Response to RC2

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

Review of "The diurnal and seasonal variability of ice nucleating particles at the High Altitude Station Jungfraujoch (3580 m a.s.l.), Switzerland" by Brunner et al.

The paper from Brunner et al, reports seasonal variability and diurnal variability of INP concentration at the JFJ site during the year 2020. This is this year the third paper in this series of INP measurements at JFJ. The first technical paper appeared earlier this year in AMT, describing the auto-HINC, a new CFDC device enabling continuous INP measurement at the JFJ. "Continuous online monitoring of ice-nucleating particles: development of the automated Horizontal Ice Nucleation Chamber (HINC-Auto)". A second paper, "The contribution of Saharan dust to the ice nucleating particle concentrations at the High Altitude Station Jungfraujoch (3580 m a.s.l.), Switzerland" currently in ACPD presents one year (2020) data of INP attributed to Sarahan dust and measured at JFJ. This current third paper is pushing the analysis further by looking more carefully at the seasonal variation of the INP during the same time, extracting the INP seasonality and diurnal variation by excluding the SDE. The data are first cleaned from local pollution (roughly 25% of the data removed) and then data is classified in 4 different air masses: FT with or without SDE and BLI with/without SDE. This paper is well written and very pleasant to read. It is a nice continuation of the first two papers published/under review this year. It presents an impressive work of high temporal resolution of INP concentration for 1 year of continuous measurement. The fact that the authors could use this high temporal resolution HINC instrument compared to "classical" daily filter measurement allow the authors to remove from the data any short local pollution, which I'm not sure would have been feasible with 24hr filter. For sure, much more of this type of high temporal INP measurement is very appreciated, and hopefully more in the future will be done (also at different locations).

I have only one main comment and a few small comments, and I recommend the paper to be published once these comments are addressed.

We thank the reviewer for their comments and positive evaluation of the paper. We respond to the reviewer concerns individually below and indicate modifications with line numbers made in the revised manuscript.

Main comment:

PL369: "Based on our observations, it is unlikely that pollen or subpollen particles are responsible for the observed high background INP concentrations in April,"Looking at the data, I would arrive at a different conclusion (or at least less affirmative about the non-influence of pollens on INP at JFJ).

We agree, and based on Reviewer 1 comments, we have modified the specific statements to be less affirmative about the absence of pollen influence on INP concentrations in April. First, we re-iterate in the conclusions that we do not expect pollen grains to contribute to the INP concentration merely because of the D_{50} size cut-off of the sampling inlet at HINC (this is already mentioned in the results section, see line 256 revised manuscript), and we now emphasize this in lines 390-393 of revised manuscript in the conclusion section. Also see comments below for back trajectory analysis and associated modifications to the manuscript.

A) Like Sarahan dust, pollens are known to be a very good candidate to act as INP as the authors explained, and this is why the authors investigated this specific source of INP. However, the authors do not have a direct measurement of pollen directly on site, so they have to speculate. The data presented here show that there is a peak of pollen measured 60 km away from the station (at Bern, 3 km lower in altitude) just a few days (1-3 days, hard to read from the figure FA1.a) before the measured "peak" of the INP at JFJ. Pollens are released first before INP increases, which therefore

does not rule out the possibility of Pollen reaching JFJ and increasing INP concentration (the other way round would not work). Similar to SDE, pollen transported to JFJ could have departed days earlier before arriving on this high altitude site?

We agree, and we have made this more clear that we cannot rule out the role of pollen fragments and sub-pollen particles contributing to the INP form other regions. Regarding Bern and Visp, since the peak in INP concentrations at JFJ was only observed around April 21-25 we ran a back trajectory analysis ending at JFJ on April 22 at 12.00 UTC on April 22 (see Fig. 1 below) with trajectories coming from the north and south with ground contact. The trajectories arrive from regions covering near Visp, but would not originate before April 15 from this region (Fig. 1 below) when pollen concentration was peaking (Fig. 3 manuscript). i.e., in Visp the pollen concentrations were already declining starting April 11-15. As such, pollen fragments from Visp will likely not contribute to the peak in INP concentrations at JFJ between April 20-25th. Furthermore, for air masses arriving at JFJ on April 22nd, the trajectories do not pass over Bern which also suggests that it could not be a source of INP to JFJ during the peak on April 20-25th. Given a second peak in pollen was observed in Bern around April 19/20, we ran back trajectories ending at JFJ on April 20 at 12.00 UTC (see Fig. 2 below) also shows that no trajectories arriving at JFJ on the 20th or 22nd of April pass through Bern or Visp. We now acknowledge this in line lines 264-277 revised manuscript.

We further explicitly state that we cannot exclude the contribution of pollen fragments and sub pollen particles coming from other parts of Europe and Switzerland, since some trajectories came from north Italy (see Fig. 1 below) during April 22nd (see lines 266-268 and 276-277 revised manuscript) and the regions in Switzerland north of JFJ (see Fig. 2 below).

B) Then there is the estimation of how many pollen particles would reach the station: P11L261 ". If every pollen grain would be ice-active at 243 K, and the same pollen concentration were present at the JFJ as measured in Bern, i.e., the PBL was perfectly mixed and the JFJ was within the PBL, pollen would only contribute up to 3.6 INP std L-1 (4 INP L-1), 5 times less than the Q95% INP concentration for BLIBG conditions during the same time period. "

However, pollen concentration measured at Bern is an average of 24hrs, whereas INP concentration measured at JFJ is a snapshot of 20 min of measurement. So for me, this will not exclude the possibility of pollens arriving in a batch at JFJ, therefore explaining this higher concentration. Also, pollen concentration measured at Bern may not be the representative concentration of pollens arriving at JFJ as another site (Visp) at a roughly similar distance from JFJ reported half of the concentration around the same day (April 20th ?).

Based on the back trajectory analysis we do not believe that the pollen concentration in Bern could have contributed to the INP concentration peak at JFJ during April 20-25 because no trajectories passed over Bern (see Figure 1 and 2 below). In addition, some air masses could come from the south of JFJ and as far south as north of Italy, or other regions north of JFJ within Switzerland as far as south of Germany (see Fig. 2) which we cannot exclude. We have modified the manuscript on lines 264-277 in the revised manuscript to clearly state this possibility.

C) Air mass origin. I wonder if an analysis of the air mass origin could help in understanding this spring peak of INP. Where the air mass is coming from during the INP peak? Could this air mass have collected pollen from somewhere in Europe? Could pollen be transported from further away than Bern or Visp (like SDE)? The authors state that an anti-cyclone was present until April 16th. Could it have influenced the non-transportation of pollen to JFJ at that time (low INP)? Could the peak of INP arriving just after the end of the anti-cyclone be a result of transportation of air mass from mainland Europe (which was full of pollen)?

Based on the reviewer comment, we conducted a back trajectory analysis for April 20th (Figure 2) and 22nd (Figure 1). We modified the manuscript to state that pollen fragments could come from the south of JFJ and north of Italy or north of Switzerland in line with the air mass trajectories shown (see line 264 revised manuscript).

Furthermore, since there was a second peak in pollen concentrations in Bern around April 19/20, which means other parts of Switzerland and Europe likely had increased pollen concentrations, we include this as a possibility for pollen fragment transport to the JFJ in the revised manuscript (see lines 266-278 and 274-275). We refrain from making much stronger statements than currently stated in the revised manuscript for two reasons: first, there are no direct pollen measurements are JFJ as we stated in line 257 - 258 initial manuscript, line 266 revised manuscript. Second, we concluded using the same data set that dust contributed to ~98% of the INP concentrations at JFJ for all the data measured as reported in [*Brunner et al.*, 2021].



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. (<u>https://www.ready.noaa.gov/HYSPLIT_traj.php</u>)



Figure 2. Back trajectories (HYSPLIT, NOAA) [Rolph et al., 2017; Stein et al., 2015]arriving JFJ on April 20th at 12.00 UTC showing no air masses arriving from Bern or Visp, suggesting that these two locations would not contribute pollen fragments to the INP concentrations peak observed at JFJ during April 20th to 25th. The colours represent different trajectories starting every 3 hours going backwards. (<u>https://www.ready.noaa.gov/HYSPLIT_traj.php</u>)

Small comments:

It would be good to have Brunner et al. accepted 2021 in ACP to use the same notation as in this paper (if it is still possible to edit the manuscript). For example table 1 in both paper show the same results but with different notation.

We thank the reviewer for the comment and agree that it would be good to have the same notation in this work and in *Brunner et al.* [2021]. And we agree that the notation from this work should be used, however, we cannot change the notation in the other manuscript anymore since it has already been published last year.

The alternative would be to change the notation in this work, but we explain why we refrain from doing so. In Brunner et al. we only had "SDE in FT" or "SDE with BLI", whereas here we would have "BG in FT" and "BG with BLI" in addition. In our opinion, using the same notation as in *Brunner et al.* [2021] reduces the legibility. One side remark: The tables do not show identical data. This work shows data from Feb 7, 2020 to Jan 31, 2021 whereas *Brunner et al.* [2021] shows data from Feb 7 – Dec. 31. Because there were no SDE in Jan 2021, the statistics for SDE periods are identical.

P3L87-88 "(CPC), TSI 3772, lower cut-off size: 14 nm) and size distribution (scanning mobility particle sizer (SMPS); optical particle sizer (OPS)" What is the size range of the SMPS (which is then used to calculate N90) and the size range of the OPS?

We have added the respective size ranges of the SMPS and the OPS to the revised manuscript (see lines 88-89 in revised manuscript)

Fig 3: "with the Q10% of PFT of a given day" what is the right axis BLI/FT %? I m a bit confused about how to read this scale. I am assuming that data close to BLI correspond to 0% and close to FT correspond to 100%. is that correct?

The data close to the BLI label correspond to 0% (or a small percent of FT), i.e. the data close to the bottom (BLI) is predominantly when JFJ was in the boundary layer, and close to the top is when JFJ was predominantly in the free troposphere. We add the respective percentages to the axis in the revised manuscript.

Fig 3: What is the meaning of the dash black line around mid-April in panel a) and b).

The dashed black line indicates the peak in INP concentration for April to show that it does not align with the peak observed in Bern or Visp. We have added the definition of the dashed black line to the figure caption in the revised manuscript.

Reference Schneider, J. et al. 2020 is from ACPD, Schneider et al. 2021 is the final version. Please correct in the text and in the reference list. Corrected

Reference Brunner et al. 2021 in ACPD might be available at the time of the publication of this article.

Corrected

if other references of manuscripts in "preparation" are now available, please add them. Powered by

We have updated Wieder et al., in prep to the final version *Wieder et al.* [2022]

<u>References</u>

Brunner, C., B. T. Brem, M. Collaud Coen, F. Conen, M. Hervo, S. Henne, M. Steinbacher, M. Gysel-Beer, and Z. A. Kanji (2021), The contribution of Saharan dust to the ice-nucleating particle concentrations at the High Altitude Station Jungfraujoch (3580 m a.s.l.), Switzerland, *Atmos. Chem. Phys.*, *21*(23), 18029-18053, doi:10.5194/acp-21-18029-2021.

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

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.