Response to reviewer

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

Anonymous Referee #3

General comments

The manuscript by Sun et al. reports INP measurements at Changbai Summit in the summer of 2021. Using results from heat and H2O2 treatments, the authors distinguish between biological INPs, other organic INPs, and inorganic INPs and conclude that the majority of INPs in the temperature range studied were of biological origin. It is an interesting study that provides new information about biological INPs. They discuss correlations of biological INPs with meteorological data as well as with some chemical tracers, suggesting that biological INPs may originate from soil dust. In addition, possible sources and transport mechanisms of INPs are discussed based on knowledge of the height of the planetary boundary layer and the concentration-weighted backward trajectories of air masses. The authors have made an effort to explain their data, but given the small number of samples (22 in total, half of which were collected during the day and the other at night) and the lack of sample dilutions to extend the freezing spectra to lower temperatures, some sections require substantial revision in my opinion. I have major and minor comments that I hope will be helpful to the authors and can be addressed during the review process.

Response: We are grateful to the reviewer for the valuable comments. During the observation period, a total of 22 samples were collected. Recognizing the potential for uncertainties due to the limited data size, we have addressed this concern in the revised manuscript. Furthermore, we have conducted dilution of the untreated and heat-treated samples and extended the freezing temperature below -25 ℃. We updated the N_{INP} spectra and corresponding analyses in the revised manuscript. We have endeavored to respond to these valuable comments and revise our manuscript accordingly. Detailed point-to-point responses are shown below.

Specific comments

Abstract

1. Line 12: "modifying" is maybe not the right word here.

Response: We change the "modifying" to "modulating" in the revised manuscript.

2. Lines 22-24: While this insight into atmospheric dynamics is welcome, I would note that this positive correlation between biological INPs with planetary boundary layer height is based on 7 data points only. 2 outliers were excluded and 2 missing data points were not mentioned. It is not stated which correlation analysis was done, something that persists throughout the study. Furthermore, it is unclear for the reviewer how these two cases of high concentrations of biological INPs differ from the other samples in terms of long-range transport. These results need thorough review, discussions about them need to be clarified, and the relevant conclusions probably need to be revised.

Response: In the revised manuscript, we performed dilutions of the samples by the factors of 30, 60, and 120 times, ensuring that all samples reached a freezing temperature of at least -25 ℃. We re-calculated the Pearson correlation coefficients that include all samples, including the two high values. The correlations between PBL height and N_{INP} are presented in Figure R1.

The two missing data corresponded to the blank filters, and we have provided clarification regarding this in the revised manuscript. Nevertheless, the data points ranged from 6 to 11 when calculating the Pearson correlation coefficients, resulting in uncertainties. The uncertainties have been addressed in the revised manuscript. Since the revised correlation analysis included the two cases with high concentrations of biological INPs, we have refrained from adding extensive discussions on these two cases.

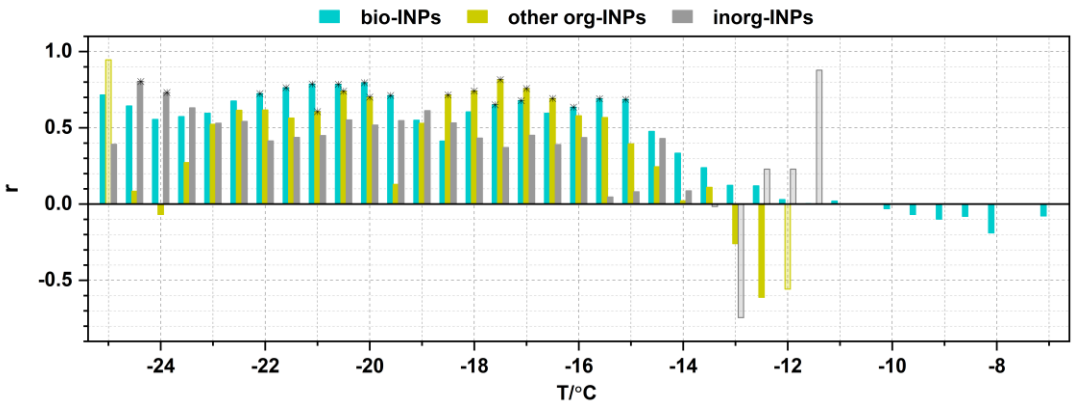

Figure R1. The relationship between PBL height and $N_{INP\text{-}bio}$, $N_{INP\text{-}other\ org}$ as well as $N_{INP\text{-}inorg}$ during daytime (8:00-17:00, m above ground level) as a function of temperature. The r denote the Pearson correlation coefficients. The asterisk indicates $p < 0.05$. The shades indicate that the data points number were less than half of all samples at each temperature.

Introduction

3. The reviewer thinks that some parts of the introduction should be worded more precisely.

Response: Thanks for the comment. We have briefly introduced the three aerosol types (biological aerosol, non-proteinaceous biological particles, and non-organic aerosols) in the revised manuscript.

4. Line 40 and line 52: Please use "etc" carefully. Try to list fully instead.

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

5. Line 40: There are also efficient proteinaceous cell-free INPs (Pummer et al., 2015) that are not embedded in cell membranes. Please, reformulate the sentence.

Response: In revision, we have revised the statement as follows: "It is generally demonstrated that biological aerosols, which have proteins present in cell-free macromolecules or outer membranes of intact cell, are the most efficient 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: I am unsure what is meant by "and [in] agricultural soils". Do you mean a site, where presumably many INPs come from particles from agricultural soils? Please reformulate.

Response: The term "and [in] agricultural soils" means the presence of INPs from regions with agricultural soils. In the revised manuscript, we have replaced "agricultural soils" with "agricultural regions" to clarity.

7. Line 68: The tree sites that are referred to are not all located in the Swiss Alps, but are located across Switzerland.

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

8. Lines 70-71: Instead of "one order of magnitude", you could keep the same units as in the previous sentence. Maybe summarize in percent per kilometer.

Response: In the revised manuscript, we replaced "one order of magnitude" with "exceed 60%".

9. Line 72: The "atmosphere" can only be in singular.

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

10. Lines 72-75: Please mention that Schrod et al. 2017 findings were obtained during specific events, i.e., a series of elevated Saharan dust plumes.

Response: In the revision, we have revised as: "In contrast, Schrod et al. (2017) reported an increase in INPs abundance of approximately 10 times over the eastern Mediterranean (2.5 km 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."

11. Lines 75-77: The citation Conen et al. 2022 is missing at the end of the sentence. I would suggest adding here the following specification: "under free-tropospheric conditions". Study could have been mentioned in L. 44-45 instead/as well.

Response: Thanks for the comment. We have cited the reference and added the content in the revised manuscript.

12. Line 87: "INPs measurements" --> "INP measurements"

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

Methods

The reviewer found that the methods are well described, and that it was helpful that the authors provided the raw data for the reviewers.

13. Line 119: Please specify that your times are meant in local time or mention the correct time zone.

Response: We used the local time and have added the description in the revised manuscript.

14. Line 120: A total of 24 samples were collected for INP analysis. The results of only 22 samples were reported in the provided data file. Could the authors please explain why the results of two samples are missing? Could it maybe be that 22 samples and 2 background filters were collected?

Response: Yes, the 24 samples include two background filters. In the revised manuscript, we have modified the statement to: "A total of 24 samples were collected on PCTE filters from August 10 to 24, 2021 in local time (LT), which includes 2 blank filters." In addition, detailed sampling information has been added in Table S1 in the Supporting Information, as presented in Table R1.

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

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

15. Line 130: It would be interesting to know how far the weather station is from the measurement site.

Response: We added the distance description in the revised manuscript: "Meteorological data, such as temperature, humidity, WS, wind direction, pressure, and precipitation, were monitored by the Tianchi weather station, a national meteorological station located approximately 20 m from the sampling site."

16. Lines 178-186: Please be more precise how you generated the air mass backwards trajectories and add information about starting time as well as number of trajectories per sample.

Response: The air mass backward trajectories cover hourly data from 10 August to 21 August, resulting in a total of 264 trajectories generated. These have been added in the revision.

17. Line 188: Which data product from the Climate Data Store did you use for the PBL data?

Response: The PBL data was obtained from the fifth-generation ECMWF global atmospheric reanalysis (ERA5 reanalysis). We have added this information in the revised manuscript.

Results and Discussion

The reviewer found that some parts of the results and discussion need thorough revision and much clearer statements.

18. Line 208 and lines 210-211: Wieder et al. (2022) contains INP concentrations from a mountain site called Weissfluhjoch (2693 m a.s.l.) and a valley site called Wolfgangpass (1631 m a.s.l.). The two sites in the Swiss Alps are only 4 km apart and are not necessarily representative of the entire Swiss Alps. Please be more specific: a) Swiss Alps and not any Alps, b) add the name of the site(s) you mean.

Response: Thanks. We have revised "Alps" to "Weissfluhjoch" in the revision.

19. Lines 208-211: Please provide the time spans for collection and averaging for these other studies. For example, Wieder et al. (2022) collected samples over short time spans (i.e., 20 minutes) and averaged them into 2-hour bins. They found a peak at 19 h UTC (i.e., 21 h CET), which is defined as nighttime based on the definition used in this manuscript, if I am not mistaken. A possible diurnal cycle at Changbai Mountain may have been averaged out due to the long sampling time of 11 hours. Please mention specifically for Changbai Mountain only the comparison of daytime and nighttime, since only two samples are taken per day. In my opinion, a conclusion over the entire diurnal cycle cannot be made here.

Response: Thanks for the comment. The time spans have been added in the revision. In this

study, our objective is to compare differences in INPs concentration between daytime and nighttime, rather than delve into an extensive analysis of diurnal variations. In the revision, we have added the statement as follow: "Additionally, the limited data size and low sampling frequency may also result in a lack of diurnal variation in this study."

20. Line 213: What is meant by "the temperature spectra showed a wider range"?

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

21. Line 217: What is meant by "the NINP value was much larger at Mt. Huang than our results"? Can't the difference between both sites be explained by the fact that your results cover a warmer temperature range than the results from Mt. Huang? Please mention the freezing temperatures while comparing INP concentrations.

Response: Based on our updated N_{INP} values after the dilution procedure, we have revised the statement as follows: "Jiang et al. (2015) reported INP concentrations at the top of Mt. Huang, where the temperature spectra of N_{INP} exhibited a much narrower range between -15.0 ℃ ~ -23.0 ℃ compared to those observed at the Weissfluhjoch. The N_{INP} value at Mt. Huang ranged from 0.1 L^{-1} to 11.9 L^{-1}, partially overlapping with our results in the temperature range from −15.0 ℃ to −23.0 ℃."

22. Line 256: Shouldn't it be "bio-INP" instead of "INP"?

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

23. Line 254-256: A mean increase in FINP-bio from 0.8 to 0.9 with decreasing temperatures between -16.5 ℃ and -20 °C doesn't make much sense and could be due to the way the data were processed here. It looks like some untreated and heat-treated samples already reached the upper detection limit at about -15 ℃ or -16 ℃. In order to draw conclusions about the proportion of bio-INPs, other org-INPs, and inorganic INPs below -15 ℃, I believe dilutions of the untreated and heat-treated samples would have been required. The number of data points of FINP-bio from maybe around -18 ℃ and lower is likely too low to draw any conclusions below that temperature threshold.

Response: In the revised manuscript, we diluted the untreated and heat-treated samples by the factors of 30, 60, and 120 times, and extended the freezing temperature below -25 ℃ (details can be found in the response to comment 2). Based on the updated N_{INP} data, we have made revisions to the boxplot, as shown in Figure R2. It can be found that as the temperature decreased from -16.5 ℃ to -21.5 ℃, the median value of $F_{INP\text{-}bio}$ increased from 0.8 to 0.9. We are confident that this increase accurately represents the presence of biogenic INPs that exhibit relatively high ice-nucleating activity in the LTR.

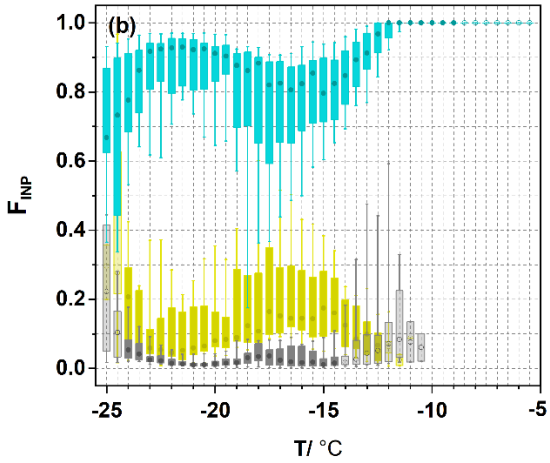

Figure R2. Boxplot of fractions of bio-INPs ($F_{INP\text{-}bio}$, blue boxplot), other org-INPs ($F_{INP\text{-}other\ org}$, yellow boxplot), and inorganic INPs ($F_{INP\text{-}inorg}$, gray boxplot) as functions of temperature. The upper and lower extents of the boxes represent the 75th and 25th percentiles, 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.

24. Lines 277-281: What about biological INPs originating from plants and oceans? Please discuss.

Response: Because N_{INP} was positively correlated with WS and Ca^{2+}, we emphasis the discussion of soil source. Section 3.4 discusses marine and plant sources.

25. Lines 279-281: Maybe instead of citing the review, authors could cite the original studies e.g. Hill et al. 2016 or others.

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

26. Line 301: Maybe instead of citing the review, authors could cite the original studies.

Response: Thanks. We cited the reference, i.e., Cozic et al. (2008) and Levin et al. (2016), in the revision.

27. Lines 315-321: Could the authors elaborate how the inclusion of these points would change the results and conclusion?

Response: We have update the N_{INP} data based on the diluted process, and the relationship between $N_{INP\text{-}bio}$ and PBL have revised as shown in Figure R1. In the HTR, there was no correlation between bio-INPs and PBL. However, upon excluding the two high INP cases at-10.5 ℃, the remaining seven cases exhibited an increasing trend in bio-INPs as PBL height increased ($r=0.77$, $p<0.05$,), as shown in Figure R4a. The two high INP cases may be source

from potential strong but infrequent local sources, which have been added in the revision.

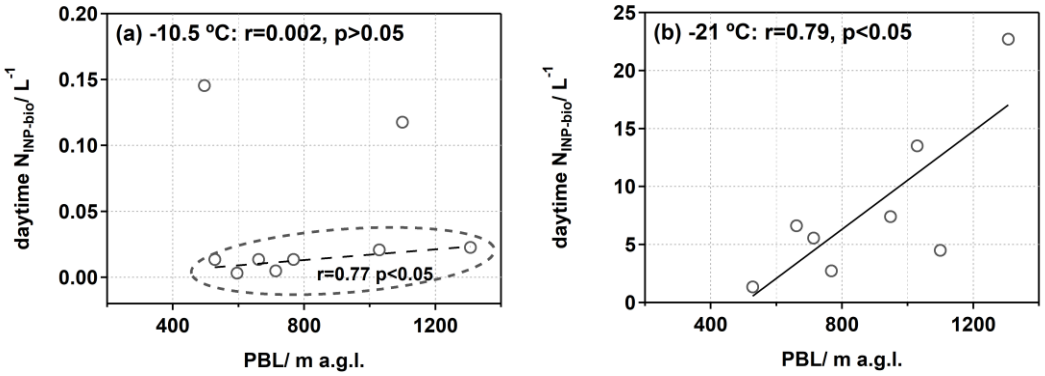

Figure R3. Relationship between $N_{INP-bio}$ and PBL height during the daytime (8:00–17:00 LT) at freezing temperature of −10.5 ℃ and −21.0 ℃. The r denotes the Pearson correlation coefficients.

28. Line 320: Please indicate here that the "freezing" temperature is meant.

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

29. Lines 323-325: How does the CWT analysis on August 18 and 25 differ from the other days?

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

Conclusion

The reviewer believes that some rewording needs to be done, especially in the last paragraph.

30. Lines 365-368: A decreasing trend in FINP-bio with decreasing temperature suggests that other organic or inorganic INPs become more important with decreasing temperatures. It does not suggest that the ice nucleation activity of bio-INPs decreases. In addition, due to the lack of data points in the low temperature regime, no conclusions can be drawn regarding FINP-bio for that temperature range, in my opinion. Dilutions would have been required, as mentioned above.

Response: We agree and delete the statement about "suggesting that the ice nucleation activity of bio-INPs decreases with decreasing temperature." We have added data for the low freezing temperature range, confirming the conclusions regarding $F_{bio-INPs}$. Please see our response to comment 23.

31. Line 379: Did you mean to write that the INP number concentration is lower than the ice crystal number concentration, especially in the warm temperature range?

Response: Considering the last paragraph was not closely aligned with the context of this paper, we have removed it in the revision.

32. Line 381: Is there a word missing between "…is second only to…"?

Response: Considering the last paragraph was not closely aligned with the context of this paper, we have removed it in the revision.

33. Line 383: Please delete "through various collisions with pre-existing ice". There are several multiplication processes, and they involve many more processes than only ice-ice collision. More information can be found for example in Korolev and Leisner, 2020.

Response: Considering the last paragraph was not closely aligned with the context of this paper, we have removed it in the revision.

34. Line 385: Bio-INPs do not impact secondary ice formation directly, please reformulate.

Response: Considering the last paragraph was not closely aligned with the context of this paper, we have removed it in the revision.

Figures

Fig.2(b)

35. Please indicate in the figure and legend the name of the mountain in the Swiss Alps from which the data originate similar to what you did with the Mt. Huang dataset.

Response: We have replaced the name to "Weissfluhjoch".

36. Should the y-axis perhaps be extended to lower values to see the lower limit of the error bar?

Response: We have modified the y-axis to display all the data. But to ensure that the information in the figure can be readily grasped, we did not display the error bar in Figure 2b. The error bar of whole samples was shown in Figure S3, as illustrated in Figure R4.

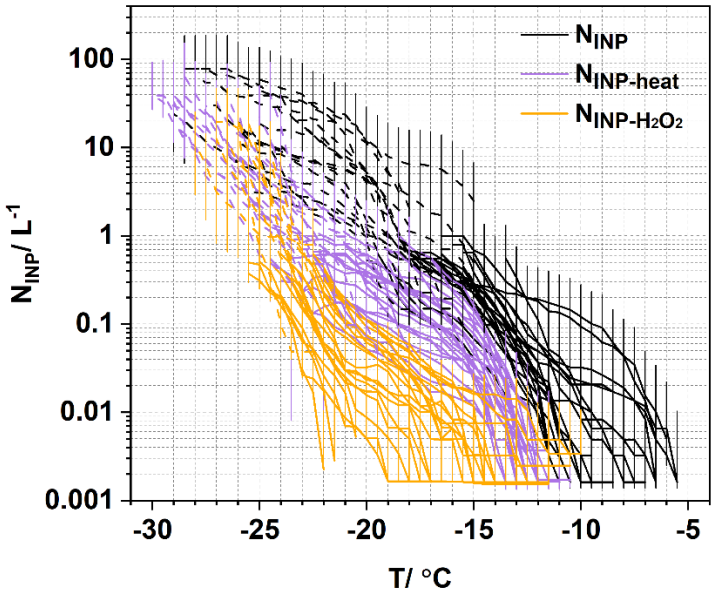

Figure R4. The N_{INP}, $N_{INP\text{-}heat}$, and $N_{INP\text{-}H2O2}$ as function of temperature. The solid line and dotted line show the sample measurement result by immersed in 5 mL MilliQ water and diluted the sample 30-120 times, respectively. The original N_{INP} is marked by black dots, $N_{INP\text{-}heat}$ is marked by purple dots, and $N_{INP\text{-}H2O2}$ is marked by pink dots, with 20% error bars indicating the 95% confidence intervals.

Fig. 3(a)

37. Why are the error bars for NINP-H2O2 below -15 ℃ extending to values outside the graph?

Response: We adjusted the y-axis range to display all error bars.

38. Please use the same axes in Fig. 3a than in Fig. 3b and Fig. 2b.

Response: We have revised the manuscript accordingly.

Fig.3(c)

39. I appreciate that there is an indication of the number of data points included in the analysis. Since the upper detection limit of about 2 INPs per liter is reached around -15 ℃ for NINP-bio, dilutions would have been necessary to extend the plot from -15 to -20 ℃ as mentioned above.

Response: In the revised manuscript, we extended the freezing temperature below -25 ℃ by diluting the samples.

40. How can the median fraction of inorganic INPs above -12 ℃ be more than 0.0 if the

median fraction of bio-INPs is 1.0?

Response: We first calculated the daily proportions and then calculated the average the proportion across all samples.

Fig.4

41. What kind of correlation analysis did you do? Please specify.

Response: We calculated the Pearson correlation coefficients, which has been added in the revised manuscript.

Fig.5(a-c)

42. What does "r" stand for? Please also describe what the red lines and the purple circles represent.

Response: The r denotes the Pearson correlation coefficients. We have described the manuscript accordingly. The purple circles represented the exclusion of two high-value data points, and the red line indicated their correlation. However, as mentioned previously, we have updated the data and no longer exclude high values. Instead, we have performed a correlation analysis on the entire dataset. Therefore, revisions have been made, as illustrated in Figure R3.

43. Is the x-axis showing the mean PBL height?

Response: Yes, the x-axis showed the mean PBL height. We have revised the figure caption to "average PBL height during the daytime".

44. Maybe add "freezing" between "three" and "temperatures".

Response: We have revised the manuscript accordingly.

Fig.5 (d-f)

45. Trajectories of which samples were included in the CWT analysis? Are the trajectories of all the samples included or only those of the daytime samples? Were the trajectories of the two outliers excluded or included here? Please be more specific.

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 $N_{INP\text{-}bio}$ at −10.5 ℃ and −21.0 ℃ during the measurement."

46. Also describe in the legend what the star and triangle represent.

Response: We have revised the manuscript accordingly.

Fig. S1

47. Please describe in the legend precisely what is shown and what is meant by "R". Is the large "R" here similar to the small "r" in Fig. 5?

Response: The "R" here was same as the "r" in Figure 5. We used “r” consistently throughout the revised manuscript.

Fig. S2

48. Authors could add the star and the triangle in these maps, similar to Fig. 5 (d-f).

Response: We have revised the manuscript accordingly.

Reference:

Conen, F., Yakutin, M. V., Yttri, K. E., and Hüglin, C.: Ice Nucleating Particle Concentrations Increase When Leaves Fall in Autumn, Atmosphere-Basel, 8, 202, https://doi.org/10.3390/atmos8100202, 2017.

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Huang, S., Hu, W., Chen, J., Wu, Z., Zhang, D., and Fu, P.: Overview of biological ice nucleating particles in the atmosphere, Environment International, 146, 106197, https://doi.org/10.1016/j.envint.2020.106197, 2021.

Jiang, H., Yin, Y., Su, H., Shan, Y. P., and Gao, R. J.: The characteristics of atmospheric ice nuclei measured at the top of Huangshan (the Yellow Mountains) in Southeast China using a newly built static vacuum water vapor diffusion chamber, Atmos Res, 153, 200-208, https://doi.org/10.1016/j.atmosres.2014.08.015, 2015.

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importance of soot aerosol, J Geophys Res-Atmos, 121, 5888-5903, https://doi.org/10.1002/2016jd024879, 2016.

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Petters, M. D. and Wright, T. P.: Revisiting ice nucleation from precipitation samples, Geophys Res Lett, 42, 8758-8766, https://doi.org/10.1002/2015GL065733, 2015.

Phelps, P., Giddings, T. H., Prochoda, M., and Fall, R.: Release of cell-free ice nuclei by Erwinia herbicola, Journal of Bacteriology, 167, 496-502, https://doi.org/10.1128/jb.167.2.496-502.1986, 1986.

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