the research engineers Birgitta Noone, Tabea Henning, and Ondrej Tesar from ACES and the staff from the Norwegian Polar Institute (NPI). NPI provides

897 invaluable on-site support including attending to the NAIS. NPI is also acknowledged for substantial long-term  
898 support in maintaining the measurements at Zeppelin Observatory (ZEP). We would also like to thank Kai  
899 Rosman for developing the software we used for the instrumentation at ZEP. We would like to thank Ove  
900 Hermansen from the Norwegian Institute for Air Research (NILU) for providing the ambient meteorological  
901 data.

## 902 Financial Support

903 Aerosol size distribution measurements are supported by the Swedish EPA. Swedish participation was supported  
904 by the Swedish Environmental Protection Agency (Naturvårdsverket), Knut and Alice Wallenberg foundation  
905 (KWA) and FORMAS funding agency.

906 Knut-and-Alice-Wallenberg Foundation within the Arctic Climate Across Scales (Project No. \,2016.0024), the  
907 Swedish EPA's (Naturvårdsverket) Environmental monitoring program (Miljöövervakning), the Swedish  
908 Research Council FORMAS (Project "Interplay between water, clouds and Aerosols in the Arctic," \#  
909 2016-01427)

910 The aerosol observations at the Zeppelin station have been supported by the KAW Stiftelse (grant  
911 no. 2016.0024), the Swedish Environmental Protection agency (Naturvårdsverket), and the ACTRIS Sweden  
912 project supported by the Swedish Research Council

913 The KAW project CLIVE and VR financed infrastructure project ACTRIS-Sweden.

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