Review of “Drivers governing the seasonality of new particle formation in the Arctic” by Heslin-Rees et al.

This paper gives an overview of new particle formation (NPF) events at Zeppelin Observatory on Svalbard in the High Arctic. It gives a comprehensive analysis of the seasonal frequency, growth and formation rates, duration and starting times, air mass origin, the factors driving NPF, and their contribution to Aitken mode particles and their ability to grow to cloud condensation nuclei (CCN) sizes. NPF events during polar night are also shown, something that is rarely observed. The authors argue that accumulated solar radiation in the past 6 hours (termed solar insolation) and the condensation sink (CS), measured locally, are the driving factors for NPF. They developed a simplified predictive model using these two parameters to estimate the likelihood of observing NPF. This model is compared to the Dal Maso NPF classification and Aliaga Nanoparticle Ranking methods for analyzing NPF activity. The authors do an excellent job of presenting the code sources throughout the text. This is the first evaluation of the Aliaga method in the Arctic. This paper fits within the scope of AR and will be of interest to the atmospheric community. This paper describes several interesting aspects of NPF and will undoubtedly add to the knowledge base of Arctic NPF, however there are some concerns that need to be addressed. Overall, I would recommend publication after major corrections.

Main Concerns:

My main concern is the lack of quantitative statistics utilized in this study, the use of rolling means with large window sizes, and comparing results visually. Two weeks and one month window sizes are very large and coupled with a logarithmic scale can make visually interpreting the results difficult. I would encourage the authors to include more quantitative statistical methods on unsmoothed or minimal smoothing the data to explore the main drivers of NPF and evaluate their simplified predictive model. To give some suggestions, this could include non-parametric methods such as Spearman rank correlations, binning variables and comparing differences between these groups using the Student’s t, Welsh’s t, or Mann-Whiteny U test (depending on the normality and homogeneity of variance of the groups) for the drivers of NPF and more classical methods such as coefficient of determination, (root) mean square error, and mean absolute error for the evaluation of the simplified predictive model, although the exact methods are up to the authors.

I would agree with the author’s interpretation that solar insolation and CS are important factors driving NPF, for the same reasons the authors nicely described, but the authors do not quantitatively demonstrate this in their manuscript. The authors should provide more convincing quantitative evidence in their manuscript.

The same can be said for their simplified predictive model, visually it agrees with the two other classifications schemes although there are notable differences in the highly smoothed time series. The authors state that April 2022 is limited in sample size which could explain this disagreement. The Aliaga method shows non-zero frequency during winter of 2022/23, I am wondering if all the blowing snow events were removed during this time, although I doubt such a large period would be missed. While I understand that a thorough evaluation of the Aliaga method is not the main purpose of this manuscript, can the authors comment on why the Aliaga method gives a non-zero frequency during the winter of 2022/2023 as opposed to the following winter? During July 2023, the simplified predictive model shows disagreement with the other methods, the authors state that this is expected to be due to the lack of adequate information regarding original reactants, do the authors have any evidence to support this expectation? There are other factors that could affect this disagreement, such as meteorology, air mass origin in relation to biologically active and sea ice regions, and wet removal. Have the authors explored other factors that could influence the observed disagreement during these periods?

Finally, the daily likelihood of NPF occurrence ($P_{NPF,D}$) is actually calculated on a weekly basis then interpolated (how?) to a daily resolution. This interpolated timeseries is then smoothed with a 30-day rolling mean. Why was the daily likelihood of NPF occurrence not calculated on a daily basis as the name implies? I would be interested in how the $P_{NPF,D}$ compares, using quantitative statistics, to the other two methods. A quantitative comparison could also be made between the Dal Maso and Aliaga methods, as this is the first time the Aliaga method has been utilized in the Arctic (to the reviewer's knowledge) and this could provide a valuable assessment of its performance in a polar environment for future studies. Although it is understandable if this assessment is deemed outside of the scope of the article.

Minor concerns:

Considering solar radiation along the back trajectory is an excellent idea as this would give a better indication of photochemical activity compared to in situ solar radiation and allows for appropriate time for oxidation of precursor gases to occur although I am now wondering if the authors considered something similar for the CS as well. Obviously, HYSPLIT cannot give an indication of the CS along the back trajectory but have the authors considered lagging the CS or possibly using accumulated precipitation along the back trajectory? They demonstrate precipitation and cloud processing has a large effect on NPF in Fig 11 and state this on 683. Tunved et al. (2013) and Khadir et al. (2023) also demonstrate the effect of accumulated precipitation on ultrafine aerosol particles nicely from ZEP. The authors demonstrate the NPF events will increase the CS, so I am wondering if the authors explored other variables, such as accumulated precipitation, to include in the simplified predictive model.

For the simplified predictive model, how did the authors calculate the line defining the ROI? Was it based on visual inspection or adjusted based on a criterion? I am very curious as to how did the authors calculate these averages “mean (median) 66% (68.4%)”? Did they simply take the mean and median of the ratios in the ROI? The large differences in the number of observations in different regions of the ROI could make such averaging give misleading results. Were the ratios weighed by the number of observations in each grid cell, i.e., weighted mean or median? I am wondering if a weighted average or calculating the sum the numerators and denominators of each grid cell in the ROI (i.e., integrating over the ROI) would be a more robust statistic. Could the authors please clarify this?

The ROI does have a higher likelihood of observing an NPF day compared to outside. However, there are grid cells with high likelihoods outside of the ROI although these grid cells do have a small sample size. I am wondering if the authors could comment on these false negatives in their model.

Line by line comments:

Line 49: Maybe it is worth mentioning ZEP is a mountain site when describing the geographic location.

Line 55: “which matches the frequency” Here would be a good place to include the performance evaluation for their model.

Line 57: I would say the authors nicely demonstrate the polar night NPF events arrive from higher altitudes (Fig. S17) therefore they are not speculating.

Lines 61-63: The two parts of this sentence are redundant. Please make this sentence more concise.

Line 63: “over 50 NPF events” Would this be more accurately expressed as a percentage of total NPF events for context? This would help contextualize the importance of NPF’s contribution to CCN in the Arctic.

Line 90-92: Could the authors include more original references for this statement? Also, Carslaw, 2022 is not in the reference list.

Lines 109-111: The authors mention both negative and positive ions but only give a reference for negative ions, can the authors include a reference for positive ions?

Line 122: Villum Research Station is the preferred name for this station.

Line 125: The town of Barrow, Alaska was renamed to its original, indigenous name of Utqiaġvik in 2016. However, the NOAA observatory located outside of the town is still referred to as the Barrow Atmospheric Baseline Observatory (BRW). I would encourage the authors to either refer to the town by its proper name or be explicit when referring to the atmospheric observatory.

Lines 128-131: Beck et al. (2021) also showed different species contributing to NPF and growth during different times of the year at these two locations, iodic acid at Villum during springtime and sulfuric acid/ammonium during summer; sulfuric acid/ammonium at ZEP during springtime and highly oxygenated molecules during summer. Could the authors please mention these important results from Beck et al. (2021)?

Line 132: Please define DMS and mention its marine, biogenic origins. The authors mention phytoplankton blooms on lines 86-87, here would be a good place to first mention DMS and its effect on NPF.

Lines 132-135: As currently constructed, this sentence implies the correlation between DMS and chlorophyll a is the reason that DMS is an important source of nucleating vapors. While none of the information in this sentence is incorrect, the way the sentence is structured is misleading, please rephrase.

Line 146: "higher end of the accumulation mode" do the authors mean Aitken mode here?

Line 176: Are two CPCs in this sentence referring to the ones on the twin DMPS system? Or the two CPCs references in the next sentence? If referring to the DMPS system, then saying "total aerosol concentration" for size segregated monodispersed aerosol is confusing.

Line 244: Would it be possible to show a figure of exemplary days for Class Ia, Ib, and II NPF event in the supplement? Given there is a degree of subjectivity in the Dal Maso method, this would help readers who are unfamiliar with the method gain context for each event.

Line 283: Was the growth rate calculated using the timestamp for the maximum rate of change for each size bin as listed in the text or the maximum concentration as listed on line 301?

Equation 1: N_{i-j} is not defined.

Line 300-301: The link does not work and there is no repository named npf_event_analyzer. on jlpl's Github page.

Equation 3: CS is not in Eq. 3 but is listed in the description. Is it part of another term in the equation?

Line 320: Was the starting height above ground level or mean sea level?

Sects. 2.4: I commend the authors on calculating ensemble trajectories as this gives a more robust estimate of air mass source regions compared to a single back trajectory.

They are also a bit more complex to work with. I am curious if the authors combined all ensemble members or used individual ensemble members for the calculation of chlorophyll a exposure, nucleation site estimation, and air mass source regions. Could the authors please explain this in the text?

Sect. 2.4.2: Could the equation for calculating chlorophyll a exposure be listed here? Readers might not be familiar with the work of Park et al. (2018). I am also curious as to why the authors calculated chlorophyll a exposure for 2.5 years of trajectories (no easy task) and do not show any of the results even though they are mentioned several times throughout the text.

Sect. 3.1.1: “Occurrence” and “frequency” are used interchangeably, I am wondering if it is better to be consistent to avoid confusion.

Lines 379-383: The authors do an excellent job of describing previous studies on NPF activity at ZEP, could the authors include studies from other Arctic sites to provide context.

Line 390: “maximum daily increase” should this be “maximum daily concentration” as listed on line 266?

Lines 400-401 and 407-408: This is where some quantitative statistical metrics could be included.

Line 411-412: What other precursor candidates are there for sulfuric acid production other than SO_2?

Lines 412-413: The CS is for condensing vapors and the CoagS is for coagulating particles, I think these terms are switched in these lines.

Lines 417-418: Can the authors simply list the changes in cloud cover or their frequencies from Maturilli and Ebell (2018)?

Lines 423-427: It is interesting to note the almost anti-correlation between DMS and $\Delta N_{max\ 2.82\text{-}5}$.

Lines 434-436: Would the polar night events be better presented as a table? What is the meaning of 3ab and x2 in these lines?

Lines 438-439: The latitudinal dependency is explained but was there any longitudinal dependency of these polar night events?

Lines 439-441: Wouldn’t the low temperatures experienced by high altitude air masses during this time of year also lower the volatility of any nucleating vapors, making the process more efficient?

Line 441: Utqiaġvik is misspelled.

Line 465: Could the authors mention the differences in the definition of the onset of nucleation between these studies?

Sect. 3.1.5: The section is titled “Seasonality of Growth Rates and formation rates” yet no seasonality of formation rates is presented. I would suggest adding a subpanel to Fig. 4 which shows the seasonality of formation rates and mentioning the seasonality in the text. There should be a space between nm and hr^{-1}, also in the figure axis label. What sites were included in Kerminen et al. (2018) for the Arctic?

Sect. 3.1.6: I am curious why the authors did not compare Cv to the results from Beck et al. (2021) as well. Observations from MOSAiC are highly valuable in this context but Beck et al. was from Gruvebadet at the base of Mount Zeppelin. The MOSAiC year was also shown to be anomalous in terms of the Arctic Oscillation and transport from Eurasia during the winter months (Boyer et al., 2023). The caption of Fig. 5 reads as if the measurements from Boyer et al. cover the years 2022-2024.

Lines 510-511: Could the other species possibly be HOMs? Which were reported by Beck et al. (2021) but not Boyer et al. (2024).

Header of Table 1: The unit for CS needs a superscript. The unit for solar insolation is given as hW m^{-2} elsewhere, also should there not be a space between hW and m^{-2}?

Caption and label of Fig. 7: I am wondering the term “Nucleation site” is confusing and would it be better to express it more generally as “Count per grid box” as expressed in previous publications (Heslin-Rees et al., 2024). Is the NAIS defining the nucleation sites or is it HYSPLIT?

Lines 575-578: I would encourage the authors to include their results here as this is an interesting finding that chlorophyll a exposure is not correlated to DMS.

Lines 578-583: Can the authors give examples of “various other meteorological and environmental factors”? Also, could there any methodological factors influencing the lack of correlation between chlorophyll a exposure and DMS, such as inaccuracies in HYSPLIT and satellite retrievals of chlorophyll a due to sea ice and cloud cover?

Lines 585-588: I would suggest removing this sentence as it does not add to the discussion nor explain their results, although this is only my opinion.

Lines 589-603: The authors do a nice job of explaining the differences between the Greenland Sea and the Barents Sea for NPF source regions, although another region highlighted in Fig. 7 is the Arctic Ocean. Could emissions from sea ice, e.g. iodine and sympagic algae be contributing to this observation? This might be worth mentioning.

Lines 606-608: Here is an example of where quantitative statistics could be used to support the results. For instance, the distributions of solar insolation on NPF vs non-NPF event days could be compared or the anomaly of the CS to the seasonal average could be analyzed for NPF event days. Such an analysis would make their justification of the simplified predictive model more robust and convincing.

Lines 613-622: The units for solar insolation are sometimes given as hWm^{-2} and sometimes as Wm^{-2} , additionally there should be a space between W and m^{-2} otherwise it reads as if both units are in the denominator. Units for CS are missing.

Line 617: I am curious how this relationship is for the Dal Maso method, does it still hold?

Fig 8: I would suggest making the two subpanels the same size and please use a color-blind friendly color bar.

Lines 627: Could the authors quantify how much less likely NPF events are outside of the ROI?

Line 693: Another example of where quantitative statistics would help as this "substantial predictive value" is not demonstrated.

Line 701-703: Figure 13 is not referenced in the text. Should it be referenced in this sentence?

Line 709: Why not be precise with an exact number of NPF events that grew over 25nm?

Line 710: In the Arctic summertime, the lack of accumulation mode particles can drive the supersaturation to high values, activating small particles as the authors detail nicely, thus 25 nm is a lower limit. Could the authors give the number of NPF events that surpass other thresholds for activation, such as 60, 70, or 80 nm (the exact thresholds are left up to the authors). This would give a range of the number of NPF events that could contribute to CCN.

Line 712-713: Figure 12a is not referenced in the text, should it be referenced in this sentence? What instruments were used to calculate the mean and median diameter and how were these calculated? Using the mean and median, it appears that the diameter reaches 25 nm after approx. 30 hours. The shading on the lines (which is not mentioned in the figure caption) appears to reach 25nm after 20 hours.

Line 753: This accuracy is not quantified.

Supplement:

Fig. S1 caption: Could the authors list the names of all the parameters in paratheses? The authors state 5 tubing parameters but more than 5 are given.

Fig. S2a: The legend significantly overlaps with the data and the abbreviations in the legend are not given.

Fig. S6 caption: What is the meaning of “These are overlapping – improvement representation”?

S2.5: The authors state “There are numerous papers which highlight this common discrepancy.” but none are cited, please cites numerous papers to support this statement.

S2.5 below Fig. S9: Fig. X? Should this be Fig. S11?

Fig. S10: The caption does not explain the figure at all. Please rewrite the caption to accurately describe the figure. Maybe this text was meant to be included elsewhere?

Y axis of Figs. S10 and S11: What is dndlDp? Should this be dN/dlogDp as used elsewhere in the manuscript? From the caption, I gathered this is the slope of the regression line, but “Coef.” is listed in the label. Finally, “(DMPS/NAIS-5)” reads as if 5 was subtracted from the ratio of the DMPS to the NAIS, although it was clear from Sect. 2.2 that it is NAIS5. Please adjust the y axis label to be clearer.

Fig. S12: There is no (b) panel in the figure. Is “N int max” in the title supposed to indicate $\Delta N_{max, 2.8\text{-}5}$?

Fig. S13: What is dNint,max and d_N_int_max? These terms are not defined. Should the ratio for the mapping of g1, g2, and g3 be 1:1:1 since there are three groups but only two numbers in the ratio?

Fig. S16: Should the units for the y label be in nm? Also, no color bar is given.

Fig. S17: What are the vertical dashed lines in this figure?

Figs. S2, S7, S8, S16 present a lot of information, therefore I would suggest making these figures larger, possibly the width of the page. This would make it easier for the reader to grasp all the details presented in these nice figures.

A bibliography is missing from the supplement.

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