Collective geographical eco-regions and precursor sources driving Arctic new particle formation, J. Brean et al. Responses to RC1.

Note: Figures in the manuscript and SI are referred to here as *"Figure X"*, figures included in these review responses, but not in the manuscript and SI are referred to as *"Response Figure Y"*. Review comments are displayed in blue, and sections that have been added to the text are coloured *green*. We thank the reviewer for their insightful comments and provide responses below.

General comments: This manuscript analyzes atmospheric particle formation and growth rates for six Arctic sites.

The manuscript provides some useful scientific contribution associated with new particle formation (NPF) in the Arctic. It is a similar idea like the paper from Sellegri et. al., 2019 [Atmosphere 2019, 10, 493; doi:10.3390/atmos10090493] for different high mountain research stations. This paper was not cited. a reference to this paper would have been very helpful here and also a short introduction of the theory to explain the key processes. This is totally missing. The manuscript concentrates too much on statistics and source area and the information on the specific aspects for the different locations are more or less missing. This is much better explained by the Sellegri paper. Some changes in the structure and presentation would be nice. The following questions/concerns should be satisfactorily addressed prior to final publication.

Thank you for providing these comments. Alongside addressing the specific comments below, we include a reference to the highly useful Sellegri paper in the manuscript in the following sentence

"Motivated by the lack of studies comparing NPF at these sites simultaneously, **and following in the stead of previous publications studying multiple sites in different environments (e.g., Sellegri et al., 2019)** with calculations of the key parameters of particle formation…"

Specific comments:

Figure 1: show the general seasonality of the different parameters: J10, GR, CS and Q. here is missing the data availability of the different stations – is the time period the same and the amount of data's for the different stations. This figure is too general and only J10 and Frequency have the same signature.

We include this figure to indicate seasonal variations in NPF frequency and formation rate, which leads firmly into Figure 2. We would not necessarily expect the same signature to be seen in the other features (GRs, CSs etc.) and the lack of a summertime maximum in growth rate across the Arctic is a useful finding. As the data availability is not included, we now include our data availability as Figure 1 (see below).

Figure 2: Here is the information, separated for the six stations and also not really clear message. Different values for the parameter at different stations. It would be better, to use Figure 1 to say the NPF Frequency is highest in June/July/August and present in Figure 2 the data sets only for these three months. Then the differences between the station can be better explained….

This is exactly what this figure is. The figure caption states this explicitly

"Figure 2: Characteristics of NPF events per site in the months May through August inclusive, showing…"

The title to section 3.2 also indicates this

"3.2 Spatial variation of summertime NPF features"

Which begins:

"The site-by-site variation in summertime NPF event characteristics is shown in Fig. 3…"

To make this clearer, we update the abstract as follows

"…and particle formation rates themselves vary greatly between sites, highest at Svalbard, and lowest in the high Arctic. **Summertime** growth rate, condensational sinks…"

Figure 3: a specific inside view in the source area seems better instead of this extreme general picture over the entire data set. For me is here Figure S5 much better and Figure 3 should remove for Figure S5.

Good point. We initially opted for the combined figure initially, but now realise it obscures some useful information. We have moved the six-panelled figure S5 now to be in the main text, and we have appropriately amended the figure captions. We agree this figure is far easier to interpret.

"The CWTs weighted by Q for each site are plotted in Fig. 4 (CWTs across the whole Arctic region in Fig. S5)"

Figure 4: is very clear and good described in the discussion (starting L253)

Thank you for the kind comment

Figure S1: this is mandatory in the manuscript, and not in the supplementary! But you see the limited data basis and also the lack of data from different station for different time period. This long measurement period from Zeppelin to compare with other station with very limited time span seems critical and should be better discussed in the text. Only 2015 show an observation overlap from 5 station. A specific discussion of this time period is here recommended. Is the result from this period similar / same for the entire period?

Very good point! We include Figure S1 in the text now as Figure 1. We also reformatted the figure so it can fit nicely in one column. We also provide some discussion of both the limited data coverage and the implications for the results, as well as showing the size distributions for these periods of overlap in the following segments, in the methodology:

"There is limited data overlap between the sites, with best overlap during 2015, where data is measured for several months at all sites except one. The mean size distribution from each site for this period, alongside the mean across all time periods is plotted in Fig S1."

In the results:

"Figure S1 shows the average size distribution during the period March – July 2015 where data was being collected at all sites except ALE, where the data for March – July 2013 is shown. All sites have two distinct modes, an Aitken mode peaking somewhere between 20 to 50 nm, and an accumulation mode peaking somewhere from 100 to 200 nm. The average across all periods is also shown, for which the distributions are similar, except ZEP which compared to the whole period of data availability, has a substantially larger mode at ~20 nm in this 2015 period. The size distribution at ALE and VRS shows overall low particle counts, especially at ALE. The two Svalbard sites, GRU and ZEP have similar size distributions, while those at TIK show a large Aitken mode, and UTQ shows a large accumulation mode."

In the discussion:

"These results cover a multi-year period across the Arctic. We highlight that some of these sites have limited data coverage (Figure 1) and the periods of data overlap between sites are limited, although the size distributions for these periods of overlap are similar to the average across all periods (Figure S1). We also note the inherent uncertainty in particle size distribution measurements between sites, especially in both the <20 nm size range, which is particularly important to these NPF studies (Wiedensohler et al., 2012)."

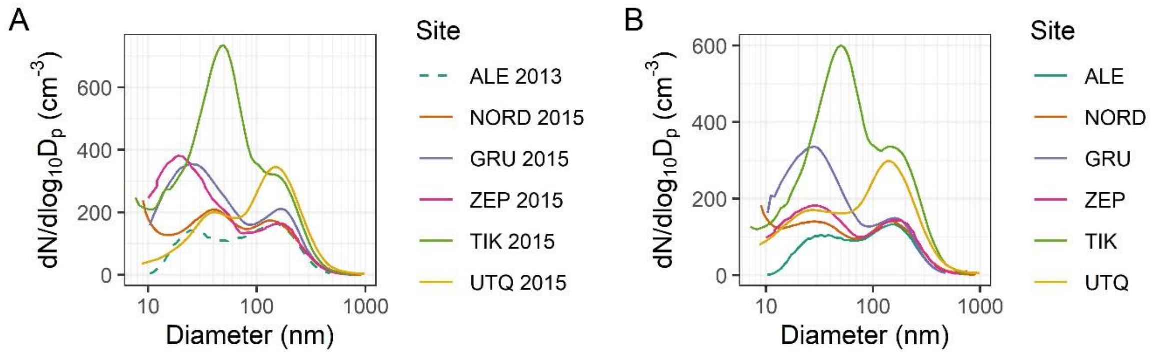

Figure S1: Average size distributions for (A) the months in 2015 with data overlap. ALE shows the data for the equivalent months in 2013, (B) shows the average size distribution for each site for all data.

Figure S3: why as example only type 1,2,3 for Tiksi – is that a typical signature or a special in compare to the stations?

We include the data from TIK here as they are rather typical of Arctic NPF across all the sites. The growth rates are slightly higher than other sites, and the difference between the three types of days is most evident. We include just one site as all of the datasets have bins at different diameters, and this saves us from any uncertainties induced when manipulating the datasets into one diameter scale to plot them on one contour.

L66: It is not enough to write, NPF is a deeply complex process – a little bit more on the theory and main processes would be very helpful

True. We include the following sentences and remove the phrase "complex processes" as it is not a particularly useful wording.

"Different measurements at Arctic sites show a strong annual cycle in aerosol characteristics, largely dictated by new particle formation (NPF) (Tunved et al., 2013; Dall'Osto et al., 2017a; 2018a; 2018b), **a process characterised by a sudden burst of nanometre sizes particles in the atmosphere, followed by their growth to larger sizes. The initial formation of these particles is driven by the clustering of gases in the atmosphere to form clusters at a rate faster than their losses due to evaporation or condensation, the second step is driven by both coagulation, and condensation of vapours with sufficiently low vapour pressures to condense down on new particles (Lee et al., 2019)**"

L81: It is here missing to mention, what are the key parameter for NPF in the Arctic. Are the same like in other regions or not.

We highlight the key parameters in the below section:

"**The key parameters driving NPF in the Arctic are not well understood. In polluted locations the surface area of pre-existing particles often dictates NPF occurrence (Lee et al., 2019), however, in remote locations condensation sinks are consistently low (Sellegri et al., 2019), and concentrations of precursors and solar radiation intensity may be key in dictating NPF frequency and intensity, however**, ~~understanding Arctic NPF is therefore a deeply complex problem~~ with multiple potential mechanisms and many poorly understood sources of precursors from the many and varied eco-regions, **Arctic NPF demands further study**"

L82: The sentence should end with dot, that is missing

This has been fixed

L89-105: The table S1 on the station is not complete, the used instruments is here also recommended, including the specific information, whether the different systems at the different stations means special constrains for the data analysis. How big are the differences between TSI 3034, TSI 3772 CPC and twin DMPS, custom built SMPS and TSI 3010 CPC. This could show very easy in a table…

This is an important thing to highlight. We have no reliable estimates of the uncertainties for each individual instrument, but these will depend on both differences between instruments, and the corrections performed on the data (CPC counting efficiency, DMA transfer function, pipe losses etc.) which are especially pertinent for the smallest size fractions. Previous intercomparison works show that different inversion routines account for some few percent difference in the size distribution, while differences between instruments from different manufacturers are within 10% difference for the 20 – 200 nm size range (Wiedensohler et al., 2012). We include the names of each instrument, as well as the appropriate size ranges measured in Table 1 as well as in the methodology section. Further, we move Table S1 into the main text. We also include a brief discussion of uncertainties in the following sentences

"Intercomparison workshops have shown differences between instruments measuring particle size distributions to be within 10%, increasing at smaller diameters (Wiedensohler et al., 2012). This produces some uncertainty when we are comparing particle formation rates and growth rates of particles in these smaller size regimes, but this uncertainty is substantially smaller than the differences in particle concentrations between sites."

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjäraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J. A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Grüning, C., Faloon, K., Beddows, D., Harrison, R., Monahan, C., Jennings, S. G., O'Dowd, C. D., Marinoni, A., Horn, H.-G., Keck, L., Jiang, J., Scheckman, J., McMurry, P. H., Deng, Z., Zhao, C. S., Moerman, M., Henzing, B., de Leeuw, G., Löschau, G., and Bastian, S.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmos. Meas. Tech., 5, 657–685, https://doi.org/10.5194/amt-5-657-2012, 2012

L114: Is the condensation sinks CS the main parameter identity NPF or the particle growth GR. I think the formulas are for this manuscript secondary. The list of priority for the Arctic site seems more attractive.

As above, we opt to move this table into the main text as we agree that it is important.

L143/144: The explanation, why only type A and type B are used for the identification of NPF, is missing.

We include the following sentence in our methods section

"Formation of particles at the smallest measured sizes is a key characteristic of NPF and is required to calculate formation rates reliably. There is also a chance that Type C events include particles not formed secondarily, but just shows growth of primary particles, and thus we neglect to include Type C events in these analyses"

L184: the discussion of the spatial variability is in general attractive, but the information in the following lines up to 245 is very unstructured. Here a table with the different mean parameter for the three regions makes the discussion on the differences and explanation of reason much easier.

We opt to not include the extra table here as these data are already presented in both figure and the main text, but we expand the discussion of spatial variability in the following

‘Here, ALE and VRS are discussed together as “high Arctic”, **as both of these sites are high latitude sites with similarly low J_{10}, GR, and CS values (Fig. 3),** GRU and ZEP are talked about together as “Svalbard” sites **as they are co-located and surrounded by the same open and ice containing ocean, with similar J_{10}, GR, and CS**, and **although dissimilar in J_{10}, GR, and CS, the low latitude** TIK and UTQ are seen to represent the “continental Arctic”’

L249/250: I see also a variation of CS at the different sites and a focus to the high frequency period of the NPF could be bring a better inside view.

As mentioned above, the analysis here does indeed focus on the summertime period. We update the sentence in question to include a reference to the condensation sink

“The results reported in this paper highlight the seasonal variation in Arctic NPF (Fig. 2), as well as the variation between different measurement sites during the summertime with J_{10}, GR, **CS,** and Q…”

Figure 5: I don’t see a big motivation for this figure, could be remove

+

L253-255: what is the motivation for Figure 5 and this sentence? Please explain it.

We include these sentences as well as Figure 5 (now Figure 6) as we feel they accurately represent the complexity of Arctic NPF. We update these sentences as follows

“We show that the vapours which drive particle growth each of these sites often, but not always ~~have strong source regions~~ **coincide with air masses flowing over particular, directional source regions** (Fig. 4). NPF in the Arctic atmospheric boundary layer is occurring within air masses flowing over vastly different Arctic eco-regions, these being regions of open ocean water, consolidated and open pack ice, snow-covered land, and non-snow-covered land (Fig. 5), **reflected in the variety of mechanisms to be seen in molecular scale measurements of new particle formation and growth (Baccarini et al., 2020; Beck et al., 2020).**”

We also update the figure caption to properly represent what is included in the figure:

“Schematic illustrations of the sea-ice, microbiota, sea-to-air emissions and New Particle Formation (NPF) occurring in the typical summertime stratus-topped Arctic boundary layer. Vertical red and grey bars broadly indicate Cloud Mixed Layer (CML) and Surface Mixed Layer (SML) as inspired by Brooks et al. (2017). The grey box indicates known possible NPF gas precursor from potential Arctic natural terrestrial and marine sources drown below.”

Brooks, I. M., Tjernström, M., Persson,P. O. G., Shupe, M. D., Atkinson, R. A., Brooks, B. J. (2017). The turbulent structure of the Arctic summer boundary layer during The Arctic Summer Cloud-Ocean Study. Journal of Geophysical Research: Atmospheres, 122, 9685–9704, https://doi.org/10.1002/2017JD027234

L303-311: Here the message is not clear enough. Figure 1 for example show a high seasonality and for the different parameter not a complete peak for summer, sometime also NPF events in winter. Too much statistics is here not perfect. A specific final statement, what is the key parameter for the NPF event in the Arctic and what are the difference between the three locations would be helpful.

We cannot identify the sole driving factor from this dataset, as it does not seem to be something evident from particle size distribution data alone (such as the condensation sink). The summertime peak in frequency and formation could be due to more intense solar radiation, a lack of Arctic haze, or higher precursor emissions due to melting sea ice and higher biological production in these warmer months, or some combination of all of these. We update our closing statement appropriately

“**While back trajectory analyses can point towards these source regions over long-terms, we still do not know the driving force behind NPF at these sites, as it is likely a combination of precursor emissions, photochemistry, ion pair production, temperature, and pre-existing**

surface area of aerosol. Measurements of particle size distributions down to critical cluster size and detailed chemical measurements are required to properly understand NPF at these sites.”