Author answer to the reviewers regarding the article "Ice-nucleating particles in Greenlandic glacial outwash plains"

Note: editor and reviewer comments are shown in black, author answers and comments in blue and adaptations referring to the manuscript are shown in red. The line numbers in our answers refer to the line numbers in the adapted manuscript including track changes.

Referee #1

The manuscript presents a comprehensive study in southern Greenland of ice nucleating particles (INPs) in surface material of glacial outwash plains. In addition, aerosol particles sampled at several locations on and near the plains provide for a thorough assessment of outwash plains as potential sources of atmospheric INPs. Analyses of sampled material include heat and hydrogen peroxide treatments to distinguish between different kinds of INPs. Various parameters, including organic carbon concentration, bacterial abundance and soil particle mineralogy provide further insights on the likely origin of INPs in outwash material. It is a technically sound study describing in great detail (28 pages main text, 73 pages in total) work that has led to new insights. Care has been taken to relate obtained results to those of similar published studies and to provide possible explanations for particular findings.

My reading of the results is that glacial outwash dust is at best a minor source of atmospheric INPs in southern Greenland during summer, for three reasons. First, the PCA analysis (Fig. 9) of cumulative INP spectra suggests that most filter samples (atmospheric INPs) contained an INP population different from bulk samples (glacial outwash dust INPs). Second, neither aerosol particle number (> 0.5 um) nor INP concentration increased at high wind speed (Fig. 8), perhaps not so surprising as Figure B1 shows biological crusts on soil surfaces. Such crusts efficiently suppress wind erosion (Zhang et al., 2006). Although it takes in the Arctic two centuries for a biological crust to develop fully, intermediate levels of development already form within a few decades (Heindel et al., 2019; Tanner, 2025). Only at Narsasuaq plain, where the surface is apparently crust-free sand and silt, as indicated by footprints left in loose material around the solar panels (Figure B1c), there is some overlap between filter and bulk samples along the PC1 axis in Fig. 9. Third, the strongest correlation of dust INPs was with colony forming units (CFU), i.e., viable microorganisms. These thrive better on biological surfaces than on loose glacial outwash material. Biological crusts, vegetation, and leaf litter seem to cover a substantial fraction of the surrounding land area, e.g., at Narsarsuaq col and NIRS (Fig B1c,). Taken together, the evidence suggests that dust from glacial outwash plains is not a major source of atmospheric INPs above these plains in summer, except perhaps within the internal boundary layer (e.g., Fig. 1 in Dupont et al., 2021) above larger patches of bare outwash material (Narsarsuaq plain). From my point of view, this outcome should guide future studies on atmospheric INPs towards focusing more on biological surfaces. Although there is tentative pointing in this direction (lines 512 and 513, 517 to 519, 533 to 535), I would recommend to put more emphasis on this issue.

We thank the reviewer for this constructive and detailed interpretation of our results. We agree with the overall conclusion that glacial outwash dust likely represents a minor rather than dominant source of atmospheric INPs in southern Greenland during summer, and have strengthened this framing throughout the manuscript. We have added more emphasis on biological surfaces as a key INP source and the importance of future investigations in this direction. Specific changes are highlighted below:

- Abstract, from line 27: Dust from the Narsarsuaq glacial outwash plain likely acted as a localized rather than regional INP source during summer, as indicated by higher atmospheric INP concentrations above -20 °C and similarities between atmospheric and bulk dust INP spectra at the outwash plain site compared to other sites in the region. The atmospheric INP population was generally dominated by organic and biological contributions, highlighting the importance of better constraining biogenic INP emissions in the Arctic.
- Section 3.1.4:

- From line 429: The stronger correlation of n_m with CFU than with FC-based total cell counts suggest that culturable, viable microorganisms are more closely linked to n_m than total cell abundance. This may reflect the importance of microbial composition, with ice-nucleating microorganisms potentially being more abundant on biological surfaces than on loose outwash material (Lindow et al., 1982).
- From line 442: Microbiota fix carbon and nitrogen, allowing the development of more complex microbial communities, soil development and plant succession, that support more diverse ecosystems (Bradley et al., 2016; Donhauser and Frey, 2018). As microbial abundance, TOC, and n_m values are higher for vegetated sites compared to unvegetated sites (Fig. B10, *microbiology* samples), higher microbial abundance may therefore be an indicator of an environment with more advanced soil and vegetation development, that can contain additional sources of INPs. These sources can include soil organic compounds (Hill et al., 2016; O'Sullivan et al., 2014), vegetation-derived INPs from plant surfaces that can be washed off during rainfall (Conen and Einbock, 2025; Seifried et al., 2020), and other organisms. Various fungal species have been shown to be ice-nucleating active (Fröhlich-Nowoisky et al., 2015; Huffman et al., 2013; Morris et al., 2013; Pouleur et al., 1992), and likely have an important contribution to the INP population in different Arctic environments (Barry et al., 2025; Gratzl et al., 2025; Jensen et al., 2025). Additionally, lichens (Kieft, 1988; Kieft and Ruscetti, 1990) and moss spores (Weber, 2016) have been found to be active as INPs, and could contribute to INPs at sites with biological crusts.

- Section 3.2.1, from line 539: In comparison to the bulk dust samples (Fig. B5), the heat labile fractions of the atmospheric filter samples are overall larger (Fig. B13, B14), as well as the magnitude of ΔT_{50} (Fig. B8), demonstrating that the atmospheric INP population is not influenced by glacial outwash dust alone, but receives substantial contributions from additional biogenic sources, for example vegetation.
- Adapted final paragraph in Section 3.2.2 (previous 3.2.3), from line 636: Overall, our observations indicate that atmospheric INP concentrations in southern Greenland were driven by a combination of regional background sources and highly localized contributions, as indicated by overlapping, but distinct INP spectra at the different locations. The glacial outwash plain in Narsarsuaq likely served as a local rather than regional HLD INP source under the observed calm summertime conditions, as supported by similar INP spectra between bulk dust and Narsarsuaq *plain* filter samples. The substantially larger heat-labile fractions in atmospheric INPs compared to bulk dust samples demonstrate that additional biogenic sources, potentially from nearby vegetated surfaces, likely contribute significantly to the atmospheric INP population and drive part of the observed variability across sampling sites. Further analysis is needed to better constrain the frequency and magnitude of potential elevated dust loading events during high-wind conditions, as well as PBAP emissions, to assess their implications for INP concentrations and cloud glaciation in this region. Improved characterization of HLD sources could focus particularly on the shoulder seasons (spring and autumn), when higher wind speeds and reduced soil moisture are expected to enhance dust emissions. The importance of biogenic INPs highlighted here motivates dedicated future investigations of biological INP sources, such as from vegetated surfaces and biological crusts. Complementary measurements such as fluorescent particles, PBAP tracers (e.g., mannitol, arabitol), pollen and spore sampling, or microscopy-based particle detection, could help to constrain their emissions and contribution to INPs in glacial outwash plains and other high-latitude environments.
- Conclusions, from line 666: The higher fractions of organic and heat-labile INPs (on average at -15 °C, 87 % heat-labile INPs, 95 % organic INPs) compared to the bulk dust samples further point to a substantial biogenic contribution in the atmosphere and motivate future studies to further investigate biological sources.

Minor issues

- I would move Tables 1 and 2 to the Appendix.
- The tables have been moved to the supplementary material (reviewer 3 recommended to place additional figures and tables in a supplement, rather than appendix).

- Line 201: "approximately" is not necessary because 98.4 °C is a precise temperature value.
- "approximately" has been removed

- Line 290: The choice of -15 °C as a good one is further supported by the findings of Hanna et al. (2008).
- The reference has been added.

- Lines 321/322: "... higher organic fractions than Tobo et al. (2019)...", change to "...higher organic fractions than reported by Tobo et al. (2019)..."
- The sentence has been adapted as suggested.

- Lines 341/342: The statement that " ice-nucleating efficiency of TOC may differ across climatic regions" could be supported by referring to Schnell and Vali (1973).
- The reference has been added.

- Correlation analysis of INP with soil parameters is convincing in numbers. However, in Figure 5 only panel b (CFU) displays a visually convincing correlation. Perhaps, it is in the cluster of data points with low values that similar patterns are hidden in panels a and c? Transforming the scale of the x-axis to log-scale might render correlations visible.
- Figure 5 has been adapted to show the x-axis in log-scale.

- In Figures 2, 6, and B12 there is a lot of overlap between symbols. Consider using open symbols to reduce cover up.
  We tested open symbols for Figs. 2, 6, and B12 (see Fig. 1 below as an example), but found that they did not improve the visibility of overlapping data points. We therefore prefer to retain the original figures.

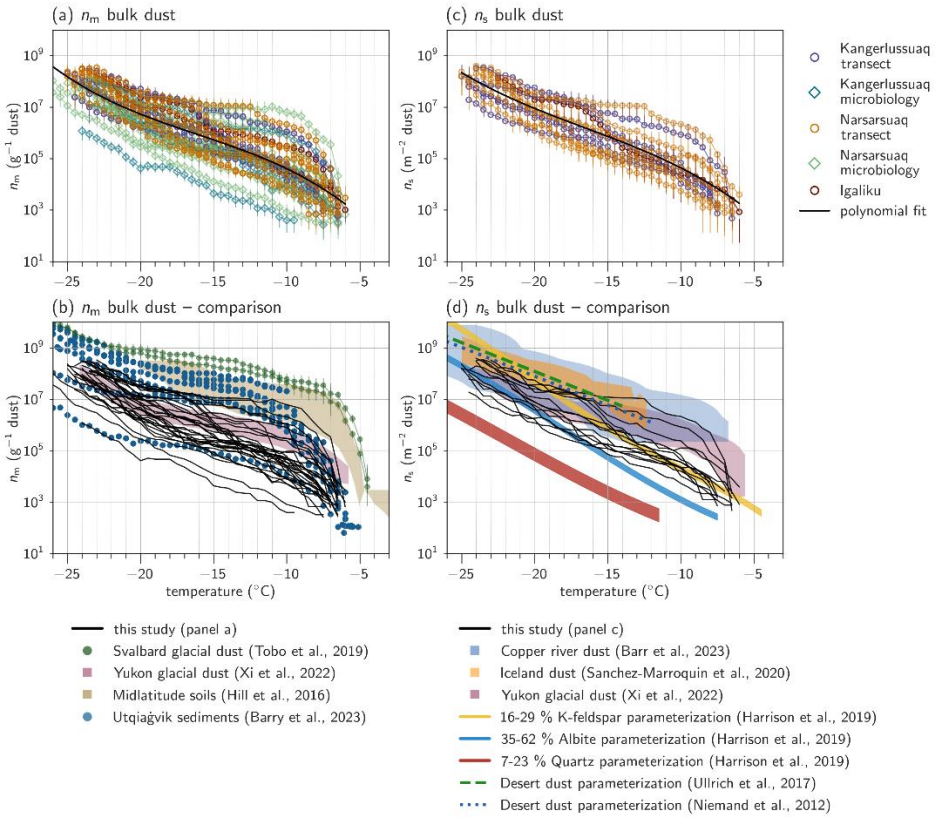

Figure 1. Test with open symbols.

- Figure B11: The morphology of the particles in panels a and b is not as typical for mineral dust as is that in panel c. Are EDX spectra available for these particles?
- Unfortunately, EDX spectra are not available for these specific particles. We have therefore updated Figure B11 (now Fig. S12) to include a larger number of particles, providing a more comprehensive overview of particle morphology. The updated figure shows that the majority of particles display irregular, angular to sub-rounded morphology consistent with mineral dust, including platy sheet-like forms indicative of clay minerals, while a minority show smoother, more rounded morphology potentially attributable to other origins. EDX elemental analysis of a particle subset confirmed the presence of silicate minerals, consistent with mineral dust from the glacial outwash plain.

References:

Dupont et al., 2021, https://doi.org/10.1029/2021JD034735

Hanna et al., 2008, https://doi.org/10.1175/2007JAMC1549.1

Heindel et al., 2019, https://doi.org/10.1007/s10021-018-0267-8

Schnell and Vali, 1973, https://doi.org/10.1038/246212a0.

Tanner, 2025, https://doi.org/10.3390/land14091827

Zhang et al., 2006, https://doi.org/10.1016/j.geod

Citation: https://doi.org/10.5194/egusphere-2026-484-RC1

Referee #2

Review: "Ice-nucleating particles in Greenlandic glacial outwash plains"

Summary:

In their manuscript, Bergner and colleagues investigated ice-nucleating particles in southern Greenland along glacier runoffs during summer field campaigns in 2023 & 2024. In general, they aimed to understand whether the number of INPs present in southern Greenland is comparable to those observed at other high latitude sites on the globe. They collected soil samples along glacier outwash zones and tested the resulting soil-suspensions in immersion freezing mode. In addition, airborne particles were collected onto filters, which were extracted in the lab and analysed in accordance to the suspensions. Their results indicate that organic and biological materials contributed significantly to the INP population in both soil and air samples. Notably, the data shows no relationship between INP abundance and distance from the glacier along the outwash zone. In contrast to previous studies, they could not identify any correlation between the amount of INPs and meteorological conditions. However, they found that the air samples were influenced by local emissions. This is comprehensive study that presents a large data set on INPs in southern Greenland, supported by detailed analytical measurements. In general, this work is of interest to the ice nucleation community and suits publication in ACP after addressing the following minor revisions.

We thank the reviewer for the positive assessment of our work and the constructive feedback. We have carefully addressed the minor revisions, which we believe have strengthened the manuscript. A detailed response to each comment is provided below.

General comment:

- How comparable are bulk dust INPs, collected from the top 1 cm of soil, with the 'microbiology' samples taken from the top 5 cm? How much does the microbial composition vary within these soil layers, and could this affect the interpretation of the results?
- The bulk dust (transect) samples were collected from the top 1 cm of soil, while the microbiology samples were taken from the top 5 cm. We acknowledge that a gradient in microbial activity across these depths is possible in principle; however, we argue that this difference is unlikely to affect the interpretation of our results for the following reasons.
    - First, the correlations between INP concentrations and microbial cell counts were derived from measurements performed on the same samples, meaning that any depth-related offset in microbial composition would affect both variables equally and therefore not bias the correlation analysis.
    - Second, the microbiology and transect samples span a similar range of ice-nucleating activity, suggesting that the different sampling depths do not introduce a systematic difference in INP concentrations between the two sample types.
    - Third, analysis of soil microbial community composition from the same field sites (Marsh et al., 2026) found no significant difference in ice-nucleating genera across soil depths between <5 cm and 10–15 cm (Wilcoxon rank sum test, $p > 0.05$, $n = 49$), suggesting that the microbial community relevant for ice nucleation is broadly homogeneous across the uppermost soil layers. This is consistent with the top 5 cm being characterised by similar oxygen and water conditions throughout.
- To reflect the above arguments in the revised version of our manuscript, we added the following to Section 2.1.1: Although transect and microbiology samples were collected from different depth intervals (top ~1 cm and top ~5 cm, respectively), analysis of microbial community composition from the same sites found no significant differences in ice-nucleating genera across these soil depths (Marsh et al., 2026), suggesting that this difference does not affect the interpretation of the results.

Specific comments

Abstract:

- Line 27-28: “Atmospheric INP concentrations above -20 °C were higher at the outwash plain sites compared to a nearby fjord site, […]” – It is unclear to me which sites are considered fjord sites? The discussion in the main text focuses on the term ‘outwash plain sites’, but the term ‘fjord sites’ is not used consistently. Please clarify.
- The term "fjord site" referred to the Narsaq sites (*NIRS* and *hill*), which are located in a more fjord-dominated setting, but we acknowledge that this terminology was not introduced or used consistently in the manuscript. To avoid confusion, we have revised the abstract to remove this phrasing and replaced it with a more general formulation (from line 27): Dust from the Narsarsuaq glacial outwash plain likely acted as a localized rather than regional INP source during summer, as indicated by higher atmospheric INP concentrations above -20 °C and similarities between atmospheric and bulk dust INP spectra at the outwash plain site compared to other sites in the region.

- Line 33-34: “[…] This highlights the importance of region-specific dust characterizations for improving representation of cloud processes and climate impacts in the Arctic.” It is unclear whether the authors mean that the physical and chemical characteristics of dust should be determined, or region-specific INP parameterizations are needed to improve our understanding of cloud processes. Please be more specific.
- Since we mean INP parameterization, we adapted the sentence as follows (line 37-38): This highlights the importance of region-specific INP parameterizations for improving representation of cloud processes and climate impacts in the Arctic.

Introduction:

- In general, the introduction gives a comprised overview of the current state of the art and identifies important knowledge gaps. Some sentences are rather long, which reduces readability. Consider dividing them into shorter, more focused statements to enhance clarity.
- We have revised the introduction and split several long sentences into shorter, more focused statements to improve readability throughout the section.

- Line 37-38: Specify that this is true for mixed-phase clouds but keep in mind that INPs can influence heterogeneous ice nucleation below the mentioned temperature threshold of -38°C.
- We have adapted the sentence as follows (from line 41):
INPs initiate heterogeneous ice-formation within the temperature range from 0 °C to -38 °C and can also aid ice formation below -38°C (Hoose and Möhler, 2012; Kanji et al., 2017; Murray et al., 2012). In mixed-phase clouds, INPs influence the partitioning between cloud liquid and ice water content through both the Wegener-Bergeron-Findeisen mechanism (Korolev, 2007) and secondary ice-production pathways (Field et al., 2016; Korolev and Leisner, 2020).

- Line 44: Please clarify if the goal is to quantify the sources of INPs or quantify INPs themselves. I’d argue that the aim is to identify the sources and quantify INPs.
- We have adapted the goal as follows (from line 48): However, identification of INP sources, quantification of INP concentrations, and understanding of their freezing characteristics and variability remain incomplete, particularly in high-latitude environments.

- Line 52: How do you define “source” in this context? Since you mentioned a few lines earlier that high latitude dust can originate from distant regions, it may be clearer to refer here to ‘location’ rather than ‘source’.
- We adapted the sentence as follows (line 56-57): More recently, increasing attention has been directed toward high-latitude dust (HLD), defined as dust from locations north of 50° N or south of 40° S […].

Methods:

- Line 115: “[…] locations farther from the glacier have been exposed longer following glacial retreat […]” Could the authors add which kind of exposure they are referring to here.
- We have adapted the sentence to (line 124-125): Dust samples were collected following a chronosequence approach (Bradley et al., 2016), where locations farther from the glacier have been exposed longer to the atmosphere and weathering processes following glacial retreat.

- Table 1: In case of a sample not being treated with neither heat nor H2O2, I would state that in the corresponding cell instead of leaving it blank.
- We have added "no treatments" in the INP treatment column for the samples without any treatments in Table 1 (now Table S1). Based on recommendations of the other reviewers, the Table has been moved to the supplementary material.

- Section 2.1.2.: It would be helpful to specify how far above the ground the filters were collected. While some photos in the Appendix provide some context about this, I think it is important to include this information in the main text.
- We have added this information in Section 2.1.2 (line 172): Inlet heights were 100 cm above ground at the plain station, 116 cm at the col station, 3 m at NIRS, and 142 cm at the hill station.

- Section 2.2.: How were the samples stored during the field campaign? Were the samples measured directly after collection or were they shipped to a laboratory? If shipment took place, under which conditions were the samples shipped? Please provide details about storage and sample handling in this section and how this could have affected the results.
- We have added the information to Section 2.2 (from line 182): All samples were stored frozen during the campaign (approximately -20 °C) and transported frozen to the laboratory (<0 °C). Filter samples remained frozen until INP analysis (approximately -20 °C), whereas transect dust samples were dried and sieved at room temperature shortly after the campaign and then stored frozen (approximately -20 °C) until INP experiments were performed. Storage and freeze-thaw cycles can affect ice nucleation activity of both mineral and biological samples, typically causing INP degradation (Beall et al., 2020; Perkins et al., 2020; Wex et al., 2015), although the effects are not fully understood. Frozen storage was chosen to minimize potential INP degradation.

- Section 2.2.: How easily were the soil samples suspendable in water? Please give more details on this and describe possible limitations.
- We have added further details on sample suspension and potential limitations to Sections 2.1.1 and 2.2. As grain size distributions are directly relevant for assessing suspension behavior and potential particle settling, we have added Figure S1 showing size distributions of the transect samples.
- Figure S1:

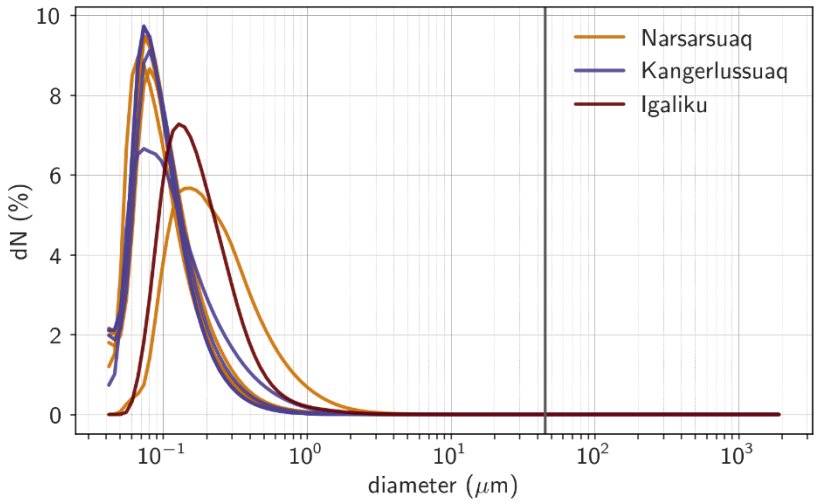

Figure S1. Dust sample grain size distributions. Particle number size distribution of *transect* dust samples from the Narsarsuaq, Kangerlussuaq, and Igaliku glacial outwash plains as measured with the laser diffraction particle size analyzer (0.017–2000 µm, LS 13 320, Beckman Coulter, USA). The samples have been sieved to < 45 µm (grey vertical line) for INP measurements.

- Addition in Section 2.1.1, from line 131: Size distributions of the transect samples, measured by a laser diffraction particle size analyzer (0.017–2000 µm, LS 13 320, Beckman Coulter, USA), show that particles are below 10 µm with a main mode below 0.1 µm for most samples (Fig. S1).

- Addition to Section 2.2*:*
  From line 193*:* The dust samples formed homogeneous suspensions in water, although some settling was observed in the sample tube with the highest dust concentration during preparation, which was addressed by repeated shaking.
- To include potential limitations regarding particle settling, we included from line 206: Particle settling during the freezing experiment is a potential concern for bulk dust samples containing coarser particles. However, size distributions of the sieved transect samples show that particles are predominantly in the fine mode with a main mode below 0.1 µm (Figure S1), making settling unlikely for this sample type. The unsieved microbiology samples follow the same methodology as Barry et al. (2023), enabling direct comparison with their dataset, but settling of larger particles cannot be excluded for this sample type, introducing additional uncertainty to the reported INP concentrations.

- Equation 1: Please add a step-by-step derivation of this equation to the Appendix. I also think Vali, 2019 would be the better reference here.
- In addition to the derivation steps that were already presented in the appendix (previous equations A1-A5), we have now added Eq. A1 for the frozen fraction calculation. We have also added the reference to Vali (2019) as suggested.

- Line 179: I believe “d” is a dilution “factor” not the ratio.
- We have changed the text to “factor”.

- Line 180: According to Vali, 2019 the terminology for ns is “active site density” and is defined as ‘[…]“the number of sites causing nucleation per unit surface area of the IN” – In the manuscript, nm is referred to as ‘ice-active mass site density’, which is slightly inconsistent with the proposed terminology. I recommend revising nm and ns, stating them as ‘ice-active site density per mass or per surface area’.

- We have adapted the terminology to the proposed terms throughout the manuscript.

- Line 198: “Heat treatment denatures proteins” – Please add more content to this statement. What is the minimum temperature required for proteins to unfold? Is this the same for all proteins? In general, I am missing the discussion on the limitations of the treatments. For example, Daily et al., 2022 showed that heat treatment is not entirely selective to proteinaceous INPs but can also alter the ice-activity of certain dusts, like quartz and plagioclase feldspar. Please add some discussion about this.
- Thank you for pointing out these important aspects, we have adapted the text as follows (from line 245):
Heat treatment denatures proteins, with proteinaceous INPs in different biological materials (e.g., bacteria, fungal spores, lichen) found to deactivate between 40-60 °C (Fröhlich-Nowoisky et al., 2016; Hara et al., 2016; Kieft and Ruscetti, 1990; Pouleur et al., 1992). The difference between untreated and heat treated spectra is therefore often used as an indicator of proteinaceous, likely biological INPs (e.g., Conen et al., 2011; Hill et al., 2016; O’Sullivan et al., 2014; Pouleur et al., 1992). Hydrogen peroxide treatment oxidizes organic matter, leaving spectra that reflect the remaining inorganic or mineral INPs (e.g., Conen et al., 2011; Hill et al., 2016; McCluskey et al., 2018a). While these treatments are useful indicators for the possible origin of INPs, they have limitations and are not entirely selective. Heat treatment can reduce the ice-nucleating activity of certain minerals, including quartz and plagioclase feldspar (Daily et al., 2022), while some thermostable biological material can remain active (Pummer et al., 2012). Similarly, hydrogen peroxide treatment can affect carbonate minerals, although other mineral components are likely unaffected (Hill et al., 2016; O’Sullivan et al., 2014; Tobo et al., 2019a).

- Line 202-204: If H2O2 is exposed to UV radiation in water, OH radicals are generated. This important detail is currently missing in the main text. Also, mention that catalase was added specifically to prevent freezing point depression, which would occur if residual H_2O_2 remained in the sample.
- We added the information (from line 258): The UV radiation generates highly reactive hydroxyl radicals that oxidize organic compounds. Remaining peroxide was neutralized by adding small quantities (90 to 130 µL) of 0.1 µm filtered catalase (Cat. No. MPB-210042910-10ML, MP Biomedicals, USA) to avoid freezing point depression.

- Line 233: Please add the reasoning why you chose a threshold of -15°C?
- The following reasoning has been added (from line 296): -15 °C was selected as a reference temperature as it falls near the center of the analysis range and is a commonly used benchmark temperature for INP comparisons relevant for mixed phase clouds (Barry et al., 2023; Morrison et al., 2012), although results are not sensitive to this choice.

Results & Discussion:

- The structure of the ‘Results & Discussion’ section is somewhat inconsistent, as discussion is often presented before the results. Typically, results should be described first, followed by their discussion.
- We have carefully reviewed the results and discussion section and find that the overall structure already follows a results-first approach, where observations are presented before their interpretation. In a combined results and discussion format, brief interpretive statements are sometimes used to motivate or contextualize subsequent results, which we believe aids readability.

- Line 362: Since you discuss how vegetation might influence your INP spectra, this aspect could be expanded further. Simply stating that fungal spores contribute is rather limited. In general, plant surfaces are well known to host INPs. For example, studies have shown that INPs can be washed off leaves during rainfall (Conen et al., 2025; Seifried et al., 2020). Conen et al. hypothesized that surfactant excretion by INP-producing bacteria leaves INPs on the plant surfaces. In addition, lichens growing on tree bark are known to be a source of vegetation-derived INPs (Kieft and Lindow, 1988).
- We thank the reviewer for the comment and agree that these are important aspects to discuss. We adapted and extended this section as follows (from line 443):
- Microbiota fix carbon and nitrogen, allowing the development of more complex microbial communities, soil development and plant succession, that support more diverse ecosystems (Bradley et al., 2016; Donhauser and Frey, 2018). As microbial abundance, TOC, and n_m values are higher for vegetated sites compared to unvegetated sites (Fig. B10, *microbiology* samples), higher microbial abundance may therefore be an indicator of an environment with more advanced soil and vegetation development, that can contain additional sources of INPs. These sources can include soil organic compounds (Hill et al., 2016; O'Sullivan et al., 2014), vegetation-derived INPs from plant surfaces that can be washed off during rainfall (Conen and Einbock, 2025; Seifried et al., 2020), and other organisms. Various fungal species have been shown to be ice-nucleating active (Fröhlich-Nowoisky et al., 2015; Huffman et al., 2013; Morris et al., 2013; Pouleur et al., 1992), and likely have an important contribution to the INP population in different Arctic environments (Barry et al., 2025; Gratzl et al., 2025; Jensen et al., 2025). Additionally, lichens (Kieft, 1988; Kieft and Ruscetti, 1990) and moss spores (Weber, 2016) have been found to be active as INPs, and could contribute to INPs at sites with biological crusts.

- Figure 4 and all figures that contain box plots: Add details explaining the box plots, specifying what the median, quartiles, and whiskers represent. Usually, outliers are visible for these types of plots. Were they excluded?
- Outliers had been excluded before but are now shown in Fig 4, Fig S11 and Figure S27. The following description has been added to the captions.
    - Fig 4: Box plots show the median (center line), first and third quartiles (box edges), and whiskers extending to the 5th and 95th percentiles; outliers beyond this range are shown as individual points.
    - Figure S11 and Figure S27: Box plots show the median (center line), first and third quartiles (box edges), and whiskers extending to the 5th and 95th percentiles; outliers beyond this range are shown as individual points. The mean is marked by the black triangles.

- Line 396-397: I don't fully agree with this statement. According to the INP spectra in Figure 6a, the hill site also shows high INP concentrations between -8 and -20°C. It is even higher at the warmer temperatures than for the col site.
- We agree that the statement as it is written is not fully accurate. We have adapted it as follows (from line 492):
Fig. 6a depicts the INP spectra colored by location. Overall, the INP concentrations at the Narsarsuaq *plain* and *col* sites, and the Narsaq *hill* site tend to be higher at relatively higher temperatures (approximately -20 °C to -8 °C), whereas the *NIRS* site shows elevated INP concentrations at lower temperatures (< -20 °C).

- Line 410: Please add Diehl et al., 2001 as a reference.
- The reference has been added.

- Line 424-429: Please add an explanation on what D10 and D15 represents.
- We have adapted the sentence as follows to make the naming clearer (from line 526):

To evaluate whether commonly used parameterizations adequately represent the observed INP spectra, we compare our data to the global aerosol parameterization of DeMott et al. (2010, hereafter referred to as *D10*, Fig. 6c) and the global desert dust parameterization of DeMott et al. (2015, hereafter referred to as *D15*, Fig. 6d), […].

- Line 469-470: What do you mean by 'threshold velocity'? Please expand this argument.
  We have adapted the explanation as follows (note that based on recommendation of Reviewer 3, we have moved this section to the supplementary material, from line 177):
  Possibly, dust emissions were limited by elevated soil moisture, which can increase particle cohesion and raise the threshold wind velocity for soil erosion. However, estimates based on Fécan et al. (1999) using measured soil moisture at the *plain* site and different soil densities (not measured) would suggest that the wind speed threshold velocity for soil erosion was exceeded (Fig. S21).

- Figure 8a-c: N_INP(L-1) represents the cumulative number of ice nuclei per litre of air. At −15 °C on day 1, there are 10^-1 L^-1 at station col, but no data is shown for col at -20 °C. Please clarify.
- The reason for this discrepancy is that the measured data did not extend to sufficiently low temperatures for the performed dilutions, with the last data point for this sample at −19.5 °C. We have added a clarifying note to the figure caption (now Fig. S17): Where INP data is shown for -15 °C, but not for -20 °C, the activated fraction reached the detection limit of the performed dilutions; no extrapolation beyond the last reliable data point is shown.

Appendix:

- Figure A6: I don't see the data for unfiltered water. In the legend it is stated as being depicted as grey lines.
  The filtered and unfiltered water data are distinguished by solid and dashed lines respectively, with colors indicating the different water types. The legend entry was incorrectly labeling line style rather than color. We have corrected the legend as this distinction is explained in the caption.

- Figure B9: For the first column, I assume nm stands for the sum of all nm? Please clarify.
- The 'nm' entry represents the correlation between TOC and ice-active site densities per mass for all samples in the dataset, while the subsequent entries ($n_{m,transect}$, $n_{m,mb}$, etc.) represent correlations for specific sample subsets. We have revised the caption to clarify this to: Figure S10. Correlation coefficient TOC and different variables. Kendall's tau correlation coefficient (τ_b) of total organic carbon (TOC) and ice-active site densities for different sample subsets and treatments, including ice-active site densities per mass (n_m) for all samples, n_m following heat treatment ($n_{m,heat}$), ice-active site densities per surface area (n_s), n_s following heat treatment ($n_{s,heat}$), n_m of *transect* samples ($n_{m,transect}$), n_m of *microbiology* samples ($n_{m,mb}$), n_m in Kangerlussuaq ($n_{m,K}$), and n_m in Narsarsuaq ($n_{m,N}$). The correlation coefficient is written in bold when statistically significant ($p < 0.05$).

- Figure B11: How did you determine that these particles are mineral dusts, come from marine sources or are from long-range transport? Did you perform any additional analysis on those particles besides SEM?
- The classification of particles shown is based on morphology as observed by SEM, with EDX performed on a small subset of particles. Angular, irregularly shaped particles were interpreted as mineral dust. We acknowledge that these assignments remain morphology-based interpretations rather than definitive identifications, and have extended the figure to include a wider range of representative particles (now Figure S12).

- Figure B12: Adjust the legend and re-name “clean marine” to a more specific description.
- The legend has been adapted as follows: pristine marine environment
- The caption has been adapted to be more specific: Figure S13. INP spectra normalized by surface area. Atmospheric INP concentrations normalized by the total aerosol surface area for particles larger than ≈190 nm, derived from optical particle counter measurements (Sect. 2.1.2). For comparison, INP spectra of bulk *transect* samples normalized by geometric surface area are shown, together with parameterizations for a pristine marine environment from measurements in the Southern Ocean (McCluskey et al., 2018b) and desert dust (Niemand et al., 2012).

- Figure B14: It is unclear what the bright brown lines indicate. Please clarify.
- The following has been added to the caption to the figure, now Fig. S15: The light brown lines represent the heat labile fractions of individual bulk dust samples.

- Are Figure C4 and Figure C2b the same? If yes, only show it once.
- Figure C4 has accidentally been included twice and the duplicate has now been removed.

Typos etc.:

- Line 263: typo for potassium feldspar (K-feldspar)
- Line 219: Tobo reference is missing a bracket
- The typos have been addressed.

References:

Conen, F., & Einbock, A. (2025). Release of ice-nucleating particles from leaves during rainfall. The Science of Nature, 112(2), 29.

Daily, M. I., Tarn, M. D., Whale, T. F., & Murray, B. J. (2022). An evaluation of the heat test for the ice-nucleating ability of minerals and biological material. Atmospheric Measurement Techniques, 15(8), 2635-2665.

Diehl, K., Matthias-Maser, S., Jaenicke, R., & Mitra, S. K. (2002). The ice nucleating ability of pollen:: Part II. Laboratory studies in immersion and contact freezing modes. Atmospheric Research, 61(2), 125-133.

Kieft, T. L. (1988). Ice nucleation activity in lichens. Applied and environmental microbiology, 54(7), 1678-1681.

Seifried, T. M., Bieber, P., Felgitsch, L., Vlasich, J., Reyzek, F., Schmale III, D. G., & Grothe, H. (2020). Surfaces of silver birch (Betula pendula) are sources of biological ice nuclei: in vivo and in situ investigations. Biogeosciences, 17(22), 5655-5667.

Vali, G. (2019). Revisiting the differential freezing nucleus spectra derived from drop-freezing experiments: methods of calculation, applications, and confidence limits. Atmospheric Measurement Techniques, 12(2), 1219-1231.

Referee #3

Review of "Ice-nucleating particles in Greenlandic glacial outwash plains" by Bergner et al.

The origin of ice-nucleating particles (INPs) in Greenland is investigated. Ice formation on dust samples collected from the ground and from airborne aerosol samples is measured using the new SPICE setup. Heat and peroxide treatments are applied to the samples to distinguish between organic and inorganic INP fractions. Ancillary analyses include TOC, mineralogy, BET surface area, microbial abundance, SEM, aerosol size distributions, meteorological data, back trajectories, specific events (Foehn, precipitation, and wildfire plumes), as well as temporal and spatial variation. Measurement results are compared with literature data and with samples collected in the Swiss Alps.

General comments:

Two main conclusions are, lower ice nucleation activity of Greenlandic dust sources compared to other HLD sources, and a dominant contribution, likely from biological substances to the ice nucleation activity. Both conclusions would benefit from further verification, taking the following aspects into consideration.

The lower ice-active surface site densities may be influenced by a normalization artefact. When comparing $n_{s,\,geo}$, normalized by geometric surface area, with $n_{s,\,BET}$ normalized by BET surface area, $n_{s,\,BET}$ is generally lower than $n_{s,\,geo}$, even for spherical particles (see, for example, Hiranuma et al., 2014, Fig. 3). For non-spherical dust particles, the difference can reach up to two orders of magnitude.

There are several aspects that may complicate the second conclusion. Dust constitutes most of the surface area of the ground samples and is known to be ice-nucleation active in the tested temperature range, and therefore likely contributes substantially more to the observed ice formation activity than small amounts of biological substances. Ambient INP concentrations can be parameterized using D15 for dust. Furthermore, the evaluation of the heat test in Daily et al. (2022) indicates that, for the minerals present in the Greenland ground samples, heat treatments can produce false positives by deactivating the minerals ice nucleation activity. In addition, peroxide treatment increases pH, and it has been shown that acid treatments of dust decrease ice nucleation activity. These aspects could be investigated more explicitly in relation to the second conclusion.

The manuscript presents a large amount of material (data dump), including data that are only marginally related to the topic of the paper (e.g., Swiss Alps samples and SPICE performance validation). In addition, many ancillary analyses are presented but not fully developed into robust findings.

I recommend major revisions of the manuscript because the material would benefit from substantial trimming and deeper analysis to support more robust and clearly articulated conclusions.

We thank the reviewer for these detailed and constructive comments, and address each point specifically:

Normalization artefact: We acknowledge that normalization effects can occur when comparing $n_{s,geo}$ and $n_{s,BET}$, and agree that this is a relevant consideration. We note that this limitation is already discussed in the manuscript (from line 310). To further assess the magnitude of this effect, we estimated geometric surface areas from measured size distributions assuming a density of 2.65 g cm^{-3}, and find that differences between $n_{s,\,geo}$ and $n_{s,\,BET}$ are comparably small for our samples (Figure 2), and do not affect the main conclusion. We note that in some cases $n_{s,\,geo}$ is smaller than $n_{s,\,BET}$, which may reflect uncertainties in the BET measurements. Given the small difference and in response to this comment, we have switched to geometric surface area normalization throughout the manuscript for consistency (Fig. 2, Fig. S10, Fig. S13). Both n_m and n_s values for our samples lie in the lower range compared to

other HLD sites, which independently supports the conclusion of lower ice-nucleating activity regardless of the normalization approach used.

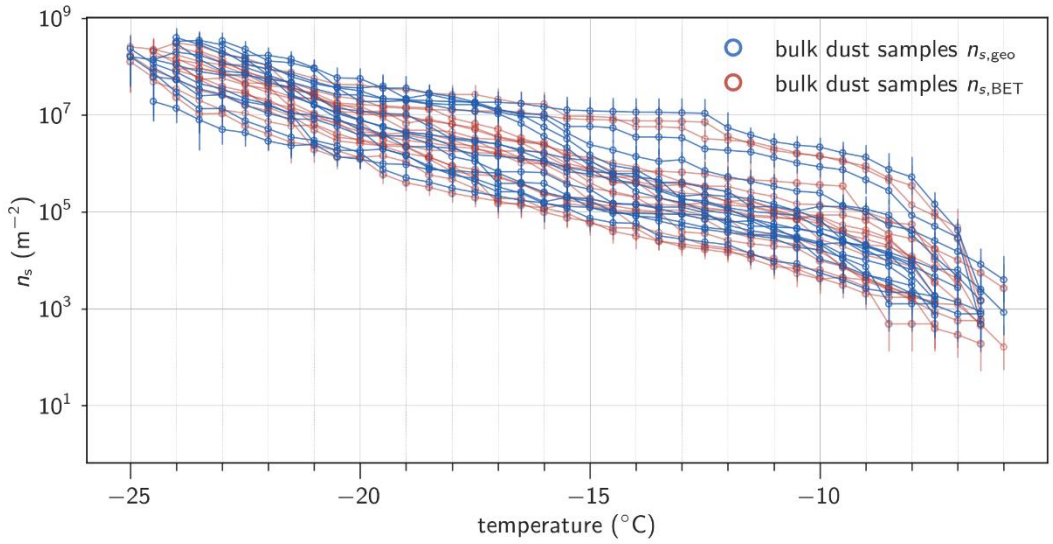

Figure 2. Comparison bulk dust samples of $n_{s,geo}$ and $n_{s,BET}$.

Role of biological substances versus mineral dust: We appreciate this nuanced point. While we agree that mineral dust constitutes the majority of surface area and is known to be ice-nucleation active in the tested temperature range, we argue that small amounts of biological and organic material can coat mineral surfaces and enhance ice-nucleating activity disproportionately to their mass or surface area fraction. Organic material, even small amounts have been reported to enhance ice-nucleating activity of dust (Hamzehpour et al., 2022; Tobo et al., 2019). Regarding the D15 parameterization: while ambient INP concentrations can be reasonably described by D15, this does not necessarily imply that the source of ice-nucleating activity is identical to mineral desert dust, as parameterizations can describe observations for different mechanistic reasons. We further acknowledge the limitations of heat and peroxide treatments raised by the reviewer, including potential false positives from mineral deactivation (Daily et al., 2022) and pH effects of peroxide treatment, and have added a more explicit discussion of these limitations in the manuscript in Section 2.2 and Section 3.1.3. Nevertheless, we note that the treatment results and the correlations with TOC and microbial abundance constitute two independent lines of evidence pointing in the same direction, which we believe supports the robustness of the conclusion despite the individual limitations of each approach.

Data volume and appendix material: We acknowledge the reviewer's concern regarding the volume of presented material. We have shortened the manuscript and addressed the specific comments below. We have moved some of the appendix material (B, C) to a supplement. While we agree that some of it is less directly relevant to the main conclusions, it still contains relevant information for interested readers. The SPICE performance validation is relevant as this study presents the first dataset obtained with this instrument. The Swiss Alps data, while geographically peripheral, provides a useful climatological contrast in TOC content and ice-nucleating activity that supports the interpretation of the Greenland results.

The specific changes made to the manuscript in response to these general comments are detailed in the point-by-point responses below and we believe that the revised manuscript presents a more focused and clearly articulated set of conclusions.

Specific comments:

- Move Appendix B and C to a Supplement. Appendices should be understandable on their own. Appendix A meets this requirement.
- We moved Appendix B and C to a Supplement.

- Line 70, 72: Define the units of "ice-nucleation activity" referred to.
- We specified as follows (from line 75): For example, Xi et al. (2022) observed dust near the Kaskawulsh Glacier in Yukon, Canada, with ice-active site densities per mass about two orders of magnitude lower than that of the Svalbard samples, showing biological contributions above -15 °C but mineral dominance at lower temperatures. Airborne dust from the Copper River Delta in Alaska exhibited high ice-active site densities per surface area, with heat-sensitive INPs active down to -25 °C (Barr et al., 2023).

- Line 98: Specify what is meant by "sources" in this context.
- We specified as follows (line 105): […], (ii) identify sources (primary biological, other organic, mineral) contributing to the ice-nucleating activity, and […].

- Table 1 and Table 2: Consider moving to the Supplementary Material. Add sample volumes to Table 2.
- We have moved the tables to the appendix and have added the sample volumes.

- Line 150–151: The size ranges of the OPCs are very specific and differ from those in the manuals. Please clarify how they were determined.
- The size ranges are the original size ranges of the instruments, except that we exclude the lowest 3 bins for the POPS (16 bin mode) and lowest 10 size bins for the FIDAS due to higher measurement uncertainty and mismatch with a comparison to a SMPS. We have added the following sentence following the size description: The reported size ranges exclude the smallest size bins due to higher measurement uncertainty (Mei et al., 2020).

- Section 2.2: This section contains repetitions from Appendix A1. Consider referring to Appendix A1 for general information and highlighting only aspects specific to the experiments presented here.
- We shortened the section and referred to the information in Appendix A for more details, particularly the equations.

- Line 165–166: Specify how much dust is used per sample and how many 10-fold dilutions are prepared (as described in Appendix A1).
- The description was adapted as follows (line 191): Initial suspensions were prepared at approximately 2 g L^{-1} by weighing a defined mass of dust (e.g., 40 mg) and adding SA water (e.g., 20 mL). Four sequential 10-fold dilutions were then prepared for bulk dust samples (0.5 mL sample, 4.5 mL SA water), resulting in a concentration range of approximately 2 to 0.0002 g L^{-1}.

- Line 168: Clarify why 8 mL instead of 12 mL are used to wash procedural filters. Using the same volume for background estimation could avoid the need for scaling.
- We used 8 mL for procedural blank filters because they were analysed first in our analytical sequence, before we determined that 12 mL was needed for the full sample treatment and dilution protocols applied to the actual filter samples. We agree that the same sample volume would be beneficial.
  We have added this info in the text for clarification (from line 195): Filter samples were suspended in 12 mL of SA water (8 mL for procedural blank filters, as these were processed earlier in the study before the protocol was optimized), […].

- Line 169: Specify that 15-fold and 225-fold dilutions are measured in addition to the undiluted suspension.
- We adapted as follows (line 198): The undiluted suspensions and corresponding dilutions were pipetted in 50 µL droplets into the wells of two single use PCR-trays (Cat. No. 781368, Brand, Germany).

- Line 171: Specify how the 192 PCR wells are distributed among samples and treatments.
- We specified as follows (from line 199): For filter samples, the 192 wells were distributed as follows: 40 wells for the initial suspension, 56 wells for the 15-fold dilution, 64 wells for the 225-fold dilution, and 32 wells for background SA water. For bulk samples, the wells were distributed in blocks of 32 for the initial suspension, for each dilution, and for the background.

- Line 184: Provide the BET surface areas of the transect samples.
- Since we have switched to geometric surface area normalization, we have added the specific surface areas to Table S1.

- Line 193–194: Explain why the water background was used instead of the procedural filter handling background. Testing the difference would help confirm that the contribution is minor.
- We use the water background correction as it is directly measured for each individual experiment. The handling blank correction is more difficult to apply consistently, as it requires selecting a representative blank filter for the sampling conditions. To assess the potential contribution of handling blank contamination to the sample INP concentration, we tested three scenarios: a worst case using the handling blank with the highest INP concentrations, a medium based on a polynomial fit to all handling blank filters, and a best case using the handling blank filter with the lowest INP concentrations. In the medium and best case, the blank contribution remains below ~3 % across the temperature range. In the worst case, contributions can be substantial at colder temperatures (up to ~50 % at -26 °C but below ~5 % above -20 °C). We have added this figure to the supplementary information (Fig. S4), and a reference in the manuscript at line 241. We acknowledge that procedural blank contamination introduces additional uncertainty to the reported INP concentrations, which is already included in the manuscript.

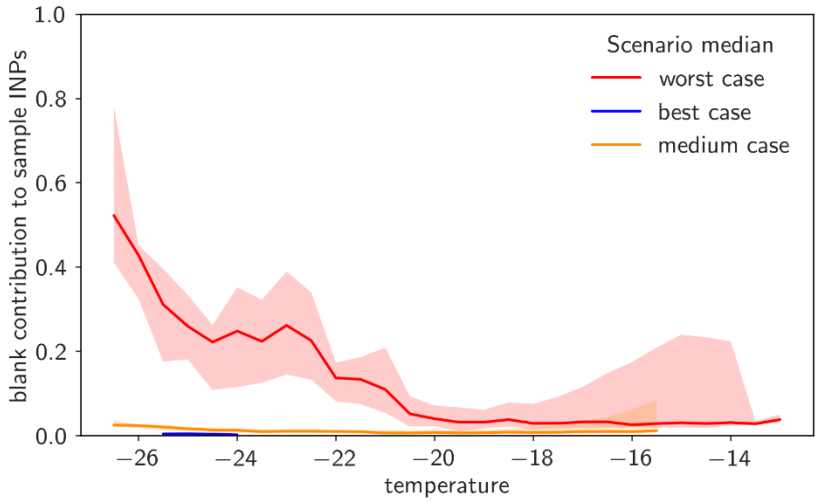

Figure S4. Contribution of procedural blank to sample INP concentrations. The fraction of procedural blank to sample INP concentrations ($N_{INP,\ procedural\ blanks}(T)$ / $N_{INP,\ samples}(T)$) is shown for three scenarios: worst case (procedural blank with maximum INP concentrations), medium case (polynomial fit to all procedural blanks), and best case (procedural blank with minimum INP concentrations). Shaded envelopes show the interquartile range across all samples, bold lines show the median.

- Line 198–199: Daily et al. (2022) may not be the most supporting reference for this statement, as it suggests nuances. Consider Hill et al. (2016) instead.
- We have changed the reference. Furthermore, we have added more discussion on the limitations of this method (from line 251).

- Section 2.4: Explain in more detail why a component analysis of slope and log concentration ratio provides information on sample similarities. Differential INP spectra could be considered as an alternative approach.
- We have added the additional explanation in the text (from line 287):
To examine similarities between bulk dust and aerosol INP spectra, we performed a principal component analysis (PCA) on derived spectral features, specifically slopes and logarithmic concentration ratios, following the approach of Barry et al. (2023). Deriving spectral features enables comparison across sample types measured in different units (n_m vs. N_{INP}), since slopes and logarithmic concentration ratios describe the shape of the INP spectra rather than absolute magnitudes. Slopes characterize the temperature-dependent steepness while logarithmic ratios capture relative concentrations across the temperature range, and both features can be indicators of the underlying INP source (e.g., Barry et al., 2023; Sze et al., 2023).

- Line 250: Explain the grain size effect.
We have removed this statement from the manuscript, as the tighter clustering is not as clearly observed with the geometric surface area normalization.

- Line 265–266: The percentages used for Fig. 2 do not appear to be averages calculated from Table B1. Instead of an average, consider providing a range.
- Thank you for pointing out this error. We have adapted Fig. 2 to include ranges of the parameterizations.

- Line 266: The surface area of K-feldspar is constant along the yellow line in Fig. 2(d). The difference in slope suggests that K-feldspar does not affect the temperature spectra of the dust samples at any temperature.
- We have adapted the text to include this consideration (line 332): While the K-Feldspar parameterization is in a similar range as the Greenland dust observations, the differences in slope suggest that other factors control the measured ice-nucleating activity.

- Line 272: Consider using the XRD results to further investigate differences in sample activity.
- We have compared the relative abundances of the mineral components as shown in Barr et al. (the other publications don't report XRD results) and have adapted the text as follows (from line 340):
The comparable or higher K-feldspar content of the Greenland samples relative to other HLD sites (Barr et al., 2023) suggests that the lower ice-nucleating activity observed in the Greenland samples is more likely driven by differences in biological or organic contributions.

- Figure 2: The BET surface area of dust can be up to two orders of magnitude larger than the geometric surface area. For a consistent comparison of n_s, either BET or geometric surface area should be used. The data in panel (d) may align with desert dust parameterizations when using a consistent surface definition. The polynomial fits could be removed, as they are not used in subsequent analysis. Consider taking specifications out of the citations.
- We estimated geometric surface areas from measured size distributions and find that the difference between $n_{s,\ BET}$ and $n_{s,\ geo}$ is small for our samples, such that the choice of normalization does not affect the main conclusions (Fig. 2 in this document). We have still switched to geometric surface area normalization and updated the affected figures (Fig. 2, Fig. S10, Fig. S13). To describe the methodology, we have added the following to the methods section (2.2): The specific surface area of the transect samples was estimated geometrically from laser diffraction particle size distributions (LS 13 320, Beckman Coulter, USA), assuming

spherical particles with a density of 2650 kg m^{-3}, using only size bins below 45 µm consistent with the sieving cutoff applied prior to measurement ($SSA = (6/\rho) \times \sum\left(\frac{f_i}{d_i}\right)$, where f_i is the normalized volume fraction and d_i the bin centre diameter). The resulting geometric specific surface area values are provided in Table S1.

We prefer to retain polynomial fits, as they allow a quick visual comparison with other datasets without the need to download the data.

- Sections 3.1.2, 3.2.2, 3.2.3: Consider moving these sections to the Supplementary Material, as they do not directly contribute to the main conclusions.
- We agree that Section 3.2.2, which examines temporal variability in INP concentrations and its relationship to meteorological variables and aerosol sources, does not directly contribute to the main conclusions, and have therefore moved it to the Supplementary Material. A brief reference to it is added to Section 3.2.1 (from line 523): Temporal variability in INP concentrations and potential links to meteorological variables and aerosol sources are further discussed in Section S2 (Supplementary Material), though no clear relationship with local meteorology or long-range transport was identified during the campaign.

However, we would argue that Sections 3.1.2 and 3.2.3 should remain in the main text. Section 3.2.3 is central to the main conclusions, as it establishes the link between bulk dust and atmospheric INP measurements through the PCA analysis, and we have revised the text to make this contribution more explicit. Section 3.1.2, while showing no systematic trend along the chronosequence or transect, illustrates the spatial variability in dust INP activity and directly motivates the analysis in Section 3.1.3; we therefore prefer to retain it in the main text.

- Line 314, Fig. B4: Most samples show an increasing heat-stable fraction from −15°C to −20°C, but sample N1GT shows the opposite behaviour. A more detailed analysis of such features would be helpful.
- In cases where heat-treated INP concentrations exceed those of the untreated sample, values are set equal to prevent negative heat-labile fractions. For sample N1GT, this occurs over a wider temperature range (-10.5 to -19.5 °C), with quality flags identified for a dilution (insufficient temperature difference between dilutions) suggesting an experimental issue. While the confidence intervals of the two spectra overlap for most of this temperature range, they do not overlap between approximately -12.5 and -14.5 °C, indicating that the difference in this case exceeds measurement uncertainty. This sample should therefore be interpreted with caution, and we have added a note to the caption of Fig. S6 accordingly:
In cases where heat treatment yielded higher INP concentrations than untreated samples, values were set equal to prevent negative heat-labile fractions. For sample N1GT, this occurs over a wider temperature range (-10.5 to -19.5 °C), possibly due to an experimental issue with a dilution; this sample should therefore be interpreted with caution.

- Line 316, Fig. B5: Clarify how Fig. B5 was constructed. The colour coding appears opposite to Fig. 4 and Fig. B4. Using consistent colours would improve clarity. Clarify whether the peroxide resistant fraction is too small to be visible. Use consistent labels in captions and legends (also for Fig. B13).
- We thank the reviewer for pointing out the inconsistencies. We have adapted the color coding of Figure B5 (now S7), B13 (now S14), and C6 (now S29) to be consistent with Figure 4 and Figure B4 (now S6).
- In addition, we have adapted the labels and captions, so it is clear how Figure S7 was constructed:

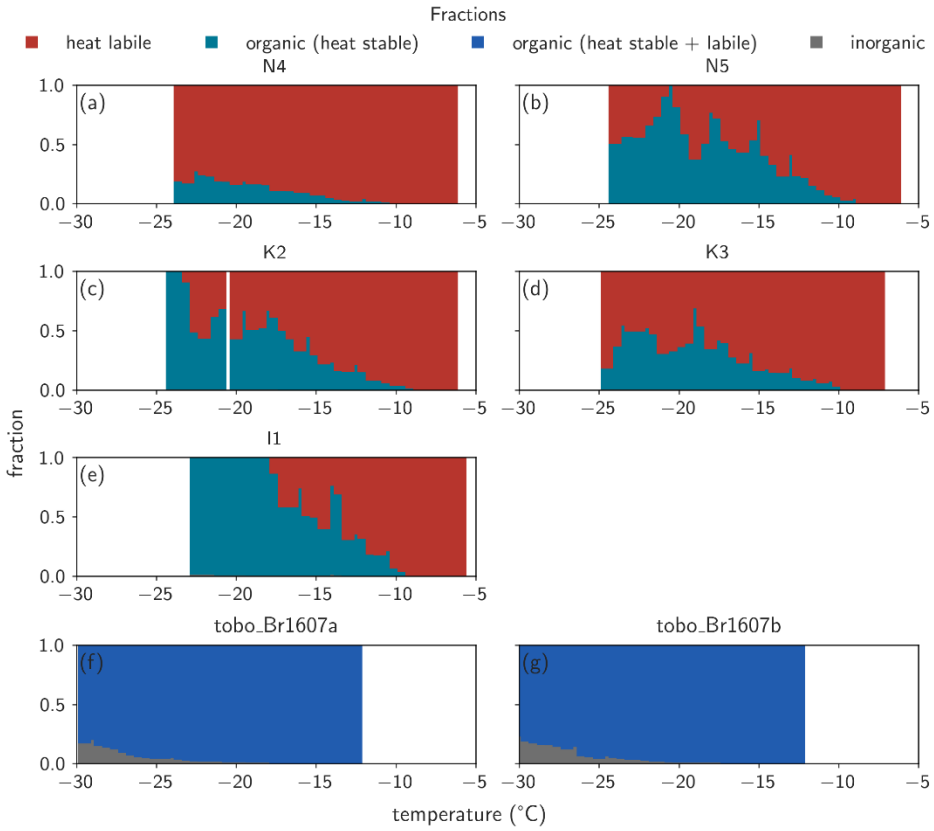

Figure S7. Composition information. Fraction of heat labile, organic (heat stable but peroxide sensitive), and inorganic (peroxide resistant) INPs per gram of dust for samples in Narsarsuaq (a, b), Kangerlussuaq (c, d), and Igaliku (e). Sample examples shown were selected without specific criteria. The inorganic fractions are too small to be visible in panels a-e. Panels (f) and (g) show the fraction of organic (which includes both heat labile and heat stable organics) and inorganic INPs per gram of dust for samples from Svalbard (Tobo et al., 2019). Fractions are calculated as: heat labile = $(n_m - n_{m,\,heat})/n_m$, organic = $(n_{m,\,heat} - n_{m,\,H2O2})/n_m$, and inorganic = $n_{m,\,H2O2}/n_m$, where n_m, $n_{m,\,heat}$, and $n_{m,\,H2O2}$ represent untreated, heat treated, and hydrogen peroxide treated samples, respectively.

- Line 317: Suppression of dust activity by treatment can be expected (see Daily et al., 2022). Consider incorporating this into the interpretation.
- We added a remark in the discussion (line 383): However, treatments may also have non-specific effects and reduce the ice-nucleating activity of minerals (Daily et al., 2022). More discussion on the limitations of heat and peroxide treatments have been added in Sect. 2.2.

- Line 320: The only significant correlations are weak and negative. Please provide an interpretation.
- The negative correlations at high temperatures occur because many samples reach 100% heat labile fraction, making the correlation sensitive to a few outlying data points with heat labile fractions < 100 %. While a positive correlation between heat-labile fraction and n_m might be expected if biological material consistently drives higher INP activity, this relationship also depends on the ice-nucleating efficiency of the specific organic compounds present, which can vary substantially between samples. After reviewing this analysis, we suggest excluding the figure and description, as it provides no meaningful insights into INP sources or mechanisms.

- Line 323: ΔT_{50} values in Barr et al. (2023) range from −1.6°C to −3.6°C, while Fig. B8 shows values down to −6°C (closer to Daily et al., 2022). Please discuss possible reasons for this difference.
- We have included the following interpretation in the text (from line 396): When comparing with the literature, the Greenlandic samples show higher organic fractions than reported by Tobo et al. (2019) (Fig. B5), and the change in median freezing temperature (Fig. B8) between untreated and heat treated samples is similar to or even higher than in Barr et al. (2023, Fig. 3), although both studies show higher overall ice-nucleating activity (Fig. 2c, d). The larger ΔT_{50} values observed in our study may be influenced by differences in sample type and size (bulk dust vs. airborne particles), and indicate a higher heat-labile fraction in the Greenlandic samples.

- Line 327: As the previous section suggests that the type and composition of organic matter are relevant, it could be noted that TOC is not sensitive to these differences.
- We have added the following sentence to note this limitation (line 407): While TOC provides only a bulk measure of organic carbon content and cannot differentiate between specific organic compounds, it can still reveal whether the overall abundance of organic matter influences INP activity across our samples.
- Line 332, Fig. B5d: Clarify why sample K3 is used as an example.
- They were no specific criteria to select the samples for treatments, except variability in space. This clarification has been added to the caption of the figure, now Fig. S7: Sample examples shown were selected without specific criteria.

- Line 332: Explain the rationale and hypothesis behind this analysis.
The rationale for this analysis is stated in the opening sentence of this paragraph, namely that organic material contributes importantly to the ice-nucleating activity of the dust samples, and TOC serves as a bulk proxy for overall organic content. We further note that TOC-INP correlations have been reported in other environments, supporting the hypothesis that organic matter abundance may drive INP activity across our samples. We have added a sentence to make this motivation more explicit (from line 404):
Given the important contribution of organic material on the ice-nucleating activity of the dust samples, we assess the correlation between TOC and n_m, as well as microbial abundance and n_m (Fig. 5). Such correlations have previously been reported for water samples (Barry et al., 2023; Wilson et al., 2015), motivating a similar analysis to assess whether organic content drives INP activity in glacial dust.

- Line 354: Microbial abundance, including the presence of *Pseudomonas*, does not necessarily imply a contribution to ice nucleation if ice-nucleating proteins are not expressed.
- We adapted the text to include this point (line 435): However, the presence of these genera does not necessarily indicate active ice-nucleating ability, as ice-nucleating proteins may not be expressed under all environmental conditions.

- Line 359: Specify which organic compounds are ice-nucleating.
- We have adapted this paragraph more generally (based on a comment by reviewer 2), which is now specified as followed (from line 442): Microbiota fix carbon and nitrogen, allowing the development of more complex microbial communities, soil development and plant succession, that support more diverse ecosystems (Bradley et al., 2016; Donhauser and Frey, 2018). As microbial abundance, TOC, and n_m values are higher for vegetated sites compared to unvegetated sites (Fig. B10, *microbiology* samples), higher microbial abundance may therefore be an indicator of an environment with more advanced soil and vegetation development, that can contain additional sources of INPs. These sources can include soil organic compounds (Hill et al., 2016; O'Sullivan et al., 2014), vegetation-derived INPs from plant surfaces that can be washed off during rainfall (Conen and Einbock, 2025; Seifried et al., 2020), and other

organisms. Various fungal species have been shown to be ice-nucleating active (Fröhlich-Nowoisky et al., 2015; Huffman et al., 2013; Morris et al., 2013; Pouleur et al., 1992), and likely have an important contribution to the INP population in different Arctic environments (Barry et al., 2025; Gratzl et al., 2025; Jensen et al., 2025). Additionally, lichens (Kieft, 1988; Kieft and Ruscetti, 1990) and moss spores (Weber, 2016) have been found to be active as INPs, and could contribute to INPs at sites with biological crusts.

- Line 396–398: Clarify how much of the difference in INP concentration is due to sample volume limiting detectable concentrations.
- To assess whether differences in sampled air volume could explain part of the variability in observed INP concentrations, we estimated the minimum air volume required to detect a given INP concentration based on our freezing assay parameters (32 wells, 50 µL droplet volume, 12 mL volume of washing water), following the formulation of Eq. A6. For the median observed INP concentration at each temperature, the sampled air volumes are sufficient for all samples across the full temperature range. The minimum detectable atmospheric INP concentration, given the assay parameters and sampled air volume, ranges from 4.5×10^{-5} to 9.9×10^{-4} L^{-1} across samples. Where data points are absent at higher temperatures, this is consistent with INP concentrations falling below the detection limit for samples with smaller sampled air volumes. We have added this information to the Section 2.2, from line 220:
  The minimum detectable atmospheric INP concentration, defined as the concentration corresponding to one frozen well given the assay parameters and sampled air volume, ranges from 4.5×10^{-5} to 9.9×10^{-4} L^{-1} across samples depending on the sampled air volume. Absent data points at higher temperatures are consistent with INP concentrations falling below this limit.

- Line 400–401: Explain how this conclusion is reached.
- The conclusion is based on the observations that the INP concentrations across all four sites span a similar order of magnitude and sometimes show quite similar INP concentrations at the same time (Fig. S17), which suggests a common regional background. The intermittent site-to-site differences could point to local sources, such as the slightly elevated INP concentrations at the plain site. We acknowledge that this interpretation is qualitative and have adapted the text as follows (from line 496):
  INP concentrations across sites fall within a similar overall range, suggesting a common regional background INP population, while the site-to-site variability may reflect the influence of local INP sources, with the glacial outwash plain potentially acting as an enhanced local source given the higher concentrations observed at the *plain* site.

- Line 401–402: This appears to repeat previous statements and could be streamlined.
- The repetition has been removed.

- Line 407: Provide supporting references for INPs emitted by each mentioned source.
- The sentence has been adapted as follows (line 503):
  At the more fjord-dominated *NIRS* site, the lower abundance of high-temperature INPs could reflect less biological or other highly active INP sources, whereas the higher INP concentrations at lower temperatures could reflect inputs from road dust or anthropogenic activities (Chen et al., 2024), or marine aerosol from the adjacent fjord (DeMott et al., 2016; Wilson et al., 2015).

- Line 410: Clarify why increases could be attributed to pollen and whether pollen concentrations at the site would be sufficiently high (>0.1 L^{-1}).
- The attribution to pollen is supported by heat lability tests presented later in this section (Fig. S13, S14), which show an increased heat-stable organic fraction in the -18 °C to -20 °C range, consistent with a polysaccharide-based source such as pollen. We acknowledge that no quantitative pollen concentration measurements were performed, but pollen was visibly observed in the field when walking through vegetation, suggesting local emissions were

present. We have adapted the text to make clear that this attribution is speculative (from line 511):

Notably, several samples from the *hill* and *col* sites exhibit a sharp increase in INP concentrations between -18 °C and -20 °C, potentially indicating sporadic emissions from a source such as pollen, which are known to become ice-active within this temperature range (Diehl et al., 2002; Duan et al., 2023), though this remains speculative in the absence of direct pollen measurements.

- Line 421: As McCluskey et al. report observations from the Southern Ocean, comparison with Arctic marine concentrations may be more appropriate.
- We agree that Arctic marine observations would provide a more direct regional comparison, and these are included among the reference datasets shown in Fig. 6b (e.g., Wex et al., 2019; Mason et al., 2015). However, we retain the McCluskey et al. (2018) parameterization in Fig. B12 (now Fig. S13) not as a regional comparison but as a common benchmark and parameterization for clean marine background INP concentrations. Its inclusion serves to illustrate that the observed INP concentrations consistently exceed clean marine background levels, supporting the interpretation of a terrestrial INP source contribution.

- Line 425: Explain how the >0.5 µm concentration was determined.
- We have added the explanation to the text (line 529): […], which are both based on the number concentrations of aerosol particles with diameters above 0.5 µm (determined by integrating the bins of the optical particle counters > 0.5 µm; specifically, > 497 nm for the POPS and > 523 nm for the Fidas Frog).

- Line 439: Clarify how the ice-nucleation mechanisms differ; both appear to involve immersion freezing.
- We have adapted the text as follows (from line 546): Interestingly, the *D15* dust parameterization captures the INP concentrations reasonably well, with 85 % of data points falling within one order of magnitude of the 1:1 line. This is despite differences in the dominant ice-nucleating components, as *D15* is based on mineral-dominated desert dust while the aerosol samples show a high heat-labile, likely biological, contribution based on the treatment results. In contrast, the D10 parameterization shows larger deviations, with only 43 % of data points within one order of magnitude.

- Figure 6(a): INP concentrations in a cumulative spectrum should not decrease toward lower temperatures. Please clarify the cause of this artefact.
- The decreasing concentrations at lower temperatures in some spectra are artifacts of background subtraction when sample concentrations approach background levels. We have added the following note in the caption: Note that some spectra show decreasing concentrations toward lower temperatures, which is an artifact of water background subtraction: when the rate of increase in frozen fraction of the water background exceeds that of the sample at a given temperature step, the background-corrected INP concentration decreases. This typically occurs when sample INP concentrations approach the water background level.

- Line 461: Consider “low INP content” instead of “ice-nucleation efficiency.”
- The wording has been adapted as suggested.

- Line 487: Specify which differences suggest a local influence.
- We have adapted the text as follows (line 604): The relatively higher INP concentrations (> -20 °C) at the *plain* site compared to the other sampling sites suggest a potential local influence of the glacial outwash plain on the atmospheric INP population in Narsarsuaq.

- Line 489: Explain why slopes and ratios in 2°C bins were chosen.

- We have added the following explanation in the text (line 295): The 2 °C window size balances spectral resolution against measurement noise, and the 1 °C step size ensures continuous coverage across the temperature range.

- Line 494: Specify the original variables.
- We have specified them as follows (line 612): original variables (slopes and logarithmic ratios)

- Line 496: The clusters are not clearly visible; consider highlighting them.
- We have added 1.5σ covariance ellipses to Fig. 8 to highlight the bulk and filter sample groups and make the overlap between the two datasets more visible.

- Line 498–500: Explain how the analysis supports this interpretation. A more detailed discussion of clusters, separations, and outliers would be helpful.
- We have extended the description and interpretation as follows (from line 611): Overall, bulk and filter samples tend to separate along PC1, indicating different INP spectra characteristics. According to the loading arrows, which indicate the contribution of the original variables (slopes and logarithmic ratios) to the principal components by their length (magnitude of contribution) and orientation (correlation with the principal components), logarithmic ratios mainly contribute to PC1, whereas slopes contribute to PC2. However, there is some overlap between bulk dust and filter samples. Interestingly, most (10 of 13) of the Narsarsuaq *plain* filter samples cluster in the same region as Narsarsuaq bulk dust samples. The overlap suggests that the atmospheric INP spectra at the plain site share the characteristic temperature dependence of the local bulk dust, supporting the interpretation that bulk dust suspended from the outwash plain could contribute to the atmospheric INP population at this location. The Narsarsuaq *col* filter samples, as well as the Narsaq samples (*NIRS* and *hill*) plot mostly separately from the Narsarsuaq bulk dust and *plain* filter samples, and could indicate more mixed INP sources, and therefore no or little influence from outwash plain dust. Outliers, such as the Narsaq *hill* filter sample (upper left in Fig. 8) may reflect a distinct and sporadic INP source, potentially consistent with the pollen signal suggested by the sharp INP increase between -20 and -18 °C (Sect. 3.2.1). The cluster of the heat treated Narsarsuaq *plain* bulk dust and filter samples persists (Fig. S16), though the result is more uncertain due to the much smaller sample size.

- Line 510–513: The statements are cautious; but consider clarifying which findings are well supported and which remain speculative.
- We adapted this section as follows (from line 636): Overall, our observations indicate that atmospheric INP concentrations in southern Greenland were driven by a combination of regional background sources and highly localized contributions, as indicated by overlapping, but distinct INP spectra at the different locations. The glacial outwash plain in Narsarsuaq likely served as a local rather than regional HLD INP source under the observed calm summertime conditions, as supported by similar INP spectra between bulk dust and Narsarsuaq plain filter samples. The substantially larger heat-labile fractions in atmospheric INPs compared to bulk dust samples demonstrate that additional biogenic sources, potentially from nearby vegetated surfaces, likely have an important contribution to the atmospheric INP population and likely drive part of the observed variability across sampling sites.

- Line 645: Quality check 3 enforces the assumption of Poisson-distributed INPs. If data fail this check, clarify whether this indicates experimental issues or contains useful information.
- We have added this clarification to check 3 (line 784): A failure of this check most likely indicates experimental issues such as sample handling errors or dilution mistakes, rather than a physically meaningful deviation from Poisson-distributed INPs, and the affected data are therefore excluded from further analysis.

- Line 659: As experiments are conducted at even faster temperature changes and down to -35°C, the calibration should be performed mirroring these specifications.
- We thank the reviewer for this valid comment. Ideally, the calibration would be performed under conditions mirroring the actual experimental temperature range and cooling rates. However, the calibration was performed using the available cooling infrastructure, which limited the accessible temperature range to -15 °C and above, and did not allow replication of the faster cooling rates used in the experiments. We acknowledge this as a limitation. Nevertheless, we argue that the impact on the reported results is limited: the calibration offsets were determined from a linear fit with slopes equal to 1, indicating that the sensor responses are linear and that a constant offset correction is appropriate across the full temperature range. These uncertainties are expected to be small relative to the variability in INP concentrations reported here, and we have added a note to the manuscript acknowledging this limitation.
- We have added the following to the paragraph (line 830): Ideally, the calibration should be extended to lower temperatures but was limited by the available infrastructure. However, the linear sensor response (slopes equal to 1) suggests that the constant offset correction is a reasonable approximation across the full experimental temperature range, with any additional uncertainty at lower temperatures expected to be minor.

- Line 695: Define minimum and maximum deviation and clarify whether a temperature gradient exists across the setup.
- We adapted the text as follow (from line 836): Due to the sensor positions and spatial differences in heat transfer from the cooling liquid, small temperature gradients exist across the block, with outer sensors showing higher temperatures than those toward the center of the block. For each of the 212 experiments, the standard deviation across the eight sensors is calculated at each time step, and the minimum and maximum standard deviations are shown in Fig. A3, illustrating that the temperature spread was not constant over time.

- Line 699: Quantify how sensitive temperature offset and variability are to dry air flow.
- We adapted the text to quantify the change in offset and variability:
- Line 841: This change coincided with improvements to the laboratory air conditioning, which created a colder and drier laboratory environment. As a result, SPICE could be operated with minimal or no filtered airflow, whereas previously stronger airflow was required to prevent condensation. This change in experimental conditions reduced the spatial temperature variability across the block (mean maximum standard deviation decreasing by 0.21 °C).
- Line 882: The smaller offset between block and well temperatures (0.51 °C smaller than for period 1), [..]

- Line 703: Reporting maximum temperature spread may be more informative than standard deviation.
- We prefer to retain the use of standard deviation as the primary measure of temperature spread, as it is the statistically appropriate quantity to characterize variability around the reported mean temperature across the eight sensors, and is consistent with the methodology reported for other ice nucleation freezing setups (e.g., David et al., 2019; Miller et al., 2021). However, to additionally convey the total spatial temperature spread across the block, we have added a second panel to Fig. A4 showing the temperature range (T_{max} - T_{min}) and adapted the text accordingly (from line 847):

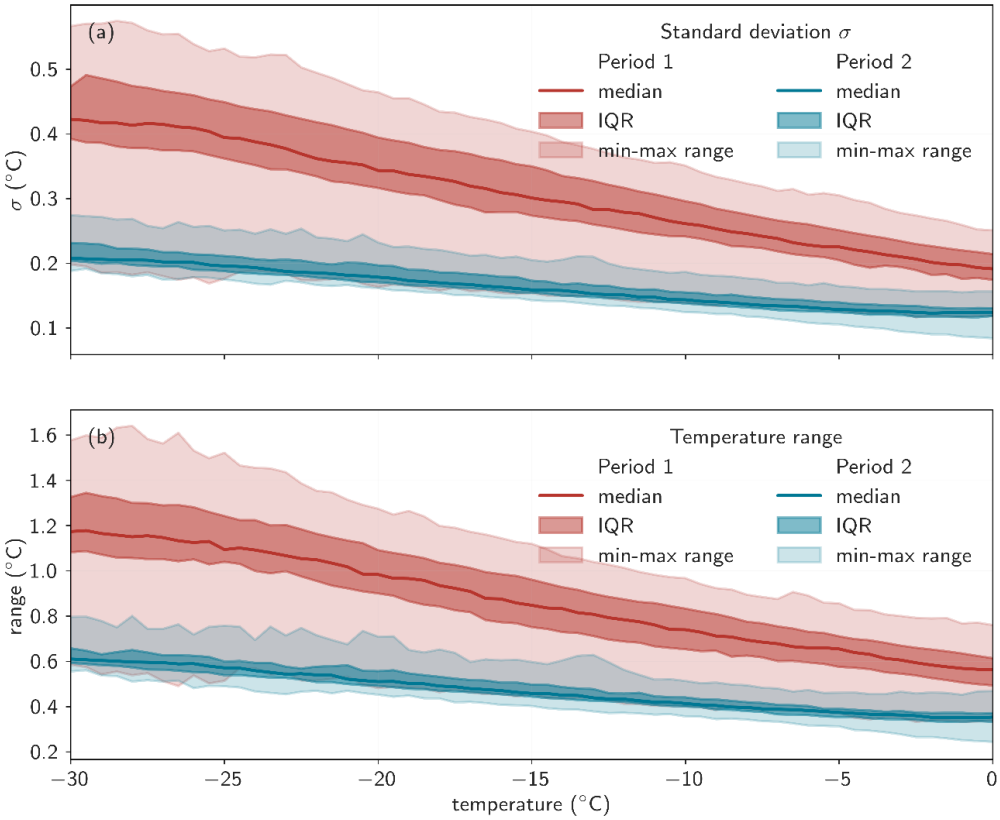

Figure A3. Temperature standard deviation and total temperature spread as a function of temperature. Median, interquartile range (IQR) and minimum-maximum range temperature of (a) standard deviations and (b) total temperature range (T_{max} - T_{min}) derived from eight PT100 temperature sensors across 212 experiments, separately for period 1 and period 2. Both panels illustrate the variability in temperature homogeneity within the aluminum block as a function of temperature.

Fig. A4 shows the median, interquartile range, and full range of temperature standard deviations and total temperature spread across the eight sensors as a function of temperature for both periods. Temperature variability increases toward lower temperatures in both cases, but more strongly in period 1. At -30 °C, the maximum standard deviation reaches 0.57 °C in period 1, compared to 0.28 °C in period 2, which is similar to comparable set-ups (e.g. 0.6 °C in David et al., 2019; 0.5 °C in Miller et al., 2021). The median total temperature spread across the block reaches 1.2 °C and 0.6 °C for period 1 and period 2, respectively.

- Line 724: Clarify that filtered airflow refers to airflow over the pane.
- We have clarified this: with filtered airflow over the glass pane

- Eq. A9: Clarify whether uncertainty includes the range between lowest and highest sample temperatures.

  As described in the text, the uncertainty in well temperatures is estimated by propagating the calibration uncertainty together with the maximum temperature standard deviation across the block ($\sigma_{T_{block}}$), as given in Eq. A9. We would argue that the standard deviation is the statistically appropriate quantity for uncertainty propagation because the reported block temperature is the mean across the eight sensors, and the standard deviation characterizes the typical deviation from this mean. Using the full temperature range would overestimate the uncertainty, as it reflects the most extreme sensor-to-sensor difference rather than the variability

around the reported value. The total temperature spread across the block is additionally reported in Fig. A4b for reference.

- Line 748: Clarify how the detection limit is applied, especially since measurements below -24.71°C are reported.
- We note that -24.71 °C represents the characteristic background freezing temperature of the water across all experiments, rather than a hard detection limit below which data are excluded. Sample INP concentrations are corrected against the water background of each individual experiment (Eq. A4), and data points are retained at temperatures where the sample frozen fraction exceeds the background frozen fraction. Measurements below -24.71 °C are therefore possible where samples show detectable INP activity above the water background. We have adapted the text and terminology accordingly, as shown below (from line 878):

A2.2 Characteristic background freezing temperature

All experiments include negative controls of the background water to quantify potential background freezing events caused by impurities in the water, contamination during handling, or the PCR tray material itself. We use SA water for our experiments, which has previously been shown to exhibit low and more reproducible freezing behavior than other purified waters (David et al., 2019; Miller et al., 2021), consistent with our comparison in the laboratory (Fig A6a). We define the characteristic background freezing temperature of the set-up as the median of the median freezing temperature ($T_{50,}$ where frozen fraction = 0.5) of 152 experiments (Fig. A6b, including both dedicated water testing experiments and negative controls within sample freezing assays), which is -24.71 °C (interquartile range of 0.88 °C) for 50 µL droplets. This characteristic background freezing temperature is therefore similar to other INP freezing set-ups (e.g., Chen et al., 2018; David et al., 2019; Miller et al., 2021).

- Figure B12: $n_{s,\ geo}$ is expected to be larger than $n_{s,\ BET}$, even for near-spherical particles (e.g., Hiranuma et al., 2014). Direct comparison should be avoided.
- We have switched from BET to geometric surface area normalization throughout the manuscript and have updated the figure (now Fig. S13).

Technical corrections:

- Line 130: Sampling sites are shown in Fig. C1, not C2.
- Corrected

- Line 152: Handix
- Corrected

- Line 156: Should it be filter “samplers”
- Corrected

- Line 323: Fig. 3 in Barr et al. (2023), not Fig. 4.
- Corrected

- Figure 8: The figure should fit on one page (print out) including the caption. Remove or move panel titles inside the boxes. (a) bar plots on a log-scale are misleading as their area does not reflect the values. Consider removing the bars.
- We have removed the bars and adapted the figure (now Figure S17) to fit in on one page.

- Line 718: Fig. A5c
- Line 726: Fig. A5a
- We have noticed that in Fig. A5, the panel subpanel labels were incorrect and have adapted them. Therefore, the in-text description remains accurate.

- Table C1: The coordinates appear inconsistent. L2a and L2b should have the same coordinates, coordinates from Ferpècle are all the same as L2b, LG2 is 48°. Please verify.
- We coordinates were indeed inconsistent, we have corrected them.

- Figure C4 appears identical to the top panel of Figure C2.
- The top panel of C4 has been removed.

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