

AR-2025-11 MS

In this report we address the concerns of the two referees in their respective reports. We want to thank both referees for the time and effort they put into reviewing our paper. We find that the scrutiny and set of recommendations has definitely helped the paper overall. In the following report we have split the responses into Report 1 and Report 2 and we have responded to each comment separately using the colour blue. For direct quotes from either the new version (i.e. this is version 3) of the paper or the previous one (i.e. version 2) we use the colour blue along with quotation marks and italics.

Report 1

accepted subject to technical corrections

The authors have done a commendable job of revising their manuscript. They have satisfied my main concern about quantitative statistical metrics and quantitatively demonstrating the relationship between NPF and solar insolation and CS. They now use the ratio of SUNFLUX to CS as a proxy for NPF in addition to their ROI which adds robustness to their methodology. They now use a binary classification for each hour and calculate daily averages instead of weekly percentages and interpolation thus alleviating one of my concerns. The authors also give some nice recommendations for the use of the Aliaga et al. method in the Arctic, which will be valuable for the community going forward. Their use of descriptive statistics now better describes the connection between the ROI and NPF. This is an important improvement as I found simply averaging ratios to be problematic. Without considering sample sizes, averaging ratios can indeed be misleading. For example, simply averaging the ratios 0.5 and 0.90 gives 0.7. Although if the fractions are $1/2$ and $90/100$, summing the numerators and denominators then taking the ratio gives a value of 0.89. Without proper descriptions of their methods readers could draw incorrect conclusions, in this case 'it is not what it is' to paraphrase the authors' response. They now mask Fig. 8 for bins that contain less than 1 data point. Their changes now better reflect how the ROI relates to NPF. They have adequately addressed my minor comments.

Main suggestions

My only remaining main concern is the selection of the 30 day window size for the rolling mean. The authors have nicely demonstrated the effect of the window size on the agreement between the SUNFLUX/CS and event frequency. They discuss how this relationship plateaus after 14 days in the main text and state in the supplement 'We argue that 10-20 days of averaging/smoothing ensures that the features (characteristic/seasonal peaks) remain present above the variability, and the data is not smoothed too much.' **So I am curious why a large window size of 30 days was still applied. Is there a specific justification for this selection?**

Whatever window is chosen will end up being somewhat subjective. Using a 30-day window at least keeps us using a monthly average, where we can compare Fig. 11 to other plots which are also using

Responses to reviewers of AR-2025-11 MS

monthly averages like Fig. 1 and Fig. 3 a) & b), Fig 4 a) & b), Fig. 5, Fig. 6, Fig. S6, Fig. S22, Fig. S23. At the end of the day, it is simply a nice unit of time and in other parts of the paper we used months - it is basically a compromise between using monthly averages and displaying the characteristic seasonal peaks.

The referee is discussing Fig. 11 I guess i.e. the following plot:

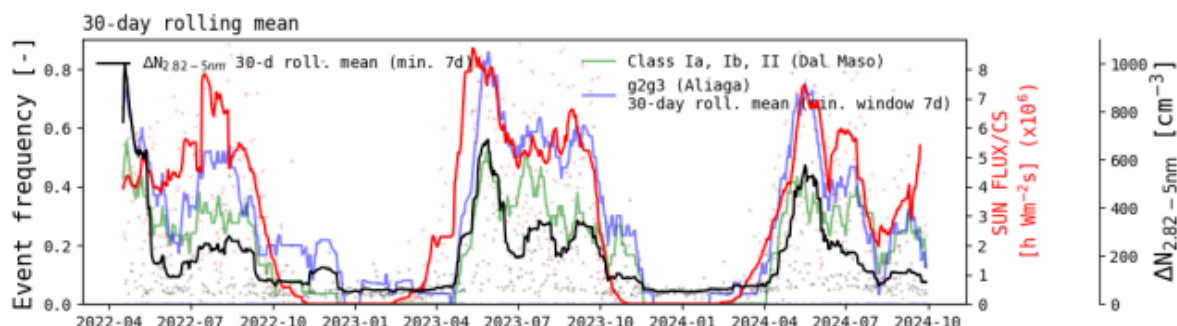


Fig 11: The red line is the solar insolation divided by the condensation sink (Solar insolation/CS) with a 30-day rolling mean. The intensity parameter ($\Delta N_{2.82-5nm}$). The green and blue lines represent the rolling averages of the NPF daily event frequencies using the Dal Maso et al. (2005) 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.

Suggested improvements

Their use of measurements from Beck et al. (2021) also adds interesting results about the importance of acids in the first part of summer vs HOMs at the end as well as the ions contributing to NPF. This could be worth mentioning in the abstract or conclusion.

Placed this in the conclusion:

“The calculated concentrations of condensable vapours required to maintain the estimated GRs were similar to the combined measured concentrations of HOMs, H₂SO₄, MSA and HI03 by Beck et al., (2021). The first peak in NPF events coincides with a larger proportion of negative ions, and over the course of the year the number of 2-4nm ions becomes more positive, and this coincides with a domination of HOMs in the later parts of the summer and into autumn alluding to a shift in composition of the small intermediate ions (2-4nm).”

Decided not to add to the abstract as it requires too much explanation.

Lines 1126-1127: Could the authors give the R² value of this agreement and reference Fig. S28.

Responses to reviewers of AR-2025-11 MS

I could not find what the reviewer was commenting on because the paper does not go past Lines 1126-1127. There are no lines 1126-1127 in the paper. However we did add a reference to Fig. S28:

“Generally, the predicted frequency of occurrence (i.e., defined by the ROI) is significantly lower than the observations (can also be observed in the correlation figure Fig. S28 in the supplement).”

S10.2: Fig. S28 is referenced here but should this be Fig. S26?

Yes, you are correct that has been changed to Fig. S26.

Fig. 12 caption seems to end abruptly.

The caption for Fig. 12 has been changed to following:

“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.”

Fig S24. Please indicate what the dotted line represents and what the circles represent. The date labels could be improved for clarity.

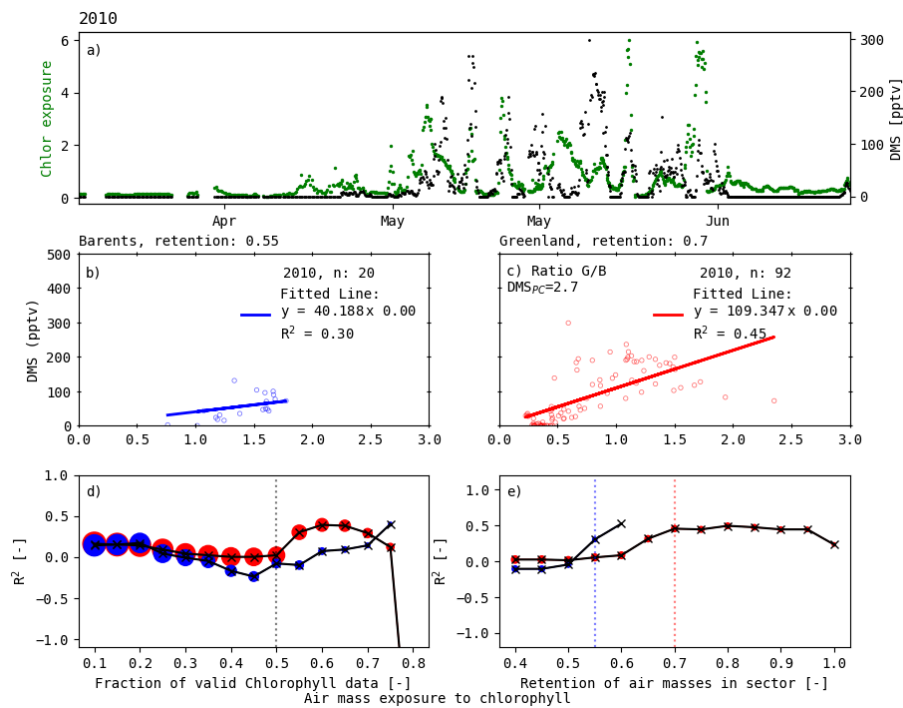
Thanks again. This has been rectified. The caption on Fig. S24 has been altered to the following:

“Fig S24: The ratio of the concentration of HOMs to the total concentration of the combined HOMs, H_2SO_4 , MSA and HIO_3 precursor gases taken from Beck et al., (2021) is represented by a black dashed line, i.e. $HOMS/Tot$. The blue circles display the ratio of the concentration of positive ions between 2-4nm with the total concentrations of 2-4nm ions. Note that the Ny-Ålesund HOMs data is from April 12 to the end of August 2017.”

Figs. S30-S37: The y-label for the two middle panels appears below the lower panels. Why is this label not below the axis it refers to? Could this be corrected?

The figures have been altered so that the x-axis label is pushed down on all Figures.

Responses to reviewers of AR-2025-11 MS



Report 2

reconsidered after major revisions

Suggestions for revision or reasons for rejection

The study provides a multi-year (2022–2024) dataset on aerosol and ion number size distributions, focusing on new particle formation (NPF) in the Arctic, thereby extending existing measurements from Svalbard (e.g., Lee et al., 2020). This dataset is valuable, as multi-year, high-resolution ion–particle observations from the Arctic are crucial for improving the limited representation of aerosols in this remote region. However, although the dataset is important, the roles of **solar insolation, CS, and DMS in driving Arctic NPF have already been extensively explored** (e.g., Tunved et al., 2013; Lee et al., 2020; Beck et al., 2021). The authors have discussed these studies in the introduction and referred to them again in the discussion. The **novelty of this study, therefore, lies primarily in the temporal extension of the data record rather than in a fundamental advancement in understanding Arctic NPF processes.**

We do not agree with this because each previous paper which focused on NPF in Svalbard had different aspects, and missed various elements. Beck et al., (2021) is missing any mention of the condensation sink (CS), the measurements in Tunved et al., (2013) start at 10nm in terms of particle size, and also do not feature CS. Tunved et al., (2013) discusses CS descriptively without qualifying it and mostly focuses on size distributions and accumulated rainfall. Beck et al., (2021) is a comparison paper and mainly discusses the two chemical pathways for the different sites. Our approach is different from all these papers - it represents a more stripped back approach.

The dimensionless criteria in Lee et al., (2020) used to predict the occurrence of NPF events, (L_T), uses chemistry - the whole point in our study was to simplify the predictive parameters, and use physical and environmental parameters only i.e., CS and solar radiation. We used solar insolation as a proxy for the photochemistry instead of H_2SO_4 . It might turn out that the Arctic is one of the few places where you can do this simplification and reduce NPF predictability down to just solar radiation and CS. Maybe it turns out that this is the only environment where it can be performed - but it is worth exploring nonetheless. The polar night and day allows for complete darkness and days when the solar is above the horizon all the time.

For instance, the reported correlation between the ratio of solar radiation to CS and the frequency of **NPF events is already well known and represents an oversimplified approach to predicting or characterising NPF**. A more process-based, multivariate analysis that includes DMS, wind speed, wind direction, and relative humidity could strengthen the robustness of NPF predictions. Alternatively, the ratio of CS to growth rate can also serve as a predictive criterion for assessing NPF likelihood, as it characterises particle growth dynamics and the survival probability of new particles against coagulation scavenging (Kulmala et al., 2017; Cai et al., 2017).

We fully understand the fact that the method described in this study is an oversimplified approach, however that was the approach from the beginning. The ratio of CS to GR for example could serve as a better predictive criterion for assessing NPF likelihood, and for sure holds more theoretical support, however, it still requires the calculation of GR and therefore the examination of NPF events - here we show a reduced method which focuses on meteorological values/parameters and a more basic knowledge of the overall particle number size distribution can be used to understand the seasonality and

Responses to reviewers of AR-2025-11 MS

likelihood of NPF. We are for sure simplifying the predictive process, trying to make it quick and simple so to say. This method would essentially allow for predictions to be performed anywhere where you have PNSD performed. SR/CS is easier to calculate just using routine measurements, which is important in the Arctic where larger set ups are not always commonplace.

Furthermore, in the case of Zeppelin Observatory, wind speed and direction are not relevant given the topography at ZEP. There are only two prevailing wind directions i.e. northerly and southerly (Pasquier et al., 2022). At ZEP you would expect a prevailing southerly component with occasional northerly ones. The Zeppelin mountain effectively helps to channel the wind creating these components. Furthermore, the mountain blocks the large-scale winds and reduces the overall wind speed observed at the observatory (i.e. mostly below 6ms⁻¹)(Pasquier et al., 2022).

We also used this paper to effectively show that DMS is not the best predictor for NPF or at least the DMS exposure. But overall, we were essentially trying to not use any more data and try to keep the study and its predictive elements as simple as possible with the aim of describing NPF frequency.

Although it is oversimplified we are effectively trying to utilise a proxies for source strength (i.e. Solar insolation) and the loss rate, or sink strength (i.e. Condensation sink, CS). A high SR/CS would indicate a source strength larger than the loss, thus favouring NPF.

Adding a measurement of the concentration of a precursor vapour e.g. SO₂ to the predictive indicator could produce better results e.g. Pierce et al. (2014) used SR-SO₂/CS. However, again adds an extra set of measurements that might not always be carried out, especially in the Arctic.

Further, the manuscript covers a broad range of topics related to Arctic NPF, such as seasonality and interannual variability, nighttime NPF events, NPF characteristics, the role of ions in NPF, geospatial extent, NPF likelihood criteria, and NPF contributions to potential CCN. However, a mechanistic understanding of the **underlying processes is largely missing**. For example, the relative contributions of **various precursor vapors** (e.g., H₂SO₄, MSA, HOMs, and iodic acid) to nucleation and growth are mentioned but not quantitatively disentangled. Similarly, while the influence of DMS and marine biogenic activity is acknowledged, there is limited discussion of the **chemical pathways or kinetics explaining the observed temporal shifts in NPF intensity**. Furthermore, it is unclear how the authors **differentiated background Aitken-mode particles from those originating from NPF events**, either upwind or downwind of the site. It is **extremely challenging to estimate the fraction of Aitken-mode particles from NPF that grow to CCN-active sizes based on a single observational location**. The decadal DMS emission trends are increasing across the Arctic due to **decreasing sea-ice coverage**, especially near the marginal ice zone. However, observations of DMS oxidation products do not show a uniform trend, highlighting the complexity of sulfur aerosol processes. If available, please include direct observations of **DMS oxidation products during the study period**. The paper could be significantly strengthened by providing a more integrated, mechanistic interpretation linking **precursor chemistry**, meteorology, and **aerosol dynamics**.

We agree with the reviewer that a mechanistic understanding of the underlying processes in terms of chemistry is largely missing, however, for this study we never set out to attempt to explore the chemical pathways. The problem is that for Zeppelin Observatory the chemical measurements are sparse - and are

Responses to reviewers of AR-2025-11 MS

generally limited to short campaigns. Generally, for our period of interest there was no precursor vapour data at Zeppelin Observatory and only during specific campaigns were there data available at Gruvabudet (closer to the research village of Ny-Ålesund).

The idea of the study was to try and simplify the process and understand and predict NPF using a more basic instrumental set up, given that many stations in the Arctic lack a complete measurement set up and that chemical measurements are sparse.

Yes it is true that some studies do show the importance of DMS (i.e. for the growth of the nucleation mode see Lee et al. (2020)), but in our study we show that DMS is not important - or at least not reliably important (can be used as predictive element every year). Gali et al. (2019) produced a nice correlation between the ice-free extent and DMS emissions for the Atlantic sector ($R^2 = 0.52$), Greenland showed a very poor correlation ($R^2 = 0.09$), and for the central Arctic sector there was <2GgS based on an increase in the ice-free extent of 6%. Yes, there are decadal DMS emission trends increasing across the Arctic however there is a large variability amongst the various seas e.g. Chukchi, Laptev, Kara, Beaufort, Baffin, Central Basin, Greenland and Barents due to the phytoplankton communities, nutrient availability, sea surface temperatures, stratification and the time of the bloom. Therefore, it will depend on the site in question as to whether decreasing sea-ice coverage within the region has led to DMS increases. DMS emissions also do not show a uniform trend see Gali et al. (2019), hence, this could also be a reason its oxidation products do not show a uniform trend either.

Unfortunately, there were no measurements of DMS oxidation products for that time period at ZEP, only DMS. If we were to add a proxy for the oxidation products of DMS using some kind of decay function (i.e. exponential function) it would add another source of uncertainty, as there is a large variability; as mentioned in the paper there is a wide range of DMS oxidation timescales depending on photochemistry and available solar insolation (Ghahreman et al., 2019). We did not want to go down that path of using too much data - we wanted to simplify the methodology due to the lack of stations with complete measurement set ups - hence adding or attempting to add more data goes directly and contrary to the nature of the paper.

At ZEP, it is not obvious what background Aitken-mode particles would consist of other than those originating from NPF events. Direct primary emissions of seasalt aerosol, mineral dust and organic and black carbon aerosols from long-transport would not contribute to the Aitken mode. We are assuming that the vast majority of Aitken mode particles measured on site have derived from regional NPF and then been advected to site. It should be noted that it is difficult to separate the Aitken mode particles formed from previous NPF events or from NPF events that happened on site. If we do not assume that the vast majority of Aitken-mode measured at ZEP are formed from NPF events then I would not know how to begin differentiating background Aitken-mode particles from those originating from NPF events.

Obviously, we have not performed a full investigation as to whether the particles that grow beyond 25nm are indeed activated and form cloud droplets. All we can do is suggest that beyond 25nm, given the right meteorological conditions i.e. high supersaturation then these particles have the potential to act as CCN. We understand the limitations and acknowledge the fact that it is extremely challenging to estimate the fraction of Aitken-mode particles from NPF that grow to CCN-active sizes based on a single observational location.

The introduction also **underemphasizes recent modeling and global aerosol–climate coupling studies (both laboratory- and field-based) that address biogenic sulfur and iodine pathways and Arctic aerosol**

Responses to reviewers of AR-2025-11 MS

processes in general. Including these would better contextualise the implications of the findings (e.g., He et al., 2023; Baccarini et al., 2020; Cala et al., 2020; Hoffmann et al., 2016; Finkenzeller et al., 2022; Xavier et al., 2024). The discussion of general aerosol processes could be shortened to make space for this.

We have included more modelling and global aerosol-climate coupling studies both laboratory and field based - biogenic sulfur and iodine pathways. But needs to be mentioned that these studies offer another approach compared to what we are trying to do in this study.

Added the following paragraph:

“Recent modelling and global aerosol-climate coupling studies both laboratory and field-based have increased our understanding of biogenic sulphur and iodine pathways. For sulphur pathways, Hoffmann et al. (2016) showed that the aqueous-phase (i.e. cloud and aerosol water chemistry), and not just the gas-phase, play an important role in the oxidation products of DMS, demonstrating the complexity of atmospheric sulphur chemistry in part due to the multiple oxidation pathways, which in turn are not represented well in climate models. Beyond sulphur-derived precursors, Finkenzeller et al. (2023) shed light on the formation mechanism for iodic acid (HIO_3) under atmospheric conditions and helps to corroborate how iodine can be produced and therefore act as a strong nucleating vapour in polar regions. Iodine-assisted NPF is also directly supported by the experimental findings shown in Baccarini et al. (2020). When iodine chemistry is included iodine acts as a missing source of low-volatility vapours, and improves the modeled Aitken mode representation (Xavier et al., 2024).”

The authors acknowledge the subjective nature of manual NPF classification and the potential observer bias. However, it is unclear why authors **still rely primarily on manual classification**. The purpose of using nanoparticle-ranking analysis is to quantitatively identify NPF days on a continuous scale (e.g., 0–1). In this study, more than 50% of the days were classified as “undefined.” I strongly recommend using the nanoparticle-ranking analysis **and correlating the NPF rank with DMA, CS, C_v, solar radiation, and other meteorological parameters**. Also, why did the **authors choose the 2.8–5 nm size range instead of 2.5–5 nm**, as suggested by Aliaga et al. (2023)?

The manual classification is simply used because of historical reasons. We still rely primarily on manual classification essentially because people understand it and the nano-ranking is new. For Nano ranking to be used in the Arctic you still have to remove the blown snow events first in order to utilise the method and there are many. Hence it is not the easiest method to use in such conditions, and this forms part of the recommendations for the Aliaga et al. (2023) method in the Arctic. Without using the manual method we also do not know how effective the Nano-ranking method is in such conditions. Windblown snow events fall into the ‘undefined’ category but still need to be checked on a daily basis, hence we might as well use the manual method as well.

The recommendation for the intensity parameter and its corresponding background value (i.e. what is removed) will vary from region to region and therefore there is not a standard predefined recommendation for all cases as far as I am aware.

Responses to reviewers of AR-2025-11 MS

In terms of correlating NPF rank with 'DMA, CS, Cv, solar radiation, and other meteorological parameters'. I have shown already that DMS is a tricky parameter to use, solar radiation I have done the correlation with and it is in the supplement i.e. intensity with SUNFLUX (Fig. S27). As I remember correlations with CS alone did not produce much, and Cv would be difficult to do because the Cv value is calculated using calculated GRs i.e. it is the Cv required to sustain a given GR, hence the correlation would be based only event days (technically NPF events strong and clear such that GRs can be estimated) as opposed to both event and non-event days - not sure what this would do to the statistics, but it may be misleading and unhelpful.

I think I chose the 2.8–5 nm size range instead of 2.5–5 nm because it compared better with the traditional Dal Maso classification. The definition of the intensity is not set in stone and depends on the region and environment.

The authors claim that NPF events "lead to an increase in Aitken-mode particles contributing to CCN" is plausible but not directly demonstrated. No CCN closure or hygroscopicity data are presented. I recommend avoiding such strong statements unless supported by direct evidence or case studies.

I think statements in the paper are not as strong as you are implying i.e. see

Lines 766-771 in version 2 in section 4.3:

"At ZEP, NPF and the subsequent growth that follows, results in particles large enough to potentially be activated and become CCN; on numerous occasions (57 times) we observed newly formed particles able to grow beyond 25nm while in the same regional event (see Fig. 15b), before being scavenged. Here, we use 25nm as the lowest bound for particles to act as CCN, however, it should be stated that this limit requires strong updrafts and extremely hygroscopic particles for particles to be activated. We observe that after ~30 hours of growth, we reach potential CCN-sizes at ZEP (see Fig. 15a))"

Lines 772-774 in version 2 in section 4.3:

"It is important to note that once produced these particles most likely continue to grow downwind of ZEP by condensation and in-cloud processing, so a large fraction of the Aitken mode should inevitably reach CCN size in due time, as long as they are not otherwise scavenged."

Lines 837-838 in version 2 in the conclusion:

"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."

We use words like '**potentially be activated**', 'this limit **requires** strong updrafts and extremely hygroscopic particles', '**not unlikely**'.

Responses to reviewers of AR-2025-11 MS

In addition the wording on the statement on line 771-772 (version 2) was altered.

“In addition, on 11, 8 and 3 respective occasions NPF-produced particles grew above beyond 60, 70 and 80nm respectively were observed (see Fig. 15b).”

We simply state that NPF events contribute to Aitken mode particles which have been shown to be large enough on some occasions to be activated. Any more would be beyond the scope of this paper. We state this in the paper **(lines 776-777 in version 2)**:

“It is difficult to ascertain the proportion of NPF-derived Aitken mode particles that actually become CCN before being scavenged or deposited, and estimating an exact fraction is beyond the scope of this study.”

Afterwards we then go on to provide evidence for this from the literature.

We added an extra qualifier to line 839 (version 2) i.e. **‘could’**. See full statement below:

*“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.”*

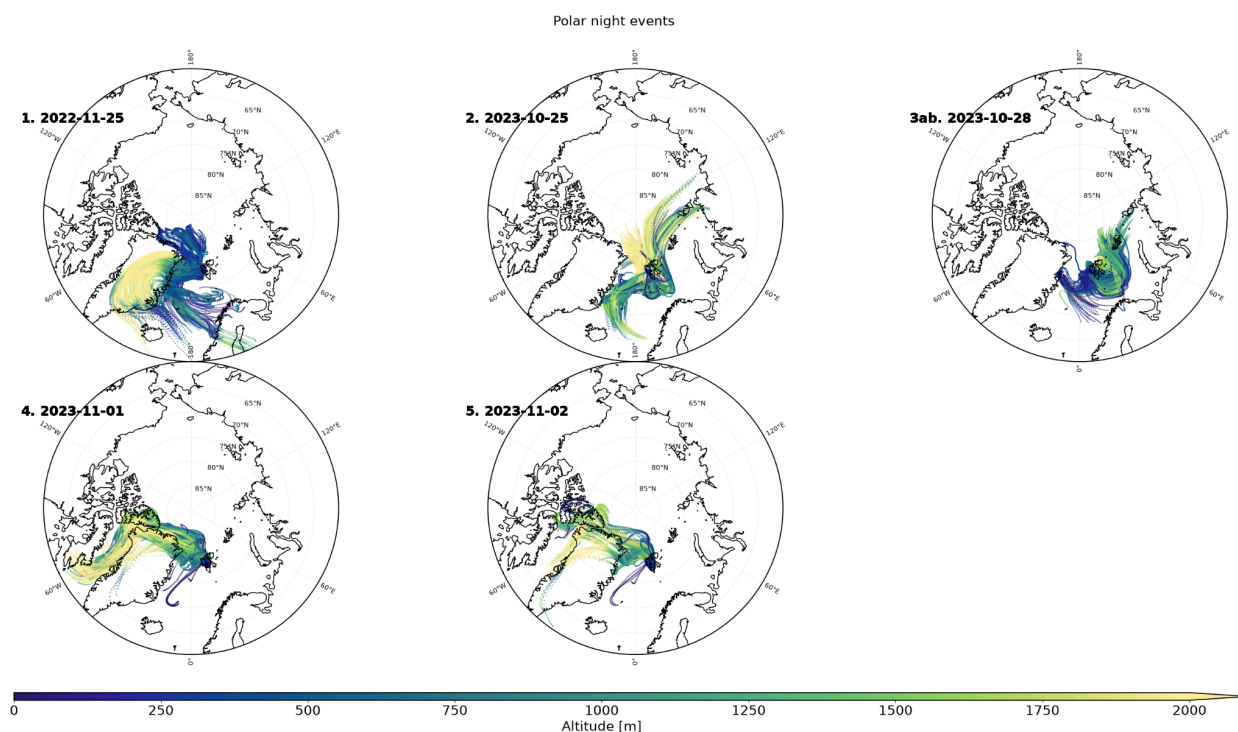
In conclusion, It is true that we do not directly demonstrate that NPF events contribute to CCN, however, considering that we do measure NPF events whereby on more than 50 occasions the particles grow beyond 25nm, a size where it has been shown by the work of Karlsson et al., (2021) for example that these particles can form CCN, then it makes sense to us that from the same station Aitken mode particles from NPF events do contribute to CCN.

If I understand correctly, the authors claim that polar-night NPF events occur under descending air masses from higher altitudes that originated in **higher latitudes**. I suggest plotting air mass back trajectory cross-sections (time/distance versus altitude) for the polar-night NPF event days (see Fig. 1b in Zha et al., <https://doi.org/10.1093/nsr/nwad138>).

No. You misunderstood - the polar-night events are associated with back trajectories from higher altitudes that originated from lower latitudes.

Airmasses have been plotted (see below):

Responses to reviewers of AR-2025-11 MS



Previous studies are cited excessively in the results, which makes it difficult to follow the paper's unique contribution. This also weakens the perceived novelty. I suggest separating the results and discussion.

I fear that having results and discussion separately will lengthen an already long paper. But I have tried to move more references to later parts i.e. have the results discussed per a section and then the literature brought in.

A mere change in diameter during an NPF event does not necessarily imply active CCN or activated CCN. **What is the background median or modal aerosol diameter?** How did the authors separate background particles exceeding the threshold diameter? From Fig. 16, the number concentration of particles >70 nm does not differ significantly between cases. The authors could **calculate survival probabilities for CCN-active particles (25 nm, 50 nm, 100 nm)** following Pierce & Adams (2007) and other methods in the literature in the absence of CCN measurements. This could be a separate paper, if I may suggest so, investigating NPF's potential to produce CCN-active particles.

As stated in the paper, studies have shown that In the Arctic particles as small as 25nm can participate in cloud formation by acting as CCN (Leaitch et al., 2016; Karlsson et al., 2020, 2021; Pöhlker et al., 2021; Gramlich et al., 2023; Motos et al., 2023).

We are only trying to show that NPF events contribute to Aitken mode - the link between CCN and NPF/Aitken is not in the scope of this paper, as you pointed out that would be a separate study in itself, and yes this would be a good idea for another paper.

Thank you for the tip about calculating survival probabilities based on Pierce & Adams (2007) but this will expand the paper beyond its scope and perhaps lead to a separate paper.

Responses to reviewers of AR-2025-11 MS

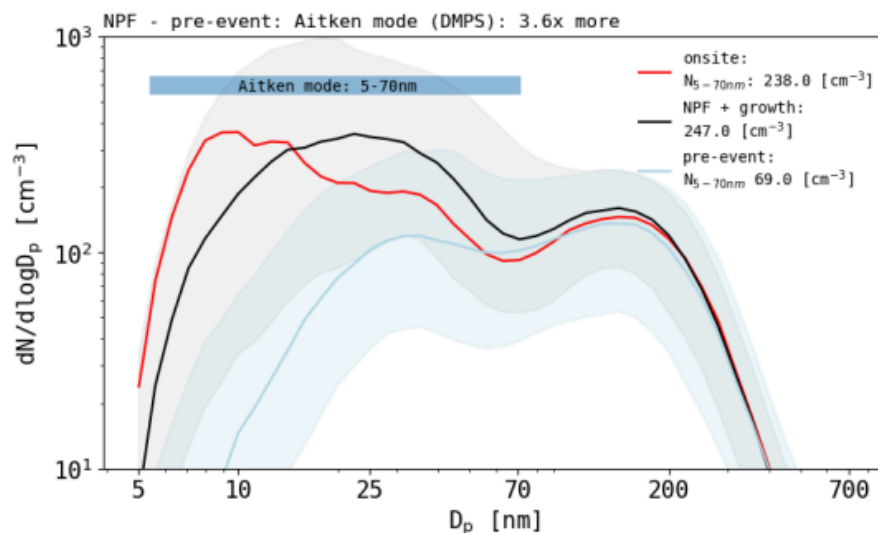


Fig 16: The particle number size distributions (DMPS) for periods before the NPF events (1-2 hours before event started) (pre-event, light blue), during NPF and growth (black) and on-site nucleation (production of 2-4nm particles, red, on-site formation).

Figure 1: Clarify what “Total” represents—is it the total number of days or total valid observation days? For instance, in April 2022, were there no measurements for approximately 18 days?

Total number of valid measurement days (i.e. Total) is written in the caption and it reads as follows:

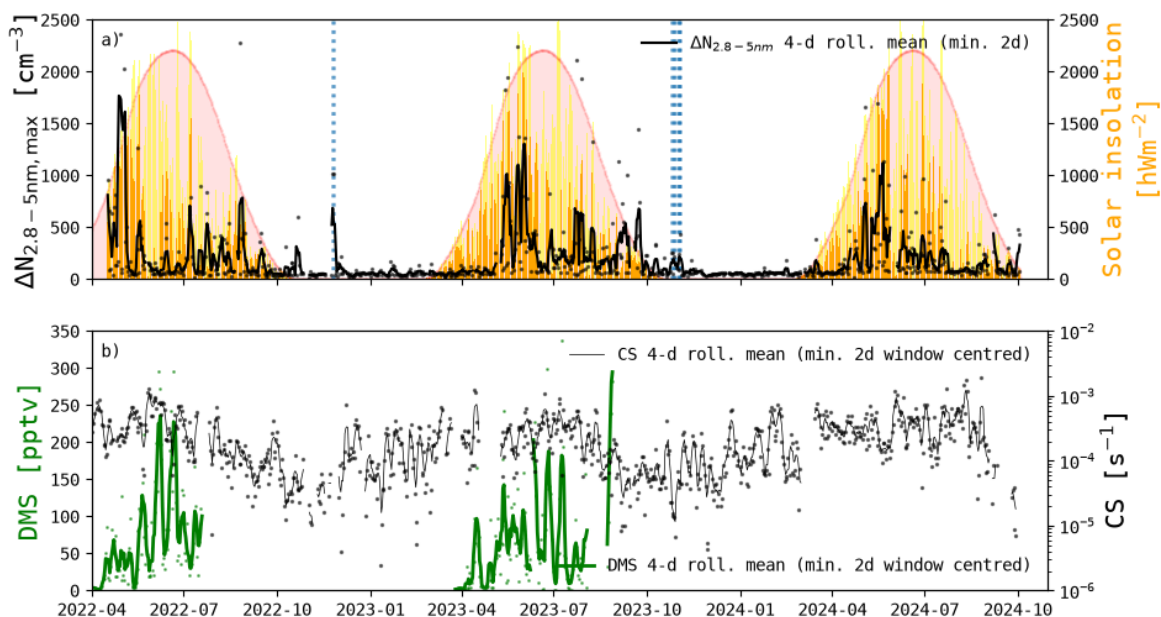
“Fig 1: Number of days per month classified as either Class Ia (red), Ib (orange), II (yellow), Undefined (green) and Non-event (grey). The left-hand axis is the number of days classified based on the total number of valid measurement days (i.e. Total)). The right-hand side is the fraction of events (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.”

Yes, for April there are 16 days of missing measurements because we only started measurements on 2022-04-17 (to 2024-10-04) as stated in the paper (i.e. Line 173 from the previous version, v2).

Figure 2: The **black dots are hardly visible**, and the legend is not fully explained. The caption is too long and should be more concise. **Avoid scaling factors where possible**; use dual axes if needed. Define **“recent 6 hours”**—from sunrise or noon? And what is the basis for using a 6-hour accumulated solar flux? How was the mixed-layer depth used? The quality of the figure is very poor.

Okay. Black dots are made larger on Fig. 2. See new version of figure 2 below:

Responses to reviewers of AR-2025-11 MS



What do you mean by avoiding scaling factors where possible do you mean for the right hand side of Fig. 2 a)?

We have added more details to the caption to define the 6-hour:

“The orange bar chart displays the daily-averaged solar insolation, which was defined as the accumulation of the most recent 6 hours of solar radiation experienced by air masses arriving at ZEP, within the mixed-layer (see Sect. 2.4.1 in the Methods for details)”

The caption reads the following now:

“Fig. 2: Time series of (a) the NPF intensity parameter ($\Delta N_{\text{max},2.8-5}$; 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.”

In the study we have already explained the use of 6-hours (see lines 334-338 in version 3)

“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

Responses to reviewers of AR-2025-11 MS

significance on solar radiation closer to the receptor; an accumulated solar flux consisting for a longer duration could lead to misleading results.”

Improve figure quality: ensure self-explanatory legends, avoid abbreviations in figure captions, and maintain consistent font type and size. For example, in Fig. S11, clarify what “AHX,” “SUM,” and “SBU” represent. Captions should be sufficiently detailed to allow interpretation without referring to the main text, and at the same time, not too long. Figure S7: **The x-axis lacks tick labels.** Please show exact hours here and in other figures.

Written in Fig. S11:

“Arctic Haze is denoted by AHZ, Summer by SUM, and Slow Build up season by SBU and the months they represent are given in the legend.”

Several figure captions (e.g., Fig. 2) are overly long; shorten them for readability.

Done for Fig. 2 - reduced quite a bit to the following:

“Fig. 2: Time series of (a) the NPF intensity parameter ($\Delta N_{\text{max},2.8-5}$; 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.”

Abstract: Replace “encourages” with “favors” or “promotes.”

Done. Changed to ‘encourages’ to ‘promotes’

Specify whether the PC version of the HYSPLIT model or tools like ZeFir were used for back trajectories.

Added the following sentence:

“HYSPLIT was run from the Desktop using the Python package pysplit (<https://github.com/mscross/pysplit>).”

Show both ion-polarity datasets from ~0.8 nm, but exclude particle data below 2 nm, as already justified.

In the paper, we mentioned that there is a loss of negative cluster ions below 2nm.

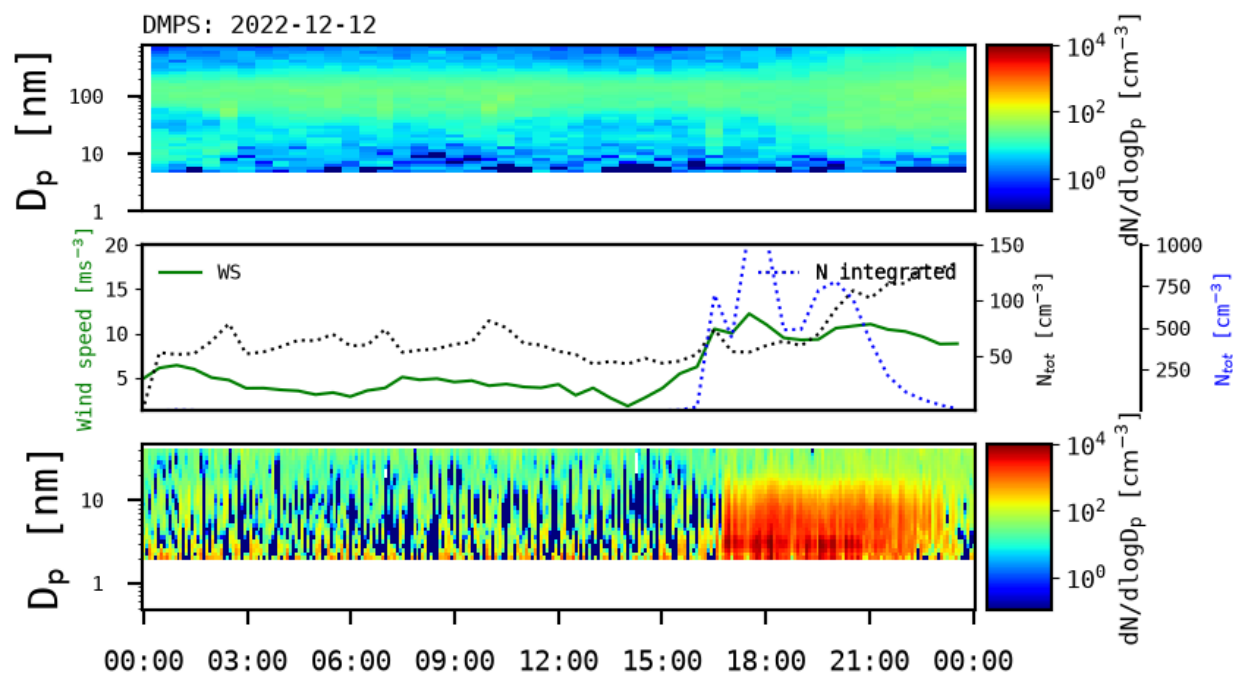
“Unfortunately, due to an instrumental problem related to a consistent loss of the negative cluster ions experienced during the measurement campaign (see Sect. S2.6 in the supplement) it was not possible to measure the concentration of negative ions less than 2nm”

So why would you like to include both ion-polarity datasets above 2nm?

Responses to reviewers of AR-2025-11 MS

Use consistent units throughout both the main text and the Supplementary Information. For example, in Figs. S2 and S4, particle diameter (D_p) is given in nm and m, respectively.

I have decided to use nm. So I changed the following figure to D_p [nm]. Added a third y-axis.



Clarify whether the CS was calculated for dry or ambient particle sizes, as hygroscopic growth affects surface area.

The condensation sink was not adjusted for hygroscopic growth effects. This is already mentioned in the MS in Sect. 2.3:

"It should be noted that the particle number size distributions presented in this study represent that of dry aerosol particle sizes. The measured particle size distributions for both the NAIS and DMPS were not corrected for hygroscopic growth, and thus do not reflect ambient conditions."

Note the spelling mistake has been corrected.

For example Massling et al. (2016) calculated *hygroscopic growth factors (HGF)* for Villum station:

"Mean particle hygroscopic growth factors ranged from 1.60 to 1.75 at 90 % relative humidity in spring, while values between 1.47 and 1.67 were observed in summer depending on the initial dry size. Although the summer aerosol number size distributions were characterized by frequent new particle formation events, the CCN population at cloud-relevant supersaturations was determined by accumulation-mode aerosols."

Responses to reviewers of AR-2025-11 MS

Applying a HGF would just change the scale and not necessarily the seasonality that might, given that we are mostly interested in the patterns and the seasonal changes such an inclusion would add another level of uncertainty.

Eq. 3 - Verify the units of the constant “A” and specify which molecular properties of sulfuric acid it represents, and provide a citation. Clarify whether dD_p/dt refers to total GR (over the full size range) or nucleation mode (<25 nm). Each equation should be self-contained with the necessary details and proper citations.

We changed the statement to the following:

‘ dD_p/dt is 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 below for further details).’

There is already a reference ‘See Dal Maso et al. (2005) for more details.’

What molecular properties of sulfuric acid does A represent?

A is derived from the accommodation coefficient of sulphuric acid i.e. the probability that a sulphuric acid molecule will permanently stick to a particle upon collision, the amount of volume H_2SO_4 will add to the particle once condensed onto it, and frequency at which H_2SO_4 molecules collide with particle, which is determined by the molecular mass (Fiedler et al., 2005).

We added the following sentence:

“The constant A incorporates the probability that sulfuric acid molecules will ‘stick’ to particles upon collision, the change in volume which occurs after collision and condensation, and also the frequency of collisions (Fiedler et al., 2005).”

Ensure that all figures (e.g., Fig. S17 and others) are cited correctly in the main text.

Added references for the following supplementary figures to the manuscript.

Fig. S3 and **Fig. S5** added to the following sentences:

“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⁻¹ (see Fig. S5).”

Following references to **Fig. S7** and **Fig. S8** is added to the sentence on line 275 of version 2.

Responses to reviewers of AR-2025-11 MS

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),

Fig. S14

“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 ($\Delta N_{2.8-5\text{ nm}}$).”

Added a reference of Fig. S15 to Line 281 (version 2)

“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).”

Added a reference of Fig. S22 to Line 624 (version 2)

“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).”

Added a reference of Fig. S25 a) to Line 642 (version 2)

“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”

Added a reference to Fig. S27 around lines 411 (based on version 2)

“Averaging the intensity parameter and correlating it to the solar insolation calculated based on the entire air mass and the parts within the ML shows that selection of air masses within the ML leads to a better correlation (see Fig. S27 in supplement).”

Fig. S28 in line 679 in version 2

“Generally, the predicted frequency of occurrence (i.e., defined by the ROI) is significantly lower than the observations (can also be observed in the correlation figure Fig. S28 in the supplement).”

Over 30 supplementary figures are excessive. Combine related figures where appropriate to reduce redundancy.

Responses to reviewers of AR-2025-11 MS

I do not know what to do about this. The other reviewers after the first round (this is the second round of reviews) asked for a lot. Originally i.e. the first version of the supplement had 18 figures and the supplement was 12 pages - it was a much shorter supplement until the last round of reviews basically.

Correct typographical inconsistencies (e.g., missing spaces in “solar insolationand” or “Condensation sink,CS”).

Corrected these typos and more

Maintain consistent language (choose either British or American English throughout).

We made sure it is all written in British English.

Format all units correctly (e.g., “nm h⁻¹” instead of “nm hr⁻¹”).

Looked for all units written incorrectly and changed.

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Xu-Cheng He et al., Iodine oxoacids enhance nucleation of sulfuric acid particles in the atmosphere. *Science* 382, 1308–1314 (2023). DOI:10.1126/science.adh2526

Other changes

The following sentence was removed on line 669-671 (version 2)

Approximately, hourly data points from 98% of the days with NPF events can be found in the non-ROI part, as opposed to 55% of the days with NPF events in the ROI.

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