

Referee comment 1: Lata et al. present in their manuscript "Measurement report: Role of Organic Coating and Chemical Composition on Ice Nucleation Potential of Atmospheric Particles in European Arctic" results of STXM/NEXAFS, SEM-EDX, and INP measurements of samples collected on Svalbard during the winter season of 2020/2021. They found indications that the properties of the organic coating influence the ice nucleation activity of their samples. While this study only features five samples, which of course limits the statistical significance of the results, one must recognize the labor that went into the analysis of these samples. STXM/NEXAFS measurements of so many individual particles are no small feat. Furthermore, STXM/NEXAFS is rarely used in studies of ambient aerosol samples in conjunction with ice-nucleating particle measurements, which makes these results valuable. Considering the limited scope of the study due to the small sample size and the value of the results, I think the publication as a measurement report is appropriate after making some revisions.

Please see my comments and suggestions below. Some comments are marked with "****"; these are the ones I deem more important and that need to be addressed. There is also a list of technical comments (typos, etc.) at the end.

Response: We sincerely thank the reviewers for their thoughtful and constructive feedback. We have carefully considered each point and revised the manuscript to improve its clarity, rigor, and overall quality. Below, reviewer comments are shown in black, followed by our responses in blue.

Comments and suggestions

L51 Immersion freezing is mentioned for the first time without further explanation. I suggest adding one or two sentences to the previous paragraph to explain that there are different freezing modes and describe immersion freezing, stating that it is the most relevant mode for MPCs.

Response: We thank the reviewer for this helpful suggestion. We have added a brief explanation of the different ice nucleation modes and clarified that immersion freezing is the most relevant freezing pathway for mixed-phase clouds (MPCs). This additional context has been incorporated into the preceding paragraph to improve clarity for readers.

"Ice formation in clouds can occur through several heterogeneous freezing modes, including deposition, condensation, contact, and immersion freezing. Among these, immersion freezing, where an ice-nucleating particle is immersed within a supercooled cloud droplet, is considered the dominant pathway in mixed-phase clouds (MPCs)."

L62 Since the em dash is now often seen as a sign of AI usage, I recommend removing it and rephrasing the sentence if AI was not actually used. Using AI is generally fine, but according to the ACP submission guidelines, if AI was used to prepare the manuscript, its use must be described in the Methods section or the Acknowledgments.

Response: We thank the reviewer for raising this point. The em dash has been removed and the sentence has been rephrased accordingly. We clarify that no generative AI tools were used in the preparation of this manuscript. Grammarly was used solely for language editing and grammar

correction, and this has now been explicitly stated in the Acknowledgments section in accordance with ACP guidelines.

“The authors used Grammarly solely for grammar and language editing.”

L91 - L94 Also specify the distance between the Gruvebadet and the AWIPEV station.

Response: We thank the reviewer for this suggestion. We have now specified the distance between the Gruvebadet Observatory and the AWIPEV research station to provide clearer spatial context for the measurements.

“The Gruvebadet Observatory is located approximately 1.5 km from the AWIPEV Arctic Research Base in Ny-Ålesund, Svalbard.”

L95 Introduce the disdrometer properly (see the 2nd technical comment for details)

Response: We thank the reviewer for this comment. The disdrometer has now been properly introduced at its first mention, including a brief description of its operating principle and its role in measuring precipitation properties relevant to this study. The revised text addresses the technical details noted in the second technical comment.

“Precipitation phase/type was determined using a Parsivel² optical disdrometer (OTT HydroMet, Germany; hereafter Parsivel²), which measures hydrometeor size and fall velocity (and associated number concentration) to characterize precipitation type and intensity during the aerosol sampling periods.”

L96 What is "Pluvio total"? Does it simply mean that the precipitation volume is derived from the OTT Pluvio² L regardless of the phase determined by the OTT Parsivel²?

Response: Yes. By “Pluvio total,” we mean the total liquid-equivalent precipitation measured by the OTT Pluvio² L weighing gauge, independent of precipitation phase. The Pluvio² L reports accumulated precipitation mass converted to liquid-equivalent depth (mm), regardless of whether the hydrometeors are rain, snow, or mixed phase. Precipitation phase information is determined separately by the OTT Parsivel² disdrometer and is not used in calculating the precipitation amount reported by the Pluvio² L.

To clarify this point, we have revised the text at Line 96 as follows:

“Precipitation was measured using an OTT Pluvio² L weighing gauge, which reports total liquid-equivalent precipitation (mm) regardless of hydrometeor phase. Precipitation phase (rain, snow, or mixed) was identified independently using an OTT Parsivel² disdrometer.”

L98 - L99 Do I understand correctly that the sampling for the INP analysis is continuous, but co-located samples for the single particle analysis were collected only on five occasions? Or were co-located samples always taken, but only five were analyzed? In either case, the text should mention the criteria used to select these five occasions.

Response: We thank the reviewer for pointing out this ambiguity. We clarify that aerosol and INP samples were collected throughout the campaign period; however, this study focuses on five selected cases (SA1-SA5) for which collocated single-particle chemical characterization and offline INP measurements were both available. These five cases were selected from the broader dataset to ensure (i) temporal overlap between single-particle and INP sampling, (ii) sufficient particle loading for microscopy and spectroscopy, and (iii) representation of distinct meteorological and air-mass conditions. This clarification and the selection criteria have now been explicitly stated in the revised manuscript. This directly answers both interpretations the reviewer raised.

“Aerosol and INP samples were collected throughout the campaign; however, among the available samples, we selected five cases (SA1-SA5) for which collocated samples for single-particle chemical composition and INP analysis were available (Table 1).”

*** Particle sizes

I am a bit surprised by the large number of supermicron particles you found on the TEM grids (Fig. 3). The TEM grids should mainly contain particles with an aerodynamic diameter between 250 and 500 nm, because you only used the last impactor stage with $d_{50} = 250$ nm and the previous stage has $d_{50} = 500$ nm. Of course, AED and aerodynamic diameter are not identical, and it is known that the AED is on average larger than the geometric diameter and also that the aerodynamic diameter is typically larger than the geometric diameter (Huang et al., 2021). But knowing this and considering typical shape factors and density of SSA, I find it hard to imagine a scenario where many particles with a significantly larger AED than aerodynamic diameter occur.

Do you have an explanation for the large number of supermicron particles? Was e.g., the impactor operated with the correct flow rate?

Response: We thank the reviewer for this careful and important comment. We confirm that the Sioutas cascade impactor was operated at the manufacturer-recommended flow rate of 9 L min^{-1} , corresponding to the nominal aerodynamic cut-off diameters ($D_{50} = 0.25 \text{ }\mu\text{m}$ for Stage D and $0.5 \text{ }\mu\text{m}$ for the preceding stage). The particle sizes reported in Fig. 3 are projected area-equivalent diameters (AEDs) derived from two-dimensional electron microscopy images and are therefore not directly comparable to aerodynamic diameters. Aerodynamic diameter depends on particle density, shape factor, and dynamic behavior in airflow, whereas AED reflects projected particle area on the substrate. Typically, AED is larger than aerodynamic diameter for non-spherical, porous, or hygroscopic particles such as sea-salt. While we cannot entirely exclude contributions from particle bounce or substrate effects, there is no indication of incorrect impactor operation or flow conditions during sampling. We have added text to the Methods section to clarify the distinction between aerodynamic diameter and microscopy-derived AED and to acknowledge this limitation in size interpretation.

“Particle sizes reported in this study are based on area-equivalent diameters (AEDs) derived from electron microscopy images and should not be interpreted as aerodynamic diameters. AED does not account for particle density or dynamic shape factor. As a result, particles collected on Stage D ($D_{50} = 0.25 \text{ }\mu\text{m}$) may exhibit AEDs exceeding the nominal aerodynamic cut-off. This limitation

should be considered when interpreting size distributions derived from microscopy.”

L108 I assume these filters are made of polycarbonate

Response: Yes, these are polycarbonate filters.

L110 The citation in parentheses is not needed if the same papers are cited within the sentence

Response: Thanks for pointing that out. I have fixed that.

L111 - L112 Does that mean that some of the samples are measured in regular intervals in Ny-Ålesund, but most samples are analyzed at the authors' home institutes? Why are not all samples measured on-site? Is it the lack of manpower or is on-site only a rudimentary freezing array setup to ensure sample integrity? Even though you cite Li et al. (2023), an additional sentence describing these on-site verification measurements would not hurt.

Response: We thank the reviewer for pointing out the ambiguity in the original text. We clarify that all ice nucleation measurements in this study were conducted exclusively offline after sample transport and cold storage; no on-site freezing measurements were performed by the authors. Li et al. (2023) was cited solely to support the validity of this offline approach, as that study demonstrated negligible differences between on-site and offline INP measurements for Arctic aerosol samples, indicating minimal loss of heat-sensitive INPs during storage and transport. We have revised the manuscript to explicitly state this and avoid any potential misunderstanding.

“All ice nucleation measurements in this study were performed offline after sample collection, transport, and cold storage. The validity of this offline analysis approach, including the preservation of heat-sensitive INPs during freezing and transport, is supported by Li et al. (2023), who reported negligible differences between on-site and offline INP measurements for similarly collected Arctic aerosol samples.”

Section 2.2.

Later, you present results from the FLEXPART back trajectory analysis. Why did you choose to use HYSPLIT to characterize the air masses and identify potential source regions, given that one could argue FLEXPART is better suited for studying aerosol particles?

Response: We thank the reviewer for this insightful question. HYSPLIT was intentionally used in Section 2.2 to provide a qualitative, synoptic-scale characterization of air-mass pathways and potential source regions, which supports meteorological context and event classification. FLEXPART, which is better suited for aerosol-specific analyses, was used separately to quantify particle residence time, surface-type influence, and free-tropospheric contributions, and these results are presented in Supplementary Information. We have clarified this distinction in the manuscript to better explain the complementary roles of the two models. To address this we have added the below lines in section 2.2 where HYSPLIT is introduced.

“HYSPLIT back trajectories were used here to provide a qualitative characterization of air-mass pathways and potential source regions for event classification, while FLEXPART was applied separately to quantify aerosol residence time, surface-type influence, and free-tropospheric contributions (presented in the Supplementary Information).”

L116 While it is known that back trajectories become significantly less accurate the farther back in time they go, and every trajectory going back more than five days should be viewed critically, 48 hours is rather short and might not reveal the full picture. I suggest reproducing Fig. 2 with five-day back trajectories for the supplement. Additionally, I suggest reproducing Fig. 2 with color-coded heights so readers can assess which regions sources close to the surface might be important in.

Response: The authors thank the reviewer for this suggestion. In response, the authors have added 5-day (120 h) HYSPLIT back trajectories to the Supplementary Information to provide a broader transport context, while keeping the 48 h trajectories in the main text as they are most relevant to near-surface conditions immediately prior to sampling. The authors also included height-resolved trajectories (multiple endpoint heights) in the Supplement so readers can assess the influence of transport altitude and potential near-surface source regions.

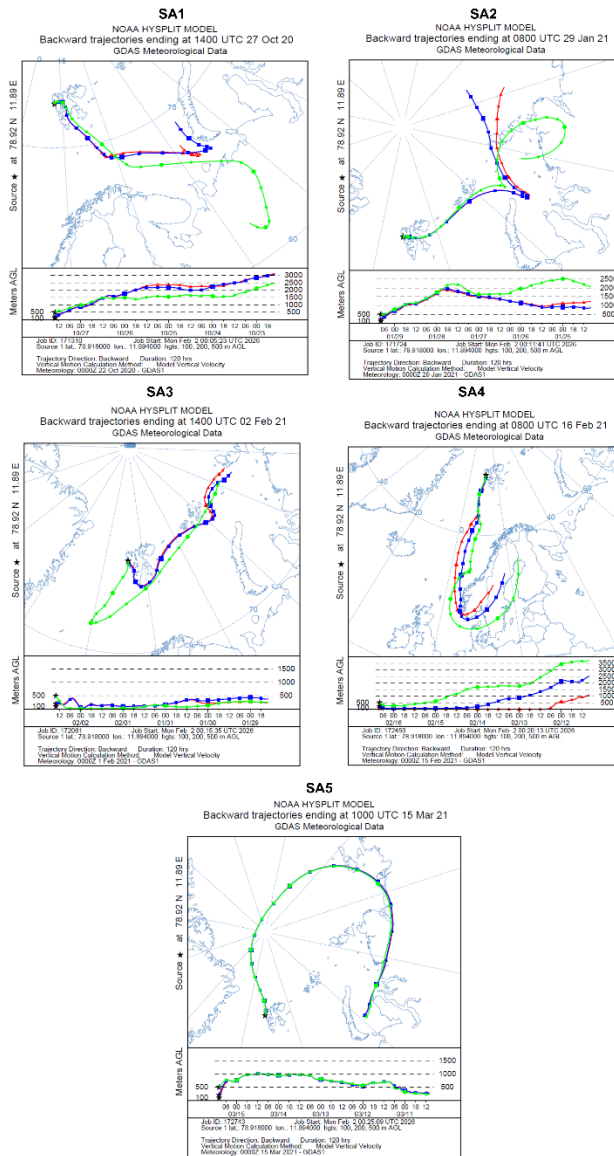


Figure S10. Height-resolved 120 h NOAA HYSPLIT backward trajectories for the five sampling periods (SA1–SA5), driven by GDAS meteorological fields. For each sample, trajectories are shown for endpoints at 100 m (red), 200 m (blue), and 500 m (green) above ground level (AGL) at the sampling location (star). 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.

L135 How does the computer identify the particles? Is it by a brightness threshold of the BSE image? If so, do you think biological particles (i.e., C-rich particles), which appear less bright in the BSE image, could be undercounted? Is there also a lower size limit for what the computer recognizes as a particle? Please mention these things in the manuscript.

Response: Thank you for raising this question regarding particle identification and potential detection bias. In this study, particle identification was performed using the automated CCSEM/EDX analysis. During CCSEM/EDX measurements, the system automatically identifies individual particles deposited on the substrate and acquires both an SEM image and the corresponding EDX spectrum for each detected particle. Image brightness and contrast were adjusted before the automated acquisition to ensure clear visualization of particles with different compositions, including low-contrast, carbon-rich particles. Particle classification was not based on image brightness or morphology. Instead, all particle classes, including those described as biogenic, were determined using a rule-based classification scheme based on elemental composition derived from EDX analysis. As a result, the chemical classification is independent of image contrast. We acknowledge that the automated CCSEM/EDX routine has a lower size detection limit. In this study, we focused on particles in the size range of 0.12 to 5 μm , and particles smaller than 0.12 μm were excluded due to limitations in reliably detecting particles and obtaining confident EDX spectra. As with any automated particle analysis, some undercounting of the smallest or lowest-contrast particles cannot be entirely ruled out. These points have now been clarified in the manuscript.

“Particle identification was performed using the automated CCSEM/EDX routine, which detects individual particles on the substrate and records an SEM image and an EDX spectrum for each detected particle; the routine also derives geometric properties (e.g., projected area and aspect ratio). During acquisition, brightness/contrast were adjusted to ensure detection of low-contrast particles; classification was based on EDX elemental composition and not on image brightness or morphology.”

“Only particles with AED between 0.12 and 5 μm were included; smaller particles were excluded because reliable detection and confident EDX spectra could not be ensured.”

L139 Please provide more information about the EDX measurement, such as how long the spectrum was collected and whether it was measured at only one central spot or if the beam scanned across the detected particle.

Response: Thank you for requesting additional details on the EDX measurement protocol. During the automated CCSEM/EDX analysis, an EDX spectrum was collected for each identified particle with an acquisition time of **10 s**. The spectrum was not acquired from a single central point. Instead, the electron beam sampled the particle across multiple locations within the detected particle region as part of the automated routine, yielding a composition that is representative of the particle as a whole.

We have revised the Methods section to explicitly state the acquisition time and clarify that the reported EDX composition represents a multi-point/area-sampled measurement for each particle.

“For each detected particle, an EDX spectrum was acquired with 10 s collection time to quantify the relative abundances of 16 elements (C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Mn, Fe, Zn, and Cu). The reported composition reflects automated multi-location sampling across the particle area rather than a single central spot.”

***L151 You write that STXM/NEXAFS was performed on a subset of particles that were already analysed with CCSEM/EDX. If you measure a particle twice, isn't it possible that a particle received beam damage, which might alter the STXM/NEXAFS results?

Response: We thank the reviewer for raising this important concern regarding potential beam damage. Although STXM/NEXAFS was performed on a subset of particles that had previously been analyzed by CCSEM/EDX, the two measurements were not conducted on the same irradiated locations. For STXM/NEXAFS measurements, particles were selected from regions of the substrate that had not been previously exposed to the electron beam during CCSEM/EDX analysis. This approach ensured that the STXM/NEXAFS measurements were performed on particles that had not experienced prior electron-beam irradiation, thereby minimizing the potential for beam-induced damage or chemical modification.

We have clarified this procedure in the Methods section of the revised manuscript.

“To investigate the organic characteristics, mixing state of aerosol particles, Scanning Transmission X-ray Microscopy coupled with Near-Edge X-ray Absorption Fine Structure (STXM/NEXAFS) was employed on particles located in substrate regions that were not previously irradiated during CCSEM/EDX analysis, minimizing potential electron-beam-induced damage.”

L165 I suppose that also at the ALS the STXM/NEXAFS measurements do not provide the convenience of computer controlled measurements such as those of the SEM. Therefore, it is valid to mention (somewhere in this paragraph) that the nearly 2000 particles were measured manually to emphasize the effort involved in these measurements.

Response: We thank the reviewer for this suggestion. Unlike the automated CCSEM/EDX analysis, STXM/NEXAFS measurements at the ALS were performed manually, with particles individually located and analyzed. We have added a statement to the manuscript to clarify that nearly 2000 particles were measured manually, highlighting the effort involved in these measurements.

“STXM/NEXAFS experiments were performed at beamline 5.3.2.2 of the Advanced Light Source, Lawrence Berkeley National Laboratory, and the analysis was performed manually by locating and measuring particles individually (1963 particles total). This synchrotron-based technique scans particles at selected photon energies to produce high-resolution maps of elemental distributions and information on chemical bonding states.”

L170 What is the upper detection limit of WT-CRAFT?

Response: The upper DL of INP concentration via WT-CRAFT for this study is 2.95 INP L⁻¹, which represents the highest possible INP concentration with x10 dilution. Note that we only report INP concentrations down to -25 °C concerning potential background contributions as stated in the manuscript, and we did not observe $N_{\text{INP}}(-25\text{ °C})$ above 1 L⁻¹ for this study.

L177 Are the "< 3%" calculated in the f_{ice} or N_{INP} space? I would also recommend showing the field blank and, if available, pure MilliQ water measurements in the supplement.

Response: It is in f_{ice} and consistent with the procedure shown in the supplemental Table S4 of Hiranuma et al. (2019)(Hiranuma et al., 2019). Below we offer frozen fraction spectra for HPLC water and field blanks as a function of freezing temperature (now available in the supplement Fig. S12).

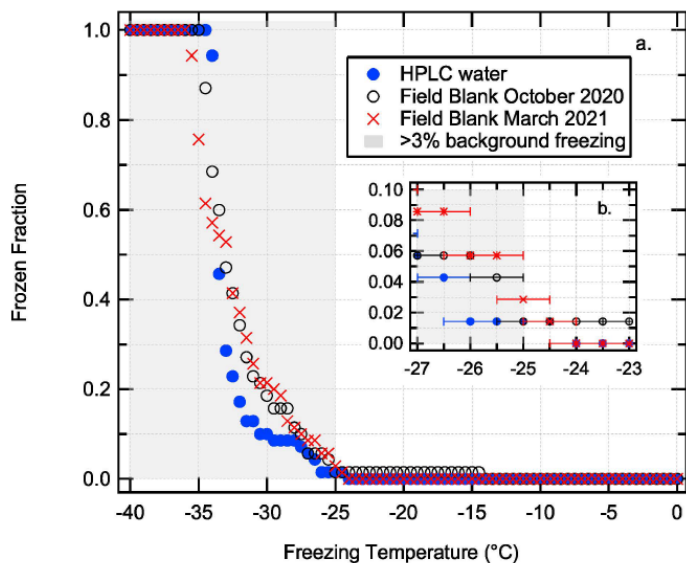


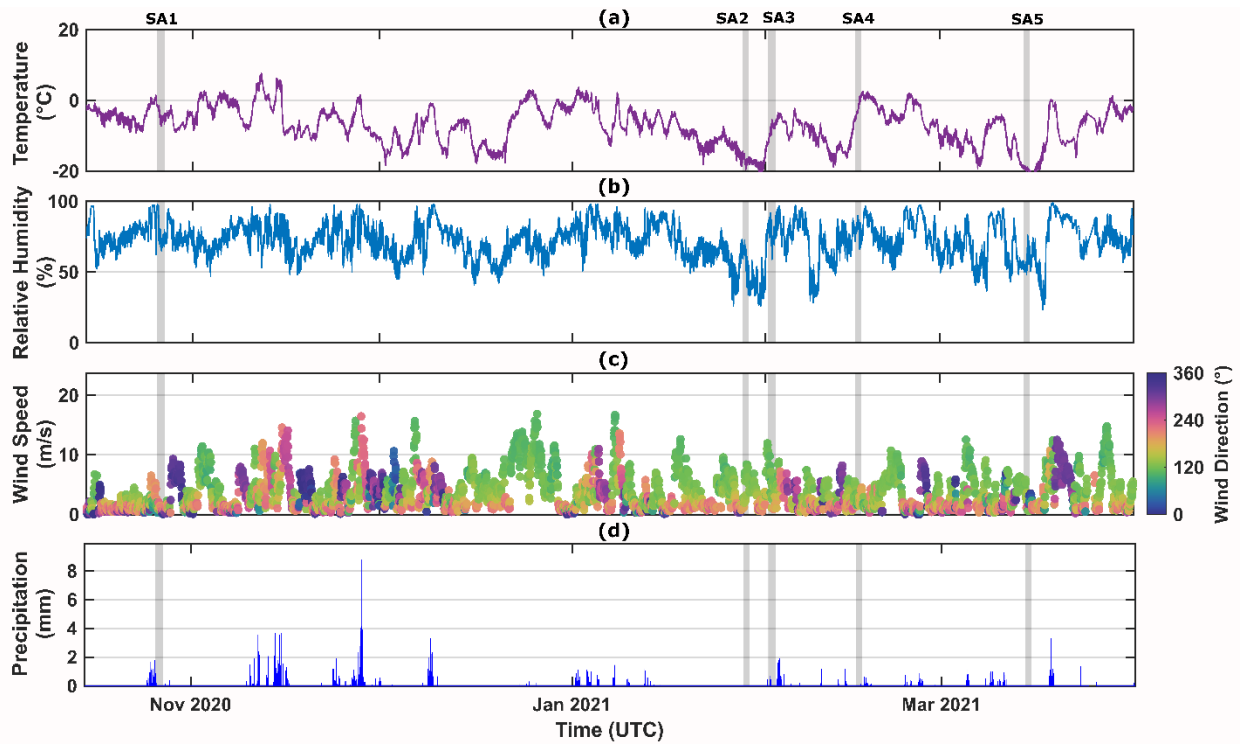
Figure S12. Frozen fraction of HPLC water, as well as field blanks suspended in 5 mL of HPLC water, as a function of freezing temperature. Two field blanks were collected prior to sampling SA1-INP and after sampling SA5-INP. Panel (b) represents a magnified frozen fraction spectra with a temperature uncertainty of ± 0.5 °C.

L181 I would not talk about "airborne samples" as this as this gives the impression of samples collected on aircrafts, balloons, drones etc.

Response: We thank the reviewer for pointing this out. We agree that the term “airborne samples” may be misleading in this context. The text has been revised to use “atmospheric aerosol samples” to avoid any implication of aircraft or platform-based in situ sampling.

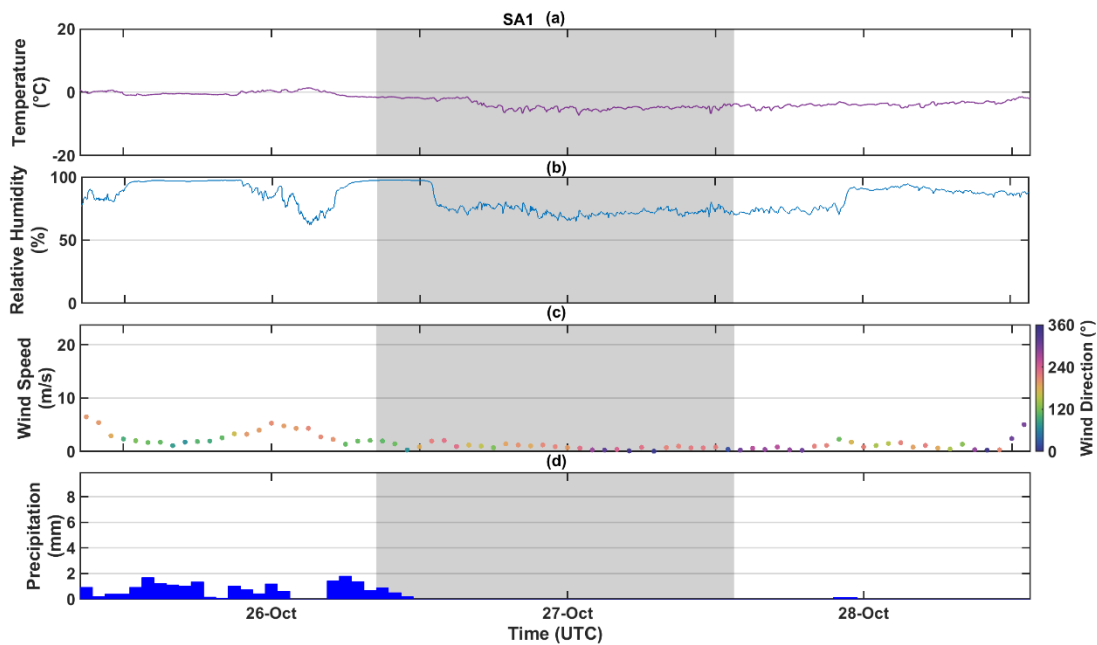
Figure 1 A cyclic colormap would be better suited for color-coding the wind direction.

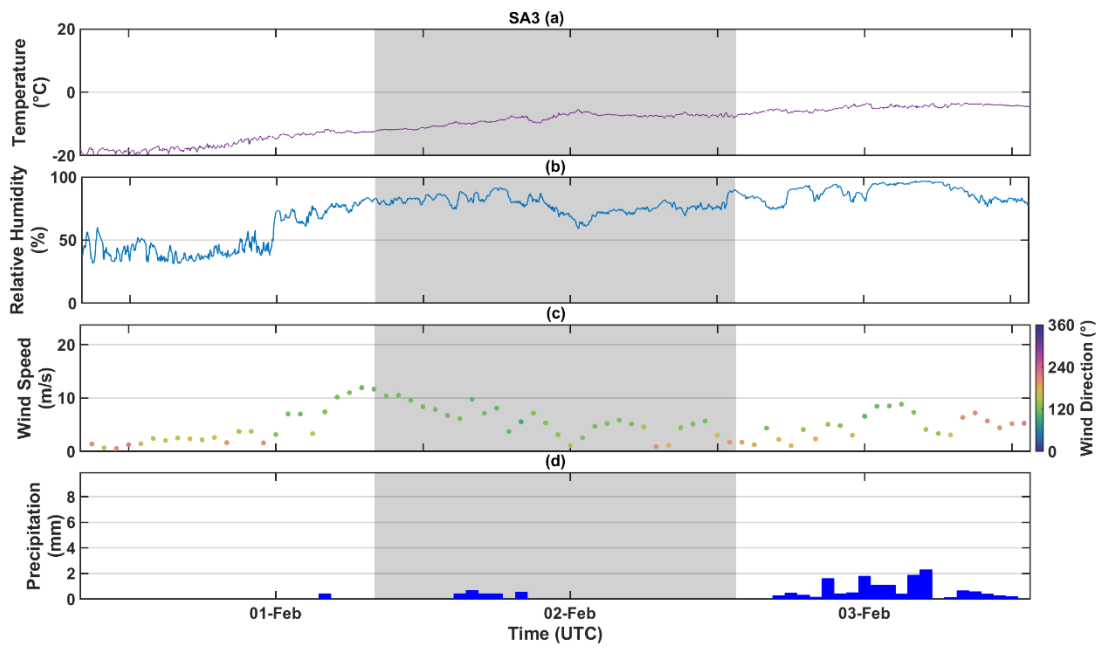
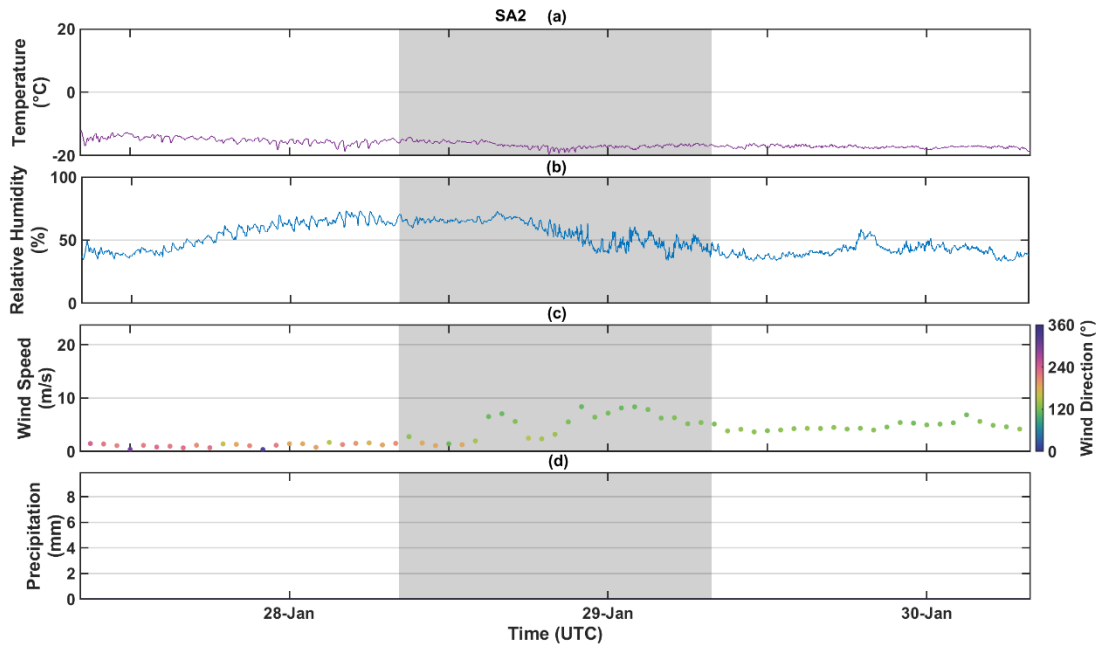
Response: To address reviewer’s comment, we revised the figure with cyclic colormap ‘twilight’.



Please also consider making as "zoomed in" version of this figure for the supplement, showing only the sampling periods

Response: To address the reviewer's comment, we added below zoomed in figures to supplementary.





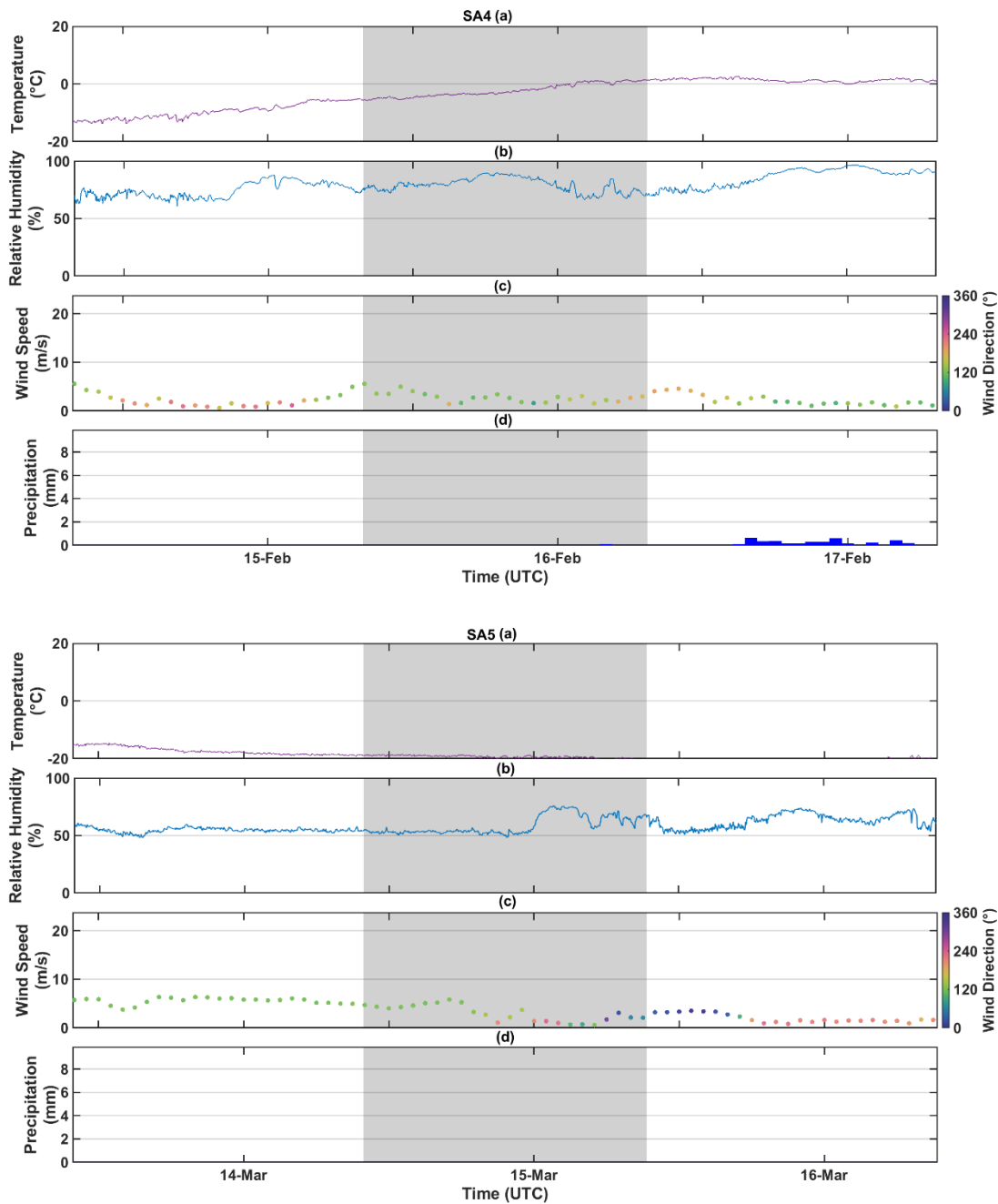


Figure 2: It would be great if the sea ice concentration or at least the sea ice edge is shown in these maps.

Response: The authors thank reviewer for this helpful suggestion. We agree that explicitly showing the sea-ice boundary provides important context for interpreting the potential influence of marine vs. cryospheric source regions during each sampling period. In the main manuscript, we kept Figure 2 focused on air-mass transport patterns (trajectory frequency) to avoid overcrowding the panels and to preserve readability across all five cases. Following your recommendation, we generated an additional figure showing the sea-ice edge for each sampling date and added it to the Supplementary Information (new figure). We also updated the Supplement text/caption to point

readers to this sea-ice-edge context when interpreting Figure 2 and the associated surface-type influence discussion.

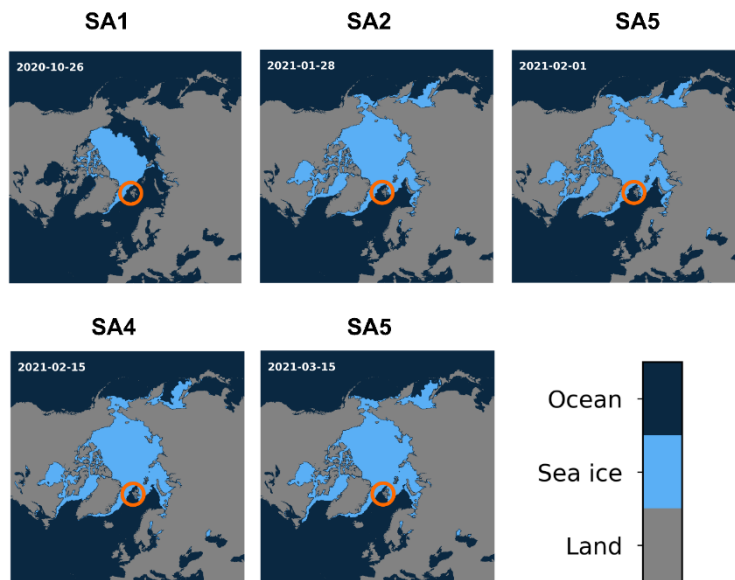


Figure S4: Sea-ice edge during sampling. Maps show sea-ice coverage on the first day of each sampling period (SA1–SA5). Light blue denotes sea ice, dark blue denotes open ocean, and gray denotes land. The red circle marks the sampling location at Gruvebadet Laboratory (GAL), Ny-Ålesund, Svalbard.

The below section is also added to supplementary-

S3. Sea-ice edge data

Sea-ice edge/coverage shown in Figure S4 was taken from the Copernicus Climate Data Store (CDS) “Sea ice edge and type daily gridded data from 1978 to present” product (DOI: 10.24381/cds.29c46d83; accessed January 2026). The dataset is derived from satellite passive-microwave brightness temperatures and provided at daily (24 h) temporal resolution on a 12.5 km grid. Each grid cell is classified into open water, open ice, or closed ice based on the sea-ice amount, and we used the field corresponding to the first sampling day for each sample to provide consistent sea-ice context across cases (Copernicus Climate Change Service (C3S), 2020).

L212 Where does the information about turbulence come from? If the statement is based on measurements, they should be also be part of the manuscript.

Response: We agree with the reviewer that the term “turbulence” implies direct measurements, which were not performed in this study. We have therefore removed the use of “turbulent” from the manuscript and revised the text to refer instead to meteorological conditions conducive to vertical mixing.

“Light precipitation (0.09 ± 0.20 mm) coincided with comparatively high wind speed, suggesting stronger boundary layer exchange than in other periods.”

L217 Can you elaborate on the possible enhanced atmospheric processing for this event? Is the cause for that primarily the farther transport (hence more time for the aging to take place), the presence of reactive species that do not exist at higher latitudes, or the availability of solar radiation?

Response: We thank the reviewer for this insightful comment. We have expanded the discussion to clarify that the enhanced atmospheric processing observed during this event likely results from multiple factors. Extended transport and longer atmospheric residence times provide greater opportunity for particle aging and internal mixing. In addition, aerosols transported over long distances may undergo cloud processing, which can promote aqueous-phase chemistry and enhance internal mixing. Contributions from reactive trace gases along the transport pathway are possible but were not directly measured, while photochemical processing driven by solar radiation is expected to be limited under Arctic conditions. The revised text reflects this multifactorial interpretation while clearly stating associated uncertainties.

“Although we cannot isolate the dominant ageing mechanism without concurrent gas-phase or radiation measurements, the warm and humid intrusion pathway implies greater cumulative exposure during transport (e.g., longer time for ageing and potential interaction with more reactive lower-latitude air masses and cloud processing along the pathway).”

Figure 3 and 4: Instead of a separate figure for CCSEM and STXM results, I think it would be better to have CCSEM and STXM sample wise side by side, eliminating the need to scroll through the document when a reader wants to assess the chemical composition of a specific sample.

Response: We thank the reviewer for this thoughtful suggestion aimed at improving figure readability. We agree that side-by-side presentation can be helpful for direct sample-wise comparison. However, CCSEM/EDX and STXM/NEXAFS provide fundamentally different types of information like size-resolved class distributions versus chemical mixing states at the single particle level and are based on different particle populations, spatial scales, and analytical assumptions. For clarity and to avoid potential misinterpretation, we therefore present these datasets in separate figures. To facilitate comparison between the two analyses, we have ensured consistent sample naming, ordering, and color schemes across Figures 3 and 4 and have clarified the cross-references in the text. This approach preserves the distinct strengths of each technique while allowing readers to assess compositional trends across samples without extensive restructuring of the figures.

Section 3.3 first paragraph: It is of course good to provide context and show what others have found, but I am missing the connection to your specific samples. Since you only have five samples from four distinct meteorological scenarios, you can directly compare your findings during, for example, the warm air intrusion with studies that also measured during a warm air intrusion.

Response: We appreciate this suggestion. The opening paragraph of Sect. 3.3 is intentionally written as a brief literature-context introduction to Fig. 5, rather than a sample-by-sample

comparison. The sample-specific linkage is provided immediately thereafter: the subsequent paragraphs interpret each sample (SA1-SA5) within the four meteorological scenarios defined in Sect. 3.1 (including the warm-air intrusion period) and discuss how those conditions relate to the observed temperature dependence and heat sensitivity. Because this connection is already addressed in the body of Sect. 3.3, we did not restructure the first paragraph.

L291 This statement should be removed. The 0.2 INP/L is simply the upper detection limit of the achievable with the used droplet freezing array. During the same expedition, also a CFDC (operated in immersion mode) was deployed and at -28°C, a max. INP concentrations of around 100 L⁻¹ were measured, which can be seen in the cited overview publication by Welti et al. (2020), but also in more detail in the expedition related publication by Hartmann et al. (2021) (which you also already cite). If you look into the latter study more carefully, you will also notice, that the name of the campaign was PASCAL, Polarstern is the name of the research vessel and PS106 is the the consecutive expedition identifier assigned by the operator of the vessel. Hence you might want to change "[...] from the Arctic expedition (Polarstern) - PS 106 [...]" accordingly so that a reader can more easily find information on that expedition online.

Response: We thank the reviewer for this clarification. We agree that the reported value of 0.2 INP L⁻¹ represents the upper detection limit of the droplet freezing assay used and should not be interpreted as a physical upper bound on INP concentrations. Accordingly, the statement at Line 291 has been removed.

We also appreciate the clarification regarding the INP measurements obtained during the same expedition using a CFDC operated in immersion mode, which reported substantially higher INP concentrations at lower temperatures, as documented in Welti et al. (2020) and Hartmann et al. (2021).

In addition, we have corrected the campaign nomenclature to accurately reflect that **PASCAL** was the campaign name, **Polarstern** refers to the research vessel, and **PS106** is the expedition identifier. The revised wording has been updated to ensure clarity –

“Similar to Creamean et al. (2022), the offline freezing assay reported by Welti et al. (2020) during the PASCAL campaign, conducted aboard the research vessel Polarstern (expedition PS106) in the vicinity of Svalbard, Norway (May-July 2017), measured INP concentrations at -28 °C that were limited by the detection threshold of the droplet freezing assay.”

L295 - L297 I suggest removing all sentences referring to the dynamic filter processing chamber measurements in the Rinaldi study. You measured immersion freezing, not condensation freezing; therefore, these results are not relevant here. Also, the discrepancy between the dynamic filter processing chamber and the droplet freezing assay is a separate topic, and mentioning it here only distracts from your work.

Response: We thank the reviewer for pointing out that. We agree that dynamic filter processing chamber/condensation-freezing results are not directly comparable to our immersion-freezing

droplet assay and that discussing the DFPC droplet assay discrepancy distracts from our study. We have therefore removed all sentences describing the DFPC measurements and the associated method comparison discussion, and we retained only the site-relevant Ny-Ålesund context based on offline droplet-freezing measurements.

L298 To which cold air outbreak are you referring to? Was it during the Rinaldi study or yours? From the phrasing, this is unclear.

Response: Thank you for noting this ambiguity. We revised the wording to make explicit that ACAO refers to the externally published Arctic Cold Air Outbreak campaign used as a reference dataset in the literature (e.g., summarized by Raif et al., 2024), rather than to the Rinaldi sampling period or to our campaign.

“Rinaldi et al. (2021) reported offline INP concentrations from samples collected at a ground-based site near Ny-Ålesund between April and August 2018, providing site-relevant context for comparison. Their highest INP concentrations were lower than those reported for the Arctic Cold Air Outbreak (ACAO) campaign and were typically 1-3 orders of magnitude below those measured in this study (Raif et al., 2024). Similarly, Li et al. (2023) reported Ny-Ålesund INP concentrations during October-November 2019 using offline droplet-freezing measurements on filter samples; these values were generally 1-4 orders of magnitude lower than those reported for the ACAO campaign (Li et al., 2023; Raif et al., 2024)”

L301 It is not clear what the difference between the preparation techniques is. If you wash off the particle, you also end up with a particle suspension.

Response: We agree with the reviewer that washing particles from a substrate also results in a particle suspension and that the distinction between preparation techniques was unclear. To avoid confusion, the sentences referring to differences between preparation techniques at Line 301 have been removed.

***L307 How do you come to the conclusion that the SML contributed? This has to be explained.

Response: We thank the reviewer for requesting clarification. Specifically, the presence of Na-rich particles internally mixed with substantial organic fractions and organic coatings are often influenced by organic enrichment from SML, as documented in previous studies. In addition, the prevailing marine air-mass influence during the sampling period suggests a potential marine source for these particles. We have revised the text to explicitly state that the possible SML contribution is inferred based on particle composition and mixing state (mixing with organics), and we now clarify the reasoning underlying this interpretation.

“Despite small inter-sample deviation in n_{INP} at the examined freezing temperatures, the samples exhibit distinctly different heat sensitivities. SA2, SA3, SA4, and SA5 contained heat-labile INPs active above -15 °C, which are consistent with marine organic INPs previously associated with sea surface microlayer (SML) influence, suggesting a possible contribution from SML-derived material.”

Figure S1: While potential readers are likely aware of the location of Svalbard, they may not know where Gruvebadet is located within the archipelago. Therefore, I suggest marking the study site on a map of Svalbard instead of the current map (or adding it to a separate panel, for example).

Response: We thank the reviewer for this suggestion. Figure S1 has been revised to include a map of the Svalbard archipelago with the Gruvebadet Observatory explicitly marked, providing clearer spatial context for the study site.

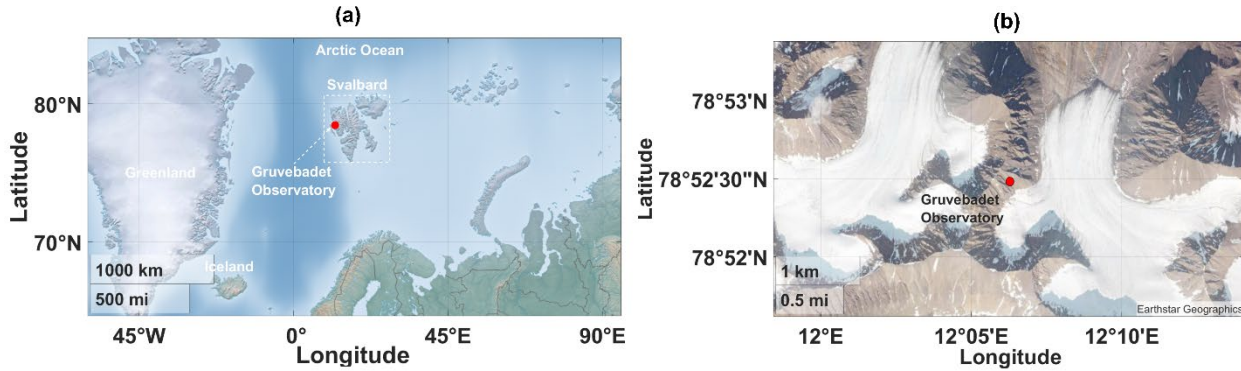


Table S3: How do you define open ice?

Response: We thank the reviewer for requesting clarification. In this study, “open ice” refers to sea ice surfaces that are exposed to the atmosphere and not covered by snow, melt ponds, or open water. We have added an explicit definition of “open ice” to Table S3 to avoid ambiguity.

“Open ice refers to exposed sea ice surfaces without snow cover or melt ponds, distinct from open water and snow-covered ice.”

*** Heat sensitivity

1. Somewhere, the calculation of the percentage of heat-labile INPs should be explained.

Response: We added below equations in supplementary-

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

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

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

2. I doubt that a percentage accurately captures the logarithmic nature of INPs.

Response: The reviewer is right. Our $n_{INP}(T)$ is an estimated product of log-scaled frozen fraction values; thereby, our $\%INP_{Heat\ Labile}(T)$ is approximated, too. In the future, comparison of heated INP via direct online INP measurement and offline result will be necessary to assess the accuracy of approximated heat labile fraction. The authors believe our analysis provides an

idea of the relative abundance of heated labile vs. stable INPs. For the current manuscript, in this context, we add this important note to supplementary section S5.

“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.”

3. Your calculation method is skewing the results since it is based on an unsound assumption. You assume that the concentration of heat-stable INPs is zero when it is simply below the detection limit of your setup. It is not only very unlikely that the concentration of heat-stable INPs goes immediately to zero when below the detection limit, but it is also not good scientific practice to make this assumption without mentioning it in the manuscript.

Unfortunately, there is no established, scientifically sound way to report the heat-labile fraction. One uncritical approach would be to report only values where the INP spectra of the heated and unheated samples overlap (see Gong et al., 2022). The disadvantage is that, especially at higher temperatures, it is not possible to report any values.

I suggest calculating the heat-labile fraction in a conservative, robust way by replacing values below the detection limit with the lowest detectable value. This method provides a conservative estimate of the heat-labile fraction that represents the case with the least amount of heat-labile INPs. Along with your existing values, which represent the maximum amount of heat-labile INPs, you can report a range for the heat-labile fraction of a sample. In my view, this is a better representation of the sample since it is impossible to determine the true value.

Response: The authors agree, and the figure and its caption have been revised accordingly.

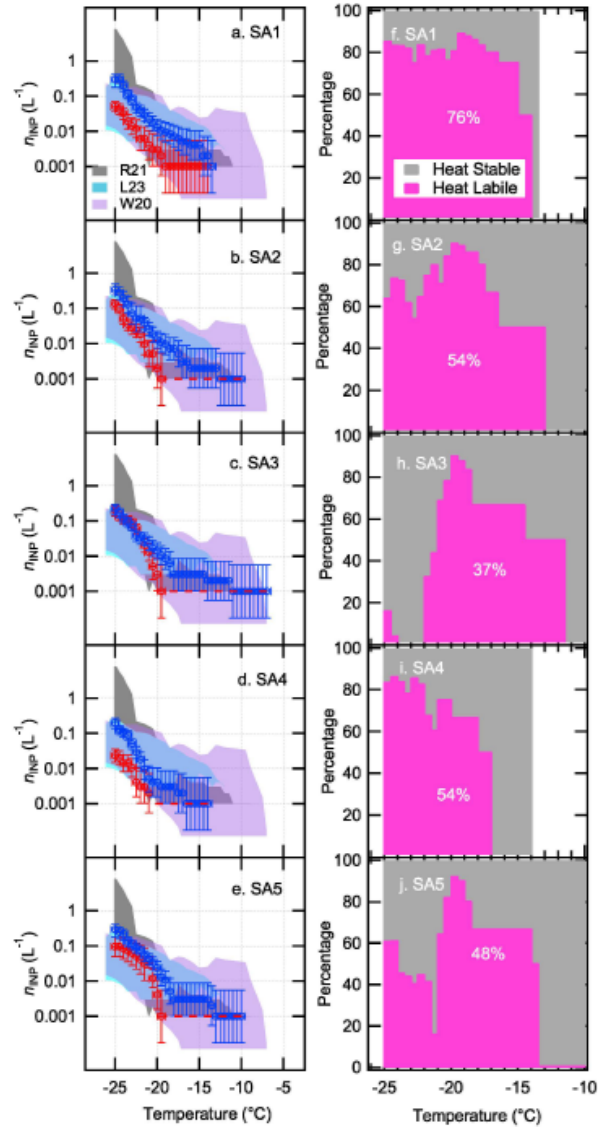


Figure 5. Immersion freezing of the particle samples collected at different time periods. In Panels (a)-(e), the blue circles indicate the ambient number concentration of INPs before heat treatment, and the red circle indicates the INP number after heat treatment. Red dashed lines show the lowest detection limit of n_{INP} for this study. Color-shaded areas show the previous results of INP measurements from GVB via the same freezing assay (Rinaldi et al., 2021; Li et al., 2023), as well as from the PS 106 Arctic expedition in the vicinity of Svalbard, Norway (May – July 2017; Welti et al., 2020). Panels (f)-(j) show the amount of heat-labile and -stable INPs (%) for SA1-SA5. We use the lowest detectable n_{INP} (i.e., 0.001 L^{-1}) for $n_{\text{INP,heated}}(T)$ in the case there were no measured values for freezing temperatures, in which $n_{\text{INP,untreated}}(T)$ was measured.

L307 What are you basing the statement that SA2 and SA4 exhibit high INA on? At -25°C, these two samples appear to have higher N_{INP} . However, at higher temperatures (>-15°C), this does not seem to be the case. Therefore, labeling them as high INA seems inaccurate.

Response: We thank the reviewer for this careful observation. We agree that the original wording was too general. As shown in Fig. X, SA2 and SA4 do not exhibit elevated ice nucleation activity across the full temperature range. Instead, these samples show relatively higher N_{INP} only at lower temperatures (around -25 °C) and are distinguished primarily by their high fraction of heat-labile INPs. We have revised the text accordingly to reflect the temperature dependence of the observed enhancement and removed the general characterization of these samples as exhibiting “high INA.”

“SA2 and SA4 show enhanced n_{INP} at lower temperatures (≈ -25 °C) and are characterized by a high fraction of heat-labile INPs, while their number concentration at warmer temperatures (> -15 °C) is comparable to the other samples.”

***L309 - L311 The phrasing in this sentence gives the impression that the cold conditions are one of the indicators pointing towards INPs that are internal mixtures of organic/biological and Na-rich particles. If that is what you meant, then this needs explanation, as I do not understand how cold conditions alone are an indicator for this. If this is not what you meant, then the sentence needs rephrasing.

Response: We thank the reviewer for pointing out this ambiguity. We agree that cold conditions alone are not diagnostic of particle mixing state. We have revised the sentence to clarify that temperature provides environmental context, while the inference of internally mixed organic/biological and Na-rich INPs is based on particle composition and heat sensitivity.

“For SA2, under cold conditions (-16.7 °C, RH \approx 57%), the combination of a high OC+IN fraction (73.6%, STXM/NEXAFS), a high Na-rich particle fraction (\sim 69%, CCSEM/EDX), and a strong heat-labile response is consistent with INPs associated with organic/biological material internally mixed with Na-rich particles.”

L315 - L316 It is certainly possible that the SML contributes biological INPs, but can you exclude other sources that are often discussed in this context, like low-INA dust acting as carrier for high-INA biological INPs? If you can, then this is also worth mentioning in the text.

Response: We thank the reviewer for raising this important point regarding alternative sources of biological INPs. Based on the CCSEM/EDX analysis (Fig. 3), mineral dust particles constitute only a minor fraction of the particle population in SA2 and SA4. As a result, dust acting as a major carrier for high-INA biological material is unlikely for these samples. While minor contributions from other sources cannot be fully excluded, the observed dominance of Na-rich particles internally mixed with organic material, together with strong heat-labile responses, supports a primarily marine-associated source. We have revised the text to clarify this point.

“Contributions from mineral dust acting as a carrier for biological INPs are unlikely to dominate, as CCSEM/EDX analysis shows only a minor dust fraction in SA4 (Fig. 3).”

L324 Do you expect the biogenic part to be from local sources or from long-range transport as well?

Response: We thank the reviewer for this question. We do not have direct measures that allow us to distinguish between local and long-range sources of the biogenic component. Therefore, we cannot attribute the observed biogenic material exclusively to either source. Both local marine and terrestrial biogenic sources as well as long-range transport are likely to contribute.

L332 Please rephrase this statement. Polysaccharidic INP certainly can be enriched in the SML, but are rather heat-resistant, so the heat treatment does not indicate their presence (see Hartmann et al., 2025).

Response: We thank the reviewer for this important clarification. We agree that polysaccharidic INPs are generally heat-resistant and that heat treatment does not indicate their presence. We have revised the text to remove the implication that the observed heat-labile response reflects polysaccharidic INPs.

“Meanwhile, SA1, characterized by marine influence and more humid conditions, shows moderate INA at -15 °C that significantly diminishes upon heating, indicating the presence of heat-labile biological INPs. Such INPs are consistent with proteinaceous or other labile biological material that can be enriched in the sea surface microlayer (SML), rather than heat-resistant polysaccharidic INPs.”

L343 Tab. S1 contains the precipitation at the measurement site. However, for your statement, the precipitation along the air mass trajectory is more important. Since precipitation is one of the HYSPLIT output variables, I recommend reporting the average accumulated precipitation along the air mass trajectory for each sample in the supplement to emphasize your point.

Response: Thank you for the suggestion. We agree that precipitation along the transport pathway is more relevant than precipitation only at the measurement site for assessing wet removal before arrival. Following your recommendation, we extracted the HYSPLIT precipitation output for each backward trajectory and added the accumulated precipitation along the trajectories for each sample to the Supplementary Information (new figure/table). We kept Table S1 as the site precipitation record, and now the supplement includes the trajectory precipitation to better support the statement in the text.

Below revised text is added to main text to respond to the reviewer’s comment-

“Precipitation influence on INP abundance and composition during the sampling periods is likely minimal, as site precipitation was dry or very low (≤ 0.1 mm; Table S1) and trajectory-based accumulated precipitation was negligible (Figure. S11), indicating limited wet scavenging prior to arrival.”

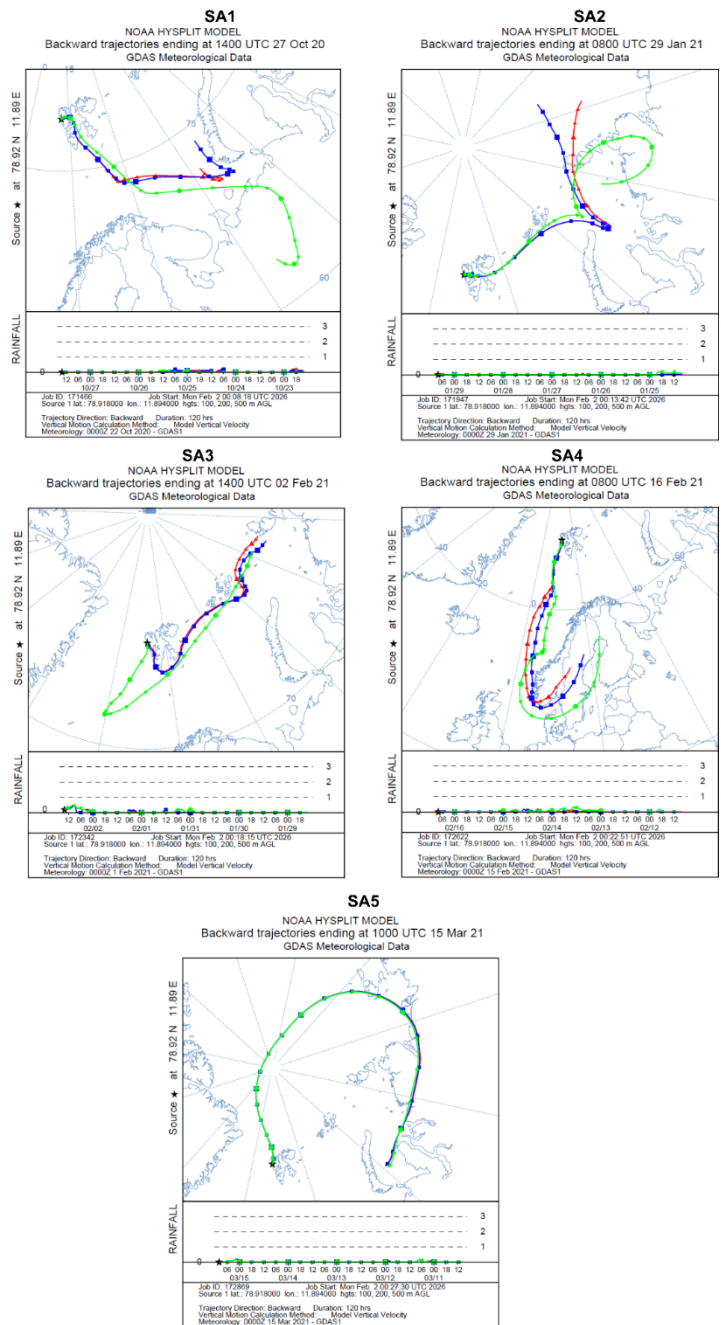


Figure S11. Backward air-mass trajectories and precipitation along transport pathways. NOAA HYSPLIT 120 h backward trajectories for samples SA1–SA5 (ending at the sampling site; star). Colored lines indicate trajectories initialized at 100, 200, and 500 m AGL. The lower panel in each plot shows the accumulated precipitation (rainfall) along each trajectory as reported by HYSPLIT, used to assess potential wet removal prior to arrival at the measurement site.

L351 At least a reference should be added to explain how the OVF is determined. Even better would be a few sentences describing it.

Response: We have revised the Methods section (around L160) to include the appropriate references and a brief description of how OVF is derived from STXM/NEXAFS data. In the revised text, we now summarize the OVF calculation approach and define how the organic and inorganic contributions are quantified following published methods (e.g., Fraund et al., 2019).

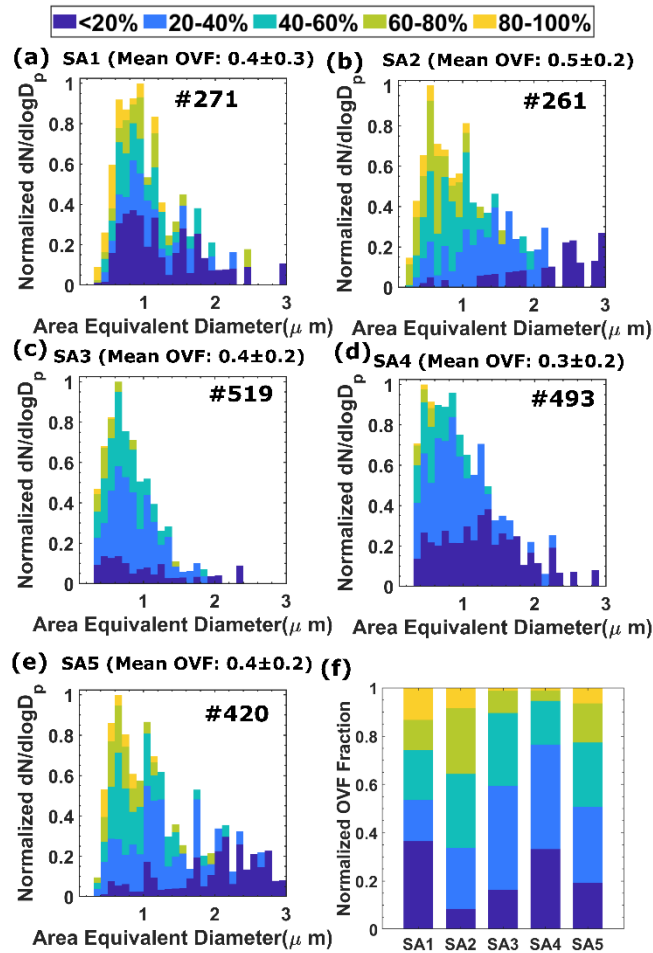
“Organic volume fraction (OVF) was determined from STXM/NEXAFS carbon K-edge measurements following the approach of Fraund et al. (2019). Briefly, transmitted-intensity images were converted to optical density and used to separate organic and inorganic contributions within each particle based on the pre-edge and post-edge absorption. OVF was then calculated taking the ratio of organic thickness contribution to the sum of organic and inorganic thickness contribution derived from STXM maps (Fraund et al., 2019, 2020; Knopf et al., 2021; Lata et al., 2021).”

L371 - L372 Are these "overall OVF values" the mean of all the individual OVF values of all the particles in a sample? If yes, would not the median be a more appropriate measure? I would also recommend adding these numbers to Fig. 7.

Response: The authors thank the reviewer for this helpful comment. The authors clarify that the “overall OVF” values refer to the mean of the particle-resolved OVF values for each sample. Following the reviewer’s suggestion, the authors now also report the median OVF together with the interquartile range (Q1-Q3) and have added these overall OVF values to Fig. 7 to make the comparison across samples clearer.

Offline chemical analysis	Type of analysis	Classes	SA1	SA2	SA3	SA4	SA5
CCSEM/EDX	Particle classes (# percentage)	Biogenic	3.9 ± 0.3	0.9 ± 0.1	0.0 ± 0.0	6.1 ± 0.6	12.8 ± 1.0
		Carbonaceous	11.2 ± 0.5	4.6 ± 0.3	18.7 ± 1.3	19.7 ± 1.0	13.3 ± 1.0
		Sulfate	5.3 ± 0.4	5.6 ± 0.3	32.1 ± 1.5	18.4 ± 0.9	1.7 ± 0.4
		Dust	0.8 ± 0.1	3.0 ± 0.3	1.2 ± 0.4	1.0 ± 0.2	1.7 ± 0.4
		Carbonaceous+Dust	0.2 ± 0.1	0.1 ± 0.0	0.3 ± 0.2	0.1 ± 0.1	0.0 ± 0.0
		Sulfate+Dust	0.1 ± 0.1	0.4 ± 0.1	0.4 ± 0.2	0.1 ± 0.1	0.0 ± 0.0
		Na-rich	70.3 ± 0.7	69.0 ± 0.7	22.9 ± 1.4	39.8 ± 1.2	66.2 ± 1.4
		Na-rich Sulfate	4.5 ± 0.3	8.3 ± 0.4	20.2 ± 1.3	5.2 ± 0.5	1.2 ± 0.3
		Other	3.8 ± 0.3	8.1 ± 0.4	4.2 ± 0.6	9.6 ± 0.7	3.1 ± 0.5
STXM/NEXAFS	Mixing state classes (# percentage)	In	2.2 ± 0.9	0.4 ± 0.4	3.3 ± 0.8	3.9 ± 0.9	0.0 ± 0.0
		OC	12.9 ± 2.0	16.1 ± 2.3	22.5 ± 1.8	12.0 ± 1.5	5.7 ± 1.1
		OCEC	4.1 ± 1.2	2.7 ± 1.0	4.8 ± 0.9	1.0 ± 0.5	0.7 ± 0.4
		OCIn	52.4 ± 3.0	73.6 ± 2.7	44.7 ± 2.2	77.1 ± 1.9	85.0 ± 1.7
		OCInEC	28.4 ± 2.7	7.3 ± 1.6	24.7 ± 1.9	6.0 ± 1.1	8.6 ± 1.4
	OVF classes (# percentage)	<20%	36.5 ± 2.9	8.4 ± 1.7	16.4 ± 1.6	33.2 ± 2.1	19.3 ± 1.9
		20-40%	17.0 ± 2.3	25.3 ± 2.7	43.0 ± 2.2	43.3 ± 2.2	31.4 ± 2.3
		40-60%	20.7 ± 2.5	30.7 ± 2.9	30.3 ± 2.0	18.1 ± 1.7	26.7 ± 2.2
		60-80%	12.5 ± 2.0	27.2 ± 2.8	9.2 ± 1.3	4.3 ± 0.9	16.2 ± 1.8
	80-100%	13.3 ± 2.1	8.4 ± 1.7	1.2 ± 0.5	1.0 ± 0.5	6.4 ± 1.2	

Overall OVF (mean)	0.4 ± 0.3	0.5 ± 0.2	0.4 ± 0.2	0.3 ± 0.2	0.4 ± 0.2
Overall OVF (median [Q1–Q3])	0.4 [0.1–0.6]	0.5 [0.3–0.7]	0.4 [0.3–0.5]	0.3 [0.2–0.4]	0.4 [0.2–0.6]



L372 - L373 In general I would recommend the use of the Spearman rank coefficient instead of the Pearson correlation coefficient, because it does not assume linearity and is not easily affected by outliers. Regardless of Pearson or Spearman, five data points are of course not a very solid basis to determine correlation. For that reason I would recommend to remove figure 8 from the main manuscript, because it highlights too much results with weak statistical basis. It can still be shown in the supplement and the values can be reported in the text, but the reader should be reminded of the small sample size and hence assess these values carefully. Also a scatter plot matrix could be shown in the supplement, as this allows the reader to assess correlation themselves.

Response: The authors thank the reviewer for these helpful statistical suggestions. The authors replaced the Pearson correlation analysis with Spearman rank correlation throughout, since it does not assume linearity and is less sensitive to outliers. The authors also revised the text to clearly

note that the correlation analysis is based on only five samples ($n = 5$) and should therefore be interpreted cautiously as an exploratory summary rather than a robust statistical relationship.

“This gradation in organic volume fraction (OVF) is reflected in the Spearman rank correlations between OVF and INP concentrations (Figure 8), which show moderate to strong positive relationships ($\rho = 0.67$ at -14 °C and $\rho = 0.89$ at -24 °C). These correlations indicate that samples enriched in organic material—particularly those dominated by higher OVF bins (60–100%), tend to exhibit higher INP concentrations under both moderately supercooled (-14 °C) and colder (-24 °C) conditions. This behavior is consistent with previous studies demonstrating the importance of organic-rich particles and organic coatings for immersion freezing, either through intrinsic biological ice-nucleating material or through modification of particle surface properties (Augustin-Bauditz et al., 2014; Ickes et al., 2020; Kanji et al., 2019; Wilson et al., 2015).

In addition to OVF, several particle classes derived from CCSEM/EDX exhibited temperature-dependent associations with INP concentrations. Biogenic and Na-rich particles showed weak to moderate positive correlations ($\rho = 0.30$ and 0.50 at -14 °C; $\rho = 0.00$ and 0.60 at -24 °C), suggesting a potential contribution from marine-influenced organic material, particularly at colder temperatures. In contrast, carbonaceous and sulfate particles displayed moderate to strong negative correlations with INP concentrations at both temperatures, indicating that these particle types are less efficient INPs in this dataset.

Dust exhibited positive correlations with n_{INP} at both temperatures ($\rho = 0.60$ at -14 °C and $\rho = 0.70$ at -24 °C), despite representing a minor fraction of the particle population based on CCSEM/EDX analysis. Given the small sample size ($n = 5$) and low dust abundance, this relationship should be interpreted cautiously and is considered exploratory.”

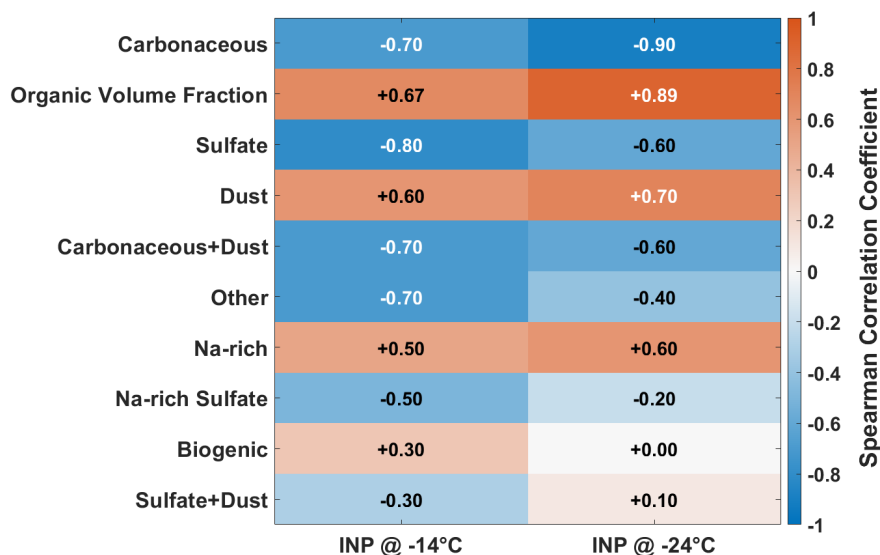


Figure 8. Spearman rank correlation coefficients between ice-nucleating particle (INP) concentrations and particle composition metrics (number fraction in each sample) at two freezing temperatures, -14 °C (left) and -24 °C (right), prior to heating. Each row represents a particle class

or metric, including biogenic, carbonaceous, sulfate, dust, mixed types (e.g., carbonaceous + dust), Na-rich categories, and organic volume fraction (OVF). Orange/red shading indicates positive correlations and blue shading indicates negative correlations, with lighter colors indicating values closer to zero.

L373 While I am not aware of fixed rules for the descriptive words used for the strength of a correlation, I never seen anyone to refer to an R of 0.5 as strong. Generally, this would be considered a moderate positive correlation.

Response: The authors thank the reviewer for this clarification. The authors agree that describing an R (or ρ) value of ~ 0.5 as “strong” is not appropriate. In the revised manuscript, the authors have replaced “strong” with “moderate” (or “moderate positive/negative correlation,” as appropriate) and adjusted the wording throughout to ensure the descriptive terms match the reported correlation magnitudes.

L394 I do not see how "promoting heterogeneous freezing via organic-induced deliquescence or restructuring" can be directly inferred from your measurements. You can of course mention this as a possible mechanism that could take place, but then the respective literature has to be cited and sentences that explain the mechanism need to be added.

Response: The authors thank the reviewer for this important point. The authors agree that “promoting heterogeneous freezing via organic-induced deliquescence or restructuring” cannot be concluded directly from the measurements presented here. In the revised manuscript, the authors have rephrased this statement as a possible mechanism and added brief explanatory text supported by literature showing that organic coatings can (i) modify water uptake/phase state (including deliquescence or glass-to-liquid transitions) and thereby change access to an underlying inorganic/ice-active surface, and (ii) undergo morphology changes (e.g., phase separation or porosity/restructuring) that can alter ice-nucleation pathways. Relevant references have been added accordingly.

“While our measurements do not directly resolve the freezing pathway, organic coatings can plausibly influence heterogeneous freezing by modifying particle water uptake and phase state. For example, transitions between liquid and highly viscous/glassy organic phases can limit water diffusion and alter when and how an underlying ice-active surface becomes accessible, which can change the apparent freezing efficiency (Berkemeier et al., 2014; Schill and Tolbert, 2013; Zobrist et al., 2008). In addition, organic coatings and internal mixing can modify ice nucleation on mineral/inorganic particles by changing surface properties or the interaction between deliquescence and ice formation under cold, humid conditions (Möhler et al., 2008; Schill and Tolbert, 2013).”

L402 I have not seen where you show and discuss potential influence of the morphology. You should either add this or remove "morphology" from this sentence.

Response: The authors thank the reviewer for pointing this out. The authors agree that the term “morphology” can imply particle shape/roughness/porosity, which is not explicitly evaluated in this study. In this sentence, the authors intended to refer to the spatial distribution of the organic

coating/mixing geometry inferred from STXM/NEXAFS mapping (mixing-state classes and OVF). To avoid ambiguity, the authors have revised the wording by replacing “morphology” with “coating distribution (mixing geometry)” and added a brief clarification in the text.

L402-403 Can this statement be supported quantitatively? Could you derive a mean organic layer thickness for the particles in a sample? Is there an existing metric for "patchiness" that could be applied to your measurements?

Response: The authors thank the reviewer for the comment. The authors clarify that the statement at L402–L403 was intended in a qualitative sense to describe the apparent coating distribution/mixing geometry inferred from the STXM/NEXAFS maps, not to imply a quantified coating thickness or a formal patchiness metric. At present, the authors did not derive a mean organic layer thickness or apply an established patchiness metric, and the available 2-D STXM/NEXAFS information does not uniquely constrain coating thickness without additional assumptions and analysis. To avoid ambiguity, the authors revised the text by replacing “morphology” with “coating distribution (mixing geometry)” and limited the statement to what is directly supported by the measured mixing-state classes and OVF distributions.

L413 – 421 In the first sentence you basically only state that there was a case of many Na-rich particles with a thin organic coating and that they had only moderate INA. I do not understand why this finding underscores the critical role of biological INPs, nor is the connection to mild, humid conditions clear. Then, you discuss the particles from cold and dry cases, which I assume are SA2 and SA5. You state that these particles have enhanced INA. However, this is not entirely consistent with what you wrote in the Results section. In L282-283, you wrote that SA1 and SA5 are less ice-active than SA2, SA3, and SA4.

Response: The authors thank the reviewer for this important comment. The authors agree that the original wording in L413–421 did not clearly explain (i) why the Na-rich, thin-coating case relates to heat-sensitive biological INPs and (ii) how meteorological conditions were being used in the interpretation. In the revised manuscript, the authors have rewritten L413–421 to clarify that Na-rich composition is presented as context for marine influence, but does not by itself demonstrate a biological contribution. Instead, the conclusion now links biological (heat-sensitive) contributions to the temperature-dependent decrease in n_{INP} after heating, which is the direct evidence in our dataset. The revised text also avoids implying that cold and dry cases are uniformly “enhanced” and consistently refers to n_{INP} rather than INA.

“Aerosols enriched in Na-rich particle types with relatively thin organic coatings showed moderate n_{INP} prior to heat treatment. However, the most direct indicator of biological (heat-sensitive) contributions is the temperature-dependent reduction in n_{INP} after heating, rather than Na-rich composition alone. Across the five samples, the heat-labile fraction varies substantially, while heat-stable INPs dominate at colder temperatures. Overall, these results indicate that Arctic n_{INP} reflects the combined influence of particle composition, coating distribution/mixing state, and air-mass history, with the relative importance of heat-labile versus heat-stable INPs depending on temperature regime.”

Fig S3 The air mass sampled on SA5 seems to have experienced the highest BC concentrations of all samples. BC emissions are also typically accompanied by the emission of sulphate species, but SA5 hardly contains sulphate in the CCSEM/EDX measurements. Do you have an explanation for this?

Response: The authors thank the reviewer for this careful observation. The authors agree that the combination of elevated BC during SA5 (Fig. S3) and low sulfate fraction in the CCSEM/EDX particle classes appears counterintuitive at first glance. However, several processes can explain this without requiring a one-to-one correspondence between the BC proxy and the CCSEM/EDX sulfate class:

1. **BC-rich air does not always imply sulfate-rich particles at the receptor.** During SA5, BC enhancements can plausibly reflect contributions from combustion sources with comparatively weak SO₂/sulfate co-emission (e.g., local shipping/diesel activity near Svalbard or other near-surface combustion influences), whereas sulfate-dominated aerosol is more strongly tied to sustained SO₂ oxidation and regional aging.
2. **Differential removal during transport can decouple BC and sulfate.** Sulfate-containing particles are typically more hygroscopic and can be more efficiently removed by cloud processing and precipitation along transport. This can reduce the sulfate fraction at the site even when the air mass history indicates combustion influence (consistent with our trajectory-precipitation context provided in the supplement).

Summary & Conclusion: With only five samples it might be a viable idea to have summary table, which contains for each sample the central results from your analysis like: meteo. conditions, dominant particles classes (EDX), mixing state, OVF, organic coating thickness/patchiness, N_INP at a high and low temperature, N_INP of heat stable INPs also at a high and low temperature.

Response: The authors thank the reviewer for this helpful suggestion. Given the limited sample set (SA1-SA5), the authors agree that a compact overview is valuable. However, the key information the reviewer lists is already compiled in the Supplementary Information across the existing summary tables (meteorological context, dominant CCSEM/EDX classes, STXM/NEXAFS mixing state and OVF metrics, and n_{INP} and heat-stable n_{INP} at representative warm and cold temperatures). To avoid duplicating the same content in an additional table, the authors did not create a new summary table.

Technical comments

L93 missing spaces: "[...] GVB(Mazzola et al., 2016) [...]" and "[...] (Pluvio2 L 400 RH)(Ebell et al., 2025) [...]"

Response: Addressed.

L93 "OTT Pluvio (Pluvio2 L 400 RH)": It is not clear what is the company name and what is the instruments name. Please follow common conventions of citing instruments. E.g., OTT Pluvio² L (OTT HydroMet GmbH, Kempten, Germany). After that, please refer to the instrument by its

correct name or introduce an abbreviation properly: "OTT Pluvio² L (OTT HydroMet GmbH, Kempten, Germany, hereafter referred to as Pluvio2, ...)"

Response: Addressed.

L94 Typo "Ny-Aalesund" should be "Ny-Ålesund"

Response: Addressed.

L94 missing article "OTT Pluvio2L weighing gauge [...]" should be "The OTT Pluvio2L weighing gauge [...]"

Response: Addressed.

L96 remove "So,"

Response: Addressed.

L99 Plural: "[...] 5 pairs of filter samples was collected [...]" should be "[...] 5 pairs of filter samples were collected [...]"

Response: Addressed.

Table 1 The headers are aligned differently. One is centered and the other is left-aligned. Please use consistent formatting. Additionally, I recommend that the authors take a look at the "APA Style manuals" and consider revising their tables. These manuals provide good guidelines for creating appealing, readable scientific tables.

Response: Addressed.

L135 "figures out" sounds colloquial. Suggestion: "determines"

Response: Addressed.

L138 missing space "[...] behavior(Lata et al., 2021, 2023)." should be "[...] behavior (Lata et al., 2021, 2023)."

Response: Addressed.

L246 Write "back trajectory" consistently with or without hyphen throughout the document.

Response: Addressed.

L286 too many parentheses "[...] range (e.g., (Hill et al. [...])", should be "[...] range (e.g., Hill et al. [...])"

Response: Addressed.

L287 missing space "[...] $N_{\text{INPmeasured}}$ [...]" should be "[...] N_{INP} measured [...]"

Response: Addressed.

L295 citation in parentheses is unnecessary since the study is already cited within the sentence

Response: Addressed.

L309 Plural. Suggestion: "Though SA2 and SA4 contain 16.1 and 12% OC-rich particles, respectively."

Response: Addressed.

L331 There seems to be a space at the beginning of this line.

Response: Addressed.

Use consistent panel identifier throughout the manuscript (Fig. 1, 2, 3 used "(a), (b), etc."; Fig. 4 used "a), b), etc."; Fig. 5 used "a., b., etc.")

Response: Addressed.

Use consistent parameter abbreviations: Fig. 5 denotes the INP concentration with a lower case "n" (n_{INP}), while the text uses the upper-case letter (N_{INP})

Response: Addressed.

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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, $C_{INP}(T)(L^{-1})$ was calculated from the droplet-freezing assay as-

$$C_{INP}(T) = -\frac{\ln(f_{unfrozen}(T))}{v_d} \quad [1]$$

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 ($V_d = 3 \mu\text{L}$).

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

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

where DF is the serial dilution factor (e.g., 1, 10, 100, ...), V_{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.

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

$$\% \text{INP}_{\text{Heat Stable}}(T) = 100 - \% \text{INP}_{\text{Heat Labile}}(T) \quad [4]$$

where $n_{\text{INP,untreated}}(T)$ is the number concentration of INPs before heat treatment and $n_{\text{INP,heated}}(T)$ 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 $n_{\text{INP}}(T)$ spectra (Eqs. 3-4) and should therefore be interpreted as an approximate and comparative metric. In particular, when $n_{\text{INP,heated}}(T)$ 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 physicochemical 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 Gruebadet 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 supermicron 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 $n_{\text{INP}}(T)$ 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 $n_{\text{INP}}(T)$ shape. The authors also revised the discussion in L307 and L321-326 to consistently describe sample-to-sample differences in $n_{\text{INP}}(T)$. To provide a quantitative comparison with uncertainties, the authors now reference the updated Fig. 5, which shows untreated and heated $n_{\text{INP}}(T)$ with uncertainty bounds for each sample and the corresponding heat-labile versus heat-stable fractions. In addition, the authors corrected the $-25\text{ }^{\circ}\text{C}$ summary statistics (mean 0.27 L^{-1} , reported as 0.3 after rounding; range $0.20\text{-}0.34\text{ L}^{-1}$) to ensure the numerical comparisons are internally consistent.

“Figure 5 displays the temperature-dependent INP number concentrations, $n_{\text{INP}}(T)$, for aerosol samples SA1-SA5 and their response to heat treatment. At $-15\text{ }^{\circ}\text{C}$, untreated n_{INP} ranged from 0.001 to 0.004 L^{-1} ($0.003 \pm 0.001\text{ L}^{-1}$; Figure. 5a–e). At $-25\text{ }^{\circ}\text{C}$, n_{INP} increased by approximately two orders of magnitude to $0.27 \pm 0.06\text{ L}^{-1}$, with a relatively narrow inter-sample range ($0.20\text{-}0.34\text{ L}^{-1}$). Overall, the $n_{\text{INP}}(T)$ 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 \pm SD across temperatures) are $76 \pm 19\%$ (SA1), $54 \pm 30\%$ (SA2), $37 \pm 33\%$ (SA3), $54 \pm 34\%$ (SA4), and $48 \pm 30\%$ (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.

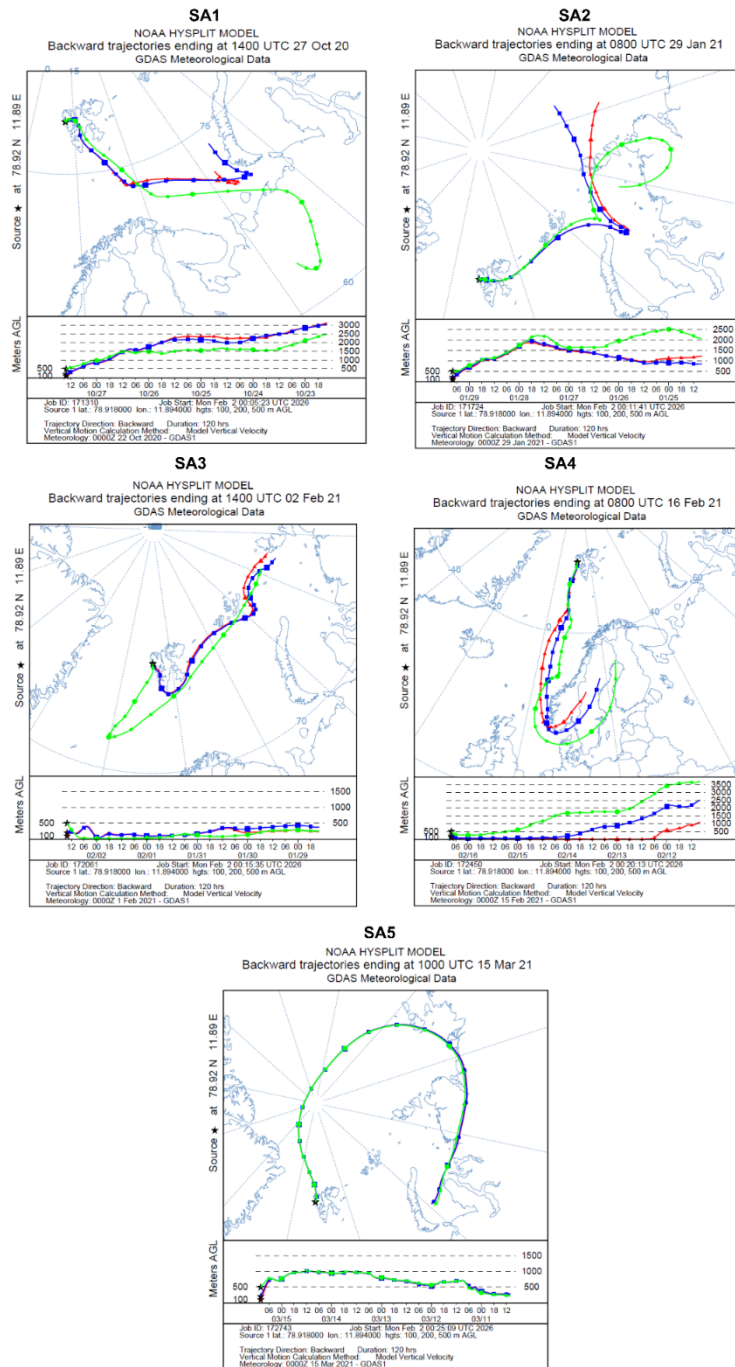


Figure S10. Height-resolved 120 h NOAA HYSPLIT backward trajectories for the five sampling periods (SA1–SA5), driven by GDAS meteorological fields. For each sample, trajectories are shown for endpoints at 100 m (red), 200 m (blue), and 500 m (green) above ground level (AGL) at the sampling location (star). 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 $-25 \text{ }^\circ\text{C}$ is **0.20–0.34 L^{-1}** . The manuscript has been updated accordingly.

3. Figure 5, how do you calculate the percentage of heat labile 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.

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

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

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

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