Referee comment 2: This manuscript presented a case study on the impacts of aerosol physiochemical characteristics on the ice nucleation activity. They conducted micro-spectroscopic single-particle analysis and derived the ice nucleation concentrations from droplet freezing experiments. Although with limited number of samples analyzed, the manuscript shows the complexity of interaction between particle mixing state and ice nucleation activity. The topic of this study fits the scope of this journal. There are several issues need to be addressed before it can be considered for publication.

Response: We thank the reviewer for the careful reading of our manuscript and the helpful suggestions provided. We have addressed each comment and revised the text to improve clarity and strengthen the presentation of our results. Reviewer comments are shown in **black**, and our responses are shown in **blue**.

Major issues:

1. Missing information on the methods for (1) INP concentration calculation, (2) fraction of heat liable stable INP, (3) FLEXPART back-trajectory analysis. Brief descriptions are important to readers and reviewers for judgement on the methodology.

   Response: The authors thank the reviewer for raising this point. We agree that brief, reader-facing method descriptions are important for evaluating the robustness and reproducibility of the approach. In the revised manuscript, we have added concise methodological text describing (1) the INP concentration calculation and (2) how heat-labile vs. heat-stable INP fractions are quantified (including the heat-treatment protocol and how the pre-/post-heating INP spectra are compared across temperature).

   For (3) FLEXPART back-trajectory analysis**,** we respectfully note that this information **is** already provided in the Supplement, where we describe how FLEXPART back-trajectories were used to estimate surface-type influence and free-tropospheric (FT) residence over a 168-hour period, with the results summarized in **Table S3** (and associated interpretation in the Supplementary text). To avoid redundancy while still addressing the reviewer's concern, we have now added an explicit cross-reference in the main text directing readers to the Supplementary section that contains the FLEXPART methodology and outputs (Supplement, **Section S2** and **Table S3**).

S4. INP concentration calculation:

First, the concentration of ice-nucleating particles in the HPLC suspension, $C_{\mathrm{INP}}(T)$(L^{-1}) was calculated from the droplet-freezing assay as-

$$C_{INp}(T) = -\frac{\ln\left(f_{unfrozen}(T)\right)}{V_d} \qquad [1]$$

where f_{unfrozen} is the fraction of droplets that remained unfrozen at temperature T(i.e., the number of unfrozen droplets divided by the total number of droplets), and $V_d$is the volume of an individual droplet ($V_d = 3$ μL).

Next, ambient INP concentrations, $n_{\mathrm{INP}}(T)$(L^{-1} air), were obtained by scaling $C_{\mathrm{INP}}(T)$using the dilution applied during analysis and the ratio of suspension volume to sampled air volume:

$$n_{INP}(T) = C_{INP}(T) \times DF \times \frac{V_1}{V_{air}} \qquad [2]$$

where $DF$is the serial dilution factor (e.g., 1, 10, 100, …), $V_{\mathrm{air}}$is the sampled air volume, and $V_1$is the (provided) total volume of the original suspension associated with the analyzed sample.

S5. Fraction of heat labile and heat stable INP calculation:

Equations 1 and 2 show the calculation of heat labile and stable INP percentages, respectively.

$$\%INP_{Heat\ Labile}(T) = \frac{n_{INP,untreated}(T) - n_{INP,heated}(T)}{n_{INP,untreated}(T)} \times 100 \qquad [3]$$

$$\%INP_{Heat\ Stable}(T) = 100 - \%INP_{Heat\ Labile}(T) \qquad [4]$$

where $n_{INP,untreated}$(T) is the number concentration of INPs before heat treatment and $n_{INP,heated}(T)$ is the number concentration of INPs after heat treatment at that temperature.

INP concentrations and their differences span orders of magnitude and are commonly interpreted on a logarithmic scale. The heat-labile fraction reported here is calculated as a linear percent difference between paired n_{INP}(T) spectra (Eqs. 3-4) and should therefore be interpreted as an approximate and comparative metric. In particular, when n_{INP},heated(T) approaches the detection limit, the inferred fraction becomes sensitive to detection-limit treatment. We use this metric to summarize relative heat sensitivity across samples rather than to define an absolute biological fraction.

2. The focus of this manuscript is connecting the particle physiochemical properties to ice nucleation activity, however, this a mismatch in particle size and sampling duration between the samples used for particle characterization and ice nucleation experiments. In addition, some organic or salt components are soluble, it is very tricky when discuss the impacts of organics what like contain soluble components on the immersion freezing ability of particles. Please comment on these and make this clear or add a few sentences to discuss these potential artifacts in the manuscript when possible.

Response: The authors thank the reviewer for raising this important point. The authors acknowledge that the particle populations used for offline physicochemical characterization (CCSEM/EDX and STXM/NEXAFS) and for immersion-freezing measurements are not perfectly matched in sampling duration and size representation; therefore, the characterization results are used to provide compositional and mixing-state context for each sampling period rather than a strictly size-resolved, time-synchronous linkage to n_{INP}. The authors also note that the immersion-freezing assay is performed on aqueous extracts, and soluble components (salts and water-soluble organics) can dissolve and redistribute during extraction and droplet preparation, which may alter surface accessibility relative to the ambient aerosol. The manuscript has been updated to explicitly state these potential artifacts/limitations. Mechanism-level discussion of how organics may influence freezing (via water uptake/phase state and related processes) is addressed separately in the revised text responding to the L394 comment from other reviewer.

"Offline particle characterization and immersion-freezing measurements were not performed on identical particle populations with perfectly matched sampling duration and size range; therefore, composition and mixing-state results are interpreted as representative context for each sampling period rather than a direct size-resolved predictor of n_{INP}*. In addition, the droplet freezing assay is conducted on aqueous extracts, and soluble salts and water-soluble organics may dissolve and redistribute during extraction and droplet preparation, potentially modifying surface accessibility compared to the ambient particle state."*

3. Ice nucleation ability (INA) and ice nucleation concentration are different concepts. Ice nucleation concentration is not only depended on the ice nucleation ability of particles but also on the total particle surface area in the sample. To compare the INA of different particle samples, it would be better to use normalized parameters, for example, ice nucleation active site density (over particle surface area or mass). Is there particle surface or mass measurements or estimation that can be used for this purpose? This is also related to previous comment. In addition, frozen fraction and freezing temperature (e.g., median freezing temperature) are usually presented in ice nucleation references. These are missing here.

   Response: The author thanks the reviewer for pointing this out. Yes, we agreed with the reviewer. Unfortunately, APS at the Gruvebadet observatory was down from July 2020 to January 2023 for its maintenance, and we are not able to report ice nucleation efficiency for our study period (i.e., October 2020 – March 2021). We have attempted to normalize our n_{INP} to submicron aerosol particle surface area concentrations estimated by an SMPS. As expected, SA2 and SA4 (corresponding to NyA_68 and _72 in Table 4.4. of Diep (2023))(Diep, 2023) exhibit higher ice nucleation active surface site densities than other samples. Regardless, since we are missing supermicon aerosol particle size distribution measurements, we would humbly like to report n_{INP} solely for this manuscript. Also, we are revising the manuscript to replace INA with INP concentration.

4. The descriptions and discussion in the differences for five samples are inconsistent, for example, in the Line 277-283, L305, it claims they are similar; however, in Line 307, 321-326, the discussion assumes that five samples have different INA. Please provide quantitative comparison with uncertainties on this comparison.

Response: The authors thank the reviewer for pointing out this inconsistency. The authors agree that the previous wording did not clearly distinguish between similarities in the overall temperature-dependent trend of n_{INP} (T) and differences in n_{INP} magnitude and heat sensitivity among the five samples. In the revised manuscript, the authors have updated the text in L277-283 and L305 to clarify that the samples are "similar" only in terms of the general n_{INP} (T) shape. The authors also revised the discussion in L307 and L321-326 to consistently describe sample-to-sample differences in n_{INP} (T). To provide a quantitative comparison with uncertainties, the authors now reference the updated Fig. 5, which shows untreated and heated n_{INP}(T) with uncertainty bounds for each sample and the corresponding heat-labile versus heat-stable fractions. In addition, the authors corrected the -25 °C summary statistics (mean 0.27 L^{-1}, reported as 0.3 after rounding; range 0.20-0.34 L^{-1}) to ensure the numerical comparisons are internally consistent.

"Figure 5 displays the temperature-dependent INP number concentrations, $n_{INP}(T)$, for aerosol samples SA1-SA5 and their response to heat treatment. At -15 °C, untreated n_{INP} ranged from 0.001 to 0.004 L^{-1} (0.003 ± 0.001 L^{-1}; Figure. 5a–e). At -25 °C, n_{INP} increased by approximately two orders of magnitude to 0.27 ± 0.06 L^{-1}, with a relatively narrow inter-sample range (0.20-0.34 L^{-1}). Overall, the $n_{INP}(T)$spectra show similar temperature dependence across samples, whereas heat sensitivity differs substantially. Using a conservative detection-limit substitution for heated spectra (Section S5), the mean heat-labile fractions (mean ± SD across temperatures) are 76 ± 19% (SA1), 54 ± 30% (SA2), 37 ± 33% (SA3), 54 ± 34% (SA4), and 48 ± 30% (SA5) (Figure. 5f–j)."

Technical comments:

1. L119, for the HYSPLIT analysis, using 50 m maybe too low, 100 or 200, 500m which are usually used. Please comment on this.

   Response: The authors thank the reviewer for this comment. The authors agree that endpoint height can influence HYSPLIT pathways, and 100-500 m AGL is commonly used for regional-scale transport assessments. In this study, the authors selected 50 m AGL to represent the near-surface air mass most relevant to the filter sampling inlet and local boundary-layer conditions at the site. Importantly, the main interpretation in the manuscript is based on consistent transport patterns rather than a single trajectory line. To address the reviewer's concern and demonstrate sensitivity to the chosen endpoint height, the authors have added additional back-trajectory calculations at multiple endpoint heights (100, 200, and 500 m AGL) in the Supplementary Information (new figure), which show that the overall source regions and qualitative conclusions are not sensitive to the endpoint height used.

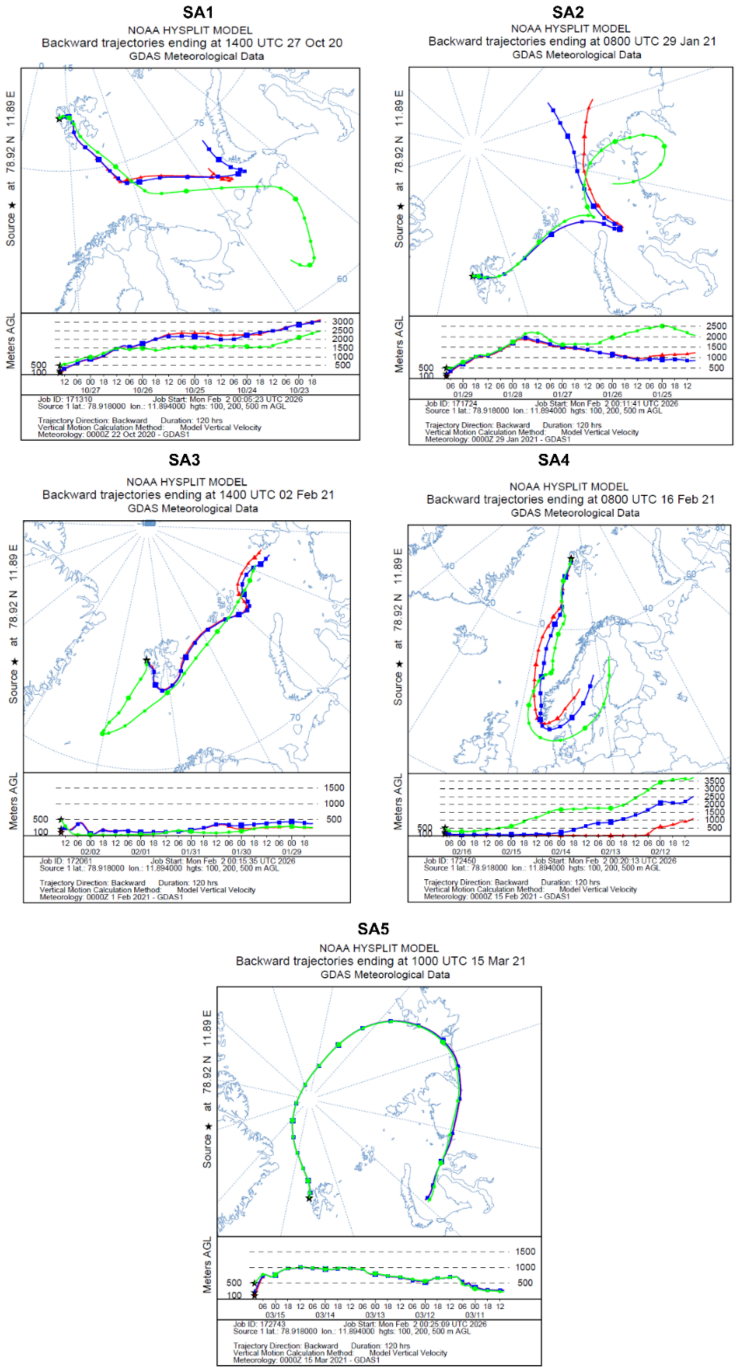

Figure S10. Height-resolved 120 h NOAA HYSPLIT backward trajectories for the five sampling periods (SA1–SA5), driven by GDAS meteorological fields. For each sample, trajectories are shown for endpoints at 100 m (red), 200 m (blue), and 500 m (green) above ground level (AGL) at the sampling location (star). The lower panels show the corresponding altitude history along each trajectory. This sensitivity analysis illustrates that the inferred transport pathways and source regions are broadly consistent across the selected endpoint heights.

2. L280, the mean is 0.3 but the range is between 0.1-0.3? these numbers don't match.

Response: The authors thank the reviewer for catching this inconsistency. The mean value is correct (0.27 L^{-1}, reported as 0.3 after rounding), but the range in the text was reported incorrectly. Based on the underlying values, the correct range at −25 °C is **0.20–0.34 L^{-1}.** The manuscript has been updated accordingly.

3. Figure 5, how do you calculate the percentage of heat liable INPs? In this figure, are these averaged over different temperatures?

The authors thank the reviewer for this question. The percentage of heat-labile INPs is calculated from paired unheated vs. heat-treated measurements at the same temperature as the fractional loss upon heating:

Equations 1 and 2 show the calculation of heat labile and stable INP percentages, respectively.

$$\%INP_{Heat\ Labile}(T) = \frac{n_{INP,untreated}(T) - n_{INP,heated}(T)}{n_{INP,untreated}(T)} \times 100 \qquad [3]$$

$$\%INP_{Heat\ Stable}(T) = 100 - \%INP_{Heat\ Labile}(T) \qquad [4]$$

The heat-stable fraction is the remaining percentage. The stacked plots on the right of Fig. 5 show the temperature-resolved fractions across the plotted temperature range; the single percentage value shown inside each panel is the mean heat-labile fraction averaged over the temperatures displayed (i.e., it is not a single-temperature value).

4. Figure 8, how many data points were used for this analysis? How about the significance level?

Response: The authors thank the reviewer for the question. Five data points ($n = 5$; one value per sample, SA1-SA5) were used to calculate each correlation coefficient in Fig. 8. The authors now state this explicitly in the figure caption and/or main text. Because of the very small sample size, the authors do not emphasize statistical significance; p-values were computed for completeness but are interpreted cautiously, and the correlations are presented as an exploratory summary rather than a robust statistical test.

References

Diep, T. T.: CHEMICAL COMPOSITION AND IMMERSION FREEZING ACTIVITIES OF AEROSOL PARTICLES IN THE EUROPEAN ARCTIC, 2023.