Referee comments 1

Review of "Physicochemical Characterization and Source Apportionment of Arctic Ice Nucleating Particles Observed in Ny-Ålesund in Autumn 2019" by Li et al., submitted to ACPD

This study presents the measurement results of ambient INP concentrations and related aerosol properties during the NASCENT campaign in Ny-Ålesund, Svalbard in October-November 2019.The paper describes the complexity of ice nucleating particles (INP) in Arctic coastal environment. Physicochemical parameters have been analyzed and tested and are ice nucleation temperature, heat-lability, and fluorescence activity, which were correlated with environmental parameters such as wind speed, wind direction, temperature, and snow coverage. The results are highly interesting and assume high-latitude dust sources from long-range transport that could be responsible for INP enrichment. This paper should be published after some minor changes:

We thank referee 1 for the valuable feedback on our manuscript acp-2023-18. In response to the questions and suggestions, please find our answers and revisions listed below. **Referee comments are reproduced in bold** and author responses in normal font; *extracts from the original manuscript are presented in red italic* and *extracts from the revised manuscript in blue italic*.

2.1 measurement location: Please provide already here the GPS coordinates for the aerosol container and the GVB station.
We now add the GPS coordinates for the aerosol container and the GVB station in the revised manuscript (see lines 84-85). In addition, we state the direction and distance relative to the container see caption of Figure 1 in the revised manuscript.

Instead of figure 1b, a map would be more helpful showing the distances and a wind rose including wind speed and wind direction frequencies.
We agree and have now added distances and a wind rose for the relevant dates discussed in this paper (see Fig. 1b in the revised manuscript). In addition, we adjust the descriptions to *"Local sources of pollution have a limited influence on the measurement sites during the measurement period, given the predominant southeasterly wind at the aerosol container and prevailing southwesterly winds close to the GVB observatory station (see detailed wind pattern in Fig. 1 b)"* (see lines 85-87 in the revised manuscript).

Figure 2 is valuable for the interested reader but might be shifted to the appendix.
We appreciate this comment, but we decided to keep Fig. 2 in the revised manuscript because it is difficult to introduce the experimental setup in Section 2 without referring to a figure and we believe it is important for reproducing such an experiment in the future.

2.2 INP sampling techniques: You might discuss the different sizes and number of droplets been investigated with the three techniques (WT-CRAFT, HINC and DRINCZ). What is the impact of both parameters on the error bars, the homogeneous ice nucleation temperature and the limit of detection? Please provide a discussion which makes these differences and their impact on the results more transparent for the reader.
We agree with the reviewer's comment. As shown in Equation (1) in the revised manuscript, both droplet size and number impact the error bars and the limit of detection of the measurement. Additionally, the homogeneous freezing temperature is impacted by the droplet size, with smaller droplets requiring lower homogeneous freezing temperature (Koop and Murray, 2016). To make the comparison clearer, we add Table 1 in the revised manuscript, summarizing the features of different INP sampling and measurement techniques along with their LOD. The table is now discussed in lines 106-111 of the revised manuscript: *"To investigate the ambient INP concentrations in immersion-freezing mode, we used different INP sampling and measurement instruments introduced in the following subsections, which provide a large range of sampled particle sizes, time resolutions, freezing temperatures, and hence different INP detection limits (see Table 1).*

In particular, the droplet-freezing techniques (see Sections 2.2.1 and 2.2.2) have different limits of detection (LOD) due to the different droplet sizes and numbers in the experimental setup."

2.3. Heat treatment: As already discussed by many authors, the impact of heat treatment is an ambiguous procedure (please quote the respective literature), e.g., some low-molecular INM from pollen can be rather heat stable (T_{on} < -15 °C), while high-molecular INM agglomerates from the same source are losing INA due to heat treatment. A more reliable technique is digestion of the INP/INMs with enzymes, which will give evidence for the presence of proteins. Treating ice nucleation active samples with enzymes (e.g. Kozloff et al., 1991; Burkart et al. 2021, Pummer et al., 2012; Felgitsch et al., 2019), chaotropic reagents (e.g. Pummer et al., 2012; Fröhlich-Nowoisky et al., 2015), or a strong oxidizer (e.g. H_2O_2, Gute et al., 2020) to investigate the nature of ice nuclei has been performed in the past.

The following references (Hill et al., 2016 and Pummer et al., 2015) suggest using heat treatment to evaluate the contribution of heat-labile materials to the INP population, representing proteinaceous IN components that are degraded after heating. Heating is a qualitative approach to investigate the presence of heat-labile/biological components, although we notice the possible ambiguity of this method suggested by the referee. As suggested by the referee, more definitive approaches e.g., Burkart et al. (2021) treated sub-pollen particles with a protease to investigate the quantitative connection between the IN activity and protein concentration. Pummer et al. (2012) treated the most efficient pollen-INPs to identify the proteinaceous and non-proteinaceous compounds. In this work, we were not aiming at identifying the type of heat-labile/biological particles. Instead, our work focused on detecting the presence vs. absence of those particles by using the heating method. Additionally, a finer assessment of the types of biological nucleators in our samples was not possible due to the limited sample volume available from the campaign. We wanted to use the samples to investigate the storage impacts and further chemical and spectroscopic analysis that are presented here. We removed statements about the specificity of biological nucleators in the revised manuscript other than implying that they are heat-labile. We also include a statement in section 2.3 (lines 197-199) to clarify this point *"We note that such heat treatment could exclude lower molecular weight samples yet still imply that proteinaceous aggregates are present (Seifried et al. 2023). Thus, any effect of heat treatment on the INP concentration would be due to the contribution of heat-labile particles from biogenic sources."*

3.1 INP concentrations: In figure 4, it is interesting to note that in the range -5 to -15 °C the INA decreases a lot, which might be interpreted as proteinaceous aggregates (Seifried et al. 2023). In figure 4c the legend with full circles is missing. Eventually, the authors might use different symbols related to the different colors. In figure 4c also error bars are missing.

In the revised manuscript, we have now used different symbols corresponding to different colors. In addition, the error bars and the legend for different symbols have been added in Fig. 4c (which has been moved to Fig. 6c in the revised manuscript also inspired by the reviewer's comments for a better comparison, see below).

In the text you mention a sensitivity of the proteins due to cryo-storage. Please provide explanation for this effect.

Two reasons are suggested in Beall et al. (2020). We now add the following description in lines 352-356 of the revised manuscript *"The number of small organic INPs could be reduced due to aggregation when enriched solute becomes incorporated into the ice phase during storage. Additionally, as the solution phase is enriched during freezing, smaller INPs may be absorbed onto the surface of larger particles, thus resulting in the coalescence of the INPs (Beall et al., 2020). However, a clear mechanism for the INP losses after cryo-storage is not reported since they lack the identities of observed INPs."* However, additional reasons are responsible for the reduction in INP concentration for PM10 filter samples. We add the following reasoning sentences in lines 358-362 of the revised manuscript *"The above reasons, however, would not explain degradation in the PM_{10} samples; as such, we believe that the lower N_{INP} in the PM_{10} samples is indicative of a size dependency since the impinger samples include particles larger than 10 μm which are excluded in the PM_{10} samples. This conclusion is also supported by the N_{INP} from impinger being systematically higher than those from the PM_{10} samples (see Fig. 3)."*

Table 1: The Pearson correlation coefficient is listed. Please, explain why a r > 0.5 indicates strong correlation. How this category has been defined? Also, for N_{INP} (T = -6 ℃) / n_{AC+ABC} the 0.63 should have two asterixis.

Thank you for pointing this out. The double asterix is added to n_{AC+ABC} = 0.63. Regarding the strength of the Pearson correlation coefficient, there is no fixed definition of the level of correlation corresponding to a specific coefficient value. In addition to the authentic correlation, the absolute correlation coefficient also depends on the quality of the data set. Considering the similarity in the format of the data set obtained from aerosol field measurements, we took the classification of correlation strength indicated in Lacher et al. (2018); Paramonov et al. (2020) and Rinaldi et al. (2021). As such we base our threshold of 0.5 from the mentioned literature. But we would support this further because 0.5 indicates that more than 50% of the data could be predicted with this correlation.

Figure 7 and 8: The labels on the axes are rather small.

The label size in Figs. 7 and 8 are increased in the revised manuscript.

Figure 9: Please use different symbols related to the different colors.

Figure 9 is changed according to the requirement in the revised manuscript.

Table 2: What means "NO"?

"NO" means number. It has been changed to *"#"* for consistency in the revised manuscript.

Figure 11 and 12: The labels in the figures are much too small and are unreadable.

We increase the label size and readability of Figs. 11 and 12 in the revised manuscript.

Figure B1: Please describe the figure in more detail. The particles have the typical shape of NaCl crystals. Please mark the spots in the SEM where the EDX has been recorded. If available an EDX mapping might be shown.

The particles have a typical sea salt crystal shape as shown in the Fig. C1 in the revised manuscript. Our CCSEM-EDX analysis has covered 0.5 mm by 0.5 mm area on the Al foil substrates to analyze sufficient particle population. Therefore, all particles in Figure C1 have been analyzed by CCSEM-EDX. The figure caption has been extended to describe the image in more detail in the revised manuscript. We have also added Fig. C2 to show the representative EDX-mapping of representative particles in the impinger samples.

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