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

Anonymous Referee #1

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In this manuscript Brennan et al. present results from INP measurements of snow samples collected at different locations, altitudes, times, and depth in the Swiss Alps. They found highly variable INP concentrations and used the data to generate a parameterization for the calculation of cloud glaciation temperatures.

The authors generated a very rich, unique and great data set of INP concentrations of 88 snow samples. This dataset is sound and the study is suited to the scope of the journal. The presented results are important for the ice nucleation community and can be useful for modelers. The experiments were well designed and were properly executed. I recommend publication after the following points have been addressed:

We thank the reviewer for their support and critical feedback.

I do not understand how the authors come to the result and very prominent message presented in the title that the INP in the Swiss Alps are chemically heterogeneous. I cannot find experiments and results in the manuscript that support this message. Filtration of the samples just allows an estimation of the (physical) size of the INP. Determination of the pH, conductivity and TOC of the snow samples does not give information on the chemical composition of the INP within the snow.

We thank the reviewer for their comment. We agree that the chemical aspect of our study is rather homogeneous instead of heterogenous and that the title was therefore ambiguous. We show that the pH, conductivity and TOC of snow samples which are chemical indicators for hydronium ions, for inorganic ions and for organic carbon, respectively are more or less constant across our samples. We agree with the reviewer that size filtration is a physical property.

We have modified the title to ,"Spatial and temporal variability in the ice nucleating ability of alpine snowmelt and extension to cloud frozen fraction".

In the abstract the authors write that they compared the INP concentrations with meteorological parameters. Which parameters were used? Where are the results?

Good point. We made a mistake. The word "meteorological" was replaced by "temporal".

Moreover, the authors highlight an alternative plotting method of INP data in the abstract. I wonder if there is another difference to Polen et al, 2018, than an extension to another sample type (field). Both studies used the same kind of data (freezing temperatures and frozen fractions). The sample type (field or laboratory) seems to be a secondary aspect, which does not explain why this is highlighted in the abstract.

We would like to advocate for the use of one-dimension visualization for freezing temperatures to avoid typically over crowded frozen fraction plots. Yet, we agree that this mention does not belong in the abstract, and have therefore removed this section. In addition, we have reworded the sentences to remove any implied novelty. The sentence in the abstract now reads,

"Boxplots of the freezing temperatures show large variability in INP occurrence, even for samples collected 10 m apart on a plain and 1 m apart in depth."

Furthermore, the mixed use of "snow" (e.g., P7L25), "snowwater" (e.g., P1L25), "melt-water" (e.g., P8L26), "melted snow samples" (e.g., P7L18), "snow meltwater" (P2L50) and "snowmelt" (e.g., P8 Table 2 caption) for the same samples in this manuscript is confusing and leads to the impression that different types of samples have been analyzed e.g., P12L26/27 "..was observed for meltwater sampled on April 4,. . ." and 4.2 "Heterogeneity of ice nucleating particles in meltwater".

The reviewer makes a very valid point and we thank them for bring it up. To help clarify, we have opted to systematically use "snowmelt" throughout the manuscript.

P2L28: "Soluble INPs have also been shown to be efficient INPs if they contain extracts from plant-based material,. . ." Please correct this statement. A soluble INP cannot contain "extracts". Moreover, for example fungal INP can nucleate at higher temperatures than plant INP (see cited reference Pummer et al 2015, ACP).

The word "soluble" can certainly be ambiguous in the field of atmospheric ice nucleation. For the specific case of P2L28, we have decided to omit the word soluble and have reworded the sentence as,

"Efficient INPs may also originate from extracts of plant-based material, including proteinaceous material and polysaccharides (Augustin et al., 2013; Dreischmeier et al., 2017; Koop and Zobrist, 2009; Pummer et al., 2012, 2015; Wilson et al., 2015)."

P4L18: Please add here the information that the tubes were sterile.

Done.

P5L3: The authors should either add some information on how the snow was compacted and/or refer to the supplement where this information is given. Did the authors check with so called "handling blanks" that no contaminations occurred during sampling, compaction, transport and further handling of the tubes?

We thank the reviewer for their clarification request. In the supplementary information, we had written that "The snow was scooped directly with the open tube, to avoid contaminating the sample with hands or spatulas. If the snow was fresh, loose powder, it was necessary to compress the snow within the tube and sample multiple times to obtain a large enough sampling volume (optimal 30 mL). This compression was accomplished by repeatedly scooping snow and banging the bottom of the tube against a firm surface (for example ski boot)." In the main text, we added that the snow was "inertially" compacted.

To address the point of a handling blank, we added the following sentence, “Since 3 of the collected 88 snowmelt samples had freezing temperature distributions at the water background of DRINCZ (Figure S2), we can suggest that no to little contamination could have been introduced during the snow sampling.”

P5L8: How did the authors avoid cross-contamination when taking the depth profile samples? Was the shovel cleaned between the different sampling sites? How was made sure that no surface or “upper layer” INP were brought into the lower snow layers during shoveling?

For further clarification, we have added the following sentence,

“The side of the freshly dug hole was then scraped with the shovel to expose untouched snow at all depths and to remove possible cross-contamination from the ground during the digging process.”

P5L35: Why are INP concentrations not available for all temperatures? A short explanation should be added here.

We agree with the reviewer that this comment was very confusing. We meant to say that for example T90 temperatures were not available for all samples, since they were recorded at the instrument background. We chose to delete that clause for clarity, since the word “detectable” was already present in the previous sentence. In the end, this section was reworded and rearranged, and in the end, this sentence no longer appeared in the text.

P5L38: Can the authors explain why the first two frozen wells were considered as contaminations? I would expect a higher risk of contamination with lower T INP if there is a contamination as shown by the three samples which overlap with the water control.

The reviewer is correct to question our discussion of “contamination”, and we agree that contamination is more likely a problem at lower temperatures in absolute terms. We use 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 discussion of section 2.3 was rewarded and reorganized to add clarity. It now reads,

“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):

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

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."

P6L8: The authors write that they used the "data without trimming" although it was described in the paragraph before (P5L40) that the data were trimmed and the first two wells were excluded. This is confusing and needs clarification.

We agree with the reviewer that those two decisions are apparently confusing. We think the confusion could have come from our unclear order of the two paragraphs (the one discussed in this point and at point P5L38 above.) For clarity, we have moved the discussing of the raw data before the discussion of the processed data. We thank the reviewer for helping us better explain these steps, and refer the reviewer to the comment above for the final version of this section.

P6L8: Please cite the final paper of Polen et al. (published in AMT in Sep 2018). Polen, M., Brubaker, T., Somers, J., and Sullivan, R. C.: Cleaning up our water: reducing interferences from nonhomogeneous freezing of "pure" water in droplet freezing assays of ice-nucleating particles, Atmos. Meas. Tech., 11, 5315-5334, https://doi.org/10.5194/amt-11-5315-2018, 2018.

It is now correctly cited.

Figure 3: This figure and caption is not clear. Two locations are displayed as refreezing's and three other locations as triplicates. If refreezing and triplicates are different things one should compare the same samples/locations. Based on the caption "refreezing triplicate data" refreezings and triplicates seem to be actually the same i.e., refreezing of triplicates? Please clarify. Omit "Finally" in the caption.

We have clarified the titles in Figure 3 as well as the caption, as there was indeed an error and we thank the reviewer for noticing this error. The caption should have read refreezing "and" triplicate data at different locations. To further clarify, we have also divided the figure into part A) and part B). The caption now reads,

"Refreezing and triplicate data at different locations show that the variability is within the instrument error of 0.9 °C. The standard deviation of the refreezings (± 0.47 °C) is comparable to the standard deviation of the triplicates (± 0.28 °C). On the box plots, the blue vertical line shows the median and is equal to T_{50}. The mean is shown as a blue circle with a concentric dot and the box ranges from the 25th to the 75th percentile. The whiskers extend from the 5th to the 95th percentile."

P7L27: Please clarify. In L26 it is said that filter with pore sizes 0.2 and 0.45 µm were used to determine the size of the INP. But then results of a 0.7 µm filtration and 0.02 are presented too. It would help to restructure the paragraph about the filtration experiments and put the filtrations in a more logical order. Please correct "Tests done with a 0.7 µm glass fiber filters". Omit the "a" or the "s" from "filters".

We agree with the reviewer that this paragraph is confusing. We have rewritten it for clarity as follows,

"Finally, to classify the size of INPs in the snow samples, the samples were filtered through cellulose acetate membrane filters with pore sizes of 0.2 and 0.45 µm (514-0063, VWR, USA). In addition, samples were filtered with a 0.02 µm pore filter (Whatman® Anotop® syringe filters, Sigma Aldrich) similarly to Irish et al, (2017). The filtered samples, including molecular biology reagent water blanks, were then measured with DRINCZ. Tests with glass fiber filters with pose sizes of 0.7 µm (SF1300-07, BGB-Analytik, USA) yielded lower freezing temperatures than with the cellulose acetate membrane filters and were thus not used further for this study."

P7L32: Omit "purchased through" - superflous

Done.

P7L35: Please introduce "SA water" when used first.

We have removed the use of SA water, and call it molecular biology reagent water instead throughout the manuscript and supplementary information.

Table 2: " and the following. . .". Nothing follows, thus omit.

Done.

P9L21: Omit "in size". "smaller than 0.2 µm" is sufficient.

Done.

P9L29: It would help to mention the sources of such proteinaceous INP such as fungi and plant pollen (see cited reference Pummer et al. 2015, ACP).

Agreed. We have modified the sentence to read,

"However, the role of bacterial fragments or proteinaceous material from sources such as fungi and plant pollen cannot be excluded (Hartmann et al., 2013; Pummer et al., 2012, 2015)."

Figure 5: "filter size"→ "pore size", missing spaces before µm, " were filtered at 450 , 200 and 20 nm". . ." → stay with µm, "filtered through";

All changed.

P10L19: The header “Sampling site characteristics” sounds more like a description of the sampling sites and thus does not fit to the subsections and results presented in this section.

Agreed. We have renamed this heading, “Spatial, altitudinal, snow age and depth variability of freezing temperatures”.

Figure 7/8: Is there a difference between TFrz as used here and Temperature used in Figure 3, which is the same type of plot?

No there is no difference. To clarify, we have changed the y-axis in Figure 3 to T_{Frz}.

Figure 13: I am not a cloud expert, but wonder if it would be better to use “frozen fraction of could droplets” instead of “frozen cloud fraction” as the INP concentrations are known.

We thank the reviewer for their comment. Unfortunately, without the droplet number within the cloud, we cannot say anything about the number (and thus the fraction) of cloud droplets which have frozen. The frozen cloud fractions were calculated from our 88 snowmelt samples. To further clarify Figure 13 (now updated to Figure 12 in the revised text), the section has been rewritten (although this revision was not directly prompted by the reviewer, we felt it was quite unclear as it was written).

P17L10: The subsection “4.2. Heterogeneity of ice nucleating particles in meltwater” is more or less redundant to the results and discussion already presented in “3.3. Sampling site characteristics” and seems not fit as a subsection in “4. Atmospheric implications”. The sections 3.3. and 4.2 should be merged in 3.3 with a new heading, as suggested above.

Agreed. We completely deleted section 4.2. And names section 3.3 to the Spatial, altitudinal, snow age and depth variability of freezing temperatures.

Figure S1: Based on the information given in the method section (P4L23) water from Sigma Aldrich was used as control. What is, with only one type of water, the difference between the untreated tube and SA (I assume this means Sigma Aldrich water) reference? Moreover, the results of the methanol, acetonitrile and HCl washings as listed on P4L21 are not presented in Figure S1, but instead there is a Milli-Q wash presented. Figure and corresponding text should be checked for consistency and completeness.

We thank the reviewer for their comment and we apologize for our mistake and ambiguity in the name of the samples. We have also relabeled Figure S1 and avoided any mention of Sigma Aldrich water and instead label is as molecular biology reagent water. We have modified the text on P4L21 and in Figure S1’s caption.

The text now read;

“In fact, preliminary pre-treatment testing showed physical degradation of the polypropylene material under heated conditions (125 °C in an oven) and when washed with organic solvents such as acetone and ethanol (Figure S1). For every Techno Plastic Products tube batch, a tube was filled with 20 mL molecular biology reagent water (89079-460, Sigma Aldrich, USA) and placed in a freezer at –20 °C (untreated tube samples in Figure S1). These reference water samples were later measured alongside the snow samples to generate background water values for measurement comparison.”

The caption now reads, "Figure S1: Comparison of freezing temperature boxplots of molecular biology reagent water (89079-460, Sigma Aldrich, USA) stored (1 day plus freeze/thaw cycle) in Techno Plastic Products tubes after different washing procedures as well as and untreated tubes. The background water refers to water directly from the molecular biology reagent water bottle without any contact with the untreated tube."

Figure S2: "The grey shaded area. . ." In my version there is no grey shaded area in this figure. The authors might want to check this.

We thank the reviewer for the heads up. It seems there was an error during the conversion from Word to PDF. We have rectified the problem.

Figure S3: "Size dependency" -> "size determination"; "filter size" -> e.g., "pore size", " were filtered at 450 , 200 and 20 nm". . ." → stay with µm, "filtered through"; omit " The T10 version. . ." as this is the T10 version.

Fixed.

Technical corrections

There are numerous other typos and inconsistencies throughout the manuscript. Many of those errors should have been caught by careful proofreading. I list the issues that caught my eye but I advise the authors to recheck their manuscript carefully to catch all of those typos.

We thank the reviewer for their careful evaluation of our manuscript. We really appreciate it.

Mixed use of l and L for Liter in text and figures. Mostly L is used but l is used in P1L15, Fig 2, P5L31, Fig 4, Fig 6, Fig 9.

We have changed all instances to the symbol of "L".

Missing spaces (Table 1 – 10m, P5L31-0.05ml, Fig 5, Fig 13, Table S1, SuppP5L7, Fig S3).

All fixed.

Inconsistent format/symbols: T50 (Text vs Table S1, caption Table 2), µ (Text, Fig 5, Fig S3) vs uS (Fig 6, Fig 9).

Fixed.

Figures have partly axes with °C and K (Fig. 11, 13), partly only with °C (Fig 12).

Figures 11, and 12 now all have both °C (bottom) and K (top) temperature axes.

Axis title capitalization (Fig 13) vs not capitalized (Fig 11).

Both y-axes have now been capitalized in figure 11.

Table 1: The “altitude” within the brackets should be “latitude”.

Fixed.

P8L31: “for” all types.
On this point unfortunately, we disagree with the reviewer. “Note that the chemical properties of the snowmelt are representative of all types of aerosols present within the collected snow sample” and not “for all types”.

Figure 4 caption: ”conductivity TOC”→ ”conductivity. TOC”? P10L25: “display”->”displays”.

Fixed.

Figure 12: Emty [] on the y-axis.

We meant to leave the brackets empty to show there were no units to this value. Since it wasn’t clear, we have removed the brackets.