

Answers to the review for our manuscript “Ice Nucleating Particles in Northern Greenland: annual cycles, biological contribution and parameterizations”

We very much thank the reviewer for helping to advance our manuscript by doing this review. Below, find the original comments in black, [our answers in blue](#) and new text added to the manuscript in “*blue, italic and quotation marks*”. All line numbers refer to the new version with tracked changes.

Major comments and suggestions:

Terminology:

1. The authors classified samples using the terms “summer,” “winter,” and “mixed” types. However, these samples were originally classified based on the exponential decay slope values and not by the sampling seasons. Indeed, they initially termed them Fletcher and Cooper types. They were termed because they were more frequent in summer or winter. Then the authors discuss the fraction of “summer” and “winter” types in each season. The terminology is confusing, e. g., line 368 “with summer and winter type dominating summer and winter months, respectively.”
2. The term “bio-ratio” is also somehow misleading. Line 170-172 says, “However, it should be noted that not all biological INPs are equally sensitive to heat. Nevertheless, overall heat-lability is thought to be more associated with biological INPs than mineral INPs, and we will use the term biological INP in this study to refer to heat-labile INP.” The “bio-ratio” is not a real bio-ratio but the fraction of heat-labile INPs. However, in section 3.4., the authors discuss the “bio-ratio” as they are actual bio fraction, e.g., line 395 “that roughly at least half of all INPs during that period were comprised of non-proteinaceous biological material.” The discussion should base on the fraction of heat-labile INPs. Otherwise, more discussion will need to prove that heat-labile INP is equal to biological INPs.

For the above two cases, I agree with the discussion and the interpretation and will be acceptable to mention the “summer” and “winter” types are dominant in summer and winter, respectively, and the bio-ratio mostly represents the fraction of biological particles. On the other hand, the terminology sounds too simplified to interpret the meanings. So, I suggest reconsidering the terminology. Or at least, the discussion should base on these real meanings (e.g., the number fraction of heat-labile INPs but not bio ratio).

[Answer to 1\) Concerning the terminology of “summer” and “winter” types, not only were “summer and winter type dominating summer and winter months, respectively” \(referring to your citation of our manuscript\), but also there was quasi no winter type in the summer months and vice versa \(see line 319 ff and Fig. 3\). To our understanding, this justifies the use of “summer” and “winter” to name the respective cases and parameterizations.](#)

Answer to 2) We were careful to define the “bio-INP” as proteinaceous INP, only. But you are right that our naming of “bio-INP” was somewhat broad and that “heat-labile INP” describes it more tightly. Following this, we renamed this parameter and also “bio-ratio” (to “heat-labile ratio”) in text and figures and changed related text accordingly (see Sec. 3.4 ff).

Section 3.5 A case study:

The case study section discusses several hypotheses and rejects the first two hypotheses by the authors (line 418 and 434). It is acceptable to discuss any hypotheses, but in my personal view, they use too much space for the discussion to decline their hypotheses, including Figures 9-11, which decreases the readability of the conclusion. At least, they can shorten, and most figures can be replaced in the appendix or supporting information. In addition, this manuscript has so many appendix figures instead of supporting information. I would suggest considering using supporting information as well. This comment is a suggestion that potentially improves the paper and is not mandatory.

We agree with your suggestion and indeed changed the appendix to a separate file with the supporting information (SI). Concerning Figs. 9 – 11, we kept them in the main text. It may have been confusing as these figures appeared within the text of the “summary and conclusion” section, while they were discussed before, but this will not be an issue in the final printed version, should this manuscript be published. Still, for the new clean version, we moved all figures up so that they are shown closer to the location where they are mentioned in the text. (Please refer to the new clean version and the new SI to see the implementation. In the version with tracked changes required for the review process, there is still the old appendix included and the figures were not moved, yet.)

As for more details on keeping Figs. 9-11: Fig. 9 already is an example consisting of two panels, and the other 15 panels were in the appendix before and are in the SI now. Fig. 9 is needed for illustrative purposes. Figs. 10 and 11 show all the parameters we used, and while there is no positive answer (we did not find the origin of the INP), we feel that also negative answers can help in showing which parameters do NOT correlate to INP concentrations. Therefore, we prefer to keep these two plots in the main text, too. Still, as before, the more space consuming figures concerning the results of the correlations panels were in the appendix before and are now shown in the SI.

Specific comments:

Line 17: “also a higher cloud ice fraction was observed in satellite data for April 2020, compared to April 2019.”

I suggest revising this last sentence in the Abstract, which is somehow awkward and is not a major conclusion of this study.

We agree that this was not a major conclusion, but it supported our finding that INP concentrations were higher in April 2020 compared to April 2019. Therefore, we moved it up so that it now appears two sentences earlier as an added observation and not as a final (concluding) sentence.

Line 249 “Hereafter the time period with a snow depth below the threshold is referred to as snow-free months,”

Similar to the general comments, they are not “snow-free month,” but there was snow with < 80 cm. I suggest using a better term here.

We made some related changes, e.g., exchanging “snow-free months” in the abstract with “*months with low to no snow cover*”, “snow-free and snow-covered months” to “*between months with differing snow-cover*” (line 242). All further occurrences related to our study were replaced by “*quasi-snow-free months*”.

Line 350 “And, these are the two slopes...”

Is this sentence “There are two slopes...” (?) Please check it.

“And these are the two slopes...” is correct. We are referring to the values used in the previous sentence, hence the “And” (i.e., to -0.3 and -0.6, i.e., the values we used to categorize the spectra). But we removed the “,” after “And” – we assume that this was what caused your confusion.

Line 371 “60% ot” typo?

Yes it is a typo. Corrected.

Line 408-409 “The altitude threshold of 250m was applied in order to locate potential source regions within the planetary boundary layer”

Over the main part of Greenland, the altitudes from the sea level are mostly > 250m, but the air mass can also pass through near the ground (ice or snow) surface. They may contribute to aerosol sources. Some explanations may be helpful.

We reformulated for clarification: “The altitude threshold of 250m *above ground* was applied ...”.

Line 475 What is PDF?

Thanks. It means probability density function. This occurrence was exchanged to “*log-normal fit*”, so PDF only occurs in Fig. 4, where it is now explained in the caption.

Line 489 what is LES?

Thanks, we added “*large eddy simulation*”.

Line 509. “...value derived from it, is statistically more certain than a low one.”

Is something missing in this sentence?

The whole sentence is “The idea behind $F(T)$ is to account for the fact that a high f_{ice} value, and the N_{INP} value derived from it, is statistically more certain than a low one.”

This is, to our understanding, a complete sentence. In the part you cite, we say that N_{INP} derived from a higher f_{ice} has a lower uncertainty compared with one derived from a lower f_{ice} . This can e.g., be seen from Fig. A1, which shows the error bars of these two instruments. And the weighting factor $F(T)$, which was first introduced in the sentence before the one we discuss here, accounts for that.

Figure 1. I suggest indicating the period used in the case study.

Done.

Figure 1. During April 2020, there were higher INPs even at -20C than in April 2019, which may suggest that there were significant numbers of aerosol particles in the month. If so, the high number of aerosol concentrations simply increases the number of INP at a high temperature rather than changing particle species. Is there any available data to check the particle concentrations?

INP are only a very minor fraction of all aerosols. There were past efforts from different groups, including ours, to simply relate the total number of aerosol particles to INP. This has never worked out well, or rather not at all (e.g., Gong et al., 2020, 2022; Hartmann et al., 2021; Li et al., 2022), except sometimes, when only accounting for particles larger than 500 nm (DeMott et al., 2010). This is likely related to the fact that, depending on the aerosol, the majority of aerosol particles will have different sources than INP: e.g., during the new particle formation period as well as during the Arctic haze period, particle concentrations will be high, but this is uncorrelated with INP concentrations (e.g., Wex et al., 2019). This is, because INP neither

originate from new particle formation, nor does aerosol emitted from burning contribute INP which are active in the here examined temperature range (as described e.g., in Chen et al., 2018; Tarn et al., 2018; Welti et al., 2020; Yadav et al., 2019; Tobo et al., 2020).

While data to do the requested comparison is not available for the two months in question here, based on past experience, we also do not expect to find a correlation.

Figure 4. I suggest adding a color legend in addition to the text.

Done.

Figure 6. It looks like not all samples were plotted (e.g., July 2018). Why?

We assume you refer to Fig. 7, here. As filter collection and related evaluation only started in July 2018, there are no heated samples available for July 2018, hence not data can be plotted. Additionally, the lower the freezing temperature, the fewer data-pairs (for untreated and heated samples) are available, hence no data can be plotted, either. This was already mentioned in the text (now in line 408): *“At -20°C , not enough data points exist to provide an insight on the fraction of biological material.”*

Figure 11. The colors in the plots are difficult to distinguish.

This plot shows data for April 2019 in dark orange, and for April 2020 in magenta, which is easy to distinguish for all authors of the study. We chose all colors in our manuscript such, that people with different types of color blindness can distinguish them, which is also the case for the colors in this plot. If you want to make an explicit suggestion, please do. Otherwise we keep it as it is.

Figure A5 (November). There are two summer spectra in November; one is the highest INP concentration, and the other is the lowest INP concentration. Is the classification correct?

You are correct that there is one very high spectrum in November. However, this does not make our classification incorrect. November is one of the three transition months, anyway. Fig. 3 shows that for November 20% of the spectra were summer-type, 20% winter-type and the remaining mix-type.

Throughout the year, our classification describes the majority of occurring cases, but a comparably simple description fitting to 100% is not to be expected in nature.

Literature:

Chen, J., Z. Wu, S. Augustin-Bauditz, S. Grawe, M. Hartmann, X. Pei, Z. Liu, D. Ji, and H. Wex (2018), Ice nucleating particle concentrations unaffected by urban air pollution in Beijing, China, *Atmos. Chem. Phys.*, 18, 3523–3539, doi:10.5194/acp-18-3523-2018.

DeMott, P. J., A. J. Prenni, X. Liu, S. M. Kreidenweis, M. D. Petters, C. H. Twohy, M. S. Richardson, T. Eidhammer, and D. C. Rogers (2010), Predicting global atmospheric ice nuclei distributions and their impact on climate, *Proc. Natl. Acad. Sci. USA*, 107(25), 11217-11222, doi:10.1073/pnas.0910818107.

Gong, X., M. Radenz, H. Wex, P. Seifert, F. Ataei, S. Henning, H. Baars, B. Barja, A. Ansmann, and F. Stratmann (2022), Significant continental source of ice-nucleating particles at the tip of Chile's southernmost Patagonia region, *Atmos. Chem. Phys.*, 22, 10505-10525, doi:10.5194/acp-22-10505-2022.

Gong, X., H. Wex, M. van Pinxteren, N. Triesch, K. W. Fomba, J. Lubitz, C. Stolle, B. Robinson, T. Müller, H. Herrmann, and F. Stratmann (2020), Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater, *Atmos. Chem. Phys.*, 20, 1451-1468, doi:10.5194/acp-20-1451-2020.

Hartmann, M., X. Gong, S. Kecorius, M. van Pinxteren, T. Vogl, A. Welti, H. Wex, S. Zeppenfeld, H. Herrmann, A. Wiedensohler, and F. Stratmann (2021), Terrestrial or marine? – Indications towards the origin of Ice Nucleating Particles during melt season in the European Arctic up to 83.7°N, *Atmos. Chem. Phys.*, 21, 11613-11636, doi:10.5194/acp-21-11613-2021.

Li, G. Y., J. Wieder, J. T. Pasquier, J. Henneberger, and Z. A. Kanji (2022), Predicting atmospheric background number concentration of ice-nucleating particles in the Arctic, *Atmos. Chem. Phys.*, 22(21), 14441-14454, doi:10.5194/acp-22-14441-2022.

Tarn, M. D., S. N. F. Sikora, G. C. E. Porter, D. O'Sullivan, M. Adams, T. F. Whale, A. D. Harrison, J. Vergara-Temprado, T. W. Wilson, J. U. Shim, and B. J. Murray (2018), The study of atmospheric ice-nucleating particles via microfluidically generated droplets, *Microfluidics and Nanofluidics*, 22(5), doi:10.1007/s10404-018-2069-x.

Tobo, Y., J. Uetake, H. Matsui, N. Moteki, Y. Uji, Y. Iwamoto, K. Miura, and R. Misumi (2020), Seasonal Trends of Atmospheric Ice Nucleating Particles Over Tokyo, *J. Geophys. Res.-Atmos.*, 125(23), doi:10.1029/2020jd033658.

Welti, A., E. K. Bigg, P. J. DeMott, X. Gong, M. Hartmann, M. Harvey, S. Henning, P. Herenz, T. C. J. Hill, B. Hornblow, C. Leck, M. Löffler, C. S. McCluskey, A. M. Rauker, J. Schmale, C. Tatzelt, M. van Pinxteren, and F. Stratmann (2020), Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern Ocean, *Atmos. Chem. Phys.*, 20, 15191-15206, doi:10.5194/acp-20-15191-2020.

Wex, H., L. Huang, W. Zhang, H. Hung, R. Traversi, S. Becagli, R. J. Sheesley, C. E. Moffett, T. E. Barrett, R. Bossi, H. Skov, A. Hünnerbein, J. Lubitz, M. Löffler, O. Linke, M. Hartmann, P. Herenz, and F. Stratmann (2019), Annual variability of ice nucleating particle concentrations at different Arctic locations, *Atmos. Chem. Phys.*, 19, 5293–5311, doi:10.5194/acp-19-5293-2019.

Yadav, S., R. E. Venezia, R. W. Paerl, and M. D. Petters (2019), Characterization of Ice-Nucleating Particles Over Northern India, *J. Geophys. Res.-Atmos.*, 124(19), 10467-10482, doi:10.1029/2019jd030702.