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.

Major comments:

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

The influence of fog and precipitation at the sampling site on the measured N_{INP} 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.

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?

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.

2) Line 22ff: Provide an explanation why PBL height could be positively correlated to N_{INP}. 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 N_{INP}.
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.
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.
5) Line 43ff: Not corrected as mentioned in response to Referee#3 question 5. Implement correction.
6) Line 45: Can lichen be considered biological aerosol? As I understand they are a symbiotic organism attached to surfaces.
7) Line 48: Double check if it is correct that pollen is non-proteinaceous. I find they contain about 30% protein.
8) Line 51: add a citation for the activity temperature of mineral dust and sea spray.
9) Line 55: inappropriate citations: references are for deposition ice nucleation, replace with references relevant for immersion freezing.
10) Line 59-62: Provide references for “numerous studies”, “spatial distribution heterogeneity”, and the altitude of mixed-phase clouds.
11) Line 69f: INP sources are not in the atmosphere but on the surface. Reformulate.
12) Line 74ff: JFJ station is not a good example for having high vegetation coverage. It is surrounded by bare rock and ice. Reformulate.
13) Line 83f: Clarify what is meant by “impact of bio INP on cloud droplets and their contribution to formation of precipitation”.
14) Line 91: Explain what boundaries are meant by “transboundary transport of air mass”.
15) Line 98: provide a reference for pollution transport to the Arctic from this region.
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 N_{INP} in the LTR with RH in Fig.4.
17) Line 116f: Elaborate on the potential impact of the surrounding (lake, dense vegetation) on N_{INP} and other variables, for example isoprene concentrations.
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?
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.
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 N_{INP}.
21) Line 126: provide model number for the $PM_{2.5}$ sampler.
22) Explain how auxiliary data was averaged for the correlation analysis with N_{INP}.
23) Line 139: is the enclosed droplet chamber part of the LTS120 cold stage?
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.

25) Line 148: “filled” might be the wrong word to describe the procedure. Do you mean covered?
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.
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.
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.
29) Line 209: As pointed out above, add equation to Sec.2.3. how the background was subtracted and refer to the equation here.
30) Line 241f: You could cite Kanji et al.,2020, here. Correct sentence structure.
    There are significant correlations between BC and N_{INP} 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.
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.
32) Line 287f: Specify where these previous studies were conducted and provide references.
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 Ca^{2+} 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} (N_{INP} in the -8°C to -11°C range) influence the measured $Ca^{2^{\circ}}$ concentration?
34) Line 298f: Provide an explanation for the correlation of N_{INP} to ambient temperature and RH.
35) Line 313f: Explain why a correlation with WS and ambient temperature indicates local sources.
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.
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.
38) Line 323: The N_{INP} was measured in this study not the IN activity. Reformulate.
39) Line 338f: Provide a reasonable explanation how PBL height can be positively correlated to N_{INP}. The opposite could be expected due to dilution of air with increasing PBL height. Total particle concentration is usually anticorrelated to PBL height.
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 N_{INP}?
41) Line 350f: Clarify how N_{INP} 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.
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 N_{INP} the occurrence of high N_{INP} at -10.5°C is correlated to high N_{INP} 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 N_{INP} colour scale indicates that dilutions were not included.

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.
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 N_{INP} occurs exclusively by night and as soon as the boundary layer forms exclusively local sources contribute to N_{INP}? This seems a critical assumption for your analysis and should be discussed in more detail and supported by stronger evidence.
45) Line 381ff: There was no diurnal cycle of N_{INP} observed in this study. Explain how conclusions can still be drawn about a diurnal cycle.
46) Line 385f: The methods applied in this study do not allow to explore properties of INPs. Only their abundance was measured. Reformulate.
47) Line 393: An increase in bio N_{INP} 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.
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 N_{INP} measurements, or at -10°C and -20°C? Would the correlation analysis, for example with Ca^{2+} differ if done separately for daytime and nighttime samples?
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.
50) Figure S6: Specify if only daytime or all samples were used to create this figure.

Technical corrections:

1) misspelled citations: DeMott, O'Sullivan, McCluskey, in several instances.
2) there is no "the" needed before Switzerland.
   Also, in Figure 2.b) remove "The" in front of Weissfluhjoch and Jungfraujoch.
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: $0.5 ugm^{-3} – 1.0 ugm^{-3} = - 0.5 ugm^{-3}$ is unphysical.
4) Line 166-168: It is unclear what is explained here. Reformulate.
5) Line 173: Do you mean: …, which is 1.96 for a 95% confidence interval? Reformulate.
6) Line 191ff: Incomprehensible sentence. Reformulate.
7) Line 198: define n_{ij} in the text.
8) Line 213: replace "three" with "up to 5" to be consistent with what is stated in the conclusion.
9) Line 233-235: Incomprehensible sentence. Reformulate.
10) Line 237: Reformulate. …magnitude at temperatures from -26.0°C to -3.0°C.
11) Line 288: replace INPs with N_{INP}
12) Line 321: define SNA.
13) Line 329: replace INPs with N_{INP}.
14) Line 336: Define HLR. Do you mean HTR?
15) Line 347: "under immersion mode" do you mean "in the immersion mode"?
16) Line 352: "occurred in" do you mean "coming from"?
17) Line 363: define "the middle layer".

18) When reporting N_{INP} ranges and consecutive the temperature range, I suggest giving the temperature range from high to low temperature if the N_{INP} range is given from low to high, so that the first number in the N_{INP} range corresponds to the first number in the temperature range.
19) Specify whenever you mean “ambient temperature” to avoid confusion to experimental temperature of IN experiments. In particular: line 299, 313.

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