Response to RC1

We have reproduced reviewer comments in **bold** font, and our responses in regular font.

General comments

The manuscript investigates the seasonal and diurnal variability in ice nucleating particle oncentration (INP) measured over a year at the High Altitude Research Station Jungfraujoch. It represents the longest continuous measurement of INPs to date with a high time resolution of 20 minutes. A easonal trend in INPs is observed with highest concentrations occurring in Spring and lowest concentrations occurring in Winter. A diurnal trend in INPs is also identified for air masses with boundary layer intrusions. The study identifies long term trends in INP concentrations and is a valuable contribution to the field of INP research. However, I believe that the discussion of the data represented in the figures could be clearer and further links between potential INP sources and with previous literature studies could be made. I therefore recommend the manuscript for publication in ACP following appropriate response to the following comments.

We thank Reviewer 1 for their positive evaluation of the manuscript, and respond to their comments individually below with corresponding modifications to the manuscript indicated by the respective line numbers in the revised version.

Specific comments

1. The results section contains very detailed analysis and lots of information is contained within each figure. It would be clearer and easier for the reader to follow the discussion if the panel or the section of the figure that is being discussed is regularly referred to in the text.

e.g. for Figure 2:

- **page 8, line 204: 'Dividing BG periods into FTBG and BLIBG…' Please refer to panel c).**
- **Page 8, line 216: '…is apparent in April for the total particle concentration.' Please refer to panel f).**
- **Page 8, line 217: '…total particle number concentrations remained at summer levels also in September' Please refer to panel d).**

We agree, and made the above recommended changes to refer to Fig. 2 (see lines 203, 213, 215, and 217 resp. in revised version)

This comment applies to all figures but especially to figures 2, 5 and 6.

We agree, and have added references to Fig. 5 in the text (see lines 329, 331, 333, 335, 337, 339 - 341, 344 in revised manuscript), Figure 6 (lines 352, 354-355, 359-360, 363) and for Figure 2 we modified the references based on the specific comments of reviewer 1 above.

Additionally it would also help the reader if colours were referred to in the text when discussing the data, e.g. in Figure 5, page 14, line 322-323: '…shows a weak diurnal cycle, with a maximum of 629 std cm-3 at 13 h UTC and a …' Please add (black line in panel a)).

We have now made references to the colour of the lines and traces as well in the revised manuscript when we refer to the specific figure panels (see response to comment above).

2. Pages 9-11 contains a detailed discussion of pollen as the potential INP source for the high INP concentrations measured in April. Whilst this discussion is interesting, I believe it could be reduced as

the overall conclusion is that it is unlikely that pollen is responsible for the high INP concentrations in April (without further pollen measurements at JFJ). Why do you not comment on any other potential sources for the high INP concentration in April? Was any back trajectory analysis of air masses performed that could inform on potential INP sources?

We believe the discussion on pages 9-11 is necessary to conclude that we cannot rule out the contribution of sub-pollen particles or pollen fragments to the peak INP concentrations observed in April, which we now make explicit (see lines 264 – 277 in revised manuscript). We now specify that we only rule out the influence of pollen grains because the HINC-auto inlet had a D_{50} size cut-off of 2.5 µm (see lines 248 initial manuscript and line 256 revised manuscript) which does not allow sampling of pollen grains. We have now made it more clear in the conclusions section that the role of sub-pollen particles and pollen fragments cannot be ruled out (see lines 391 – 393 in revised manuscript).

We have conducted a back trajectory analysis (see Fig. 1 below) for and exemplary time during the peak INP concentrations. Fig. 1 below shows the back trajectory analysis initiated for April 22nd at 1200 hrs UTC. The analysis shows that the air masses with recent ground contact were advected from either side of the main Alpine ridge, i.e., from both north or the south of JFJ, thus it is possible that sub-pollen particles could be transported to the JFJ from the south (Italy) or other regions of Switzerland. However, it is clear that trajectories do not arrive from Bern. Trajectories could have arrived at JFJ on April 22nd from the region near Visp, however, only after April 19, when the pollen concentrations had already declined (i.e. after April 11-15). Thus, we do not believe these two locations would contribute to the INP concentration peak observed (see Figure 3 in revised manuscript) during April 20-25th. The sources of sub-pollen particles could have been from elsewhere in Europe as also suggested by RC2. We now add this modification in lines 266-270 of the revised manuscript.

Were any samples collected (gas or filters) and analysed for chemical composition?
No filters or gas samples were collected that matched the time resolution of HINC-Auto for this period or analysed. Filter chemical composition could provide insight, but the resolution is on the order 24-hours, whereas the analysis here is obtained by having the higher 20 min time resolution. However, after a longer-term data set has been collected by HINC-Auto, the PM filter composition with resolution of 24-hrs could provide beneficial knowledge for future studies.

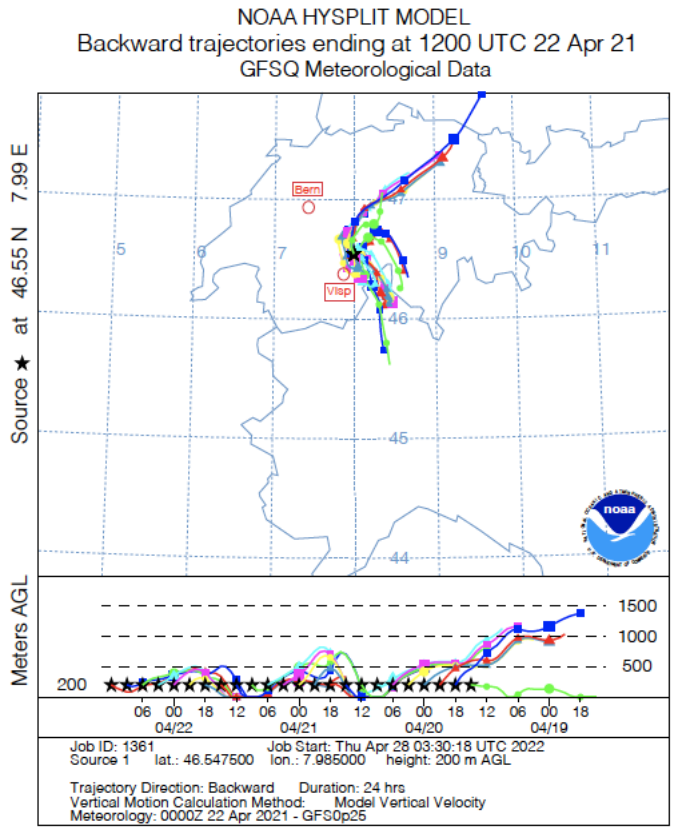

Figure 1. Back trajectories(HYSPLIT, NOAA) [Rolph et al., 2017; Stein et al., 2015] ending at JFJ at 12.00 UTC on April 22nd showing no air masses arrive from Bern and Air masses arriving from Visp would only have passed over Visp after April 19, when pollen concentrations in Visp had already declined. The colours represent different trajectories starting every 3 hours going backwards. (https://www.ready.noaa.gov/HYSPLIT_traj.php).

3. The introduction discusses trends in seasonal and diurnal variability in INP measurements in the literature from various studies using mostly offline analysis. It would be good to make links back to the findings of these studies during the results section for comparison i.e. similar seasonal dependences were observed.

We have now added a paragraph in section 3.1 (see lines 222-230 in revised manuscript) to compare the seasonal cycle observed in this study compared to the other studied mentioned in the introduction.

For the comparison of the diurnal INP cycle to the literature, see comment 5 below.

4. The introduction states that knowledge of seasonal and diurnal variability will help to understand the sources and sinks of INPs. The conclusion only briefly mentions that the observed seasonal variation of INP concentrations could be linked to partitioning of particles in different seasons. As this appears to be the main motivation for the measurements, this discussion should be expanded in either the results or conclusion section.

We have dedicated the entire section 3.1 of the manuscript to the discussion of the seasonality of INP at JFJ. Motivated by the reviewer comments we have added a statement in the conclusions elaborating further on the seasonality observations (see lines 381-389 in revised manuscript).

Can any further information on sources and sinks of INPs at JFJ be obtained from this study?

Recently *Lacher et al.* [2021] showed the need for a significant number of additional necessary instruments in addition to an INP counter at Jungfraujoch during a field campaign to categorically and correlationally support the presence of other sources contributing to INP populations at JFJ. Indeed these other sources included particles of marine origin, dust and anthropogenic sources. However, this required a suite of mass spectrometers and offline chemical composition measurements of ice residuals to be running in parallel. A much bigger effort with chemical composition identification during the continuous monitoring for 12 months would be necessary. As such, a detailed assessment of sources is beyond the scope of the data set of monitoring presented here. We now make a statement in the conclusions referencing these sources and a study conducted at the same site with an instrument similar to the one used here, but a non-automated version (see line 393 revised manuscript). We also add to the conclusions that majority of the particles observed as INPs at -30 °C at JFJ should be from dust (Brunner et al., 2021) (see line 396 revised manuscript)

5. Comment: The only other study to have observed diurnal variation in INPs over a longer time period is mentioned on page 3, lines 65-69 (Wieder et al., 2021 in prep.). It would be useful to make further comparisons between this study (data in Figure 5) and that of Wieder et al., however, as the manuscript is in prep this is not possible.

We thank the reviewer for this comment. Indeed *Wieder et al.* [now 2022] is no longer in review and instead published. A comparison between the two studies is in principle possible but needs to be caveated. The diurnal cycle in *Wieder et al.* [2022] caused by advection and orographic lifting could generally apply to the JFJ site as well. However, the meteorological transport assessed in much greater detail and extremely specific to the topography/valley system of the Davos region with data during the field campaign taken from a wide variety of observations set-up at several sites adjacent to the site observing INP concentrations. In *Wieder et al.* [2022], we measured INP concentrations at a mountaintop and a high valley site, simultaneously and comparing these two sites, allowed us to reach a conclusion on how topography-influenced meteorology impacted the diurnal cycle at the mountaintop but not in the valley.

We now mention the agreement in the conclusions section (see lines 402-404 in revised manuscript) and add a few sentences to discuss the nature of the diurnal concentration in section 3.2 (see lines 370-374 in the revised manuscript).

<u>Technical corrections</u>

Page 7, line 196-7: the text states that 'June had the most active SDE of the investigated period with a duration of 116 h' whereas in Figure 2 it appears that the SDE in June lasts for 123 hours. Please correct.

Thank you for the correction, now corrected (see line 196 in revised manuscript)

Page 16, line 352: the text states 'The large particle concentrations continue to decrease between 9-12 h UTC…' which I think should be 21-24 h UTC from the data presented in Figure 6, panel f). Please correct.

We have corrected this (see line 366 in revised manuscript)

Figure A3 is not mentioned in any part of the paper. Is this needed?
Thank you for catching that, the reference to Figure A3 is now made in the results section to show the annual overview time series of INP concentrations and related parameters used to assess the seasonal and diurnal variability of INP concentrations (see lines 169 – 171 in revised manuscript)

Typing errors/grammar:

Page 1, line 13: '…is with a factor of…' should be changed to 'is within a factor of'.
Corrected (line 13 revised manuscript)

Page 3, line 81: 'Furthermore, the remote location allows to study…' should be 'Furthermore, the remote location allows the study of…'
Corrected (line 81 revised manuscript)

Page 4, line 98: unites should be units.
Corrected (line 98 revised manuscript)

Page 6, line 181: 'There were two exceptionally dry period in the end…' should be 'There were two exceptionally dry periods at the end…'
Corrected (line 180 revised manuscript)

Page 11, line 291: '…uncertainty can alter the frequency distributing…' should be '…uncertainty can alter the frequency distribution…'
Corrected (line 303 revised manuscript)

References

Lacher, L., H. C. Clemen, X. Shen, S. Mertes, M. Gysel-Beer, A. Moallemi, M. Steinbacher, S. Henne, H. Saathoff, O. Möhler, K. Höhler, T. Schiebel, D. Weber, J. Schrod, J. Schneider, and Z. A. Kanji (2021), Sources and nature of ice-nucleating particles in the free troposphere at Jungfraujoch in winter 2017, *Atmos. Chem. Phys.*, *21*(22), 16925-16953, doi:10.5194/acp-21-16925-2021.

Rolph, G., A. Stein, and B. Stunder (2017), Real-time Environmental Applications and Display sYstem: READY, *Environmental Modelling & Software*, *95*, 210-228, doi:https://doi.org/10.1016/j.envsoft.2017.06.025.

Stein, A. F., R. R. Draxler, G. D. Rolph, B. J. B. Stunder, M. D. Cohen, and F. Ngan (2015), NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, *Bull. Amer. Meteorol. Soc.*, *96*(12), 2059-2077, doi:10.1175/BAMS-D-14-00110.1.

Wieder, J., C. Mignani, M. Schär, L. Roth, M. Sprenger, J. Henneberger, U. Lohmann, C. Brunner, and Z. A. Kanji (2022), Unveiling atmospheric transport and mixing mechanisms of ice-nucleating particles over the Alps, *Atmos. Chem. Phys.*, *22*(5), 3111-3130, doi:10.5194/acp-22-3111-2022.