Response to reviewer

We gratefully thank the reviewer for the constructive comments and suggestions to improve the manuscript. Below are the detailed point-to-point responses to the reviewer's comments. For clarity, the reviewer's comments are listed below in *black italics*, while our responses and changes in the manuscript are shown in blue. The changes in the revised manuscript and supporting materials are also highlighted.

Anonymous Referee #1

Review of Sun et al. 2023: Measurement report: Atmospheric Ice Nuclei at Changbai Mountain (2623 m a.s.l.) in Northeastern Asia

Sun et al. 2023 present INP measurement results from the Changbai mountain in summer 2021. Changes in INP concentration are investigated towards diurnal variability, composition, source, and transport mechanism. It is an interesting study given the location of the measurement site, methodology and efforts taken in the analysis. However, major adjustments are needed to streamline and support the claims of the manuscript.

Response: We are grateful to the reviewer for the comments and have endeavored to respond to these and revise our manuscript accordingly.

General Comments:

Major doubts concern the influence of the PBL height to Changbai mountain in Section 3.4. To the reviewer the analysis and interpretation are inconclusive. Especially it is not entirely traceable how the PBL height was derived and the involved error. Given the points raised below, the reviewer is not entirely sure if some claims need to be removed if not being support by further evidence from in-situ observations. In addition, the local wind system and dynamics should be better described or referenced.

Overall, it appears that frequently there are statements in the manuscript that should be supported by a reference. The authors may want to consider this aspect, while working through the manuscript. In addition, the figures of the manuscript could also be linked to the made statements more often. Furthermore, there appear some side information at some point, which not necessarily add to the flow of the manuscript. The authors may want to critically read through the paper, deciding which information is needed to reach the presented conclusions.

Some strong statements are made without reference or presenting data. For additional support of some of the made claims particle size measurements may be helpful. Have there not measurements been available for the study?

Changbai mountain is referred as 2623 m a.s.l. (Tianchi site) and 2740 m a.s.l. (highest point). To avoid confusion the two locations and heights should be consistently appear combined.

For deeper explanations, see specific comments below.

Response: Thanks for the comment. In this study, the PBL data was obtained from the fifth-generation ECMWF global atmospheric reanalysis (ERA5 reanalysis). The reliability of ERA5 dataset has been substantiated in previous studies, such as Le et al. (2020), Tornow et al. (2021), and Slattberg et al. (2022). We made more specified statements regarding the source and data reliability of the PBL in the following response. Further details regarding the source and the robustness of the PBL data can be found in the subsequent responses.

We calculated the 72-hour backward trajectories, and found that prevailing air masses predominantly approached the sampling site from the east. However, the local winds exhibited a prevailing pattern from the west and south. The Changbai Mountains feature a topography characterized by higher elevations in the southeast and lower elevations in the northwest, and our observation site is located in the northwest direction. We inferred that air masses arriving from the east encounter obstruction by the Changbai Mountains, resulting in a lifting along the southern to western slopes. Combined with the trajectory heights of the air masses, it is evident that as the air mass approached the observation sites, their trajectories inclined upward along the southern or southwestern mountainsides. These findings suggested that the air masses underwent a noticeable lifting process prior to reaching the sampling site, potentially attributed to orographic lifting along the mountain slopes in the south and westward directions. We have added this information into the revised manuscript to provide a clearer understanding of the wind dynamics in this region.

We have added essential references to provide more comprehensive support for some of the statements. Furthermore, we have improved the integration of figure citations within the context. To enhance the overall flow of the article, we have removed the discussion of secondary ice formation from the conclusion section.

We apologize for the absence of parallel size measurements to distinguish particle chemical concentrations between particles larger than 2.5 µm and those smaller than 2.5 µm. This may result in some uncertainties in the investigation of the sources of INPs based on $PM_{2.5}$ chemical composition. These uncertainties have been addressed in the revised manuscript.

Our sampling site is situated at an elevation of 2623 m a.s.l. on Changbai Mountain. Notably, Lu et al. (2016) collected rainwater samples at the peak of Changbai mountain, which stands at an elevation of 2740 m a.s.l. We have clarified this in the revised manuscript.

Moreover, we diluted the untreated and heat-treated samples by the factors of 30, 60, and 120 times, and extended the freezing temperature below -25°C. We have updated the N_{INP} spectra and analysis in the whole revised manuscript.

Detailed point-to-point responses are shown below.

Specific Comments:

Introduction

1. L29: 'As most precipitation in clouds initiates via the ice phase', this statement could be refined in regard of which cloud types and regions are affected and direct studies could be cited.

Response: Thanks for the comment. We have revised this statement as follows: "Global precipitation is predominantly produced by clouds containing the ice phase, especially in continental regions and mid-latitude oceans, emphasizing the paramount significance of investigating ice formation within clouds (Mulmenstadt et al., 2015; Lau and Wu, 2003; Demott et al., 2010; Kanji et al., 2017)."

2. L63: 'At present, it is unclear whether…', in the reviewer's opinion the current knowledge gap is not whether but to which extend and through which transport pathway INPs are brought to MPC relevant heights.

Response: We agree and have therefore revised the statement as follows: "At present, there remain uncertainties how INPs can be extended and transported to the altitudes of mixed-phase cloud formation (approximately 3–7 km)."

3. L68: One station in Conen et al. 2017 was located outside the Swiss Alps.

Response: Thanks for the comment. We have revised the statement as follows: "For example, in the Switzerland, simultaneous measurements taken at different-altitude stations revealed a reduction of approximately 50% per kilometer in the abundance of INPs in the vertical gradient (ranging from 489 m above sea level (a.s.l.) to 3580 m a.s.l. in the Swiss Alps) in the warm season (Conen et al., 2017)."

4. L71: Wieder et al. 2022 sampled frequently for short time spans throughout the day, whereas Conen et al. 2017 used filters sampling over longer timespans. Wieder et al. 2022 observed also a diurnal cycle with INP concentrations seeming to equilibrate over the course of a day. Further it may be important to point out to the reader that this was only observed in a wind direction where the topography promoted vertical transport of air masses from lower elevation.

Response: Thanks for the comment. We have added "Note that variations in sampling methods and the influence of wind directions can also exert an impact on INP concentrations." in the revised manuscript.

5. L75-77: "For example, at the Jungfraujoch station (3580 m a.s.l.) in the Swiss Alps, approximately 80% of INPs were biological aerosols at freezing temperatures above −15 °C" – add reference.

Response: We apologize for missing this reference and cited it in the revised manuscript (Conen et al., 2022).

6. L81-82: "… and establishing a parametric equation that depends on temperature and ice supersaturation for predicting the INPs concentration" – this information seems irrelevant for the current study.

Response: We agree and have removed it in the revised manuscript.

7. L83: Here, Changbai mountain is attributed with a height of 2740 m a.s.l., whereas in L87 a height of 2623 m a.s.l. is given. If understood correct, the latter height refers to the INP measurement station of the current study. To avoid confusion the distinction between the peak and the measurement station should be made (also throughout the manuscript).

Response: Yes, the 2740 m a.s.l. in Line 83 was the peak height of Changbai mountain, and the 2623 m a.s.l. in Line 87 was the height of our sampling site. To avoid confusion, we have revised as "Changbai Mountain (at the peak of 2740 m a.s.l.)".

Methods

8. L102: Shouldn't the elevations around the mountain decrease to all directions?

Response: The elevations gradient around the mountain generally decreases to all directions. However, we aimed to describe the comprehensive pattern of height changes across the entire mountain, which is higher elevation in the southeast compared to the northwest directions. Furthermore, we have included the appropriate reference citation in the revised manuscript (Wang et al., 2014).

9. L104: Is 150m correct? Looking at maps it seems like more.

Response: Thanks. We carefully checked the distance from our observation site to Tianchi Lake, and revised the distance to 410 m in the manuscript.

10. L106-112: Adding a timeseries plot (appendix or supplement) of the general meteorological parameters throughout the campaign including exemplary INP concentration would be helpful to understand the sampling conditions.

Response: Figure R1 has been added to the Supporting Information and mentioned in section 2.1 as follows: "Figure S1 presents the timeseries of meteorological parameter (i.e. wind, temperature, and RH), NO_x concentration, and the concentration of INPs at -12℃, -17℃ and -21℃, as measured during the field campaign."

In addition, detailed sampling information has been added in Table S1 in the Supporting Information, as presented in Table R1.

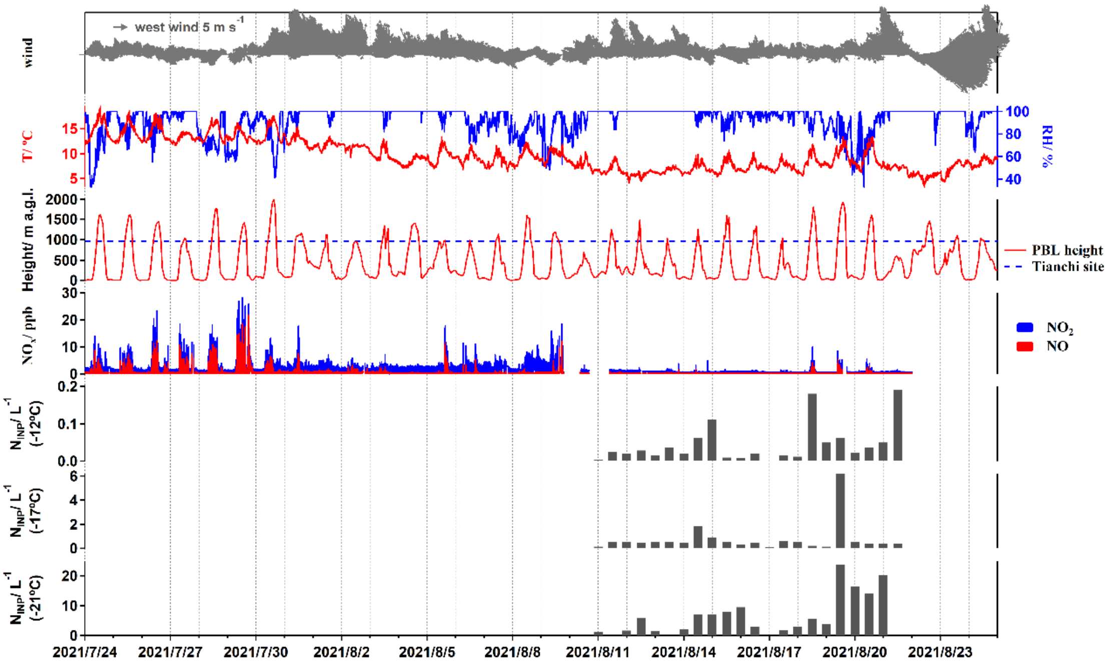

Figure R1. Time series of meteorological parameter (i.e., wind, temperature, and RH), the height of PBL and Tianchi site (above ground level), NO_x and the cumulative number concentration of INPs (N_{INP}) at -12 ℃, -17 ℃ and -21 ℃ measured during the campaign.

Table R1. Detail information including sampling date, duration and total sampling volume in this study.

Sample Date	Start time	End time	Duration/ min	Total Volume/ L
2021.8.10-Night	2021/8/10 18:00	2021/8/11 5:30	294	34500
2021.8.11-Day	2021/8/11 10:30	2021/8/11 17:30	168	21000
2021.8.11-Night	2021/8/11 18:00 2021/8/11 20:32	2021/8/11 20:26 2021/8/12 3:32	242	28300
2021.8.12-Day	2021/8/12 6:00	2021/8/12 17:30	294	34500
2021.8.12-Night	2021/8/12 18:00	2021/8/13 5:30	294	34500
2021.8.13-Day	2021/8/13 6:00	2021/8/13 17:30	294	34500
2021.8.13-Night	2021/8/13 18:00	2021/8/14 5:30	294	34500
2021.8.14-Day	2021/8/14 6:00	2021/8/14 17:30	294	34500
2021.8.14-Night	2021/8/14 18:00	2021/8/15 5:30	294	34500
2021.8.15-Day	2021/8/15 6:00	2021/8/15 18:00	288	36000
2021.8.15-Night	2021/8/15 18:30	2021/8/16 5:30	264	33000
2021.8.16-Day	2021/8/16 6:00	2021/8/16 17:30	294	34500
2021.8.16-Night	2021/8/16 18:00	2021/8/17 5:30	294	34500
2021.8.17-Day	2021/8/17 6:00	2021/8/17 17:30	294	34500
2021.8.17-Night	2021/8/17 18:00	2021/8/18 5:30	294	34500
2021.8.18-Day	2021/8/18 10:00	2021/8/18 17:30	198	22500
2021.8.18-Night	2021/8/18 18:00	2021/8/19 5:30	294	34500
2021.8.19-Day	2021/8/19 6:00	2021/8/19 17:30	294	34500
2021.8.19-Night	2021/8/19 18:00	2021/8/20 5:30	294	34500
2021.8.20-Day	2021/8/20 6:00	2021/8/20 17:30	294	34500
2021.8.20-Night	2021/8/20 18:00	2021/8/21 5:30	294	34500
2021.8.21-Day	2021/8/21 6:00	2021/8/21 17:30	294	34500
Blank-Night	2021/8/23 17:29	2021/8/24 5:36	295	-
Blank-Day	2021/8/24 7:30	2021/8/24 18:50	284	-

11. L112: What about touristic activities?

Response: We have added the touristic activities in the revised manuscript: "Changbai Mountain is a national nature reserve with no large industrial facilities nearby, and tourism is the important economic activity in the region. Due to the emergence of novel coronavirus (COVID-19) cases, strict lockdown measures have been implemented from August 10, 2021, resulting in a substantial reduction in visitor numbers, as indicated by the marked decrease in NOx concentration (Figure S1)."

12. L114: How was differentiated whether the sampling site was in the free troposphere or influenced by the PBL? Please elaborate and add data or reference.

Response: In Figure R1 (and also in Figure S1), we have included PBL data represented by the red line, while the blue dashed line corresponds to the height of the sampling site. Throughout the observation period, the sampling site experiences alternations between the free troposphere and the boundary layer due to changes in the PBL.

*13. L116: A dedicated reader may be interested in a picture of the setup (also possibly in the appendix or supplement)***.**

Response: We added the picture of the TH-150D medium flow sampler in Figure 1 in the revised manuscript, as show in Figure R2b.

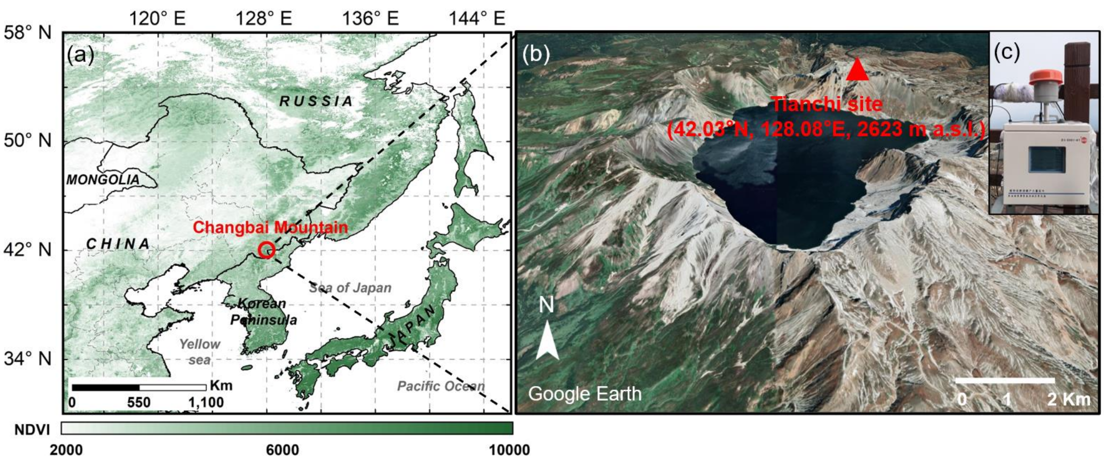

Figure R2. Geographical maps showing the location of Changbai Mountain. (a) This map is color-coded according to the normalized difference vegetation index (NDVI) in 2015, which was downloaded from the Geospatial Data Cloud (https://www.gscloud.cn/search). (b) This map shows the three-dimensional shape of the sampling site, which was obtained from Google Earth. (c) The ice nuclei sampler (The TH-150D medium flow sampler, Wuhan Tianhong Corporation, China).

14. L117: Was there any pretreatment of the PCTE filters?

Response: We did not pretreat the PCTE filters prior to sampling.

15. L117-124: For clarity, maybe specify what each filter sample type was used for, i.e. PCTE for INP analysis, PM2.5 for chemical composition.

Response: Thanks. We have added specific descriptions in the revised manuscript.

16.L120: There were 24 samples taken covering roughly 30% of the entire measurement campaign. The authors may want to comment on the underlying measurement strategy and providing more concrete information under which meteorological conditions samples were taken.

Response: Thanks for the comments. In this study, a total of 24 samples were collected on PCTE filters from August 10 to 24, 2021, which included 2 blank filters. Notably, our observation period coincided with a significant reduction in human activities following the strict lockdown measures enforced after August 10 (refer to response to comment 10). As demonstrated in Figure R1 (and also in Figure S1), the concentration of NOx decreased markedly from 3.0±2.1 ppb during July 24 to August 9 to 0.9±0.3 ppb between August 10 and August 24. This effectively minimizes the influence of human activities on the collection of INPs samples.

17. L120: The chemical analysis was conducted on the samples collected using a PM2.5 inlet. Can the authors estimate the number of particles larger than 2.5 µm which contributed to the INP analysis samples, but would not be covered in the chemical analysis? Has there been any parallel size measurements supporting that claim?

Response: We apologized that we did not collect the INPs at different stages and we had no parallel size measurement to differentiate the particle chemical concentrations between particles larger than 2.5 µm and those smaller than 2.5 µm. This may result in some uncertainties in the investigation of the sources of INPs based on $PM_{2.5}$ chemical composition. These uncertainties have been addressed in the revised manuscript.

18. L130: For consistency, specify the type of weather station.

Response: The Tianchi weather station is an institution under the Changbai Mountain Meteorological Bureau and is affiliated with the National Meteorological Station. We added the information in the revised manuscript: "Meteorological data, such as temperature, humidity, WS, wind direction, pressure, and precipitation, were monitored by the Tianchi weather station, a national meteorological station located approximately 20 m from the sampling site."

19. L153: above -> at

Response: Thanks, we had revised the manuscript accordingly.

20. L158: What was the value of Vair?

Response: V_{air} ranged from 2.1×10^4 to 3.6×10^4 L during the sampling period. Details of the total sampling volume was shown in Table R1.

21. L159: If the INP concentrations were given in standard liters, the reviewer would advise to indicate this by using e.g., sL-1, or stdL-1 in Equation 3 and corresponding INP data plots.

Response: In the explanation of the formula for NINP calculation, we described the computations conducted in standard liters. Consistent with the approach used in Chen et al. (2018; 2021), we intend to retain this explanation in the main text while excluding it from the INP data plots.

22. L178: Which PBL data product was used? The reviewer could not find a unique record. Furthermore, what is the uncertainty and sensitivity of the PBL data product? Is Changbai mountain centered in a grid box that data was taken from or were different adjacent grid boxes averaged? Given the complex terrain of the mountainous region, how reliable does a 25km x 20km grid box represent the PBL height? Generally, the reviewer is a bit skeptical of the representativeness for the presented application. The authors should elaborate why this data is applicable. In addition, are there any direct meteorological (especially wind) observations along the mountain slope that would support the later claim of vertical transport due to orographic lifting?

Responds: The PBL data was obtained from the fifth-generation ECMWF global atmospheric reanalysis (ERA5 reanalysis). This dataset has been widely used in numberous studies, such as Le et al. (2020), Tornow et al. (2021), and Slattberg et al. (2022). Guo et al. (2021) conducted a comparative analysis of ERA5 reanalysis products against other widely used products, i.e., MERRA-2, JRA-55, and NCEP-2. The results showed that the ERA5 exhibited the smallest bias. Therefore, we have confidence in the reliability of the PBL data sourced from ERA5 for our analysis.

In the input meteorological dataset, specifically the Global Data Analysis System (GDAS) data, Changbai Mountain's terrain height is recorded at 1656 m. In our simulation, we utilize a trajectory ending height of 967 m (above ground level), to achieve a sampling station elevation of 2623 m.

As show in Figure R3b, the 72-hour backward trajectories showed that prevailing air masses predominantly approached the sampling site from the east. However, local winds exhibited a prevailing pattern from the west and south. As elucidated in the manuscript, the Changbai Mountains exhibit a topography characterized by a southeastern high and a northwestern low. The observation site is situated in the northwest of Tianchi Lake. Air masses arriving from the east encounter obstruction by the Changbai Mountains, resulting in a lifting along the southern to western slopes. Figure R3c showed the trajectory heights of the air masses during the daytime. It is evident that as they approached the observation sites, their trajectories inclined upward along the southern or southwestern mountainsides (Figure R3c). This suggests that the air masses underwent a noticeable lifting process prior to reaching the sampling site, potentially attributed to orographic lifting along the mountain slopes in the south and westward directions.

Air masses arriving from the east encounter obstruction by the Changbai Mountains, resulting in upward displacement along the southern to western slopes. Figure R3c illustrates the trajectory heights of these air masses during daytime. It is evident that as they approached the observation site, their trajectories exhibited an upward trend along the southern or southwestern mountainsides (Figure R3c). This suggests that the air masses underwent notably lifting process prior to reaching the sampling site, likely attributed to orographic lifting along the mountain slopes in the south and west directions.

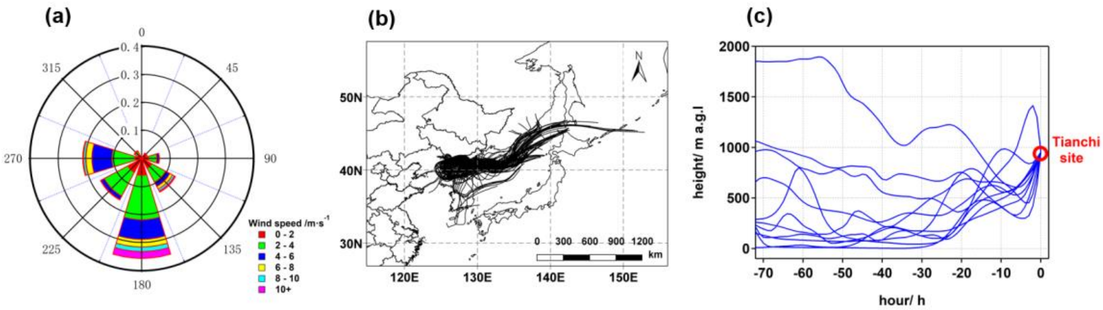

Figure R3. Wind rose illustrating one-minute wind speed and directions measured at the sampling site (a). Air mass trajectories over the entire campaign duration (b) and the average daytime air mass trajectory heights (c).

23. L184: The authors may want to briefly describe the principle of CWT.

Responds: We have added the description of CWT in Section 2.5 as follows:

"The CWT assigns the average weighted concentration by trajectories were divided into grids. The calculation was used Equation 5 according to the method of Hsu et al. (2003):

$$C_{ij} = \frac{1}{\sum_{k=1}^{M} \tau_{ijk}} \sum_{k=1}^{M} C_k \tau_{ijk}, \quad (5)$$

where C_{ij} is the average weighted concentration in the ij cell, k is the index of the trajectory, M is the total number of trajectories, C_k is the concentration observed on arrival of trajectory k in the ij cell, and τ_{ijk} is the time spent in the ij cell by trajectory. The weight function W_{ij} was also applied to the CWT analysis to reduce the uncertainty in the cells with small values of n_{ij}:

$$WCWT_{ij} = C_{ij} \times W(n_{ij}), \quad (6)$$"

Results and Discussion

24. L190: "characterize situation of droplet freezing" sounds odd.

Responds: In the revised manuscript, we have revised this sentence to make it more readable: "A metric was applied to compare the droplet freezing results, i.e., the freezing temperature at which 50% of the droplets are frozen (T_{50})."

25. L192: As there are only two MilliQ water backgrounds displayed, rephrase to "were -30°C and -28.5°C".

Response: We conducted measurements on more than two MilliQ water samples, as displayed by the purple lines in Figure R4.

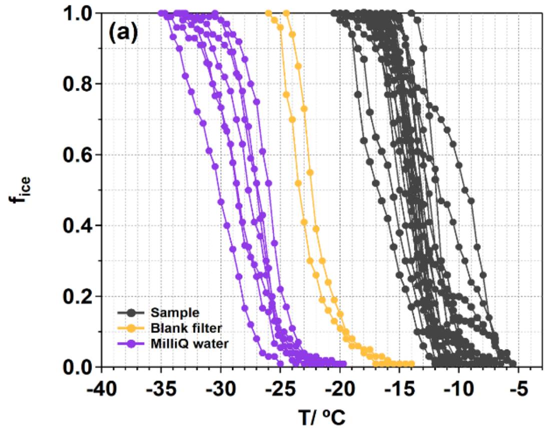

Figure R4. Frozen fractions (f_{ice}) as functions of temperature. The f_{ice} of collected samples measured by GIGINA is shown by the black curves, and presented together with blank filters (orange curves) and MilliQ water (purple curves) as background signals.

26. L193-196: This seems contradicting. If the contaminants could be ignored, one would not need to correct for it.

Response: We agree and therefore revised the statement as follows: "For the two blank filters, T_{50} was averaged to −24.2 ± 2.12 °C, which was slightly higher than that of MilliQ water, but much lower than that of the collected samples (for which T_{50} was −17.0 ± 4.1 °C), indicating the presence of minimal contaminants stemming from the filter membrane.

27. L195-196: Were averaged concentrations at each temperature step of the two blank filter samples subtracted as correction – please specify. As two samples are not many, the authors may want to comment on the overall obtained repeatability of blank filter measurements obtained which are not presented in this study.

Response: We collected blank filters during both daytime and nighttime, as listed in Table R1. The sampling duration lasted for 11 hours, and we believe that the two blank samples could adequately represents the background values of the filters. During the calculation process, we applied corrections by subtracting the values obtained from the two blank filter samples at each freezing temperature.

28. L199: -26.0°C -> -20°C

Response: Thank you for the comment. In the revised manuscript, we have updated the temperature value from "-26.0°C" to "-29°C." This modification was made because we diluted the samples to obtain the N_{INP} spectra at lower freezing temperature.

29. L200: What it the values of T50? -13°C from above? The authors should repeat this value here, or introduce another variable to avoid the nomenclature confusion of T50 representing the result of one sample (L191) or the average of all samples (L194).

Response: Thank you for the comment. The T_{50} was -17°C based on the full freezing temperature spectra from −29.0 °C to −5.5 °C. We have made the corrections in the revised manuscript.

30. L201: It is not necessarily the diversity of different INPs but could also just relate to different emission strengths.

Response: Thanks for the comment. We have removed the statement in the revision.

31. L202: What would that local source be?

Response: We apologize for the vague wording. In the revision, it has been modified to read as follows: "Some of the N_{INP} curves exhibited bumps in the HTR, which has been previously reported at a coastal site (the Cape Verde Atmospheric Observatory, Africa) in air samples by Welti et al. (2018), as well as in the upper bound of the composite nucleus spectrum of cloud water and precipitation samples by Petters and Wright (2015). Welti et al. (2018) reported that when the IN properties are narrower, the steeper slope can be observed in a temperature spectrum".

32. L205-206: This statement cannot be made, as it seems that there have no dilutions of samples been made. This sets the upper limit in detectable NINP = 1/Vair i.e., all droplets frozen.

Response: In the revised manuscript, we extended the freezing temperature below -25°C by diluting the samples. Firstly, we re-measured INP concentrations for the original samples, and found that the concentration of N_{INP}, $N_{INP\text{-}heat}$, and $N_{INP\text{-}H2O2}$ were basically consistent between the latest measurements and previously recorded concentrations (Figure R5). Subsequently, we diluted the suspension liquid by the factors of 30, 60, and 120 times, ensuring that all samples reached a freezing temperature of at least -25°C. Consequently, we have updated the freezing temperature spectra of N_{INP} in the revised manuscript.

Based on the updated data, we are confident in this statement. Here, we have modified the statement in the revised manuscript: "In contrast, in the low-temperature region (LTR, freezing temperature below T_{50}, -17.0 °C ~ -29.0 °C), N_{INP} showed a relatively narrow variation than LTR, from 0.1 L^{-1} to 78.3 L^{-1}."

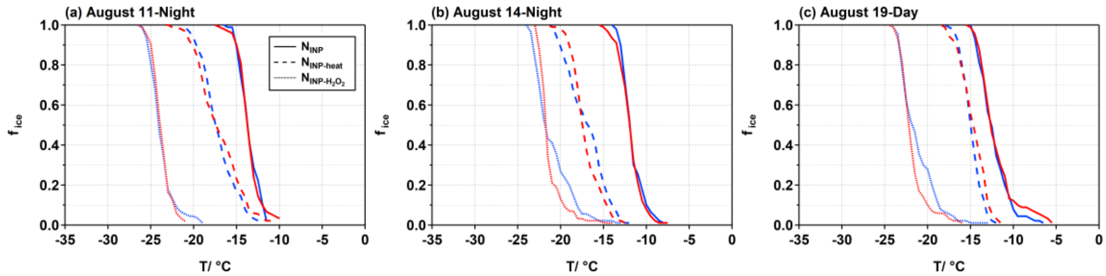

Figure R5. Comparison of frozen fractions (f_{ice}) as functions of temperature between the latest measurements and previously recorded concentrations. The blue and red lines represent the experiment conducted on January 2022 (previous experiment) and September 2023 (latest experiment), respectively.

33. L206: To the reviewer, the temperature dependence results especially from the fact that NINP is cumulative. A direct connection to the complexity of sources cannot be made.

Response: We agree and have removed the statement in the revision.

34. L207: What test was used to check for differences between daytime and nighttime samples?

Response: We conducted an independent t-test, yielding a significance level is 0.61 which is higher than 0.05. Therefore, we concluded that there are no significant differences in N_{INP} between the daytime and nighttime samples.

35. L207-211: It remains questionable if the sampling intervals used in the study (maximally two per day, 20 samples total spread out unknown over a month) allow for the detection of a diurnal cycle. It is conceivable that if, e.g., the minimum in INP concentration occurs at 6:00 and the maximum at 18:00, no difference would be observed on the filters. The described scenario could be likely if the transport is facilitated by convection. In addition, are there any potential local sources located on Changbai mountain, e.g., from the lake, which could cover up a potential diurnal cycle? The authors should elaborate on these aspects.

Response: Our objective is to compare differences in INPs concentration between daytime and nighttime, rather than delve into an extensive analysis of diurnal variations. Factors such as changes in valley breezes, the evolution of the PBL, photochemical reactions, and other variables may exert distinct effects on INPs concentration during these two time periods. Additionally, local sources such as vegetation and the lake could also influence INPs concentration, with biogenic emissions potentially differing between daytime and nighttime.

We agree that our dataset was limited in size, resulting in uncertainties when comparing INPs concentrations between daytime and nighttime. These uncertainties have been addressed in the revised manuscript.

36. L212-226: In the following discussion, references to the data figure could be beneficial. Furthermore, when discussing the obtained results to previous studies, more precision is needed, what is the difference between "narrowly" (L214) and "much narrower" (L216)? More quantitative expressions are needed. In addition, the present study motivated the need for measurements on high altitude sites. It is not entirely clear, how the sites of Cerro Mirador and Beijing relate to this. Maybe an extra motivation for this comparison would be needed. Studies from high altitude stations like Storm Peak (US), Jungfraujoch (Switzerland), or Altzomoni (Mexico) might be interesting additions, which could also complement Figure 2b, but the reviewer leaves this up to the authors.

Response: In the revised manuscript, we have included temperature range data to provide a more precise explanation of the terms "narrow" and "much narrower."

In the revised Figure 2b, we have excluded the N_{INP} data for Cerro Mirador and Beijing, and added the N_{INP} data for Jungfraujoch and Colorado Rocky Mountains. The corresponding discussions have been added as follows: "In the LTR, our results were comparable with the measurements conducted at the Storm Peak Laboratory in the northwestern Colorado Rocky Mountains (Hodshire et al., 2022). However, in the HTR, N_{INP} measured in Switzerland were approximately 1-3 orders of magnitude higher than in our study. The high concentration of N_{INP} was primarily attributed to the aerosolized epiphytic microorganisms, which contributed most of the INPs to primary ice formation in Switzerland (Conen et al., 2022)."

37. L213: Precise to Swiss Alps.

Response: Thanks for the comment. We have made the corrections in the revised manuscript.

38. L214: Shouldn't it be high-temperature and low-concentration region and vice versa?

Response: Thanks for the comment. We have revised the statement as follows: "In our observations, the spectra range of N_{INP} were narrowly located in the relatively high-concentration regions."

39. Section 3.2: The reviewer perceived the usage of the abbreviations bio-INPs, other org-INPs, and inorg-INPs at times a bit odd. If the authors want to keep them, they may want to consider introducing them already in the introduction.

Response: Thanks for the comment. In the introduction, we provided a brief overview of the categories of bio-INPs, other org-INPs, and inorg-INPs to facilitate a better understanding for readers in following sections.

40. L236: Specify type of H2O2

Response: Thanks for the comment. We have added this information in the revised manuscript.

41. L254-256: Was this increase significant and isn't it rather a bias due to the measurement limitations (no dilutions)?

Response: We have updated Figure 3b based on the dilution procedure, and have significantly expanded the dataset at lower temperatures below -25°C. Figure R6 illustrated the revised boxplot depicting fractions of biological INPs, other organic INPs, and inorganic INPs. It can be found that as the temperature decreased from -16.5°C to -21.5°C, the median value of $F_{INP\text{-}bio}$ increased from 0.8 to 0.9. We are confident that this increase reflects the presence of biological INPs exhibiting relatively high ice-nucleating activity in the LTR.

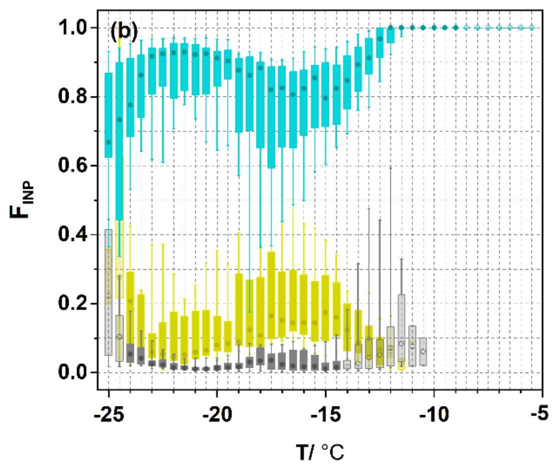

Figure R6. Boxplot of fractions of bio-INPs ($F_{INP\text{-}bio}$, blue boxplot), other org-INPs ($F_{INP\text{-}other\ org}$, yellow boxplot), and inorganic INPs ($F_{INP\text{-}inorg}$, gray boxplot) as functions of temperature. The upper and lower extents of the boxes represent the 75th and 25th percentiles, respectively, while the whiskers indicate the 10th and 90th values. The circle in each boxplot represents the median value. The light-colored boxes indicate that the number of data points is less than half (the sample number is less than 11) of all samples at each temperature.

42. L264: Add the actual number of samples that make up 50% (also in the caption of Fig. 3).

Response: Thanks for the comment. We have added the actual number of samples in the revised manuscript.

43. L269-270: Again, how significant was this increase and is the decrease to low temperatures owed to a measurement bias?

Response: Please see our response to Comment 41.

44. Section 3.3: The data used in this section is unavailable. As the raw data for creating Figure 4 is already quite digested in Figure 4, the authors may want to add a table with the data written out per sample in the appendix. In addition, the found correlations are described as "good" or "weak". As the definitions for these terms may vary, the value and type of used *correlation should be named throughout the manuscript.*

Response: Thanks for the comment. We have added a table to show the Pearson correlation coefficients in the supplementary materials, as shown in Table R3. In the table caption, we provided a description of the correlations as follows: "When r is below 0.5, the correlation is considered weak; when r exceeds 0.5, the correlation is considered good." In addition, we have added the r and p values in the revised manuscript.

Table R3. The Pearson correlation coefficients (r) between (a) N_{INP}, (b) $N_{INP\text{-}bio}$, (c) $N_{INP\text{-}other\ org}$, (d) $N_{INP\text{-}inorg}$ and meteorological parameters, chemical compositions, as functions of temperature. Coefficients reported in bold are statistically significant at $p < 0.05$, while the shades indicate that the number of data points is less than half (the sample number is less than 11) of all samples at each temperature. When r is below 0.5, the correlation is considered weak; when r exceeds 0.5, the correlation is considered good.

	-25	-24	-23	-22	-21	-20	-19	-18	-17	-16	-15	-14	-13	-12	-11	-10	-9	-8	-7
(a)																			
T	**0.63**	**0.61**	**0.63**	**0.64**	**0.60**	**0.51**	0.43	**0.47**	**0.49**	**0.52**	**0.58**	**0.43**	0.16	0.28	0.25	0.19	0.12	0.08	-0.14
RH	**-0.66**	**-0.63**	**-0.69**	**-0.69**	**-0.65**	**-0.6**	-0.44	-0.37	-0.28	-0.26	-0.31	-0.04	0.07	0.03	0.08	0.16	0.09	0.17	0.14
WS	-0.22	-0.28	-0.24	-0.25	-0.2	-0.12	-0.08	-0.2	-0.23	-0.22	-0.23	0.04	0.20	0.36	**0.52**	**0.57**	**0.74**	0.57	0.44
BC	**0.84**	**0.8**	**0.83**	**0.76**	**0.63**	**0.53**	0.34	0.23	0.16	0.1	0.13	-0.11	0.07	-0.06	-0.14	-0.32	-0.28	-0.54	-0.37
$PM_{2.5}$	**0.63**	**0.56**	**0.54**	0.47	0.43	0.42	0.27	0.11	-0.02	-0.04	-0.01	0.04	0.19	0.17	0.07	0	0.17	-0.1	-0.04
NH_4^++NO_3^-+SO_4^{2-}	0.13	0.01	0.09	0.05	0.02	0.05	-0.01	-0.14	-0.18	-0.22	-0.26	-0.43	-0.20	-0.29	-0.18	-0.24	-0.19	-0.57	-0.64
Ca^{2+}	-0.22	-0.27	-0.31	-0.30	-0.24	-0.14	-0.09	-0.13	-0.19	-0.09	-0.14	0.05	-0.05	0.08	**0.64**	0.67	**0.94**	**0.93**	0.71
(b)																			
T	**0.53**	**0.59**	**0.62**	**0.64**	**0.59**	**0.48**	0.37	0.48	**0.54**	**0.5**	**0.57**	**0.44**	0.16	0.27	0.25	0.19	0.12	0.08	-0.14
RH	**-0.58**	**-0.68**	**-0.68**	**-0.69**	**-0.64**	**-0.57**	-0.37	-0.34	-0.28	-0.27	-0.31	-0.05	0.06	0.04	0.08	0.16	0.09	0.17	0.14
WS	-0.17	-0.23	-0.24	-0.24	-0.19	-0.1	-0.1	-0.28	-0.25	-0.21	-0.2	0.11	0.2	0.37	**0.52**	**0.57**	**0.74**	0.57	0.44
Isoprene	0.66	0.45	0.37	0.45	0.5	0.42	0.44	-0.14	-0.13	0.01	0.43	0.44	0.49	0.53	0.63	0.61	0.7	0.66	0.94
Isoprene	0.95	0.71	0.7	0.71	0.69	0.51	0.47	-	-	-	0.13	0.03	0.09	0.33	0.65	-	-	-	-

$\times O_3$								0.09	0.16	0.44									
Cl^-	-0.24	-0.32	-0.38	-0.35	-0.31	-0.31	-0.22	-0.15	-0.12	-0.03	-0.05	-0.01	-0.15	-0.19	-0.22	-0.31	-0.32	-0.51	-0.43
Ca^{2+}	-0.23	-0.25	-0.32	-0.31	-0.24	-0.15	-0.12	-0.15	-0.18	-0.11	-0.13	0.09	-0.04	0.08	**0.64**	0.67	**0.94**	**0.93**	0.71
(c)																			
T	**0.96**	0.15	**0.51**	0.29	0.28	**0.62**	**0.61**	**0.7**	**0.68**	**0.51**	0.32	0.1	0.16	-0.3	-	-	-	-	-
RH	-	0.02	-0.17	-0.18	-0.23	-0.42	**-0.67**	**-0.44**	-0.33	-0.18	-0.04	0.01	-0.01	0.22	-	-	-	-	-
WS	-0.4	-0.15	-0.12	-0.21	-0.27	-0.33	-0.21	**-0.45**	**-0.46**	**-0.48**	**-0.42**	-0.32	-0.3	-0.17	-	-	-	-	-
Isoprene	-	0.68	0.33	0.24	0.24	0.28	-0.07	0.69	0.7	**0.76**	**0.76**	**0.73**	-0.14	-0.96	-	-	-	-	-
Isoprene $\times O_3$	-	0.32	-0.6	-0.33	-0.13	-0.13	0.09	0.54	0.58	0.61	0.59	0.64	-0.15	-	-	-	-	-	-
OC	0.85	0.35	0.12	0.32	0.42	0.07	0.2	-0.15	-0.12	-0.16	-0.18	-0.06	0.28	0.43	-	-	-	-	-
EC	0.93	0.36	0.07	0.13	0.25	0	0.18	0.22	0.16	0.27	0.28	0.35	0.31	0.43	-	-	-	-	-
(d)																			
T	0.06	0.26	**0.59**	0.34	0.34	0.36	**0.47**	0.32	0.34	0.1	0.04	-0.28	-0.59	-0.45	-0.56	-	-	-	-
RH	-0.33	-0.32	**-0.7**	**-0.46**	**-0.52**	**-0.53**	**-0.62**	**-0.63**	**-0.64**	-0.5	-0.16	0.38	0.48	0.42	0.65	-	-	-	-
WS	0.28	-0.19	-0.31	-0.16	-0.08	-0.1	-0.13	-0.09	-0.14	0.24	0.08	0.01	0.47	0.49	0.91	-	-	-	-
$NH_4^+ + NO_3^- + SO_4^{2-}$	**0.78**	**0.65**	0.46	**0.63**	**0.62**	**0.49**	0.43	0.34	0.36	0.49	0.23	0.41	0.64	0.56	-	-	-	-	-
Cl^-	-0.36	-0.27	-0.37	-0.35	-0.3	-0.43	-0.46	-0.38	-0.32	-0.31	-0.3	-0.32	-0.19	-0.18	-	-	-	-	-
Ca^{2+}	0.34	-0.03	-0.26	-0.31	-0.24	-0.35	-0.32	-0.39	-0.29	0.27	0.39	0.21	-0.18	0.03	-	-	-	-	-

BC	0.28	0.46	**0.85**	**0.74**	**0.74**	**0.78**	**0.8**	**0.83**	**0.74**	0.48	0.24	-0.2	-0.14	-0.16	0.82	-	-	-	-

45. L278-279: Even though the authors selected a mild formulation for their interpretation, this statement should be toned down a little bit more for having only found a good correlation with one element. Furthermore, could there be other sources as well?

Response: We have revised the statement as follows: "Moreover, N_{INP} and Ca^{2+} showed a good positive correlation (r = 0.6-0.9) in the HTR within the range of -11.0 ℃ to -9.0 ℃, leading us to speculate that soil dust may play an important role in ice nucleation in this temperature range."

In addition to Ca^{2+}, we did not conduct the analysis of other ions originating from soil dust.

46. L281: Kanji et al. 2017 is a summary, maybe use direct study for this reference.

Response: Thanks. We have cited the references, i.e., Hill et al. (2016) and O'sullivan et al., (2014), in the revision.

47. L288-289: Is there reference for this methodology? What was the used threshold in concentration to come to this conclusion?

Response: The dust event is defined as the day when the peak PM_{10} concentration exceeds 150 μg m^{-3} and the $PM_{2.5}/PM_{10}$ ratio falls below 0.4, in accordance with previous studies (Wu et al., 2020; Liu et al., 2006). During our sampling period, $PM_{2.5}$ concentrations ranged from 1.5 μg m^{-3} to 31.6 μg m^{-3}, with an average of 9.3±6.0 μg m^{-3}. Notably, these values remained significantly lower than the established threshold.

48. L306-309: Despite being textbook knowledge, the authors may want to give a reference to read up in these topics. Furthermore, a short description of the essential processes for Changbai mountain could be given in the introduction.

Response: We have cited the references, i.e., Chow et al. (2013) and Wieder et al. (2022) in the revised manuscript.

In the method section, we have added a description of the meteorological conditions as follows: "Changbai Mountain is situated within the westerly wind belt and experiences a typical temperate continental mountain climate influenced by the monsoon, characterized by long cold winters and short temperate summers. The prevailing winds in this region are the westerly and northwesterly winds in the spring, autumn, and winter seasons, and the southeasterly and southwesterly winds in the summer season (Zhao et al., 2015)."

49. L313: Specify "moderate-to-good".

Response: We have added the r values in the revision.

50. L315: "exceptionally high NINP-bio values" add e.g., 'as discussed below' for *readability.*

Response: Thanks for the comment. We have made revisions in the revised manuscript.

51. L315-316: Even though being only a suggestion, in the reviewer's opinion this statement cannot be made given too many assumptions and misinterpretation. First, the two high INP cases are excluded and given the argument should coincide with height. Is that an indication for a potential strong but infrequent local source? This might well be pure coincidence, but given the dates being a week apart, was there some periodic event near the measurement site? Second, while the correlation (which type of coefficient?) for some temperature is comparably large, there does not seem to be a significant increase in INP concentration. In addition, if the transport of bio-INP was the underlaying process, one could expect that the correlation should be expressed at a broad range of INP concentrations at a wide range of temperature in the HTR – has this been observed and could the correlation at all temperature been shown, e.g., in a table? Lastly, the PBL height never extends to the mountain top. For transport there is further evidence needed like wind speed and direction along the slope to support this claim. Ultimately, the analysis also bases only on 6-9 datapoints and the uncertainty in the PBL product remains undiscussed.

Response: In the revision, we extended the freezing temperatures below -25°C by diluting the samples, resulting a larger dataset for N_{INP}, especially in the low-temperature region (LTR). Therefore, we re-calculated the correlation that include all samples, without excluding the two high values. The correlation between PBL height and N_{INP} is illustrated in Figure R7. It is evident that the Pearson correlation coefficient between bio-INPs and PBL in the LTR has notably increased compared to the initial calculation. However, in the HTR, there was no correlation between bio-INPs and PBL. For example, Figure R8 (also Figure 5ab in the revision) showed the relationship between bio-INPs and PBL at temperatures at -10.5°C and -21°C. During daytime sampling, bio-INPs and PBL exhibited a good positive correlation at -21°C, but showed no correlation at -10.5°C. However, upon excluding the two high INP cases at-10.5°C, the remaining seven cases exhibited an increasing trend in bio-INPs as PBL height increased ($r=0.77$, $p<0.05$). The two high INP cases may be source from potential strong but infrequent local sources, which have been added in the revision.

The backward trajectory in Figure R3c showed that the air mass underwent a noticeable lifting process prior to reaching the sampling site, which could be associated with orographic lifting along the mountain slopes in the south and westward directions. We agree the presence of uncertainties in both the simulation of air mass backward trajectory and the determination of PBL height. These uncertainties have been added in the revised manuscript.

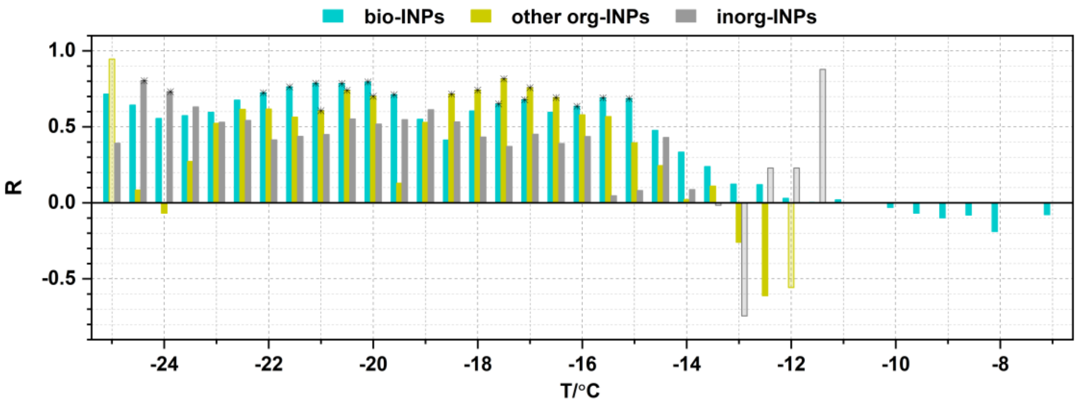

Figure R7. The relationship between PBL height and $N_{INP\text{-}bio}$, $N_{\text{INP-other org}}$ as well as $N_{\text{INP-inorg}}$ during daytime (8:00-17:00, m above ground level) as a function of temperature. The r denote the Pearson correlation coefficients. The asterisk indicates $p < 0.05$. The shades indicate that the data points number were less than half of all samples at each temperature.

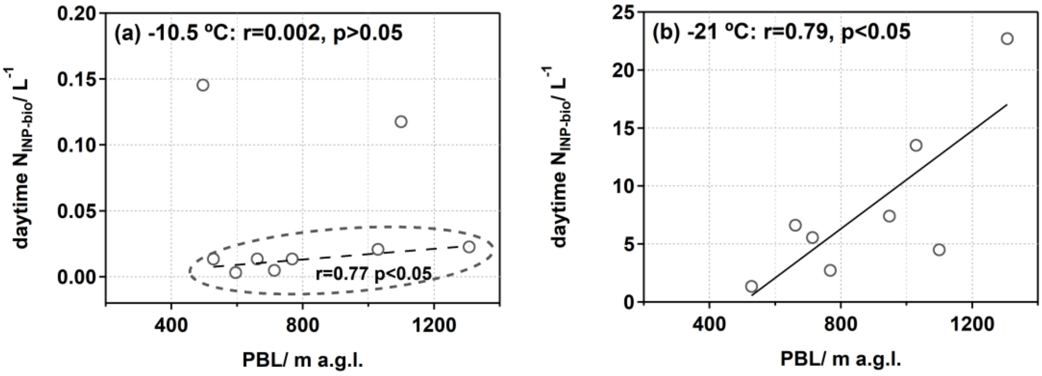

Figure R8. Relationship between $N_{INP\text{-}bio}$ and PBL height during the daytime (8:00–17:00 LT) at freezing temperature of −10.5 °C and −21.0 °C. The r denotes the Pearson correlation coefficients.

52. L316-319: For this claim, data should be presented.

Response: We have provided data in the supplementary materials as Figure S6, as shown in Figure R3c.

53. L328: Add reference for phytoplankton blooms occurrence.

Response: We have conducted a correlation analysis on the entire dataset and did not delve further into the origin of the two high values.

54. L348: Were there still enough datapoints available below

Response: After the dilution experiment, there were enough datapoints below -19.5℃.

55. L357-358: Following Figure S1, it appears to the reviewer, that the correlations for other *org-INPs seem similarly or even more consistent throughout the temperature range than for the presented bio-INPs data.*

Response: After the dilution experiment, we updated the N_{INP} values and revised Figure S1 (as shown in Figure R7. Generally, bio-INPs and PBL showed good correlations (r=0.4-0.8) in the LTR within the range of -25.0 °C to -15.0 °C, while other org-INPs and PBL showed good correlations (r=0.5-0.8) within the range of -23.0°C to -20.0 °C and -19.0 °C to -15.5 °C. Detail description can be found in the revised manuscript.

Conclusion

56. L364-371: Maybe repeat introduced variables such as FINP-bio, LTR, WS.

Response: We have added an explanation to the abbreviation in the revised manuscript.

57. L377-378: "With larger contributions observed from local and oceanic sources" - was this shown in the results section?

Response: The conclusion has been included in the revised Section 3.4.

58. L379-389: The reviewer is unsure whether diving in to the topic of secondary ice formation in the last paragraph without any prior mention of the topic is within the scope of this publication.

Response: The last paragraph has been removed in the revision.

59. L384: "confirm" seems to be the wrong word, the statement should be weakened.

Response: We have changed the word to "indicate".

Figures

60. Figure 1: Is it essential to indicate Beijing? If so, it should be named in the caption. To stay consistent between (b) and (a) it could be beneficial to indicate Changbai mountain (check spelling in figure) in red in (a).

Response: We removed the labeling for Beijing and adjusted the text color for the stations in (a) and (b), as shown in Figure R2.

61. Figure 2a: What is the error in T and f?

Response: The error of f_{ice} was 0.002-1.0. To ensure the readability of the information in the figure, we did not display the error bars in Figure 2a.

62. Figure 2b: Specify the data of Wieder et al. 2022 to Weissfluhjoch instead of Alps. The *data of Wieder et al. 2022 is not fully visible. Beijing data is hard to read. For better comparability, maybe an average or median of each data set could be added.*

Response: We have revised the site names and adjusted the concentration display range on the spectra. The data from Beijing has been removed, and data from the high-altitude stations have been added into Figure 2b, as shown in Figure R9.

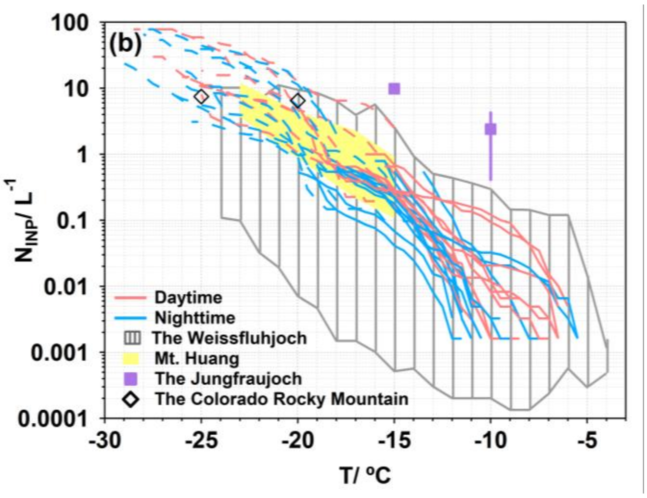

Figure R9. The concentrations of INPs (N_{INP}) as functions of temperature. The dark gray shaded area represents the upper and lower limits of N_{INP} over the Weissfluhjoch (2693 m a.s.l.) (Wieder et al., 2022). The yellow shaded area represents the atmospheric N_{INP} ranges at Mt. Huang (1840 m a.s.l) (Jiang et al., 2015). The purple square represents the median N_{INP} at −15 °C and −10 °C in the Jungfraujoch (3580 m a.s.l.) (Conen et al., 2022). And the black rhombus represents the median N_{INP} at −25 °C and −20 °C at the Storm Peak Laboratory in the northwestern Colorado Rocky Mountains (3220 m a.s.l.) (Hodshire et al., 2022).

63. Figure 3a and 3b: Using thin lines instead of dots per spectrum may enhance the readability.

Response: We made the changes in the revised manuscript.

64. Figure 3a: Error bars extend beyond axis limits. As NINP, NINP-heat, and NINP-H2O2 are only used to calculate NINP-bio, NINP-other org, and NINP-inorg Figure 3a could be moved to appendix to focus on the essential plots 3b and 3c.

Response: We modified the N_{INP} concentration display range on the spectra, and moved the figure to Supplementary Information.

65. Figure 3b: Are all the calculated differences in concentration significant? Could (exemplary) error bars be added?

Response: The presence of error bars can sometimes obscure the information in the figure. Consequently, we have presented the error bars in Figure R10, and added it in the supporting information.

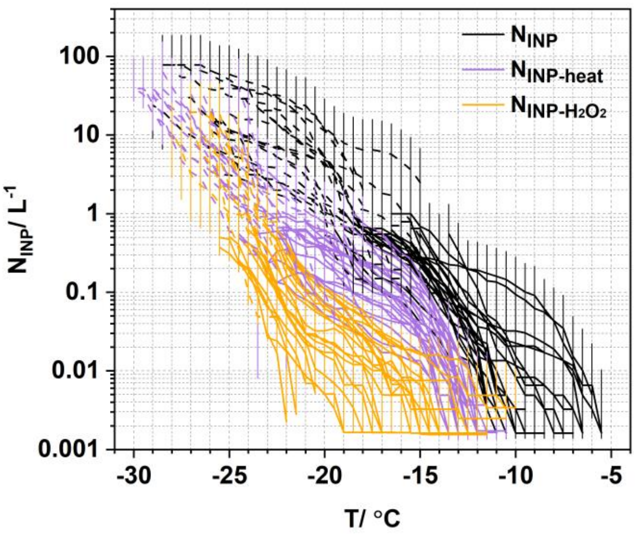

Figure R10. The N_{INP}, $N_{INP\text{-}heat}$, and $N_{INP\text{-}H2O2}$ as function of temperature. The solid line and dotted line show the sample measurement result by immersed in 5 mL MilliQ water and diluted the sample 30-120 times, respectively. The original N_{INP} is marked by black dots, $N_{INP\text{-}heat}$ is marked by purple dots, and $N_{INP\text{-}H2O2}$ is marked by pink dots, with 20% error bars indicating the 95% confidence intervals.

66. Figure 4a: Figure missing.

Response: Thanks, we added this figure in revised manuscript, as shown in Figure R11.

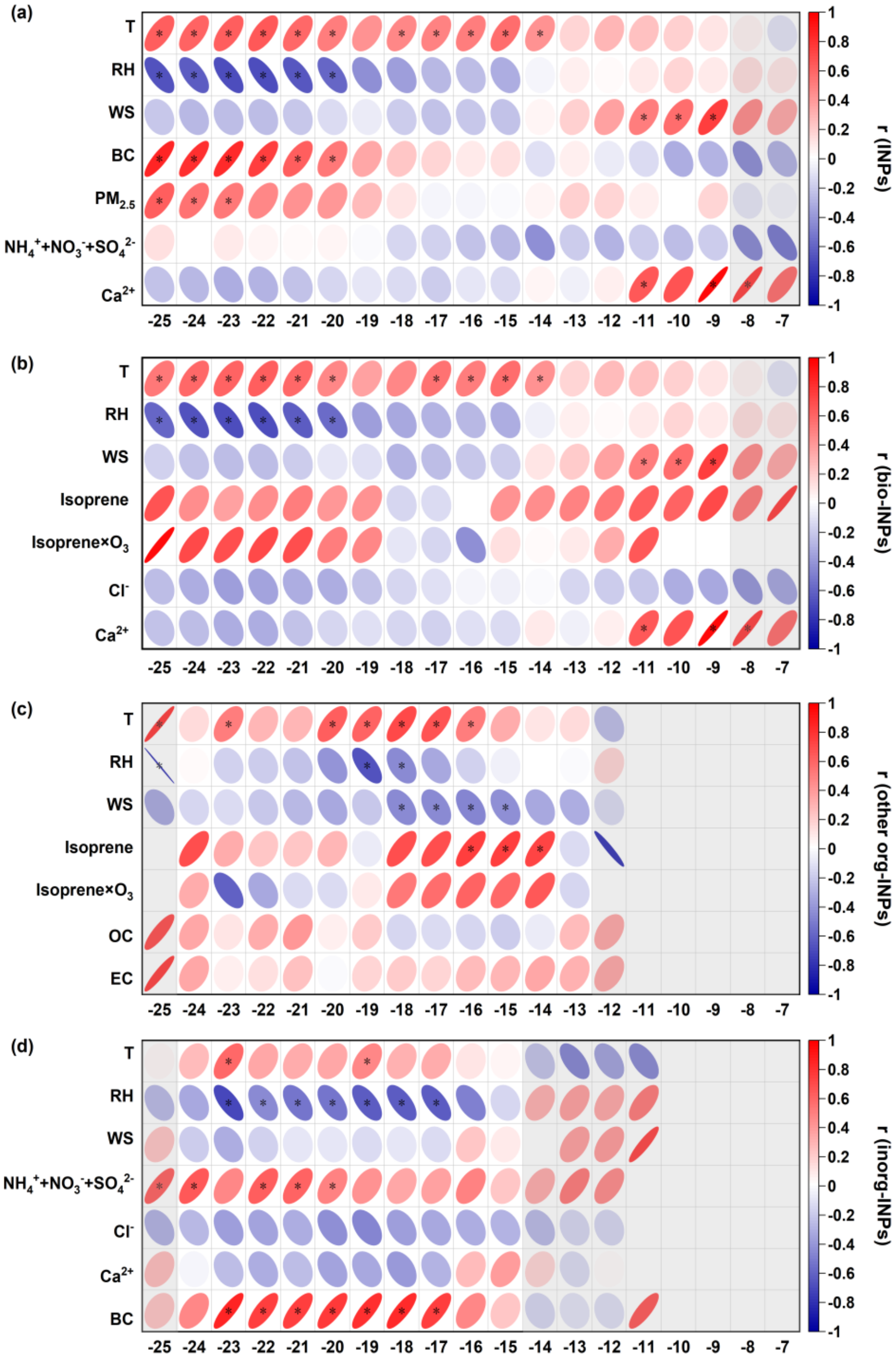

Figure R11. Correlation analysis between (a) N_{INP}, (b) $N_{INP\text{-}bio}$, (c) $N_{INP\text{-}other\ org}$, (d) $N_{INP\text{-}inorg}$, and meteorological parameters, chemical compositions, as functions of temperature. The r denotes the Pearson correlation coefficients. The asterisk indicates $p < 0.05$, while the shades indicate that the number of data points is less than half of all samples at each temperature.

67. Figure 4: As all other figures temperature increases to the right, the x axis of Figure 4 could be flipped for consistency. Which correlation coefficient was used? Specify in caption.

Response: We adjusted the x axis in Figure 4, as illustrated in Figure R11.

68. Figure 5: What is the red star? Is there a specific reason for the y-axis not being logarithmic in contrast to the other plots of INP concentrations?

Response: The red star represents the city of Beijing, which has been removed from the figure. The logarithmic axe is not used because INP concentration is shown at specific freezing temperature, where the range of values is not broad.

Reference:

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