Public justification (visible to the public if the article is accepted and published):
I thank the authors for addressing most of the comments and suggestions provided by Reviewers #1 and #3; however, the two additional reviewers are concerned about the readability of the manuscript. Additionally, Reviewer #4 has several Major, Specific, and Technical comments and I also added a list of Minor/Technical comments. Please improve the grammar of entire manuscript and properly address each comment listed below.

Response: 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.

Reviewer #2:
Please carefully go through the manuscript to meet the standard of ACP.

Response: Thanks for the comments. We carefully go through the manuscript and make the necessary revisions to ensure that it meets the standards of ACP.

Reviewer #4:
Review of "Measurement report: Atmospheric Ice Nuclei at Changbai Mountain (2623 m a.s.l.) in Northeastern Asia" by Sun et al.
The paper reports on the outcome of a one-month field campaign conducted at the Tianchi site on Mt. Changbai during the summer of 2021. Filter samples collected during 10 days at the end of the campaign are used to measure the concentration of immersion freezing INPs and additional heat treatment and H2O2 degradation of the sample are used to infer the contribution of biological INP to the INP concentration. In addition, the INP concentration data is correlated to axillary meteorological data, bulk aerosol composition and trace gas concentration, and back trajectories are used to find potential source regions of INP active in a certain temperature regime. The authors find that biological INP contribute the majority to the INP concentration and stress the importance of soil dust as a regional source as well as long range transport of biological marine INP and biological INP from vegetation.
While the conducted measurements and analysis are state of the art, the interpretation is mostly speculative and not explained clear enough. Instead of deducing conclusions from signals in the current data the reader is pointed to literature sources to back up interpretations. An additional shortcoming is the very limited number of samples collected, making it more of a preliminary study hinting at several interesting aspects on the sources of INPs in Northeast Asia that deserve deeper investigation. The authors have given insightful replies to some of the comments in the previous round of review but have not managed to implement all necessary clarifications and improvements into the manuscript. On some occasions the changes the authors mention in the reply have not been transferred to the revised manuscript, for example reply to question 5 of Referee #3.
Because the manuscript has not been substantially improved in reply to the previous round of review and the many additional comments below, I recommend major revision before the

manuscript can be considered for publication. I also recommend a thorough language check by a native English speaker to avoid misunderstandings of the scientific content due to poor phrasing.

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

Major comments:

The surrounding of Tianchi station next to Tianchi Lake, inside a crater could have a major impact on the measured NINP. Provide an explain in the manuscript how the contribution of long-distant transport can be clearly distinguished from local or regional sources of INP.

Response: Thanks for the comment. Changbai Mountain is a dormant volcano, unaffected by local volcanic eruptions. Human activities exhibit minimal impact, with no nearby industrial facilities, and a significant reduction in tourism activities during our sampling period. Therefore, our sampling site is an ideal site for studying the regional background atmosphere of Northeast Asia, as extensively elaborated in the section 2.1. In the regional background atmosphere, the impact of long-range transport on aerosols and INPs were determined using HYSPLIT model.

The influence of fog and precipitation at the sampling site on the measured NINP should be analysed in more detail. In addition, the influence of precipitation along trajectories should be considered in the CWT analysis. For INPs to form ice in mixed-phase clouds they need to act as cloud condensation nuclei as well. Cloud formation and precipitation along trajectories should therefore wash them out preventing their long-range transport.

Response: We distinguished three different meteorological conditions, i.e., sunny days, foggy days, and rainy days, and conducted a comparative analysis of INPs spectra (see Figure R1). Within the HTR, no significant difference was observed among the three weather conditions. In the LTR, the INP concentrations on rainy days were slightly lower than those on sunny and foggy days. These have been added in the revision.

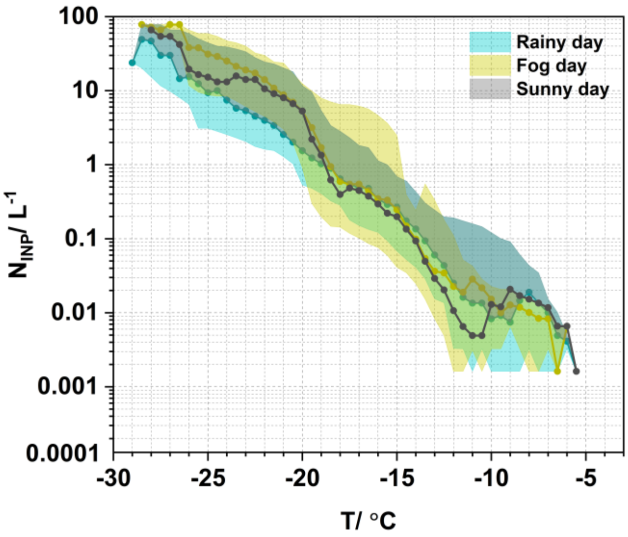

Figure R1. N_{INP} for sunny, foggy, and rainy day as functions of temperature. Each point represents the median value and the shadow area represents the maximum and minimum value.

The CWT analysis does not account for the impacts of wet deposition, including cloud droplets and precipitation along the trajectories. We acknowledge this limitation in this study, and added this in section 2.5.

For non-precipitation clouds, particulate matter with a dry diameter in the range of 80 to 300 nm is deemed to exhibit a more efficient activation as cloud condensation nuclei (CCN) (Ma et al., 2016). For particles with diameters exceeding 1 μm, they are less likely to undergo activation in a cloud due to their large activation radius (> 10 μm) (Grabowski et al., 2022). In contrast, particulate matter with the potential to serve as ice nuclei typically exhibits larger diameters, generally surpassing 500 nm, owing to the presence of a broader spectrum of nucleation-active sites (Demott et al., 2010). This distinction in particle radius underscores the variability in their ability to active as either CCN or IN.

As pointed out in the previous round of reviews, the appropriateness of the PBL height from ERA5 for the Tianchi station site is questionable and needs further confirmation. The authors acknowledged in their response, that the GDAS topography in the region is off by over 900m. Also, complex effects from the mountain and its crater on the formation of the boundary layer cannot be neglected. Are additional measurements for example from balloon soundings available to provide evidence that the PBL height from ERA5 are representative for the location?

Response: We apologize that the balloon-derived PBL data is unavailable around our sampling site. Instead, we conducted a comparison of PBL heights between ERA5 and GDAS products, respectively. As shown in Figure R2, the two datasets showed high consistency. This further demonstrates the robustness and reliability of PBL heights in both products. Hence, employing ERA5 PBL data in our analysis is deemed reasonable.

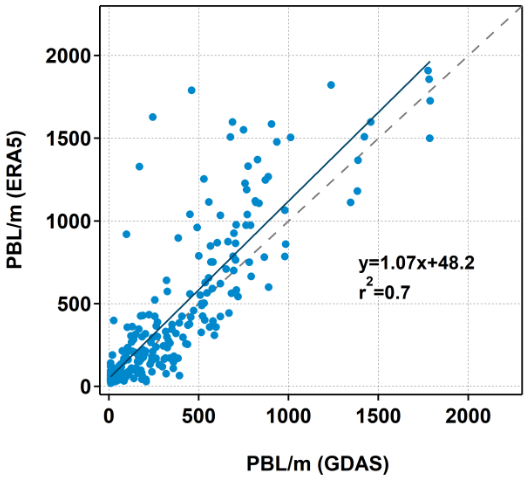

Figure R2. The relationship of PBL data obtained from ERA5 and GDAS, from Aug. 10 to 21, 2021, at our sampling site.

Specific comments:

1) Line 21: explain why correlation to windspeed, Ca^{2+} and isoprene suggests bio aerosol is attached to soil dust and act as INP.

Response: Thanks for the comment. In this study, the origin of Ca^{2+} was considered to be mainly of natural sources due to calcareous nature of the soil. The presence of isoprene indicates a biological source. Previous studies have reported that biological materials may attach to or mix with dust particles and promote INPs formation. Therefore, we speculate that the higher windspeed may have facilitated the exposure of the local soil dust and bioaerosol containing bio-INPs to the air. We have modified the statements in the abstract and section 3.3.

2) Line 22ff: Provide an explanation why PBL height could be positively correlated to NINP. The opposite could be expected because of dilution of aerosol concentration with increasing PBL height. Explain based on what data it is found that valley breeze influences NINP.

Response: Thanks for the comment. Our sampling site is situated atop Changbai Mountain, at an elevation of 2623 m above sea level. The impact of dilution within the boundary layer is negligible at this location, as it consistently above the PBL for the majority of the time. According to Ketterer et al. (2014), the upslope valley wind could increase the altitude of the PBL height locally, and if strong enough, trigger the vertical exchange of PBL air into the free troposphere. In our investigation, the positive correlation between the PBL and INPs may be elucidated by the presence of a valley breeze.

3) Line 33ff: inappropriate citations. Koop et al., 2000, Murray et al., 2010, Cziczo et al., 2013 report on homogeneous ice nucleation and ice nucleation under cirrus cloud conditions *not immersion freezing. For example, Murray et al., 2012 would be a better reference.*

Response: Thanks. We have replaced the references accordingly in the revision.

4) Line 37ff: By definition, mixed-phase clouds contain droplets and therefore only exist at water saturated conditions. Water saturation is not a prerequisite for ice formation. Reformulate.

Response: Thanks for the comment. We have revised the statement as follows: "Recent studies have concluded that water saturation is a prerequisite for ice formation in low- and mid-level clouds, and that contact and immersion freezing are the most primary pathways for ice formation (Sassen and Khvorostyanov, 2008; Phillips et al., 2007; Murray et al., 2012)."

5) Line 43ff: Not corrected as mentioned in response to Referee#3 question 5. Implement correction.

Response: Thanks for the comment. We have revised the statement as follows: "Biological aerosols from microbial and proteinaceous origin, containing ice nucleation active protein, demonstrate significant efficiency as INPs at temperatures above −15 °C (Phelps et al., 1986; Petters and Wright, 2015; Murray et al., 2012; Kunert et al., 2019; Huang et al., 2021)."

6) Line 45: Can lichen be considered biological aerosol? As I understand they are a symbiotic organism attached to surfaces.

Response: Lichens are recognized as extraordinary biological ice nucleators, and the ice nucleation activity is primarily resided in the mycobiont, which is the proteinaceous biological INPs. We have revised the statement and added reference as follows: "For example, biological components in lichens can induce freezing above −10 °C (Kieft, 1988; Moffett et al., 2015), and certain bacterial organisms such as Pseudomonas syringae can facilitate droplet freezing even at extremely high temperatures (above −2 °C) (Maki et al., 1974)."

7) Line 48: Double check if it is correct that pollen is non-proteinaceous. I find they contain about 30% protein.

Response: Although pollen contains proteins, its non-proteinaceous compounds, such as polysaccharides, that induce ice formation. We rewrote this sentence to make it more precisely: "The other biological aerosols with non-proteinaceous compounds act as ice nucleation catalysts, such as pollen, cellulose, and other macromolecular organic particles, can also induce ice formation through heat-resistant polysaccharides on their surfaces, but at lower temperatures than proteinaceous biological particles (Knopf et al., 2010; Pummer et al., 2012)."

8) Line 51: add a citation for the activity temperature of mineral dust and sea spray.

Response: Thanks. We have cited the references, i.e., Alpert et al. (2022); Atkinson et al. (2013); Ladino et al. (2019), in the revision.

9) Line 55: inappropriate citations: references are for deposition ice nucleation, replace with references relevant for immersion freezing.

Response: Thanks. We have revised the citations in the revision.

10) Line 59-62: Provide references for "numerous studies", "spatial distribution heterogeneity", and the altitude of mixed-phase clouds.

Response: Thanks. We have cited the references in the revision.

11) Line 69f: INP sources are not in the atmosphere but on the surface. Reformulate.

Response: To avoid ambiguity, we have revised as "This decline in INPs could exceed 60% per kilometer during the cold season (from 1631 m a.s.l. to 2693 m a.s.l.) (Wieder et al., 2022), which was attributed to the scarcity of effective INPs sources in high-altitudes".

12) Line 74ff: JFJ station is not a good example for having high vegetation coverage. It is surrounded by bare rock and ice. Reformulate.

Response: In the revised manuscript, we have revised this sentence to make it more accurate: "In some mountainous areas, biogenic aerosols can act as the most abundant type of INPs."

13) Line 83f: Clarify what is meant by "impact of bio INP on cloud droplets and their contribution to formation of precipitation".

Response: We have revised this sentence to make it more accurate: "Because the number of rainwater samples was limited, further research is necessary to explore the impact of biological INPs on ice formation and their contribution to the formation of precipitation."

14) Line 91: Explain what boundaries are meant by "transboundary transport of air mass".

Response: The term "boundaries" refers specifically to national boundaries. We have added this information as follows: "Given the high-altitude of Changbai Mountain, it is an ideal location to capture the characteristics of the regional atmospheric background and transboundary transport of air masses from nearby countries."

15) Line 98: provide a reference for pollution transport to the Arctic from this region.

Response: Thanks. We have cited the references in the revision.

16) Line 112: Extend the discussion on weather conditions. What is meant by humid weather?

Was it raining, or was there fog? Fog and rain can have an impact on the sampling as well as on the transport of INP. Potential evidence for this impact can be seen from the anticorrelation of NINP in the LTR with RH in Fig.4.

Response: We distinguished three different meteorological conditions, i.e., sunny days, foggy days, and rainy days, and conducted a comparative analysis of INPs spectra (see Figure R1). Within the HTR, no significant difference was observed among the three weather conditions. In the LTR, the INP concentrations on rainy days were slightly lower than those on sunny and foggy days. These have been added in the revision.

17) Line 116f: Elaborate on the potential impact of the surrounding (lake, dense vegetation) on NINP and other variables, for example isoprene concentrations.

Response: We have added the influence of the surrounding in section 3.1, read as follows: "In this study, local sources such as vegetation and the lake may impact INPs concentration, with biogenic emissions potentially exhibiting variations between daytime and nighttime."

18) Line 121ff: Provide characteristics on the sampler inlet cut-off. Is it a total inlet? As RH was high during the campaign and often at 100%, was there fog or precipitation? Does the sampler collect cloud droplets as well?

Response: We use a TSP sampler with a 100 μm cut-point. The sampling head design effectively prevents the collection of rainwater during rainy days. But fog and cloud droplets remain unavoidable. It is a common challenge encountered with all types of samplers.

19) Line 123: Table S1 lists 25 sample intervals. Was the filter not changes for the two intervals during nighttime on the 11.8.? If not, maybe the 6min gap in sampling can be neglected and doesn't need to be listed. Otherwise specify what happened.

Response: On August 11, a momentary power outage led to a temporary interruption in instrument measurements lasting approximately 6 minutes. This information has been added in Table S1.

20) Line 124, Tab.S1: The sample duration in Tab.S1 do not agree with the time difference between start to end time. Where does the duration come from? Does the sampler turn off if the pressure-drop over the filter is too high? The sample volume has been calculated using the set flow rate and start to end time. Does the sampler not provide a more precise measurement of the sample volume? If possible, use measured and not calculated sample volume to calculate NINP.

Response: Thank you for the comment. The variation between the instrument-recorded volume and the calculated volume is within 2%. We adopted the recorded volume in Table S1 accordingly. Throughout the sampling period, there were no instances of the sampler shutting down due to excessive pressure.

21) Line 126: provide model number for the PM2.5 sampler.

Response: Thank you for the comment. We added the instrument model in the revision.

22) Explain how auxiliary data was averaged for the correlation analysis with NINP.

Response: The calculate method has been added in the revision as follows: “The parameters mentioned above were analyzed by determining the average value over the corresponding INPs sampling period.”

23) Line 139: is the enclosed droplet chamber part of the LTS120 cold stage?

Response: Yes, the cold stage is located inside the droplet chamber. We revised the statement as follows: “GIGINA is a cold-stage-based ice nucleation array that consists of an enclosed droplet chamber with a commercial cold stage inside (LTS120, Linkam, Epsom Downs, UK), an external refrigerated water circulator (VIVO RT4, Julabo, Seelbach, Germany), and a charge-coupled device (CCD) camera (DMK33G274, The Imaging Source, Bremen, Germany).”

24) Line 147ff: Clarify if sample droplets rest inside the oil or if the oil is between the cold stage and the glass slide? Are droplets in contact with the aluminium spacer? Revise the step-by-step description in the manuscript.

Response: The silicone oil was between the hydrophobic glass slide and cold stage, and the droplets cannot contact with the aluminium spacer. To avoid confusion, we have revised as “First, a hydrophobic glass slide was placed on a cold stage and covered with silicone oil between them to achieve good thermal contact. Second, a round aluminum spacer with 90 round compartments was placed on the glass slide, and the particle suspension was sequentially pipetted into the center region of each compartment."

25) Line 148: “filled” might be the wrong word to describe the procedure. Do you mean covered?

Response: Yes, we have replaced this word by “covered” in the revision.

26) Line 160 and Eq.2: As Equation 2 is irrelevant for the rest of the manuscript I suggest deleting it. If you want to keep it in, define K(T) in the text and explain what is meant by “cumulative concentration of each droplet above K(T)”. Add how Vair is calculated from the sample volume, volume of washing water and droplet volume. Add the equation how the background signal is subtracted.

Response: We agree and delete the Equation of K(T). The equation for calculating N_{INP} has been revised to include the background signal and is modified as follows: “The cumulative number concentration of INPs (N_{INP}) per unit volume of sampled air were calculated following the method of Vali (1971, 2015):

$$N_{INP}(T) = -\frac{ln[1-f_{ice,\ sample}(T)]-ln[1-f_{ice,\ blank}(T)]}{V_{air}}(L^{-1}\ air), \quad (2)$$

where V_{air} is the total volume of sampled air per droplet converted to standard conditions (0 °C and 1013 hPa). The $f_{ice,\ sample}$ and $f_{ice,\ blank}$ are the measured frozen fractions for the filter samples and the field blanks, respectively. The calculation for Vair entails multiplying the droplet volume (1 μl) by the sample volume and then dividing by the volume of wash water.

27) Line 183-184: As pointed out by Referee#1 comment 22. in the previous round of reviews, the authors should elaborate in the text why the ERA5 PBL height data is applicable specific to their analysis.

Response: The PBL data obtained from ERA5 has been widely used in numerous studies, such as Le et al. (2020), Tornow et al. (2021), and Slattberg et al. (2022). In this analysis, we compared PBL heights obtained from ERA5 and the Global Data Assimilation System (GDAS) meteorological fields, and find a high consistency between the two datasets, as illustrated in Figure R2. This further demonstrates the robustness and reliability of PBL data of the two products. Therefore, it is reasonable for us to use ERA5 PBL data.

28) Figure 2a: add dilution data to this figure to show that measurements were not affected by the water background instead of having an additional Figure S2.

Response: We incorporated Figure 2a into the Figure S2 in the Supporting Information, as illustrated in Figure R3.

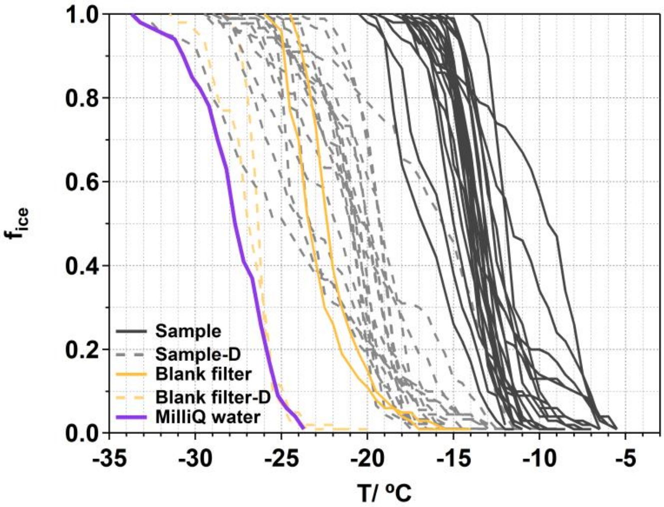

Figure R3. Frozen fractions (f_{ice}) of collected samples measured by GIGINA as functions of temperature is shown by the black curves, and presented together with blank filters (orange curves) and MilliQ water (purple curves) as background signals. The dashed line presented the diluted sample suspension.

29) Line 209: As pointed out above, add equation to Sec.2.3. how the background was *subtracted and refer to the equation here.*

Response: Please see our response to Comment 26.

30) Line 241f: You could cite Kanji et al.,2020, here. Correct sentence structure. There are significant correlations between BC and NINP below -20°C shown in Fig.4 not supporting this statement. The discrepancy to literature is worth adding a discussion of these data here or line 300.

Response: Thanks. We have cited this reference in the revised manuscript.

We have added a brief discussion in section 3.3. "Previous studies have suggested that carbonaceous particles may not act as efficient INPs in the immersion mode or could decrease ice nucleation activity in polluted urban environments, which is attribute to the formation of organic coatings (Kanji et al., 2020; Schill et al., 2020; Nichman et al., 2019; Hammer et al., 2018). However, our observations revealed a positive correlation between BC and both N_{INP} and $N_{INP\text{-}inorg}$ in the LTR. The discrepancy may be attributed to the different sources and aging degrees of BC, which remains unclear."

31) Line 271f: The fraction of bio-INP is affected by the concentration of these INP and much less by their moderate or high ice activity. Reformulate.

Response: We agree and revised as follow: "Interestingly, when the temperature decreased from -16.5 °C to -21.5 °C, the median of $F_{INP\text{-}bio}$ increased from 0.8 to 0.9, indicating the high concentration of bio-INPs in the LTR."

32) Line 287f: Specify where these previous studies were conducted and provide references.

Response: We have removed this sentence in the revision.

33) Line 295f, 309f: Please justify speculations by explaining the deductive chain of logical steps that lead to them. It is not obvious to me here. Is Ca2+ a proxy for soil dust? Are there sources of soil dust downwind of the sampling site? Considering the weather situation, wouldn't soil be wet and therefore not prone to wind erosion? Isn't soil covered by vegetation in this season that prevents erosion? Clarify in the text. Is it realistic that particle concentrations below 0.1 L-1 (NINP in the -8°C to -11°C range) influence the measured Ca2+ concentration?

Response: The vegetation types across Changbai Mountain exhibits obvious variations from the foothills to its summit. The transition encompasses tall temperate broad-leaved forests at the mountain's foothills, evolving into Changbai Pinus bungeana (coniferous forests), and further to the low creeping birch (shrub), alpine meadows, mossy lichens, and exposed rocks (Xu et al., 2004). Vegetated soil acts as a mitigating factor against erosion. However, the prevalence of low-lying vegetation around Tianchi Lake may facilitate soil erosion during periods of high wind speed (up to 25.7 m/s in our observation). Numerous studies show that the origin of Ca^{2+} is primarily attributed to natural sources due to the calcareous nature of the soil (Al-Momani, 2003; Al-Khashman, 2005). The Ca^{2+} was extracted from $PM_{2.5}$ samples and the concentration was analyzed using an ion chromatograph, which is unaffected by INP concentrations.

34) Line 298f: Provide an explanation for the correlation of NINP to ambient temperature and RH.

Response: Please see our response to Comment 16.

35) Line 313f: Explain why a correlation with WS and ambient temperature indicates local sources.

Response: We apologize for the ambiguity and have removed the statement in the revision.

36) Line 316f, Line 396: Oxidation products of isoprene should be water soluble and not contribute to immersion freezing. Correlation does not imply direct causation.

Response: Thanks. We have revised the statement as "Although the oxidation products of isoprene are expected to be water-soluble and unable to induce immersion freezing, the observed positive correlation suggests a potential role of secondary organic compounds associate with vegetation or other biogenic sources in ice nucleation."

37) Line 318: It could be expected that SOA activate as CCN when RH>100%. Did the sampler also collect cloud droplets? An anticorrelation to RH could provide a hint if the INP are also CCN. For INP to generate ice in mixed-phase clouds CCN activation is a prerequisite. I encourage a reanalysis of the data considering the weather situation at the time of sampling.

Response: Particulate matter with a dry diameter in the range of 80 to 300 nm is deemed to exhibit a more efficient activation as CCN (Ma et al., 2016). For particles with diameters exceeding 1 μm, they are less likely to undergo activation in a cloud due to their large activation radius (> 10 μm) (Grabowski et al., 2022). In contrast, particulate matter with the potential to serve as ice nuclei typically exhibits larger diameters, generally surpassing 500 nm, owing to the presence of a broader spectrum of nucleation-active sites (Demott et al., 2010). This distinction in particle radius underscores the variability in their ability to active as either CCN or IN. In addition, most ice nuclei have appreciable fractions of soluble materials, so the same aerosol particles may serve both as IN and CCN. But the immersion freezing mode is the dominant freezing mechanism for insoluble particles in mixed-phase clouds. Therefore, we think the INP cannot act as CCN in the immersion freezing mode.

38) Line 323: The NINP was measured in this study not the IN activity. Reformulate.

Response: We have revised the statement as "Note that $PM_{2.5}$ chemical composition was used in this study, which may lead to uncertainties in the interpretation of the INPs in TSP."

39) Line 338f: Provide a reasonable explanation how PBL height can be positively correlated to NINP. The opposite could be expected due to dilution of air with increasing PBL height. Total particle concentration is usually anticorrelated to PBL height.

Response: Our sampling site is situated atop Changbai Mountain, at an elevation of 2623 m above sea level. The impact of dilution within the boundary layer is negligible at this location, as it consistently above the PBL for the majority of the time. According to Ketterer et al. (2014), the upslope valley wind could increase the altitude of the PBL height locally, and if strong enough, trigger the vertical exchange of PBL air into the free troposphere. In our investigation, the positive correlation between the PBL and INPs may be elucidated by the presence of a valley breeze.

40) Line 341f: This is not supported by the data. If valley breeze transported INPs to the station during daytime, why is there not a difference between night and day NINP?

Response: Upon examining the diurnal variation of N_{INP} on individual days, notable differences between the daytime and nighttime can be observed on certain days, such as August 18 and 21, 2021, as shown in Figure R4. However, there was no statistical difference when considering the entire sampling period. The absence of diurnal variations may be attributed to the limited dataset size and low sampling frequency, as explained in section 3.1.

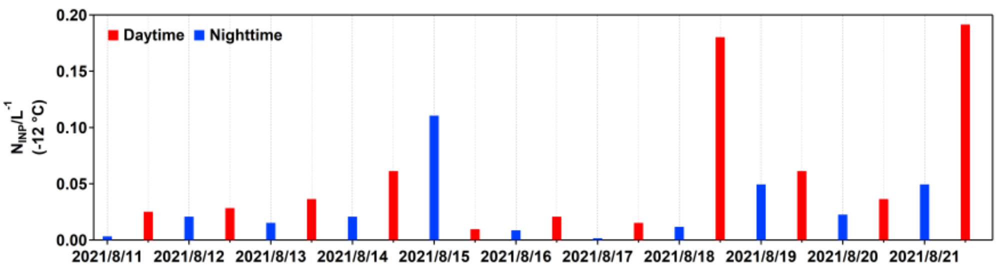

Figure R4. Time series of N_{INP} at -12 °C measured during the campaign.

41) Line 350f: Clarify how NINP is related to phytoplankton bloom and growth. The Japanese Sea experiences a phytoplankton bloom twice a year, one in spring and one in fall (Wang et al., 2022). Clarify the relevance for the current observations.

Response: We identified elevated concentrations of bio-INPs originating from the Japan Sea and inferred that marine biogenic organic materials contribute to INPs. It is worth noting that large amounts of organic matter are present in the sea surface microlayer, regardless of whether there are phytoplankton blooms or not.

42) Line 352-355, Figure 5: It is surprising that two distinct origins for HTR and LTR bio INPs are found, as it could be assumed that for cumulative NINP the occurrence of high NINP *at -10.5°C is correlated to high NINP at -20°C. Is this not the case? Could there be an artifact from the measuring range? For example, how were samples with a frozen fraction =1 at temperatures above -20°C included in the CWT analysis in the LTR? Did you use the data of their dilutions? The NINP colour scale indicates that dilutions were not included.*

Response: We clarify that Figure 5d includes the dilution data. The CWT analysis illustrates weight concentration of the target species. In order to reduce the uncertainty caused by the smaller number of trajectories through a grid, the CWT value is multiplied by the weight function as well. Specifically, Korea is an important source of biological INP in the HTR; however, in the LTR, its contribution is reduced relative to other sources. Consequently, its relative contribution is too small to be displayed, considering the calculation of the weight concentration in the CWT.

43) Line 370ff: Explain how it can be concluded that long-range bio-INP were less prominent in your measurements? It seems contradictory to say there was no qualitative or quantitative analysis of bio INP and then state that they were less prominent from long-range transport.

Response: Thanks for the comment. To avoid confusion, this sentence has been removed in the revision.

44) Line 376: Clarify the interpretation between PBL height and local sources or long-range transport. Are you inferring that long-range transport contribution to NINP occurs exclusively by night and as soon as the boundary layer forms exclusively local sources contribute to NINP? This seems a critical assumption for your analysis and should be discussed in more detail and supported by stronger evidence.

Response: We apologize for the ambiguity regarding this aspect. The influence of PBL on INPs can be elucidated by the presence of valley breezes, as detailed in our response to comment 2. This phenomenon was recognized as the mountain–valley breeze circulations in the vicinity of Changbai mountain. The CWT calculation encompassed the entire day, including both daytime and nighttime. The results from CWT analysis revealed the influence of long-range transport.

45) Line 381ff: There was no diurnal cycle of NINP observed in this study. Explain how conclusions can still be drawn about a diurnal cycle.

Response: Thanks for the comment. We have revised this statement as follows: "In summary, our findings suggest that valley breezes and the long-distance transport of air mass from the Japan Sea influence the abundance of INPs at Changbai Mountain."

46) Line 385f: The methods applied in this study do not allow to explore properties of INPs. Only their abundance was measured. Reformulate.

Response: Thanks for the comment. We have revised this statement as follows:

"Measurements of INPs were carried out at the Changbai Mountain in northeastern Asia to explore the abundance and source of INPs in the immersion freezing mode."

47) Line 393: An increase in bio NINP indicates that the concentration of bio-INP increased, not the activity. The type of measurements does not provide information if, for example, 1% or 100% of a certain INP type was active at a certain temperature. Reformulate.

Response: Thanks for the comment. We have revised this statement as follows: "Notably, a turning point occurred at −16.5 °C, where FINP-bio increased from 0.8 to 0.9 as the temperature decreased from −16.5 °C to −21.5 °C, indicating an enrichment of active bio-INPs in the low-temperature region (LTR, freezing temperature below T_{50}, -17.0 °C ~ -29.0 °C)."

48) Line 398ff: Please resolve the apparent contradiction between local soil dust sources and CWT pointing to long range transport by clarifying how long-distance sources can be disentangled from local sources. In addition, would the CWT analysis reveal different patterns for the nighttime NINP measurements, or at -10°C and -20°C? Would the correlation analysis, for example with Ca2+ differ if done separately for daytime and nighttime samples?

Response: Thanks for the comment. Local soil dust sources and long-range transport both contribute to the INPs concentrations. However, we cannot completely distinguish between the two aspects based on the field investigation measurements; instead, we analyze them separately. We don't think the two aspects as contradictory.

We examined the relationship between N_{INP} and both Ca^{2+} and WS separately for daytime and nighttime samples, and found the more pronounced correlations during the daytime (Figure R5). This further substantiates our findings, demonstrating the positive correlations between the PBL and INPs during the daytime, which is associated with the influence of valley breeze. These have been added in the revision.

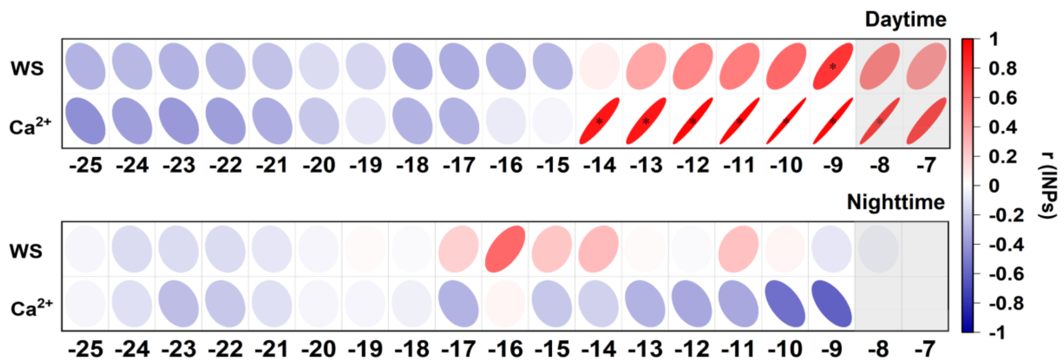

Figure R5. Correlation analysis between N_{INP} with wind speed (WS) and Ca^{2+} as functions of temperature. The r denotes the Pearson correlation coefficients. The asterisk indicates $p < 0.05$, while the shades indicate that the number of data points is less than half of samples at each temperature.

49) Figure S5: replace with Fig. R3 from the Author's Response and include the reply to *question 22 from Referee #1 explaining how airmasses approach the measurement site and why the trajectory endpoint was chosen at 967m a.g.l.*

Response: Thanks. We have replaced the Figure S5 by Figure R3 and provided additional explanations about the influence of the valley breezes in the revised manuscript. The trajectory ending height of 967 m a.g.l. was selected because the terrain height of Changbai Mountain was approximately 1656 m in the GDAS data. These have been added in the revision.

50) Figure S6: Specify if only daytime or all samples were used to create this figure.

Response: All samples were included in the CWT analysis. We have revised the figure caption to "The concentration-weighted trajectory (CWT) analysis for the distribution of NINP-other org (a, b) and $N_{INP\text{-}inorg}$ (c, d) at -15 and -21 °C during the measurement."

Technical corrections:

1) misspelled citations: DeMott, O'Sullivan, McCluskey, in several instances.

Response: Thanks. We checked and removed inappropriate citations in the revised manuscript.

2) there is no "the" needed before Switzerland. Also, in Figure 2.b) remove "The" in front of Weissfluhjoch and Jungfraujoch.

Response: Thanks. We revised them in the revised manuscript.

3) Line 110: Maybe use standard error of the mean instead of standard deviation to avoid range of RH (92.4%+11.8%=104,2%) to extend beyond the range of measurement. Also, line 309: 0.5ugm-3 – 1.0ugm-3 = - 0.5ugm-3 is unphysical.

Response: Thanks. In the revision, we have used the standard error of the mean. The average RH stands at 92.4 ± 0.4%, and the average concentration of Ca^{2+} is 0.5 ± 0.2 μg m^{-3}. Further adjustments to additional values have been made and elucidated in the revision.

4) Line 166-168: It is unclear what is explained here. Reformulate.

Response: We have revised this statement as follows: "The rarity of INPs in the atmosphere leads to their low concentration in the suspension. Because the suspension used in the measurement contained a limited number of droplets, we need to consider the resulting uncertainty caused by statistical errors. Therefore, we calculated the confidence intervals of the apparatus for f_{ice} according to the method of Gong et al. (2022) and Agresti and Coull (1998) to address uncertainty associated with the droplet-freezing apparatus".

5) Line 173: Do you mean: ..., which is 1.96 for a 95% confidence interval? Reformulate.

Response: Yes. We have revised the sentence.

6) Line 191ff: Incomprehensible sentence. Reformulate.

Response: Thanks. We have revised the sentence.

7) Line 198: define nij in the text.

Response: We have added the define of n_{ij} in the revised manuscript.

8) Line 213: replace "three" with "up to 5" to be consistent with what is stated in the conclusion.

Response: Thanks. We have revised the sentence.

9) Line 233-235: Incomprehensible sentence. Reformulate.

Response: Thanks. We have revised the sentence.

10) Line 237: Reformulate. ...magnitude at temperatures from -26.0°C to -3.0°C.

Response: We revised the statement as follows: "Gong et al. (2022) measured INPs at the mountain station at Cerro Mirador (622 m a.s.l., Chile), and reported N_{INP} values lower than those in our study by around one order of magnitude at similar freezing temperatures from -26.0 °C to -3.0 °C."

11) Line 288: replace INPs with NINP

Response: Thanks, we revised the statement as "N_{INP}" in the revised manuscript.

12) Line 321: define SNA.

Response: We define the SNA as "sulfate, nitrate, and ammonium" in the revised manuscript.

13) Line 329: replace INPs with NINP.

Response: Thanks, we revised the statement as "N_{INP}" in the revised manuscript.

14) Line 336: Define HLR. Do you mean HTR?

Response: Yes, we corrected it to "HTR" in the revised manuscript.

15) Line 347: "under immersion mode" do you mean "in the immersion mode"?

Response: Yes, we revised the statement as "in the immersion mode" in the revised manuscript.

16) Line 352: "occurred in" do you mean "coming from"?

Response: Yes, we revised the statement as "originating from" in the revised manuscript.

17) Line 363: define "the middle layer".

Response: Thanks, we corrected the "middle layer" to "Mesosphere" in the revised manuscript.

18) When reporting NINP ranges and consecutive the temperature range, I suggest giving the temperature range from high to low temperature if the NINP range is given from low to high, so that the first number in the NINP range corresponds to the first number in the temperature range.

Response: Thanks, we have revised in the revised manuscript.

19) Specify whenever you mean "ambient temperature" to avoid confusion to experimental temperature of IN experiments. In particular: line 299, 313.

Response: Thanks, we revised the statement as "ambient temperature" in the revised manuscript.

References:

Kanji, Z. A., Welti, A., Corbin, J. C., & Mensah, A. A. (2020). Black carbon particles do not matter for immersion mode ice nucleation. Geophysical Research Letters, 46, e2019GL086764. https://doi.org/10.1029/2019GL086764

Wang D, Fang G, Jiang S, Xu Q, Wang G, Wei Z, Wang Y and Xu T (2022). Satellite-detected phytoplankton blooms in the Japan/East Sea during the past two decades: Magnitude and timing. Front. Mar. Sci. 9:1065066. doi: 10.3389/fmars.2022.1065066

Editor:

L14: Replace "concentrations" with "concentration"

Response: Thanks, we have revised the manuscript accordingly.

L20-L21: Replace "not significant correlate with isoprene" with "not significant correlation *was found with isoprene"*

Response: Thanks, we revised the statement as "not significant correlation was found with isoprene" in the revised manuscript.

L42: Replace "al., 2009), volcanic ashes" with "al., 2009), and volcanic ashes"

Response: Thanks, we have revised the manuscript accordingly.

L43: I think "such as" should be "from"

Response: Thanks, we have revised the manuscript accordingly.

L47: Delete "For the"

Response: Thanks, we have revised the manuscript accordingly.

L59: I think "atmosphere" should be "atmospheric"

Response: Thanks, we have revised the manuscript accordingly.

L62: What do the authors mean with "INPs can be extended"?

Response: Thanks, we have revised the statement as "there remain uncertainties about how INPs transported to the altitudes of mixed-phase cloud formation (approximately 3–7 km)" in the revised manuscript.

L65: Add a reference after "site".

Response: We have added the reference "(Wieder et al., 2022)" in the revised manuscript.

L70: Replace "high-altitude atmosphere" with "high-altitudes"

Response: Thanks, we have revised it accordingly.

L71: Delete "In contrast"

Response: Thanks, we have removed it in the revised manuscript.

L81: Replace "mountains" with "high-altitude sites"

Response: Thanks, we have revised the manuscript accordingly.

L81: Replace "m a.s.l.), Wuling" with "m a.s.l.), and Wuling"

Response: Thanks, we have revised it accordingly.

L82: I suggest replacing "Dinghu Mountain (1000 m a.s.l.), and found that the initiated freezing temperature" with "Dinghu Mountain (1000 m a.s.l.). The authors found that the onset freezing temperature was..."

Response: Thanks, we have revised it accordingly.

L84: "cloud droplets"? Do the authors mean "ice formation"?

Response: Thanks, we revised the statement as "ice formation" in the revised manuscript.

L89: Add a reference after "yields"

Response: Thanks. We have cited a reference in the revision.

L89 and along the text: "high altitude". Either use "high altitude" or "high-altitude"

Response: Thanks, we revised the statement as "high-altitude" in the revised manuscript.

L91: Replace "air mass" with "air masses"

Response: Thanks, we have revised it accordingly.

L93: Replace "pathways of INPs" with "pathways on the INP population"

Response: Thanks, we have revised it accordingly.

L93: Replace "understanding of INPs sources" with "understanding of their sources"

Response: Thanks, we have revised the manuscript accordingly.

L94: I think "clouds" should be "mixed-phase clouds"

Response: Thanks, we revised the statement as "mixed-phase clouds" in the revised manuscript.

L98: Add a reference after "Arctic"

Response: Thanks. We have cited the references in the revision.

L110: Replace "field measurements" with "field campaign"

Response: Thanks, we have revised it accordingly.

L113: Should "national nature reserve" be "national natural reserve"?

Response: Thanks, we revised the statement as "national natural reserve" in the revised manuscript.

L114-115: Replace "Due to the emergence of novel coronavirus (COVID-19) cases, strict" with something like "Due to the COVID-19 pandemic, strict"

Response: Thanks, we have revised the manuscript accordingly.

L115: Replace "measures have been implemented" with "measures were implemented"

Response: Thanks, we have revised it accordingly.

L124: Replace "analysis and detailed" with "analysis with the detailed"

Response: Thanks, we have revised it accordingly.

L124: Delete "is" in "information is provided"

Response: Thanks, we have removed it in the revised manuscript.

L133: Replace "frequency increased to every 3 h during" with "frequency was increased from 2h to 3h"

Response: Thanks, we have revised it as: "sampling frequency was increased to every 3 h (5 samples per day)".

L135: "national meteorological station". Please add the model and manufacturer.

Response: The national meteorological station doesn't have model and manufacturer.

L135: Replace "from" with "away from"

Response: Thanks, we have revised it accordingly.

L140: Replace "Germany), a charge-coupled" with "Germany), and a charge-coupled"

Response: Thanks, we have revised it accordingly.

L140-141: "The Imaging Source, 140 Bremen, Germany), a ring LED light, and a computer system" is unclear.

Response: This segment does not influence the comprehension of the apparatus's structure and the execution of its performance. To enhance clarity, we have removed “a ring LED light, and a computer system” in the revised manuscript.

L144: Replace “wash off particles” with “wash off the particles”

Response: Thanks, we have revised it accordingly.

L146: Replace “spectra” with “a spectrum”

Response: Thanks, we have revised it accordingly.

L147: “Figure S3”. This figure cannot be called before Figure S2. Please reorganize the order of the figures to call them sequentially

Response: We changed Figure S3 to Figure S2 in the revised manuscript.

L149: Replace “and the particle suspension” with something like “and the suspension containing the particles”

Response: Thanks, we have revised it accordingly.

L160-161: “the unit volume of sampled air”. This is unclear.

Response: Thanks, we have revised the statement as “The cumulative concentration of each droplet above K (T), and the cumulative number concentration of INPs (N_{INP}) per the unit volume of sampled air were calculated following the method of Vali (1971, 2015):” in the revised manuscript.

L185: Replace “during our sampling” with “during the sampling”

Response: Thanks, we have revised it accordingly.

L203: Delete “A metric was applied to evaluate the freezing of droplets, i.e.,”

Response: Thanks, we have revised it accordingly.

L204: Replace “frozen (T50)” with “frozen (T50) was calculated for each sample”

Response: Thanks, we have revised it accordingly.

L207: “(for which T50 was -17.0 ± 4.1 ℃)." Please double check this. From figure 2 it seems that the average T50 is round -14C with T50 values as warm as -10C

Response: We diluted all samples and extended the freezing temperature below -25°C. The T_{50} was recalculated as 17.0 ± 0.6 °C.

L212: Replace "The freezing of" with "The onset freezing of"

Response: Thanks, we have revised it accordingly.

L214: Should "freezing temperatures above T50" be "freezing temperatures above the average T50"?

Response: Yes, we have added "average" before T_{50} in the revised manuscript.

L225-226: "and the limited dataset size and low sampling frequency may have contributed to the absence of diurnal variations." Is this related to the present study?

Response: Reviewer #1 and #3 suggested that the lack of diurnal variation might be related to sampling intervals and a small number of samples. Therefore, we have provided an explanation of the uncertainty here.

L228: Units are missing.

Response: Thanks, we added the units in the revision.

L237: Should "magnitude during the measured" be "magnitude at similar"

Response: Thanks, we have revised it accordingly.

L240: Replace "nuclei" with "INP"

Response: Thanks, we have revised it accordingly.

L272: Replace "ice nuclei" with "INP"

Response: Thanks, we have revised it accordingly.

L288: Replace "atmospheres" with "environments"

Response: Thanks, we have revised it accordingly.

L293: Replace "with temperature range from -11.0 ℃ to -9.0 ℃." With "at temperatures ranging between -11.0C and -9.0C"

Response: Thanks, we have revised it accordingly.

L296: Replace "within the range of" with "between"

Response: Thanks, we have revised it accordingly.

L299: Replace "with temperature" with "with ambient air temperature"

Response: Thanks, we have revised it accordingly.

L303: Replace "And a significant" with "A significant"

Response: Thanks, we have revised it accordingly.

L303: Replace "was showed" with "was found"

Response: Thanks, we have revised it accordingly.

L303-307: Improve the grammar in this paragraph.

Response: Thanks, we have corrected the grammar in this paragraph.

L308: Replace "with average" with "with an average"

Response: Thanks, we have revised it accordingly.

L309: "metal ions with Ca2+". This is unclear

Response: Thanks, we have deleted the "metal ions with" in the revised manuscript.

L313: Replace "did observe that" with "observed"

Response: Thanks, we have revised it accordingly.

L316: Add a reference after "aerosols"

Response: Thanks, we added a reference, i.e. Carlton et al., 2009 in the revised manuscript.

L322: Replace "role in INPs formation" with "role as INPs"

Response: Thanks, we have revised it accordingly.

L322-323: "2016). Note that PM2.5 chemical composition was used in this study, which may lead to uncertainties in the interpretation of the bulk IN activities". This is unclear

Response: We have revised the statement as follows: "Note that $PM_{2.5}$ chemical composition was used in this study, which may lead to uncertainties in the interpretation of the INPs in TSP."

L334: Replace "significant spanning temperatures from" with "at temperatures ranging between xx and xx."

Response: Thanks, we have revised it accordingly.

L335: Replace "observed when the" with "observed at the"

Response: Thanks, we have revised it accordingly.

L338: Replace "discussed in the following paragraph" with "discussed below"

Response: Thanks, we have revised it accordingly.

L341: Figure S5. This figure cannot be called before S4.

Response: We changed the order of Figure S4 and S5 in the revised manuscript.

L357: "as -2~-5 ℃". Please fix this.

Response: Thanks, we revised it as "higher temperatures, ranging from −2 °C to −5 °C" in the revised manuscript.

L358-359: Replace "with biological aerosols produced there able to reach our sampling site through long-distance transport" with "."

Response: Thanks, we have revised it accordingly.

L365: Should "In global transmission" be "On a global scale"?

Response: Thanks, we revised it as "On a global scale" in the revised manuscript.

L366: Add a reference after "ocean"

Response: Thanks, we added a reference, i.e. Mayol et al. (2017), in the revised manuscript.

L369: What do the authors mean with "aerosol transmission"

Response: We revised the statement as follows: "This process can enable biological aerosol to be transported over greater distances, significantly enhancing the ice nucleation activity of dust."

L369-370: Replace "with the ice nucleation activity of dust significantly enhanced" with "enhancing the ice nucleation activity of dust significantly"

Response: Thanks, we have revised it accordingly.

L371-272: "activity. Although long-range transported bio-INPs were less prominent in our study". What do the authors mean and how is this supported?

Response: Thanks, we have removed the statement.

L375: Replace "However, when" with "However, at"

Response: Thanks, we have revised it accordingly.

L378: Replace "But" with "However"

Response: Thanks, we have revised it accordingly.

L382: Replace "influence the diurnal cycles of INPs" with "influence the diurnal INPs"

Response: Thanks, we have revised it accordingly.

L390-391: Please improve the grammar.

Response: Thanks, we have corrected the grammar.

Figure S1. Why there is not INP data between 2021/7/24 and 2021/8/11?

Response: Thanks for the comments. As shown 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. Therefore, the collection of samples was carried out from August 11th, 2021.

Figure 1: Replace "(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)." With "(b) Map with the three-dimensional shape of the sampling site, which was obtained from Google Earth. (c) The INP sampler (The TH-150D medium flow sampler, Wuhan Tianhong Corporation, China)."

Response: Thanks, we have revised it as "(b) Map with the three-dimensional shape of the sampling site, which was obtained from Google Earth. (c) The INP sampler (The TH-150D medium flow sampler, Wuhan Tianhong Corporation, China)." in the revised manuscript.

Figure 2: Replace "(b) NINP was measured during the daytime and nighttime. The dark gray shaded area represents the upper and lower limits of NINP over the Weissfluhjoch (2693 m a.s.l.) (Wieder et al., 2022). The yellow shaded area represents the atmospheric NINP ranges at Mt. Huang (1840 m a.s.l) (Jiang et al., 2015). The purple square represents the median NINP at -15 ℃ and -10 ℃ in the Jungfraujoch (3580 m a.s.l.) (Conen et al., 2022). And the black" with "(b) NINP measured during the daytime and nighttime. The dark gray shaded area represents the upper and lower limits of NINP over the Weissfluhjoch (2693 m a.s.l.) (Wieder et al., 2022), the yellow shaded area represents the atmospheric NINP ranges at Mt. Huang (1840 m a.s.l) (Jiang et al., 2015), the purple square represents the median NINP at -15 ℃ and -10 ℃ in the Jungfraujoch (3580 m a.s.l.) (Conen et al., 2022), and the black"

Response: Thanks, we have revised it as "(b) N_{INP} measured during the daytime and nighttime. 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" in the revised manuscript.

Figure 3: Replace "dots). The point plot represents the median value. The shadow area represents" with "dots). Each point represents the median value and the shadow area represents"

Response: Thanks, we have revised it accordingly.

Figure 4: Replace "and meteorological parameters, chemical compositions, as functions of temperature." With "with meteorological parameters and chemical compositions as functions of temperature."

Response: Thanks, we have revised it accordingly.

Reference:

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Measurement report: Atmospheric Ice Nuclei at Changbai Mountain (2623 m a.s.l.) in Northeastern Asia

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Abstract. Atmospheric ice nucleation plays an important role in modulating the global hydrological cycle and atmospheric radiation balance. To date, few comprehensive field observations of ice nuclei have been carried out at high-altitude sites, which is close to the height of mixed-phase cloud formation. In this study, we measured the concentration of ice-nucleating particles (INPs) in the immersion freezing mode at the summit of Changbai Mountain (2623 m above sea level), Northeast Asia, in summer 2021. The cumulative number concentration of INPs varied from 1.6×10^{-3} L−1 to 78.3 L^{-1} over the temperature range from −5.5 ℃ to −29.0 ℃. Proteinaceous-based biological materials accounted for the majority of INPs, with the proportion of biological INPs (bio-INPs) exceeding 67% across the entire freezing temperature range, with this proportion even exceeding 90% above −13.0 ℃. At freezing temperatures ranging from −11.0 ℃ to −8.0 ℃, bio-INPs were found to significantly correlate with wind speed (r = 0.5-0.8, $p < 0.05$) and Ca^{2+} (r = 0.6-0.9), and good but not significant correlation was found with isoprene (r = 0.6-0.7) and its oxidation products (isoprene × O_3) (r = 0.7), suggesting that biological aerosols may attach to or mix with soil dust and contribute to INPs. During the daytime, bio-INPs showed a positive correlation with the planetary boundary layer height at freezing temperatures ranging from −22.0 ℃ to −19.5 ℃ ($r > 0.7$, $p < 0.05$), with the valley breezes from southern mountainous regions also influencing the concentration of INPs. Moreover, the long-distance transport of air mass from the Japan Sea and South Korea significantly contributed to the high concentrations of bio-INPs. Our study emphasizes the important role of biological sources of INPs in the high-altitudes atmosphere of northeastern Asia, as well as the significant contribution of long-range transport to the INP concentrations in this region.

1 Introduction

Clouds play a crucial role in regulating the Earth's energy balance by absorbing, reflecting, and scattering solar and terrestrial radiation (Zhou et al., 2016; Bjordal et al., 2020). 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). Atmospheric aerosols can act as ice-nucleating particles (INPs), triggering the freezing of cloud droplets through heterogeneous nucleation processes (Rinaldi et al., 2017; Rosenfeld and Woodley, 2000; Demott et al., 2003; Murray et al., 2012). Currently, the four main mechanisms of heterogeneous ice nucleation are considered: deposition nucleation, condensation freezing, immersion freezing, and contact freezing (Demott et al., 2003; Vali et al., 2015). Recent studies have concluded that water saturation is a prerequisite for ice formation in low- and mid-level clouds, and that contact and immersion freezing are the most primary pathways for ice formation (Sassen and Khvorostyanov, 2008; Phillips et al., 2007; Murray et al., 2012).

Various aerosol particles are potential INPs, such as biological aerosols (Pratt et al., 2009; Tobo et al., 2013), mineral dusts (Pratt et al., 2009; Atkinson et al., 2013), sea spray aerosols (Mccluskey et al., 2017; Alpert et al., 2022), carbonaceous aerosols (Grawe et al., 2018; Demott, 1990; Diehl and Mitra, 1998; Fornea et al., 2009), and volcanic ashes (Grawe et al., 2016; Umo et al., 2015). Biological aerosols from microbial and proteinaceous origin, containing ice nucleation active protein, demonstrate significant efficiency as INPs at temperatures above −15 ℃ (Phelps et al., 1986; Petters and Wright, 2015; Murray et al., 2012; Kunert et al., 2019; Huang et al., 2021). For example, biological components in lichens can induce freezing above −10 ℃ (Kieft, 1988; Moffett et al., 2015), and certain bacterial organisms such as *Pseudomonas syringae* can facilitate droplet freezing even at extremely high temperatures (above −2 ℃) (Maki et al., 1974). The other biological aerosols with non-proteinaceous compounds act as ice nucleation catalysts, such as pollen, cellulose, and other macromolecular organic particles, can also induce ice formation through heat-resistant polysaccharides on their surfaces, but at lower temperatures than proteinaceous biological particles (Knopf et al., 2010; Pummer et al., 2012). Mineral dust and sea spray aerosols predominantly consist of inorganic compounds and serve as effective INPs at temperatures below −15°C (Alpert et al., 2022; Atkinson et al., 2013; Ladino et al., 2019). The ice-nucleating properties of aerosols are affected by many factors. For instance, the size of particles is a crucial factor for providing active sites for ice formation, with larger particles containing more efficient ice nucleation sites than smaller ones (Chen et al., 2018; Demott et al., 2010; Demott et al., 2015). In addition, the chemical composition and surface properties of aerosol particles, such as their surface topology, defects, roughness, and functional groups, also influence their activity as INPs (Hiranuma et al., 2014; Marcolli, 2014; Roudsari et al., 2022; Yun et al., 2021; Zolles et al., 2015). Furthermore, the ice-nucleating properties of aerosol particles can be modified through chemical reactions with trace gases or organic/inorganic component or through physical processes such as efflorescence or deliquescence (Cziczo et al., 2009; Hoose and Möhler, 2012; Creamean et al., 2013; Tang et al., 2016; Tang et al., 2018).

Over recent decades, numerous studies have focused on investigating heterogeneous ice nucleation in various atmosphere environments (Gong et al., 2022; Chen et al., 2021; Testa et al., 2021; Knopf et al., 2023; Harrison et al., 2022; Ren et al., 2023; Sanchez-Marroquin et al., 2023). In low-altitude atmospheric environments, the abundance of ground-based sources and sinks results in spatial distribution heterogeneity (Larsen, 2007), which restricts the characterization of INPs properties on a regional scale. At present, there remain uncertainties about how INPs transported to the altitudes of mixed-phase cloud formation (approximately 3–7 km) (Knopf et al., 2018). High-altitude sites provide favorable conditions for in situ observations to investigate INPs characteristics, as they can represent tropospheric background conditions and reflect long-distance transport and vertical mixing processes prior to arriving at the ground sampling site (Wieder et al., 2022). Therefore, field experiments have been conducted in several high-altitude sites. For example, in 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). This decline in INPs could exceed 60% per kilometer during the cold season (from 1631 m a.s.l. to 2693 m a.s.l.) (Wieder et al., 2022), which was attributed to the scarcity of effective INPs sources in high-altitudes. Note that variations in sampling methods and the influence of wind directions can also exert an impact on INP concentrations. Schrod et al. (2017) reported an increase in INPs abundance of approximately 10 times over the eastern Mediterranean (2500 m a.s.l.) relative to ground level using unmanned aircraft systems, with this difference attributed to the long-distance transport of a series of elevated Saharan dust plumes at the height of a few kilometers. In some mountainous areas, biogenic aerosols can act as the most abundant type of INPs. 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 ℃ under free-tropospheric conditions (Conen et al., 2022). Similarly, at the Puy de Dôme station (1465 m a.s.l.) in France, the average contribution of biological aerosols in cloud water could reach up to 85% at freezing temperatures above −10 ℃ (Joly et al., 2014). To date, fewer field observations have been carried out in high-altitude regions in China. For example, Jiang et al. (2014, 2015) performed measurements at Mt. Huangshan (1840 m a.s.l.) in Southeast China, finding that larger particles were more likely to be effective INPs. Lu et al. (2016) collected seven rainwater samples from three high-altitude sites in eastern China, i.e., Changbai Mountain (at the peak of 2740 m a.s.l.), Wuling Mountain (900 m a.s.l.), and Dinghu Mountain (1000 m a.s.l.). The authors found that the onset freezing temperature was approximately −6 ℃, but bacteria played minor roles in the overall INP activity. Because the number of rainwater samples was limited, further research is necessary to explore the impact of biological INPs on ice formation and their contribution to the formation of precipitation.

In this study, we conducted offline INP measurements at the top of Changbai Mountain (2623 m a.s.l.) in Jilin province, China, which is located in Northeast Asia. This region is particularly vulnerable to climate change because of the presence of distinct ecotones caused by land type changes, as well as the influence of the North Atlantic Oscillation and the Northern Hemisphere circulation (Sugita et al., 2007; Zhang et al., 2021). Moreover, Northeast Asia is densely populated and serves as a crucial breadbasket for the world, making rainfall an essential factor for determining crop yields (Zhang et al., 2021). Given the high-altitude of Changbai Mountain, it is an ideal location to capture the characteristics of the regional atmospheric background and transboundary transport of air masses from nearby countries. Our main objective was to investigate the concentration levels of INPs and identify their major sources at the height of the mountain’s peak.

Additionally, we evaluated the impact of the planetary boundary layer (PBL) height, valley breezes, and transport pathways on the INP population to gain a better understanding of their sources in this region. Our findings could provide valuable insights into the formation and behaviour of mixed-phase clouds over this region.

2 Methods

2.1 Site description

Changbai Mountain is the highest mountain in the border region between China and the Korean Peninsula. It is situated on the transport pathways of continental air pollutants from Asia to the North Pacific Ocean and even as far as the Arctic (Ikeda et al., 2021). The regional topography is characterized by forests and mountains, with elevations ranging from 410 m a.s.l. to 2740 m a.s.l. The southeast exhibits higher elevations compared to the northwest (Wang et al., 2014). At the top of Changbai Mountain, there is a vast dormant crater known as Tianchi Lake, which has a depth of 373 m and covers an area of 9.82 km^2. In this study, a field campaign was carried out at the Tianchi Meteorological Station (Tianchi Site, 42.03°N, 128.07°E, 2623 m a.s.l., Figure 1), which is approximately 410 m north of Tianchi Lake, from July 24 to August 24, 2021.

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 season and the southeasterly and southwesterly winds in the summer season (Zhao et al., 2015). The annual average temperature is typically lower than −7.4 ℃ (Jin et al., 2018), with the mountain summit always covered by snow and ice for approximately three quarters of the year. Figure S1 presents the timeseries of meteorological parameter, NOx concentration, and the of INP concentrations during the field campaign. During the campaign, the relative humidity (RH) ranged from 33% to 100%, with a mean of 92.4 ± 0.4% (average ± standard error of the mean (SEM)). Notably, seventy percent of the RH exceeded 90% throughout the campaign, indicating that the campaign was performed under humid weather conditions. The sampling site was predominantly affected by southerly and westerly winds, with wind speed (WS) ranging from 0.1 m s^{-1} to 25.7 m s^{-1}. Changbai Mountain is a national natural reserve with no large industrial facilities nearby, and tourism is the important economic activity in the region. Due to the COVID-19 pandemic, strict lockdown measures were implemented from August 10, 2021, resulting in a substantial reduction in visitor numbers, as indicated by the marked decrease in NOx concentration (Figure S1). The surroundings of the observation site are covered by dense vegetation, such as shrubs and perennial herbs. Most of the time, the site is above the PBL and in the free troposphere, making it an ideal site for studying the regional background atmosphere of Northeast Asia.

2.2 Sample collection

Total suspended particulate (TSP) was collected on polycarbonate (PCTE) membrane filters (Sterlitech 1870, nominal porosity 0.45 μm) using a TH-150D medium flow sampler (Wuhan Tianhong Corporation, China, Figure 1c) at a flow rate of 50 L min^{-1} for the INPs analysis. Samples were collected during the daytime (06:00 to 17:30) and nighttime (18:00 to 05:30 in the following day) in local time. A total of 24 PCTE filters were collected, including 22 aerosol samples and 2 blank filters. These samples were used for INP analysis with the detailed sampling information provided in Table S1. Meanwhile, fine particulate matter ($PM_{2.5}$) samples were collected on quartz microfiber filters (PALL Pallflex, 7204）, which were heated at 560 ℃ for 4 h before sampling to remove any adsorbed organics, using a TH-150A medium flow sampler (Wuhan Tianhong Corporation, China) with a 2.5 μm impactor at a flow rate of 100 L min^{-1}. A total of 157 samples were collected on quartz filters every 3 h and used for chemical composition analysis. After sampling, all filter samples were kept frozen at ≤ −18 ℃ until analysis.

Real-time measurements of $PM_{2.5}$ and black carbon (BC) were recorded at 1 min intervals by using SHARP 5012 (Thermo Scientific, USA) and SHARP 5030 (Thermo Scientific, USA), respectively. Trace gases including CO, SO_2, NO_x, and O_3, were detected using Thermo Scientific 48i, 43i, 42i, and 49i, respectively. Ambient volatile organic compounds (VOCs) were collected by taking air samples using stainless-steel canisters at two specific time intervals (i.e., 11:00-13:00 and 20:00-22:00) on clean days, and the sampling frequency was increased to every 3h (5 samples per day) during air pollution episodes. 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 away from the sampling site. The parameters mentioned above were analyzed by determining the average value over the corresponding INPs sampling period.

2.3 INPs analysis

INP measurements in the immersion mode were conducted using the Guangzhou Institute of Geochemistry Ice Nucleation Apparatus (GIGINA) from −40 ℃ to 0 ℃. GIGINA is a cold-stage-based ice nucleation array that consists of an enclosed droplet chamber with a commercial cold stage inside (LTS120, Linkam, Epsom Downs, UK), an external refrigerated water circulator (VIVO RT4, Julabo, Seelbach, Germany), and a charge-coupled device (CCD) camera (DMK33G274, The Imaging Source, Bremen, Germany). Further details regarding GIGINA have been published by Chen et al. (2023).

Each polycarbonate filter was immersed in 5 mL MilliQ water (resistivity of 18.2 MΩ cm^{-1} at 25 ℃) and sonicated for 30 min to wash off the particles (Chen et al., 2021). Note that an ice water bath was utilized during ultrasonic extraction to mitigate any potential alterations in protein properties and biogenic activities. In addition, the suspension underwent dilution at multiple levels: 30-fold, 60-fold, and 120-fold, in order to generate a spectrum that encompass freezing temperature below

−25°C, as illustrated in Figure S2. The INPs measurement process is briefly described as follows. First, a hydrophobic glass slide was placed on a cold stage and covered with silicone oil between them to achieve good thermal contact. Second, a round aluminum spacer with 90 round compartments was placed on the glass slide, and the particle suspension was sequentially pipetted into the center region of each compartment. Then, another glass slide was placed above the spacer to avoid the Wegener–Bergeron–Findeisen process (Jung et al., 2012). Afterward, the temperature of the droplets was cooled down to 0 °C at a cooling rate of 10 °C min^{-1}, after which the cooling of the droplets continued at a rate of 1 °C min^{-1} until all the droplets were frozen. During the freezing experiment, high-purity nitrogen was continuously delivered onto the cold stage to prevent frost from forming on the surface of the glass slide. Meanwhile, real-time images of the droplets were photographed by the CCD camera and recorded by the LINK software every 6 s. After the experiment, the phase transition of each droplet was identified by analyzing the changes in image brightness, which distinguished between unfrozen (white) and frozen (dark) droplets.

The frozen fraction, f_{ice}, was calculated according to Eq. (1):

$$f_{ice}(T) = \frac{n_{ice}}{n_{tot}} \ , \qquad (1)$$

where n_{ice} is the number of frozen droplets at a certain temperature T, and n_{tot} is the total number of droplets (90 droplets). The cumulative number concentration of INPs (N_{INP}) per unit volume of sampled air were calculated following the method of Vali (1971, 2015):

$$N_{INP}(T) = -\frac{ln[1-f_{ice,\ sample}(T)]-ln[1-f_{ice,\ blank}(T)]}{V_{air}}(L^{-1}\ air), \qquad (2)$$

where V_{air} is the total volume of sampled air per droplet converted to standard conditions (0 °C and 1013 hPa). The $f_{ice,\ sample}$ and $f_{ice,\ blank}$ are the measured frozen fractions for the filter samples and the field blanks, respectively. The calculation for V_{air} entails multiplying the droplet volume (1 μl) by the sample volume and then dividing by the volume of wash water. In our study, the N_{INP} values were significantly larger in filter samples than in the field blanks. In the following analysis, the concentrations of the two blank filters were subtracted from the daytime and nighttime samples at each freezing temperature, respectively. The rarity of INPs in the atmosphere leads to their low concentration in the suspension. Because the suspension used in the measurement contained a limited number of droplets, we need to consider the resulting uncertainty caused by statistical errors. Therefore, we calculated the confidence intervals of the apparatus for f_{ice} according to the method of Gong et al. (2022) and Agresti and Coull (1998) to address uncertainty associated with the droplet-freezing apparatus:

$$\left(f_{ice} + \frac{Z^2_{\alpha/2}}{2n_{tot}} \pm Z_{\propto/2}\sqrt{[f_{ice}(1-f_{ice}) + Z^2_{\alpha/2}/(4n_{tot})]/n_{tot}}\right) / \left(1 + Z^2_{\alpha/2}/n_{tot}\right), \qquad (3)$$

where $Z_{\alpha/2}$ is the standard score at a confidence level $\alpha/2$, which is 1.96 for a 95% confidence interval.

2.4 Chemical analysis

The $PM_{2.5}$ samples collected by quartz membranes were used to analyze the particle chemical composition. For each sample, an eighth of the filter was ultrasonically extracted using 15 mL MilliQ water for 30 min to make a suspension. The concentrations of inorganic water-soluble anions (Cl^-, SO_4^{2-}, and NO_3^-) and cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) were identified using the ICS 1100 ion chromatograph (Thermo Scientific). In addition, the concentrations of organic carbon (OC) and elemental carbon (EC) were measured using the Sunset Laboratory Model-5 semi-continuous OC/EC field analyzer. The VOCs canister samples were analyzed using online gas chromatography–mass spectrometry (TT24xr, Makers, UK; GC–MS, Thermo Scientific, USA) in the laboratory. A total of 106 target VOCs, including 29 alkanes, 11 alkenes, one alkyne, 17 aromatics, 35 halogenated hydrocarbons and 13 oxygenated VOCs (OVOCs), were quantified.

2.5 The PBL data and air mass back trajectory model

The PBL data were downloaded from the fifth-generation ECMWF global atmospheric reanalysis product (ERA5 https://cds.climate.copernicus.eu), which provides hourly records on latitude–longitude grids at 0.25° × 0.25° resolution. The 72-h air mass backward trajectories at the sampling site were calculated on an hourly basis during the sampling days. These calculations were performed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://ready.arl.noaa.gov/HYSPLIT.php), which is developed by the National Oceanic and Atmospheric Administration Air Resources Laboratory (NOAA ARL) (Stein et al., 2015). The simulations were based on meteorological data from the Global Data Assimilation System (GDAS) with a spatial resolution of 1° × 1°. A trajectory ending height of 967 m above ground level (a.g.l.) was selected because the terrain height of Changbai Mountain was approximately 1656 m in the GDAS data. Using the open-source software of MeteoInfo, concentration-weighted trajectory (CWT) analysis was conducted to explore the potential sources of INPs based on the air mass backward trajectories and N_{INP}. The CWT reflects the pollution levels of different trajectories by calculating the weight concentration of the airmass trajectory in potential source areas. The calculation was used Equation 4 according to the method of Hsu et al. (2003):

$$C_{ij} = \frac{1}{\sum_{k=1}^{M} \tau_{ijk}} \sum_{k=1}^{M} C_k \tau_{ijk}, \quad (4)$$

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 number of endpoints that fall in the cell is defined as nij, and the weight function W_{ij} is used to reduce the uncertainty in the cells with small values of n_{ij}:

$$WCWT_{ij} = C_{ij} \times W(n_{ij}), \quad (5)$$

Note that CWT analysis does not account for the impacts of wet deposition such as precipitation along the trajectories. Uncertainties may exist in the CWT analysis due to the relatively small dataset of INPs in this study.

3 Results and Discussion

3.1 INP concentrations

The freezing temperature at which 50% of the droplets are frozen (T_{50}) was calculated for each sample. The frozen fractions (f_{ice}) of all freezing curves containing the collected samples and MilliQ water are shown in Figure S2. For the blank filters, T_{50} was averaged to −24.9 ± 1.1 °C, which was slightly higher than that of MilliQ water, but significantly lower than that of the collected samples (for which T_{50} was −17.0 ± 0.6 °C). This result suggests the presence of minimal contaminants stemming from the filter membrane.

The N_{INP} values as a function of temperature are presented in Figure 2, where the pink and blue circles represent the samples collected during the daytime and nighttime, respectively. The onset freezing of ambient samples was observed in the temperature range of −5.5 °C to −29.0 °C, with N_{INP} spanning up to five orders of magnitude from 1.6×10^{-3} L^{-1} to 78.3 L^{-1}. For freezing temperatures above average T_{50} (−17.0 °C), the temperature region is referred to as the high-temperature region (HTR), where N_{INP} spans three orders of magnitude from 1.6×10^{-3} L^{-1} to 6.2 L^{-1}. Some of the N_{INP} curves exhibited bumps in the HTR, which has been also observed at a coastal site (the Cape Verde Atmospheric Observatory, Africa) by Welti et al. (2018) in air samples and 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 the narrower the IN properties, the steeper slope can be observed in a temperature spectrum. In contrast, in the low-temperature region (LTR, freezing temperature below average T_{50}, −17.0 °C ~ −29.0 °C), N_{INP} showed a relatively narrow variation from 0.1 L^{-1} to 78.3 L^{-1}. Furthermore, there were no significant differences observed in N_{INP} between daytime and nighttime (the significance level is 0.61). However, in some mountainous sites, such as Mt. Huang, where samples were collected twice daily at 08:00 and 14:00 (Jiang et al., 2015), and the Weissfluhjoch in the Swiss Alps, where samples were collected at 20-minute intervals (Wieder et al., 2022), N_{INP} displayed a distinct diurnal cycle induced by the orographically lifted air masses containing high INP concentrations from low elevation upstream during the daytime. In this study, local sources such as vegetation and the lake may impact INP concentrations, with biogenic emissions potentially exhibiting variations between daytime and nighttime. However, we collected two samples per day, and the limited dataset size and low sampling frequency may have contributed to the absence of diurnal variations.

We compared our N_{INP} measurements with previous results from diverse sites. For instance, in mountainous regions, the N_{INP} value at the Weissfluhjoch varied from 10^{-4} L^{-1} to 10^{1} L^{-1} in the temperature range of −4.0 °C to −24.0 °C (Wieder et al., 2022). In our observations, the spectra range of N_{INP} were narrowly located in the relatively high-concentration regions at overlapping freezing temperatures compared to the measurements of Wieder et al. (2022). Jiang et al. (2015) reported that the INP concentrations at the top of Mt. Huang spanning from 0.1 L^{-1} to 11.9 L^{-1} over a temperature range from −15.0 °C to −23.0 °C, which overlapped with our results. In the LTR, our results were comparable to the measurements conducted at the Storm Peak Laboratory in the northwestern Colorado Rocky Mountains by Hodshire et al. (2022). But in HTR, Conen et al. (2022) recorded results in Switzerland were 1-3 orders of magnitude higher than our study. Gong et al. (2022) measured

INPs at the mountain station at Cerro Mirador (622 m a.s.l., Chile), and reported N_{INP} values lower than those in our study by around one order of magnitude at similar freezing temperatures from −26.0 ℃ to −3.0 ℃. In heavily polluted urban sites, such as Beijing (Chen et al., 2018) and Tai'an (Jiang et al., 2020) in China, the INP concentrations were comparable to our measurements at overlapping freezing temperatures. Chen et al. (2018) reported that INP concentrations might not be influenced by urban air pollution because no correlation was found between the immersion-freezing INP concentrations and the $PM_{2.5}$ or BC concentration.

3.2 Contribution of biological particles, other organics, and inorganics to INPs

Generally, biological particles can induce ice nucleation in the immersion mode at relatively high temperatures above −15.0 ℃ (Murray et al., 2012). Proteinaceous components mainly induce biological ice nucleation, and wet heat treatment (i.e., heating the particle suspension to 95.0 ℃ for 30 min) is used to identify the protein-based biological ice nucleation activity (Beall et al., 2022; Chen et al., 2021). We measured the N_{INP} values of the suspensions after heat treatment, which we refer to as heat-resistant N_{INP} ($N_{INP\text{-}heat}$, as shown in Figure S3), and the difference between the original N_{INP} and $N_{INP\text{-}heat}$ was considered to be mainly due to the proteinaceous biological N_{INP} ($N_{INP\text{-}bio}$). However, some biological aerosols, such as pollen, cellulose, or other macromolecular organic particles, are insensitive to heat treatment at 95.0 ℃ (Daily et al., 2022). Therefore, we also measured the heat-stable organic INPs, which are defined as other organic INPs (other org-INPs), following the methods of Suski et al. (2018) and Testa et al. (2021). We added 30% H_2O_2 (guaranteed reagent) to the suspension to obtain a final concentration of 10%, and then heated it at 95 ℃ for 20 min under UVB fluorescent bulbs. To prevent freezing point depression, we neutralized the remaining H_2O_2 in the suspension with catalase. The N_{INP} value following treatment by this procedure was denoted H_2O_2-resistant N_{INP} ($N_{INP\text{-}H2O2}$), which is the concentration of inorganic INPs ($N_{INP\text{-}inorg}$). The difference between heat-resistant N_{INP} and H_2O_2-resistant N_{INP} was considered to be equivalent to the concentration of other organic INPs ($N_{INP\text{-}other\ org}$).

Figure 3 illustrates the concentrations and fractions of the three types of INPs. The biological INPs (bio-INPs) showed ice nucleation activity at temperatures between −28.5 °C and −5.5 ℃. After the ice nucleation activity of the bio-INPs was destroyed, $N_{INP\text{-}heat}$ decreased by around 1–2 orders of magnitude compared with the original N_{INP}, indicating a significant contribution of $N_{INP\text{-}bio}$, as shown in Figure S3. The initial freezing temperature of other org-INPs was −11.0 ℃, which was approximately 5.5 ℃ lower than that of bio-INPs. Inorganic INPs exhibited ice nucleation activity at temperatures between −28.0 ℃ and −10.0 ℃. Interestingly, the initial freezing temperatures of some inorganic INPs were slightly higher than those of other org-INPs, indicating that some inorganic aerosols could trigger freezing at relatively high temperatures.

The proportions of the three types of INPs as functions of temperature are presented in Figure 3(b). Here, the fractions of $N_{INP\text{-}bio}$ ($F_{INP\text{-}bio}$) account for 100% of the N_{INP} value above −11 ℃, and show a decreasing trend as the temperature decreases from −11.5 ℃ to −16.5 ℃. This decreasing trend of $F_{INP\text{-}bio}$ is consistent with trends observed in other areas dominated by bio-INPs in similar temperature regions (Gong et al., 2022; Testa et al., 2021), suggesting that the importance of bio-INPs decreases with decreasing temperature. Interestingly, when the temperature decreased from −16.5 °C to −21.5 °C, the median of $F_{\text{INP-bio}}$ increased from 0.8 to 0.9, indicating the high concentration of bio-INPs in the LTR. Previous observational studies have indicated that although most bio-INPs act as INPs at high freezing temperature, some heat-sensitive biological aerosols, such as fungal cloths, exhibit ice-nucleating activity at low temperatures (Iannone et al., 2011; Kanji et al., 2017). Modelling studies have also shown that bio-INPs can influence the ice phase of clouds and produce ice crystals when the cloud-top temperature is below −15 °C (Hummel et al., 2018). This phenomenon may also be related to the sensitivity of different species of biological aerosol to heating conditions. In wet heat treatment it is assumed that the ice-nucleating active protein in bio-INPs is completely destroyed and denatured, thus losing any ice formation potential. However, this method may lead to decrease in the freezing temperature of bacteria and fungi, but their ice-forming activity still cannot be ignored (Daily et al., 2022). As the temperature dropped to −25.0 °C, $F_{INP\text{-}bio}$ began to decrease significantly to 0.7. Overall, the median value of $F_{\text{INP-bio}}$ was more than 67% in the entire temperature range from −25.0 °C to −5.5 °C, with the value exceeding 90% above −13.0 °C, which was much higher than in some mountainous areas in southwestern South America (Gong et al., 2022) and urban areas in China (Chen et al., 2021). The fractions of $N_{\text{INP-other org}}$ ($F_{\text{INP-other org}}$) showed an opposite trend from that of $F_{\text{INP-bio}}$ at freezing temperatures between −25.0 °C and −11.0 °C. First, $F_{\text{INP-other org}}$ increased from 0.08 to 0.2 as the temperature decreased from −11.0 °C to −15.0 °C, and then sharply decreased from 0.2 to 0.05 as the temperature decreased further from −15.0 °C to −22.0 °C. When the temperature was lower than −22.0 °C, $F_{\text{INP-other org}}$ gradually increased to 0.3. The fractions of $N_{\text{INP-inorg}}$ ($F_{\text{INP-inorg}}$) remained below 0.22 throughout the entire temperature range, with an increasing trend observed below −22.0 °C. Overall, our results showed that protein-based biological aerosols contribute the most to N_{INP} at Changbai Mountain.

3.3 Source analysis of different types of INPs

We investigated the relationship between different types of INPs and various environmental conditions, as well as the gases and particle compositions, as show in Figure 4 (details can be found in Table S2). In the HTR, our results showed a significant positive correlation (r = 0.5-0.8, $p < 0.05$) between N_{INP} and WS at temperature ranging between −11. 0 °C and −9.0 °C. High WS can enhance the uplift of soil dust and the long-distance transport of aerosols. Moreover, N_{INP} and Ca^{2+} showed a good positive correlation (r = 0.6-0.9) between −11.0 °C and −8.0 °C, leading us to speculate that soil dust may play an important role in ice nucleation in this temperature range. The correlations between N_{INP} and both Ca^{2+} and WS were more pronounced during the daytime (Figure S4), potentially linked to the evolution of PBL and the presence of valley breeze (refer to details in section 3.4). Previous studies have shown that when soil dusts mix with biological components, their freezing temperatures can increase to as high as −6 °C, which is much higher than that of natural dust (below −20 °C) (Hill et al. 2016; O'sullivan et al., 2014). In the LTR, N_{INP} demonstrated a significant positive correlation with ambient air temperature (r = 0.5-0.6, $p < 0.05$), but showed a significant negative correlation with RH (r = 0.6-0.7, $p < 0.05$). We further examined the N_{INP} under various weather conditions, including sunny, foggy, and rainy days, which characterized by significantly different RH levels. As shown in Figure S5, it was observed that on rainy days, the concentrations of INPs were slightly lower compared to the other two weather types in the LTR. This phenomenon may be attributed to the wet deposition of coarse particles (Olszowski, 2016; Kumar et al., 2020). When the temperature falls below -20.0 ℃, N_{INP} exhibits a significant positive correlation with $PM_{2.5}$ and BC, implying that inorganic components may serve as active INPs in lower freezing temperature.

We also investigated the potential sources of different types of INPs, as shown in Figure 4(b–d). Similar to the total INP concentrations, $N_{\mathrm{INP\text{-}bio}}$ were more abundant during the high freezing temperature and low RH. A significant positive correlation was found between $N_{\mathrm{INP\text{-}bio}}$ with WS (r = 0.5-0.7) and Ca^{2+} (r = 0.6-0.9) within a temperature range of −11 ℃ to −8 ℃. Additionally, a good but not significant positive correlation emerged between $N_{\mathrm{INP\text{-}bio}}$ and isoprene (r = 0.6-0.7, p > 0.05) along with its oxidation products (isoprene × O_3, r = 0.7, p > 0.05). O'sullivan et al. (2016) and Augustin-Bauditz et al. (2016) previously reported that biological materials may attach to or mix with dust particles, thus promoting the formation of INPs. However, no mineral dust events were observed during our sampling period, based on the low mass concentrations of $PM_{2.5}$ (the range from 1.5 μg m^{-3} to 31.6 μg m^{-3} with average of 9.3 ± 0.4 μg m^{-3}) and Ca^{2+} (the range from 0.007 μg m^{-3} to 3.6 μg m^{-3} with average of 0.5 ± 0.2 μg m^{-3}). We speculate that the higher WS may have facilitated the exposure of the local soil dust and bioaerosol containing bio-INPs to the air. Alternatively, the long-distance transport of biological aerosol attached to soil dust surfaces may also contribute to bio-INPs, leading to the high N_{INP} accompanied by high WS and Ca^{2+}.

The positive correlation was more obvious between $N_{\mathrm{INP\text{-}other\ org}}$ and isoprene, especially at temperatures ranging from −16.0 ℃ to −14.0 ℃ (r = 0.7-0.8, p < 0.05). It is considered to be an important natural gaseous precursor to the formation of secondary organic aerosol (Carlton et al., 2009). Additionally, $N_{\mathrm{INP\text{-}other\ org}}$ was positively correlated with the oxidation of isoprene bio-INPs (r = 0.5-0.6) at temperatures ranging from −18.0 ℃ to −14.0 ℃. Although the oxidation products of isoprene are expected to be water-soluble and unable to induce immersion freezing, the observed positive correlation suggests a potential role of secondary organic compounds associate with vegetation or other biogenic sources in ice nucleation.

In the temperature range of −23.0 °C to −17.0 °C within the LTR, $N_{INP\text{-}inorg}$ exhibited a significant negative correlation with RH (r = 0.5-0.7, p < 0.05), indicating an enrichment of inorg-INPs under low RH conditions. $N_{\mathrm{INP\text{-}inorg}}$ showed a significant positive correlation with BC and SNA (sulfate, nitrate andammonium) in the LTR. Previous studies have suggested that carbonaceous particles may not act as efficient INPs in the immersion mode or could decrease ice nucleation activity in polluted urban environments, which is attribute to the formation of organic coatings (Kanji et al., 2020; Schill et al., 2020; Nichman et al., 2019; Hammer et al., 2018). However, our observations revealed a positive correlation between BC and both N_{INP} and $N_{\mathrm{INP\text{-}inorg}}$ in the LTR. The discrepancy may be attributed to the different sources and aging degrees of BC, which remains unclear. Note that $PM_{2.5}$ chemical composition was used in this study, which may lead to uncertainties in the interpretation of the INPs in TSP.

3.4 Transport pathways of INPs

At the mountaintop site, the horizontal and vertical transport of air mass are important pathways for INPs under favorable conditions, such as valley breezes, variations in mixing layer height, and long-range transport processes (Chow et al., 2013; Wieder et al., 2022). Understanding the coupling between the PBL changes and the air mass transport process can help us comprehend the characteristics of the target aerosols. Therefore, we conducted further analysis to examine the relationship between the PBL height and N_{INP}, and combined it with CWT analysis to explore the effect of transport on N_{INP} at the sampling site.

At Changbai Mountain, changes in the PBL are also complicated by a variety of processes, such as orographic gravity waves, moist convection, and turbulent transport. Figure 5(a, b) shows the relationship between bio-INPs and the PBL height during the daytime. We found a positive correlation between PBL height and $N_{INP\text{-}bio}$ in the freezing temperature ranging from −25.0 °C to −15.0 °C (r = 0.4-0.8), especially at temperatures ranging between −22.0 °C and −19.5 °C ($r > 0.7$, $p < 0.05$). However, the correlation was no longer observed at the freezing temperature above −15 ℃ ($r < 0.5$, $p > 0.05$). Notably, this correlation increased in the HTR when we excluded two outliers with exceptionally high $N_{\mathrm{INP\text{-}bio}}$ values (as shown in Figure 5a, r increased to 0.77, $p < 0.05$). The two high values may be related to ocean and vegetation emissions, and they will be further discussed below. In brief, our findings suggest that an increase in the PBL height may cause a corresponding increase in $N_{\mathrm{INP\text{-}bio}}$ in the clean mountaintop atmosphere. Moreover, based on the analysis of the air mass backward trajectory, our analysis revealed a significant elevation in the height of the trajectory as it moved through the southern mountainous regions, indicative of an upslope valley wind effect (Figure S6d). According to Ketterer et al. (2014), such upslope valley winds have the potential to locally raise the altitude of the PBL, and could even trigger the vertical exchange of PBL air into the free troposphere. In our investigation, the observed positive correlation between the PBL and INPs suggests that valley breezes may facilitate the upward transport of INPs from the foothill to the top of Changbai Mountain during the daytime.

The CWT analysis revealed the transport pathways and potential source regions of bio-INPs, as shown in Figure 5(c, d). Ocean was identified as an important INPs source for long-range transport, as previous studies have reported that bubble bursting processes can release marine microorganisms (Burrows et al., 2013; Kwak et al., 2014; Vergara-Temprado et al., 2017). Different ice nucleating entities can trigger droplets to freeze at various temperatures in the marine environment. For example, Wilson et al. (2015) found that the biogenic organic materials within the sea surface microlayer could induce droplet freezing in the immersion mode, with a broad freezing temperature range of −7.0 °C to −35.0 °C. Laboratory experiments have further revealed that aerosols generated by phytoplankton are particularly effective at triggering ice nucleation at temperatures below −15.0 °C, with a notable increase in INP concentration within the range of −15.0 °C to −23.0 °C, which was related to the unique dynamic processes of phytoplankton bloom and growth (Brooks and Thornton, 2018; Mccluskey et al., 2017; Thornton et al., 2023; Wilbourn et al., 2020). Our study detected the high concentrations of bio-INPs in the LTR originating from the Japan Sea (Figure 5d), implying that the air mass passing over the Japan Sea surface might have carried marine bio-INPs, contributing to their presence at our sampling site. In contrast, in the HTR, bio-INPs are mainly originate from the southern part of the Korean Peninsula. Previous studies demonstrated that vegetation contains a substantial density of microorganisms (10^6-10^7 cm^{-2}) and serves as a recognized reservoir of highly efficient biological INPs (Moore et al., 2021; Lindow and Brandl, 2003). These bio-INPs typically induce freezing at relatively higher temperatures, ranging from −2 °C to −5 °C (Schneider et al., 2021; Maki et al., 1974). South Korea has a large vegetation coverage area, as shown in Figure 1(a), with a high potential to emit biological aerosols those of which may be able to reach our sampling site through long-distance transport.

The residence time of various biological particles in the atmosphere can range from less than a day to a few weeks, depending on their size and aerodynamic properties (Despres et al., 2012). The long-range transport of biological aerosols has been observed in previous studies. For example, abundant microbial components originating from the ocean or land have been found in the troposphere, even extending to the stratosphere and the Mesosphere (Burrows et al., 2009; Smith et al., 2013). High concentrations of microbial populations have also been identified in the background atmosphere during trans-Pacific intercontinental transport (Smith et al., 2013). On a global scale, microorganisms have been found to travel thousands of kilometers, with approximately 33%–68% originating in the ocean (Mayol et al., 2017). This suggests that the ocean's bubble bursting processes play a significant role in the generation of biological aerosols. In addition, bio-INPs can attach to dust particles for long-distance transmission, with an adhesion rate that can even exceed 99.9% (Creamean et al., 2013; Yahya et al., 2019). This process can enable biological aerosol to be transported over greater distances, significantly enhancing the ice nucleation activity of dust (O'sullivan et al., 2016; Augustin-Bauditz et al., 2016).

In addition, a positive correlation was found between the PBL height and other org-INPs during the daytime, with significant correlations observed between −18.5 °C and −16.5 °C ($r > 0.7$, $p < 0.05$), as shown in Figure S7. However, at the freezing temperatures greater than −15.0 °C, no correlation was observed between the PBL and $N_{\text{INP-other org}}$, suggesting that local sources may be an important source for other org-INPs. For the inorg-INPs, a weak correlation with the PBL height was observed at temperatures greater than −23.0 °C and was not statistically significant ($p > 0.05$). However, at −24.5 °C and −24.0 °C, the correlation is more significant (r is 0.73 and 0.80, $p < 0.05$). The CWT simulation also indicated that high values of $N_{\text{INP-other org}}$ and $N_{INP\text{-}inorg}$ appeared in both local areas and adjacent Japan Sea regions (See Figure S8).

In summary, our findings suggest that valley breezes and the long-distance transport of air mass from the Japan Sea influence the abundance of INPs at Changbai Mountain. However, the impact of the PBL and valley breezes on the transport of inorg-INPs was found to be less significant than the contributions of bio-INPs and other org-INPs.

4 Conclusion

Measurements of INPs were carried out at the Changbai Mountain in northeastern Asia to explore the abundance and source of INPs in the immersion freezing mode. Our results showed that N_{INP} spanned up to five orders of magnitude between 1.6 × 10^{-3} L^{-1} and 78.3 L^{-1} over the freezing temperature range from −5.5 ℃ to −29.0 ℃, with these values corresponding to previously reported measurements for mountain sites.

The observed INPs predominantly comprised protein-based bio-INPs. The fractions of proteinaceous biological N_{INP} ($F_{INP\text{-}bio}$) constituted 100% of N_{INP} above −11 ℃, gradually decreasing as the temperature decreased from −11.5 ℃ to −16.5 ℃. Notably, a turning point occurred at −16.5 ℃, where $F_{INP\text{-}bio}$ increased from 0.8 to 0.9 as the temperature decreased from −16.5 ℃ to −21.5 ℃, indicating an enrichment of active bio-INPs in the low-temperature region (LTR, freezing temperature below T_{50}, −17.0 ℃ ~ −29.0 ℃). When the temperature falls below -22.0 ℃, $F_{INP\text{-}bio}$ exhibits a pronounced declining trend. We also found a significant positive correlation between biological INPs and both wind speed (WS) and Ca^{2+}, whereas there was only a weak positive correlation for biological INPs with isoprene and its oxidation products (isoprene × O_3). We speculate that the higher WS may facilitate the exposure of the local soil dust and bioaerosols containing bio-INPs to the atmosphere.

Our study also suggests that an increase in the planetary boundary layer (PBL) during the observation period may lead to a corresponding increase of diverse types of N_{INP} in the clean mountaintop atmosphere. During the daytime, valley breezes facilitate the orographic lifting of INPs from the foothill to the top of mountain. However, for the high values of $N_{INP\text{-}bio}$, it may originate from long-distance transport from the Japan Sea and South Korea areas. Our findings suggested that the oceanic and vegetation biogenic aerosols from these areas make significant contributions to the INPs at the top of the Changbai Mountain.

Our measurements in the high-altitude atmosphere above Northeast Asia indicate the predominant role of bio-INPs. However, our study has limitation in terms of dataset size. Further observational and modelling studies employing high-resolution instruments are urgently needed to analyze the characteristics of INPs and their influence on ice crystal formation as well as the cloud properties in the high-altitude atmosphere.

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Competing interests. The authors declare that they have no conflict of interest.

Data availability. The datasets related to this work can be accessed via https://doi.org/10.17632/b9y6pfw39n.1 (Sun et al., 2023).

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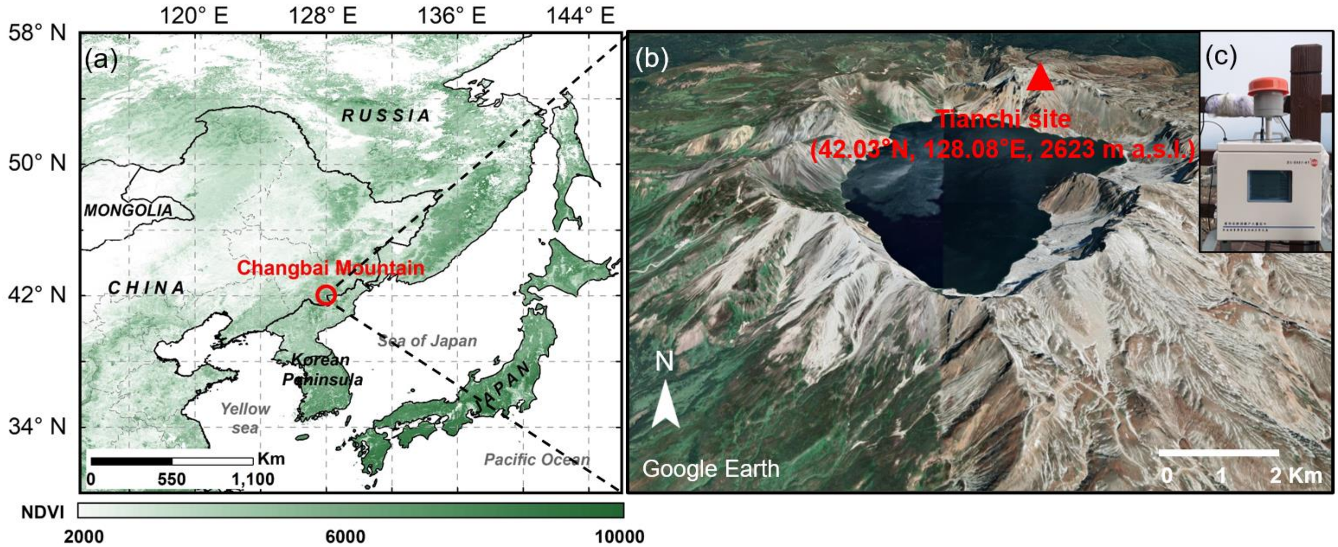

Figure 1. Geographical maps showing the location of Changbai Mountain. (a) This map is color-coded according to the normalized difference vegetation index (NDVI) in 2015, which was downloaded from the Geospatial Data Cloud (https://www.gscloud.cn/search). (b) Map with the three-dimensional shape of the sampling site, which was obtained from Google Earth. (c) The INP sampler (The TH-150D medium flow sampler, Wuhan Tianhong Corporation, China).

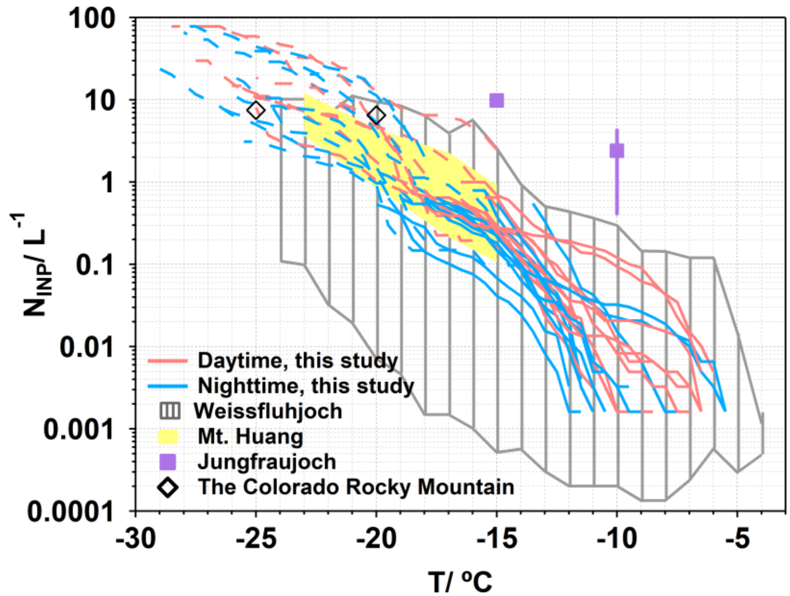

Figure 2. The concentrations of INPs (N_{INP}) as functions of temperature. The dark gray shaded area represents the upper and lower limits of N_{INP} over Weissfluhjoch (2693 m a.s.l.) (Wieder et al., 2022), the yellow shaded area represents the atmospheric N_{INP} ranges at Mt. Huang (1840 m a.s.l) (Jiang et al., 2015), the purple square represents the median N_{INP} at −15 ℃ and −10 ℃ in Jungfraujoch (3580 m a.s.l.) (Conen et al., 2022), and the black rhombus represents the median N_{INP} at −25 ℃ and −20 ℃ at the Storm Peak Laboratory in the northwestern Colorado Rocky Mountains (3220 m a.s.l.) (Hodshire et al., 2022).

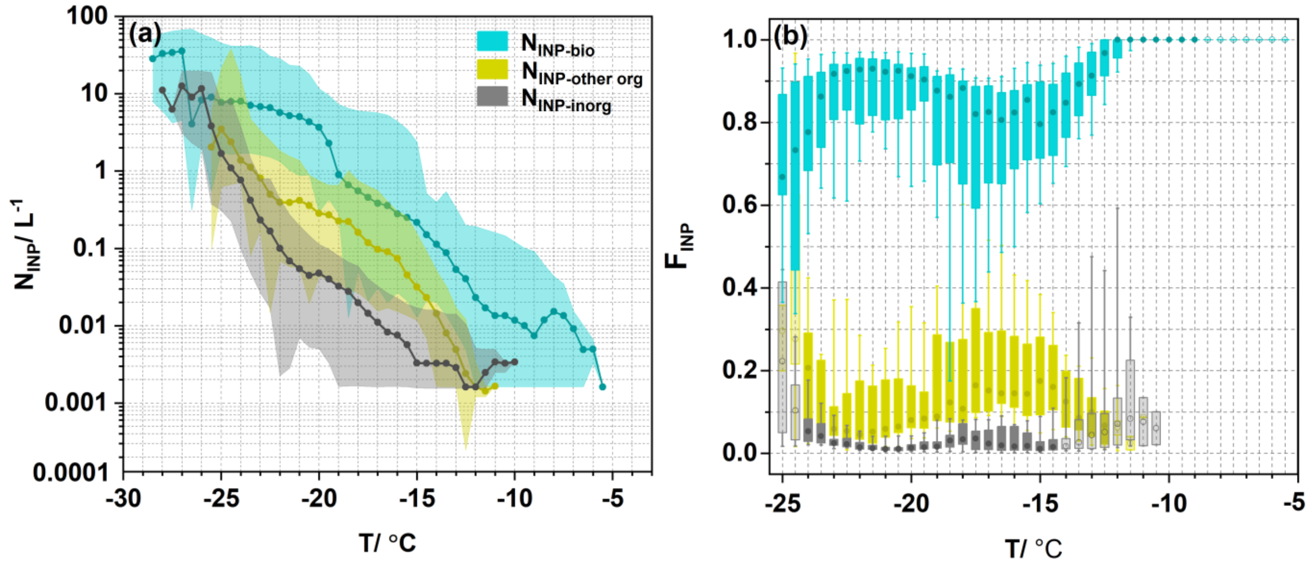

Figure 3. N_{INP} for different types of INPs and their fractions as functions of temperature. (a) The INPs spectra of biological INPs ($N_{INP\text{-}bio}$, blue dots), other organic INPs ($N_{INP\text{-}other\ org}$, yellow dots), and inorganic INPs ($N_{INP\text{-}inorg}$, gray dots). Each point represents the median value and the shadow area represents the maximum and minimum value. (b) 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.

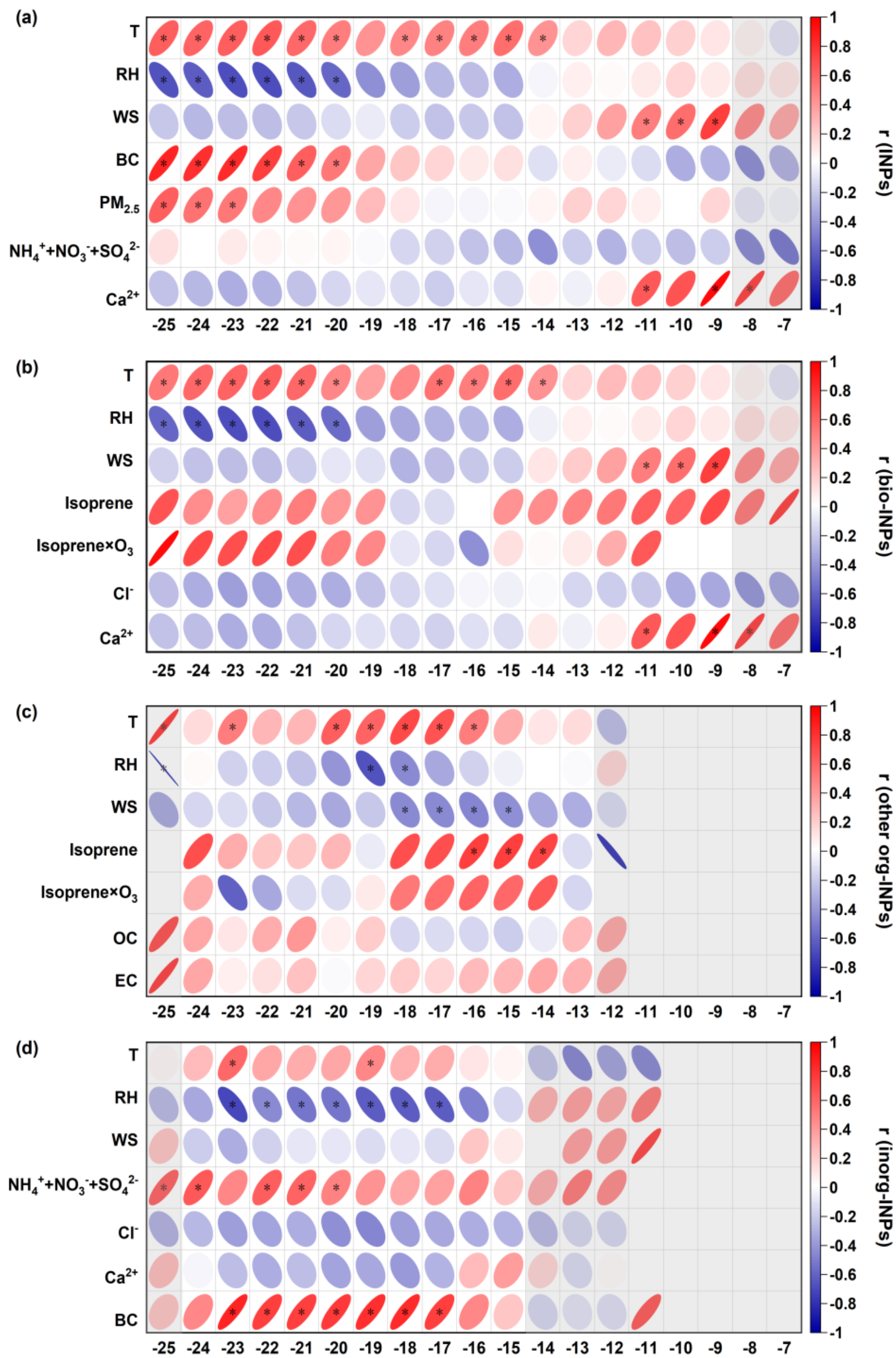

Figure 4. Correlation analysis between (a) N_{INP}, (b) $N_{\mathrm{INP\text{-}bio}}$, (c) $N_{\mathrm{INP\text{-}other\ org}}$, (d) $N_{\mathrm{INP\text{-}inorg}}$, with meteorological parameters and chemical compositions as functions of temperature. The r denotes the Pearson correlation coefficients. 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.

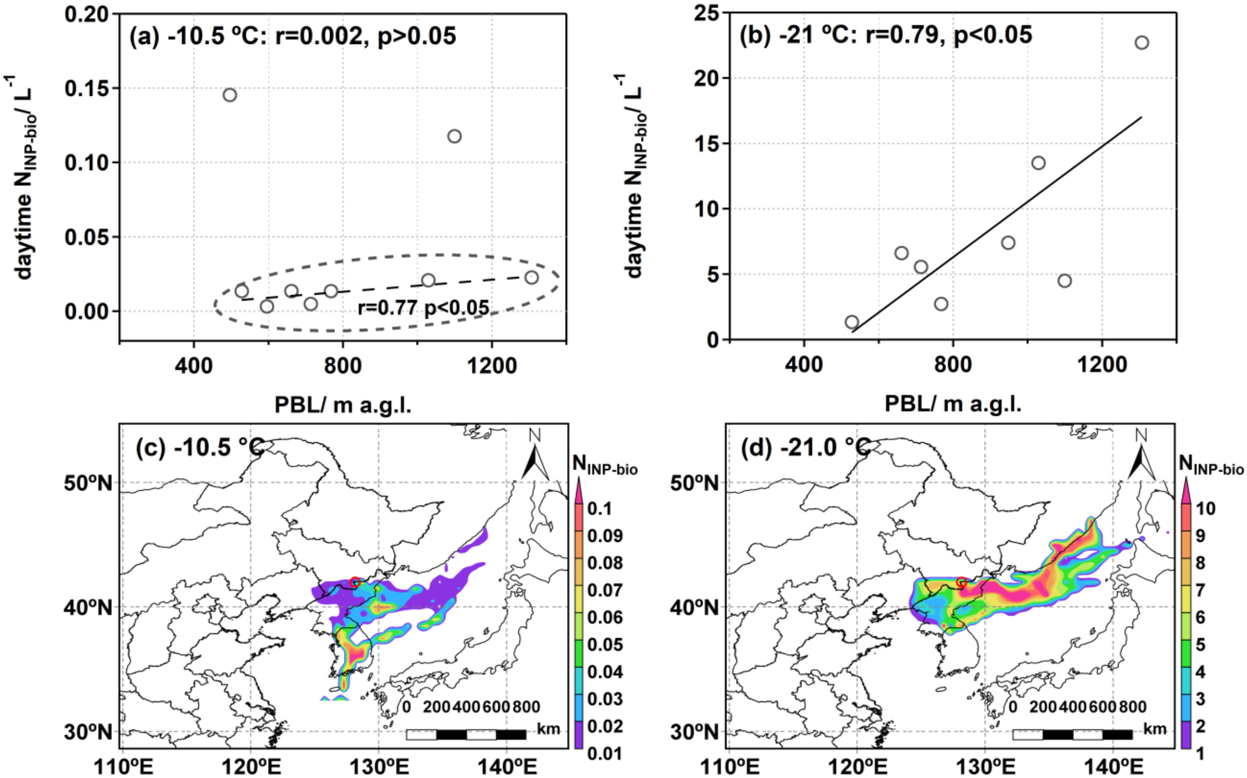

Figure 5. (a-b) Relationship between $N_{INP\text{-}bio}$ and average PBL height during the daytime (8:00–17:00 LT) at freezing temperature of −10.5 °C and −21.0 °C. The *r* denotes the Pearson correlation coefficients. (c-d) The concentration-weighted trajectory (CWT) analysis for the distribution of $N_{INP\text{-}bio}$ at −10.5 °C and −21.0 °C during the measurement. The red circle represents the Tianchi site.