Editorial Note:
Referee comments are in black.
Author replies are in blue.
Text modified are in red.

Anonymous Referee #2

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In their study Brennan et al. investigate INP concentrations from snow samples taken in the Swiss Alps. In total 88 samples were collected during the winter of 2018. Samples were obtained from 17 locations covering a vast area of the Swiss Alps. Attention was paid to terrain characteristics, elevation, snow age, snow depth and distance to Jungfraujoch. INP concentrations were determined in the lab together with other physicochemical parameters of the bulk meltwater. Based on the INP concentration per ml meltwater the authors also create a parameterisation to calculate cloud glaciation temperature.

The study is well conducted and scientifically sound. Sampling procedures are described in detail in the supplement material and are appropriate. To measure INP concentrations the authors use a newly developed method that has been tested and is extensively discussed in a recent publication referred to in the manuscript. While the analysis of the data is rather descriptive, a large benefit of the study is that samples were taken at 17 different locations in Switzerland spanning a large area. Most other studies focus on single locations. Sampling was also done from snow depth profiles and the local variability was investigated. Such data are very useful because they are rare.

Overall the manuscript provides a large dataset that is very beneficial for the ice nucleation community. The study fits the scope of ACP and I recommend publication after the points below have been addressed.

We sincerely thank the reviewer for their positive feedback and critical assessment of our work.

General comments

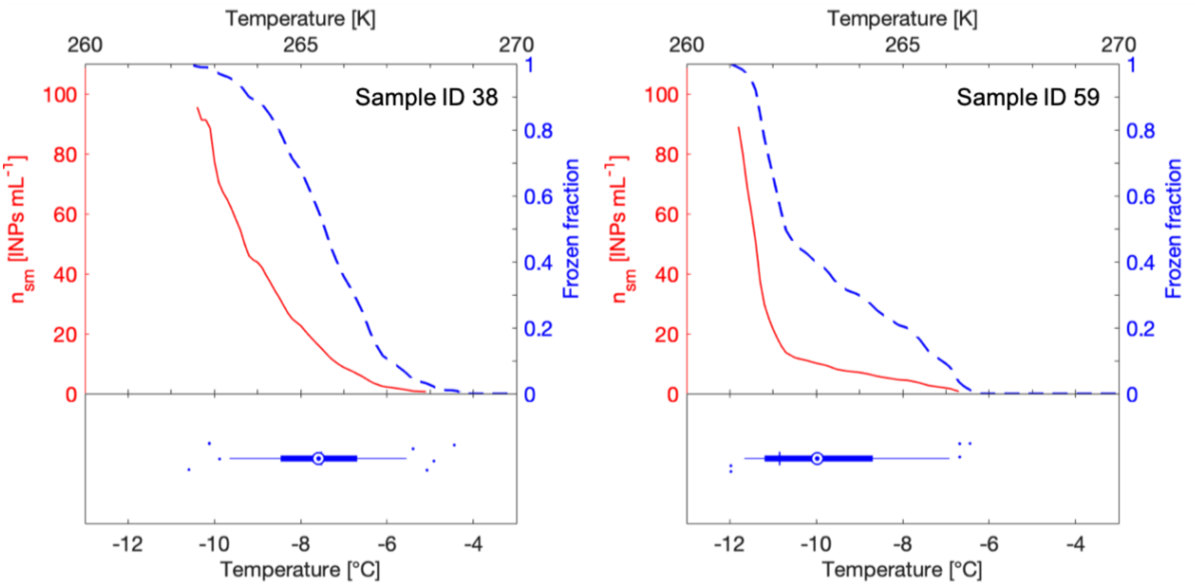
Title – The title should be changed. The current title suggests that the nature of INPs was studied. However, I don't see that much about the nature of INPs can be said from the study. E.g. chemical analyses were done on bulk samples and as the authors point out I agree that "the chemical signature of an INP is probably lost among total aerosol chemistry" (P834-35). The INP size was addressed but actually seems to be within the same range for all samples, so not heterogeneous. Consider addressing the variability of INP concentrations in the title rather than the INPs directly.

We thank the reviewer for their insightful comment, and we agree that the title misrepresented our study. We have modified the title as follows, "Spatial and temporal variability in the ice nucleating ability of alpine snowmelt and extension to cloud frozen fraction".

Why did the authors not calculate differential freezing nucleus spectra? They present a very useful picture of the entire INP population (Vali 2019) and INPs could be qualitatively classified (warm mode and cold mode INP, see also Creamean et al. 2019). Looking only at T_50 values might disguise the presence of a few INP at high temperatures. Comparing T_50 values is a rather limited approach when investigating heterogeneous environmental samples. The authors should consider adding freezing curves to the supplement material. Their current data representation in form of box plots looks nice but omits potentially relevant information.

We thank the reviewer for their comment. We believe we didn't explain graphically and within the text the value of the boxplot method adequately. To further clarify the value of freezing temperatures as boxplots as alternatives to differential freezing nucleus spectra, we have remade Figure 2 and written a new paragraph in the methods section to clarify the interpretation of the freezing temperature boxplots. We believe that in the revised manuscript we now explicitly state the comparison with Creamean et al. 2019 as well as better highlight the value of the boxplots to depict warm modes and cold modes.

Revised Figure 2:



Revised text in the methods section:

"In addition to showing *FF* versus temperature in a two-dimensional line graph (blue line in **Error! Reference source not found.**), we show all 96 raw data points as freezing temperatures in a boxplot (bottom half of **Error! Reference source not found.**). The blue box ranges from the 25th to the 75th percentile of freezing temperatures, whereas the whiskers extend from the 5th to the 95th percentile. Within the boxplot, the median, equal to T_{50}, is shown as a thin perpendicular blue line to the box and the mean is shown as a blue circle with a concentric dot (**Error! Reference source not found.**). When the mean and the median values overlap, the FF curve is more or less linear (Figure 2 - left). However, when the mean and median values differ by one degree or more, the FF curve has a kink or bump in its slope (Figure 2 - right), often observed for biological INPs (Creamean et al., 2019). The boxplot graphing method reduces from two to one the required number of dimensions for displaying a single experiment. This visualization allows for the clear comparison of many samples side by side for every sample measured in this study and uses all 96 data points without trimming (Figure S2).

In order to extrapolate the *FF* determined by DRINCZ into an INP concentration, Poisson distribution calculations were used as in Eq. (2) (Vali, 1971b, 2019):

$$n_{sm}(T) = -\frac{1}{V_d}\ln(1 - FF(T)) \qquad (2)$$

where $n_{mw}(T)$ is the cumulative INP concentration per mL of snowmelt as a function of temperature and V_d is the droplet volume in mL (0.05 mL) (**Error! Reference source not found.**). Although different arguments on omitting the first two wells exist (DeMott et al., 2016; Polen et al., 2018), we argue that trimming enhances representativeness, reproducibility and confidence in the processing of the data from FF to INP concentrations. Thus, the first two wells to freeze out of the 96 wells were omitted for calculating cumulative INP concentrations."

Specific Comments

P1L19-23: The abstract seems rather long. I suggest that the authors cut out L19-23. This describes just another way of plotting data (box plots). I don't see why this is really novel. See also my general comment.

We agree with the reviewer and we have edited this section in addition to shortening the overall abstract length from 412 words to 351 words.

P1L19: As far as I can see meteorological parameters were not used. Delete "meteorological".

Agreed. Done.

P1L28: The equation stated refers to c_air and not the INP concentration per ml melt- water. Please correct.

The reviewer is correct. We modified the expression to, "cumulative concentrations of INPs per m^{-3} of air".

P3L19: The elevation of both sites is about 2800m. Why is this well above the altitude of artificial snow production?

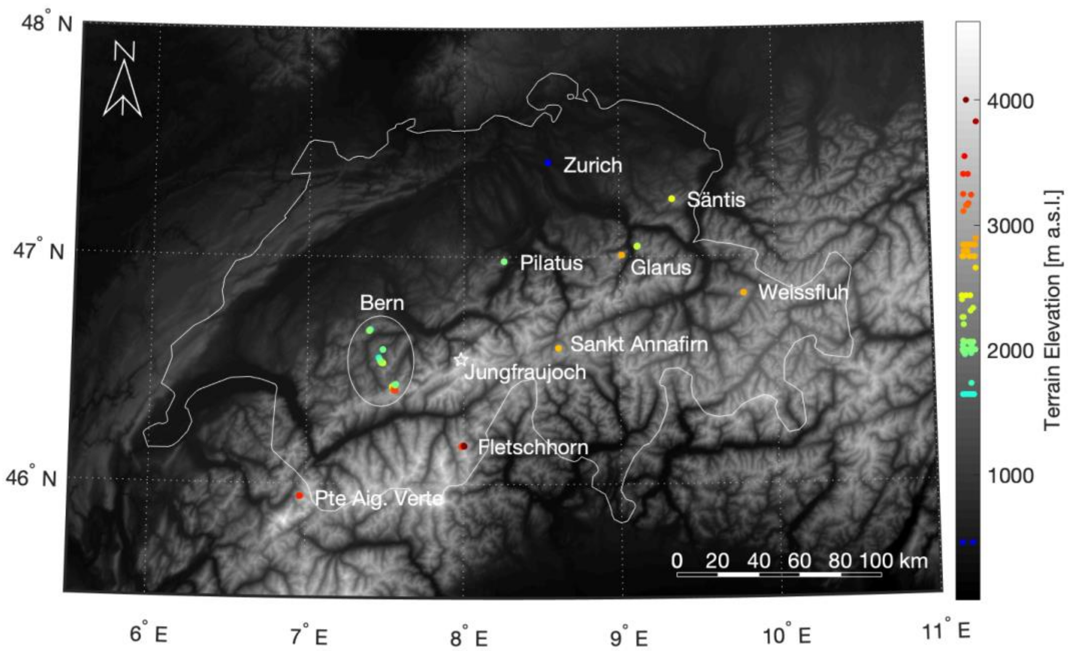
These samples were collected where no evidence (as far as we can tell) of snow making was observed during the ski day at those resorts. We clarified our statement which now reads,

"Two of these sites were within the boundaries of two different ski resorts, Davos (Weissfluh) and Andermatt (Sankt Annafirn). Considering the altitudes at which these samples where obtained as well as an absence of operating snow canons during the sampling days, we expect to not have sampled any artificial snow."

P4 Figure1: Please color code the sampling locations by altitude. At the moment it is not possible to attribute a certain altitude to a specific location, which would be useful and can be easily added.

We thank the reviewer for their recommendations and have modified Figure 1 to add more information using an altitude colour scale. We have also added site labels to the Figure. We think the figure is much improved now in relating information about the sampling sites.

Revised Figure 1:



P4L32: First, I am not familiar with biology reagent water. What is the purpose of biology reagent water? Is there a difference to ultrapure (MilliQ) water? Please explain. Second, what was done with the so determined background values? Were they subtracted from the respective snow sample freezing curves?

Molecular biology reagent water is high purity water purchased from Sigma-Aldrich. In our method validation and technique optimization in DRINCZ (See David et al., AMTD, 2019), we obtained better reproducibility with the water from Sigma-Aldrich then with MilliQ water. We wonder if the Milli-Q water quality is affected by the lifetime of the UV lamp inside the instrument or by the warm up time of the lamp when the dispenser is turned off. In any case, the molecular biology reagent water continues to be a more reliable background for our drop freezing measurements.

No further corrections were made with the background. We did not subtract them from our obtained freezing temperatures, as that mathematical operation would not be consistent with Poisson statistics. We added a sentence to the text to clarify;

"Finally, background corrections for the freezing temperatures were not necessary, as all of the T_{50} values were statistically above the water background of the instrument. Only three of the 88 samples had 75th percentile freezing temperatures overlapping with the mean of the background water: samples 21, 24 and 62 (Table S1). No further data manipulation was done for these samples as the conclusions drawn from these freezing temperatures were the same with or without a correction (Table S1)."

P6L4-10: This is a nice idea but I think this approach is not ideal for field samples with most likely heterogeneous INP populations. See also my general comment.

We appreciate the reviewer's comment. We think we did not do an effective job at communicating the value of a boxplot to display freezing temperatures. In an effort to better communicate the boxplot display of freezing temperatures, we have rewritten this paragraph almost entirely as well as moved the paragraph sooner in the section. We have also modified Figure 2 to give the boxplot more prominence. By doing so, we hope to have better communicated its value, as we argue that no to little information is lost in the boxplot when one considers the difference between the freezing temperatures of the median and of the mean. In other words, "warm mode" can still be identified in the boxplots as it has been identified with differential freezing spectra in Creamean et al., ACP, 2019.

"In addition to showing *FF* versus temperature in a two-dimensional line graph (blue line in **Error! Reference source not found.**), we show all 96 raw data points as freezing temperatures in a boxplot (bottom half of **Error! Reference source not found.**). The blue box ranges from the 25th to the 75th percentile of freezing temperatures, whereas the whiskers extend from the 5th to the 95th percentile. Within the boxplot, the median, equal to T_{50}, is shown as a thin perpendicular blue line to the box and the mean is shown as a blue circle with a concentric dot (**Error! Reference source not found.**). When the mean and the median values overlap, the FF curve is more or less linear (Figure 2 - left). However, when the mean and median values differ by one degree or more, the FF curve has a kink or bump in its slope (Figure 2 - right), often observed for biological INPs in a so-called warm mode (Creamean et al., 2019). The boxplot graphing method reduces from two to one the required number of dimensions for displaying a single experiment. This visualization allows for the clear comparison of many samples side by side for every sample measured in this study and uses all 96 data points without trimming (Figure S2)."

P6L7-8: Here it is stated that the data was not trimmed and all 96 data points are used, while the previous paragraph explains that the data was trimmed (omitting 2 wells). This is confusing.

We agree with the reviewer that this section was misleading. To clarify, we used all 96 raw data points for our presentation of freezing temperatures and boxplot, but for the data processing, in other words, from the raw data to cumulative INP concentrations, we chose to trim the first two wells to enhance reproducibility. We therefore make a distinction between raw data with no trimming, and INP concentrations with trimming.

The text was modified accordingly,

"Although different arguments on omitting the first two wells exist (DeMott et al., 2016; Polen et al., 2018a), we argue that trimming enhances representativeness, reproducibility and confidence in the INP concentrations. Thus, the first two wells to freeze out of 96 were omitted for calculating INP concentrations."

P6L22: add “horizontally” . . .and scattered “horizontally” to avoid overlapping.

Done.

P7L35-37: I wonder whether blank measurements were done with all filters? Figure 5 suggests so. Add this information here.

Yes, molecular biology reagent water was used with all the filters and we have modified the sentence as follows,

“The filtered samples, including molecular biology reagent water blanks, were then measured with DRINCZ”

P10L1-5: “SA background T_50” What is SA?

SA means Sigma-Aldrich water background. The use of this acronym is ambiguous, and so we have now changed all instances of background water mentions, to specifically state that it is molecular biology reagent water instead.

P11L14-15: I don’t understand the conclusion that INP were abundant but inhomogeneously spread. Is there evidence for less snow drift at the St. Anna Firn site? Did the authors compare wind speeds during the last snow fall at the sites?

Unfortunately, we do not have wind speeds at any of the sites. We are not looking for sources, but rather we wanted to study the variability, and look for correlations with different parameters. Wind speed might not be a clear indicator of blowing snow. For clarity we reworded the concluding sentence of this paragraph as,

“The narrow spread suggests that the INPs responsible for the observed freezing in these samples (at origin and at 2 m) were at abundant at these locations, but inhomogeneous across the plain (**Error! Reference source not found.**).”

P11 Paragraph “Altitude Dependence”: In order to evaluate the influence of the boundary layer, airmass trajectories should be analyzed. A site at e.g. 2000m can be in or out of the boundary layer depending on meteorological conditions.

Airmass trajectories required to analyze the height of the boundary layer would require high resolution not available with HYSPLIT (0.25 degrees equivalent to roughly 25 km resolution has only be available since June 2019 and only 0.5 degrees is available for our sampling days in the winter of 2018). We are concerned about the impact of the terrain not being well represented in such a coarse model. To better answer the reviewer, we did HYSPLIT trajectories and the results are below. Unfortunately, we are unable to draw conclusions from these analyses as to whether the airmasses were in or out of the boundary layer. To further complicate this type of analysis, the time between the snowfall (recorded for these back trajectories in Figure RC1) and the sampling sites varied.

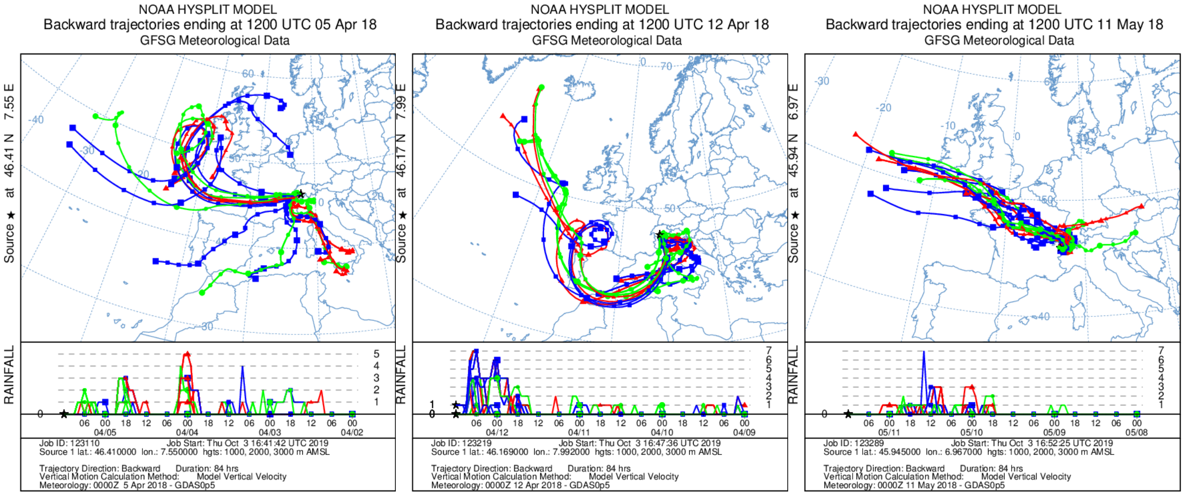


Figure RC1: HYSPLIT backward trajectories were generated at the last snowfall at the site using GDAS at 0.5 degrees resolution. The model parameters employed included model vertical velocity for the year, month, day and hour of sampling at Gorssstrubel (14.04.2019), Fletschhorn (22.04.2019), and Pointe Aiguille Verte (12.05.2019). Total run time was 84 hours at heights of 1000 m, 2000 m and 3000 m AMSL.

No further change to the text was made.

P14L32-34: I don't see in what way source regions or microphysical pathways up-stream of the sampling locations were analyzed, but the statement suggests so. Neither meteorological data nor airmass trajectories were included.

We acknowledge that we didn't look into HYSPLIT data. Because of resolution constraint, and because we saw high variability in the INP data.

P17L10: This section reads more like "Conclusions" and should not be a subsection to "4. Atmospheric implications".

Agreed. We have deleted section 4.2 since it was redundant and since ACP doesn't have a conclusion section.

References

Vali, G.: Revisiting the differential freezing nucleus spectra derived from drop-freezing experiments: methods of calculation, applications, and confidence limits, Atmos. Meas. Tech., 12, 1219–1231, https://doi.org/10.5194/amt-12-1219-2019, 2019

Creamean, J. M., Mignani, C., Bukowiecki, N., and Conen, F.: Using freezing spectra characteristics to identify ice-nucleating particle populations during the winter in the Alps, Atmos. Chem. Phys., 19, 8123–8140, https://doi.org/10.5194/acp-19-8123-2019, 2019

Both references are now correctly part of our manuscript.