Answers to the review from Reviewer 1 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.

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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.

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Answers to the review from Reviewer 2 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

- Parameterization: Which fraction of your sample is within a certain factor of the developed parameterization? E.g., are you able to predict the INP concentration accurately within a factor of 10, as deviations larger than this value can impact cloud microphysical and radiative properties (Phillips et al., 2003)?

Variations in INP concentrations may indeed influence cloud glaciation, particularly when ice multiplication processes are assumed, as in the simulations in the study you refer to here. However, both, ice multiplication processes and with this the relevance of a change in INP concentration (relative or absolute) are still a matter of scientific research today and not well constrained. As such, the influence of e.g., a change of a factor of 10 on cloud gladiation is uncertain. Still, we added some text and a related table to the study, and added a plot to the SI, Fig. S12. (We replaced the appendix by an SI, please check for the figure there.) The text is as follows (line 383 ff) (for the table, please refer to the revised version of the manuscript):

“*We furthermore examined which fraction of the corresponding samples falls within a range covering one order of magnitude above and below our parameterizations. This fraction is generally well above 90%, as shown at three selected temperatures of −12, −16 and −20°C in Table 3 (for more see SI, Fig. S12), emphasizing the representativeness of our parameterizations.*“

- Case study: The linkage of an observed higher cloud ice fraction to an increased INP concentration in April 2020 as compared to the previous year is insufficient. First, to my knowledge, there is no evidence that ground-based INP concentrations can impact cloud properties observed on top of the cloud. Second, other parameters differ between April 2019 and 2020, for example, mean wind speed (potential impact from blowing snow) or a lower surface temperature in April 2019 (e.g., a greater impact of pre-activation of INPs; Conen et al., 2015). Moreover, it would be interesting to investigate if there was an enhanced impact from glacial dust sources or a higher biological activity in the ocean. In addition, in the late winter months, the Arctic haze phenomena (e.g., Shaw, 1995) can impact aerosol populations, which is not discussed here.

The impression that this part may be insufficient may arise from the fact that we were not able to pin down INP sources. Still, we did examine a broad range of meteorological data as well as back-trajectories. It is also valuable information to the research community to see what did not work out. Also, past studies show a connection from ground based INP measurements to glaciation of clouds under coupling conditions (Griesche et al., 2021) and to the boundary layer in general (Gong et al., 2022), connecting ground based and lidar derived data. Further examinations are for sure needed, but this is not the topic of our study.

As for parameters differing between April 2019 and April 2020, none of the parameters differs throughout the whole month (the parameters you mentioned differ for the second half of the month, but not for the first half), while INP concentrations are higher throughout the month. So no conclusions can be drawn from that.

We agree with the reviewer that an examination of additional factors such as glacial dust, blowing snow or oceanic activity would be interesting. But this is far beyond the scope of this study. We did examine the effect of biological activity in the ocean, and did not see an effect on a first glance, which lead us to drop this and not include it. Arctic haze can be excluded as a source for INP in the here examined temperature range, as we have described previously (Hartmann et al., 2019; Wex et al., 2019). On that, we added in the introduction (line 89/90):

"in line with the observation that Arctic haze (Wex et al., 2019; Hartmann et al., 2019), or anthropogenic pollution in general (Chen et al., 2018; Tarn et al., 2018; Welti et al., 2020), do not contribute INP that are ice active in the temperature range down to≈-30°C."

Blowing snow is now mentioned in the text, as we elaborate on in our answer to your second minor comment.

As our data will be available on Pangaea, maybe you or others can pick up the thread and add additional examinations to this two year long data-set.

Minor comments

- What is the relevance of mixed-phase clouds in the Arctic winter regarding the radiative forcing?

  We do not see what this has to do with the topic of our study and hence did not elaborate on this in order to not lengthen the text unnecessarily.

- Is there an impact of blowing snow on the aerosol filters?

  For the atmosphere, INP may be resuspended through blowing snow. However, due to the use of a PM10 inlet for the low volume aerosol sampler, snow will not have been collected. Still, we added a comment on blowing snow (line 456 ff):

  "*Blowing snow occurring in the Arctic (Yang et al., 2019) may contribute to atmospheric INP. However, in our case, due to the use of a PM_{10} inlet during filter sampling, we do* *not expect that snow was collected in considerable amounts and hence we do not expect an influence of blowing snow on our measured INP concentrations."*

- In some cases, the mentioned publications are examples and do not represent all existing literature. Please check and make use of "e.g." in such cases or complete the cited literature

  You are right: certainly not all publications we could have cited on all the mentioned topics are included. But your request would mean to add an "e.g." to quasi all citations, or at least to those where we added citations concerning more general statements. This would flood particularly the introduction with "e.g." which reduces the readability.

  We would want to leave it up to the editor to decide if we should really add this. For the time being, nothing was changed.

- You mention that blank filters were collected weekly, but do not present them here. How high were the INP concentrations from those filters and did you consider using them for a background correction of the INP concentration?

  Thanks for pointing this out. We now added respective figures and some explanatory text in the supplemental information (SI, Fig. S4 and corresponding text). (As said before: We replaced the appendix by an SI.) We refer to them in the main text at the end of section 2.2.5: "*Regarding the blank filters, measured f_{ice} were generally clearly below the respective values of the atmospheric samples and a background subtraction was not done (for details see SI, section S5)."*

  Text in the SI:

  Figure caption:

  "*Measured f_{ice} for blank filters and respective atmospheric samples for LINA (left panels) and INDA (right panels) for the month of January (upper panels) and July (lower panels). Blank filter data are always shown in black, atmospheric sample data in gray, except for INDA data for July (panel C, lower left side), for which data for the blank filters with the highest values are given in blue and the corresponding filters in cyan."*

  Main text:

  "Fig. S4 exemplarily shows measured frozen fractions (f_{ice}) for blank filters and respective atmospheric samples. Data are given for both measurement devices LINA and INDA (see main text for more details), and for all samples collected in the months of January and July. Data are shown separately for the two months, as blank filter values are generally higher in summer than in winter, an observation which was made before (Wex et al., 2019). Even within July, blank filters with the highest values correspond to atmospheric samples with higher concentrations.

The blank filter values were clearly below the values from the atmospheric samples. It also can be seen that the temperature range covered by the data from the blank filters is at lower temperatures than that of the measurements. Therefore, correcting the measurements with background data would only be partially possible. Also, a correction is done based on concentrations (Vali et al., 2019), i.e., using the logarithms of f_{ice}, such that corrections for the data used in this study are small and no influence on the outcome of our overall results is observed. Therefore, a background subtraction was not done."

- Lines 35 – 36: Reference for this statement missing.

  The sentence was adjusted slightly as given below, and the following references have been added.

  "Also, exposure of non snow-covered surfaces to the atmosphere *likely* will enhance emission of soil dust, *and therewith could* largely contribute to ice-nucleating particles (INPs) *(Šantl-Temkiv et al., 2019; Tobo et al., 2019; Sanches-Marroquin et al., 2020)."*

- Lines 94 – 97: It might be worth mentioning that also the impact of glacial dust is increasing due to retreating glaciers (e.g., AMAP, 2021).

  We adjusted the text and added the citation as follows:

  "… where snow and sea ice cover are expected to decline in the upcoming years, *glacial dust can also increase due to the retreating glaciers (AMAP, 2021)*, which will … "

- Lines 125: What is the pore size of the polycarbonate filters?

  The pore size was 200 nm. Information was added to the text.

- Line 152: What is the uncertainty in temperature regarding the 6 second time resolution of the camera and using a 1 °C/min cooling rate?

  A picture is taken every 0.1 K, but that is independent of the temperature uncertainty. Both LINA and INDA are regularly calibrated, and this is accounted for in the data. The temperature uncertainty per se is 0.5 K, determined from these regularly executed calibration measurements, for both LINA and INDA. Information was added to the text.

- Line 253: Can you quantify the variability in INP concentration better, e.g., using the standard deviation?

  We already quantified the variability of N_{INP} by giving the span between the 10% and the 90% percentile (based on values given in Table 1). Therefore, we do not think it is necessary to add also standard deviation. However, in the respective text we made it clearer that the "1 to 2 orders of magnitude" mentioned in the respective sentence refer to these percentile values. Additionally, a better quantification is now also given with respect to our answer to your first major comment.

- Line 277: Statement „Their respective INP parameterizations are often used in atmospheric models“ needs a reference.

  We added DeMott et al. (2010) and Curry & K Khvorostyanov (2012).

- Lines 333 – 334: Are there publications that can strengthen this statement („A common background of mineral dust particles throughout the year may exist“)?

  Mineral dust as a contribution to the atmosphere in the Arctic has been described. We carefully expressed this statement by using “may”, and now added the following citations: *“(e.g., Si et al., 2019; Tobo et al., 2019; Sanches-Marroquin et al., 2020)*”

- Lines 354 – 355: It might be worth explaining on which measurements the parameterizations from Cooper (1986) and Fletcher (1962) are based to understand the difference of three orders of magnitudes as compared to your parameterization.

  Concerning the data by Cooper (1986), we wrote before (line 292 ff): “In Cooper (1986), a selection of previously made measurements from literature was examined. However, it was not well described based on what criteria certain data was included or rejected. Still, when sighting this literature, it can be seen that data at higher temperatures up to −5°C was included in Cooper (1986), …”. Indeed, after repeated careful reading of Cooper (1986), we are still unable to give more detail. Should you be aware of more, please let us know and we will add it happily.

  As for Fletcher (1962), he collected literature data from a number of different cloud chambers used for atmospheric measurements. We already wrote (line 291 ff), “that Fletcher (1962) reported the value of −0.6 as the usual value, but commented that values between −0.4 and −0.8 were still common.” Similarly, concerning the value given for A (the y-intercept), he said that it is “more variable, typically being about 10^{-5} L^{-1}, with variations of several orders of magnitude sometimes occurring.” We added this information to our text at the location you indicated (see line 368 ff). Overall, this shows that the parameterization often cited as originating from Fletcher (1962) is merely a small fraction of what was really reported in this older study, which we hope to clarify.

- Line 426: In April 2019 there is a correlation coefficient of 0.86 between INP concentrations at -18°C and surface temperature.

  We had to update some meteorological data due to a new quality control check. This changed values slightly. The value you are referring to is now 0.73.

Technical comments

- Line 5: The abbreviation „VRS“ is not used in the abstract, thus should not be introduced here.

Done.

- Line 473: „biolgocial“ should be „biological“.

  Done.

- Check the use of hyphens: I believe it should be „ice-active“, „temperature-dependent“, etc.

  Done. (Also, language check by ACP will take care of those we did not see.)

References:

AMAP, 2021. Arctic Climate Change Update 2021: Key Trends and Impacts. Summary for Policy-makers. Arctic Monitoring and Assessment Programme (AMAP), Tromsø, Norway. 16 pp

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