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

Anonymous Referee #1

Received and published: 16 August 2019

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):

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

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."

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 25_{th} to the 75_{th} percentile. The whiskers extend from the 5_{th} to the 95_{th} 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.

Anonymous Referee #2

Received and published: 10 September 2019

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

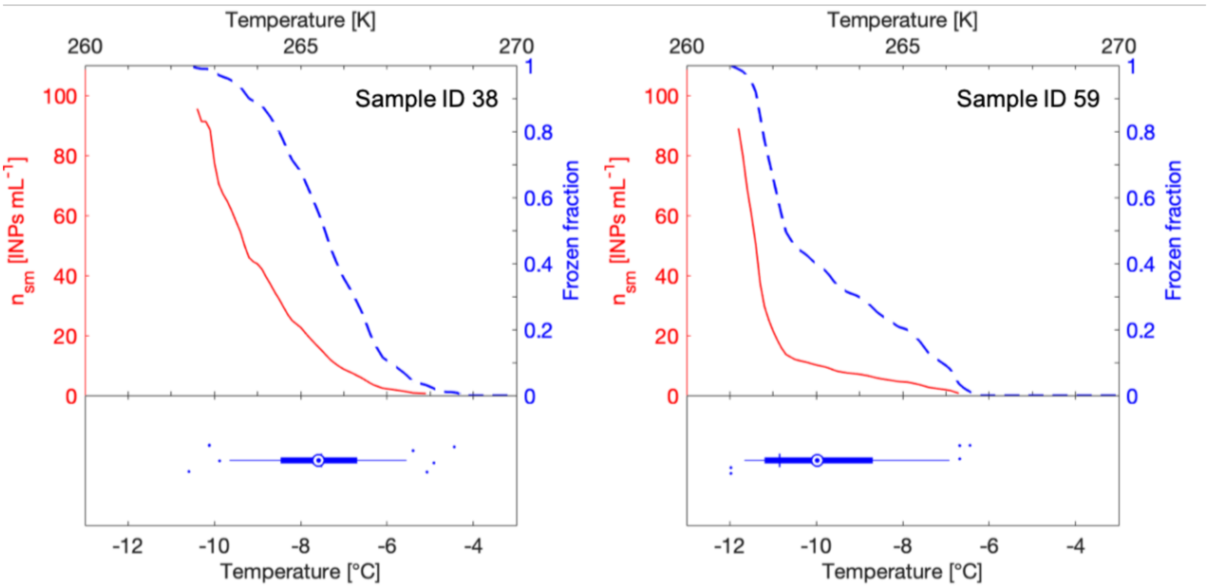
Title – The title should be changed. The current title suggests that the nature of INPs was studied. However, I don't see that much about the nature of INPs can be said from the study. E.g. chemical analyses were done on bulk samples and as the authors point out I agree that "the chemical signature of 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?

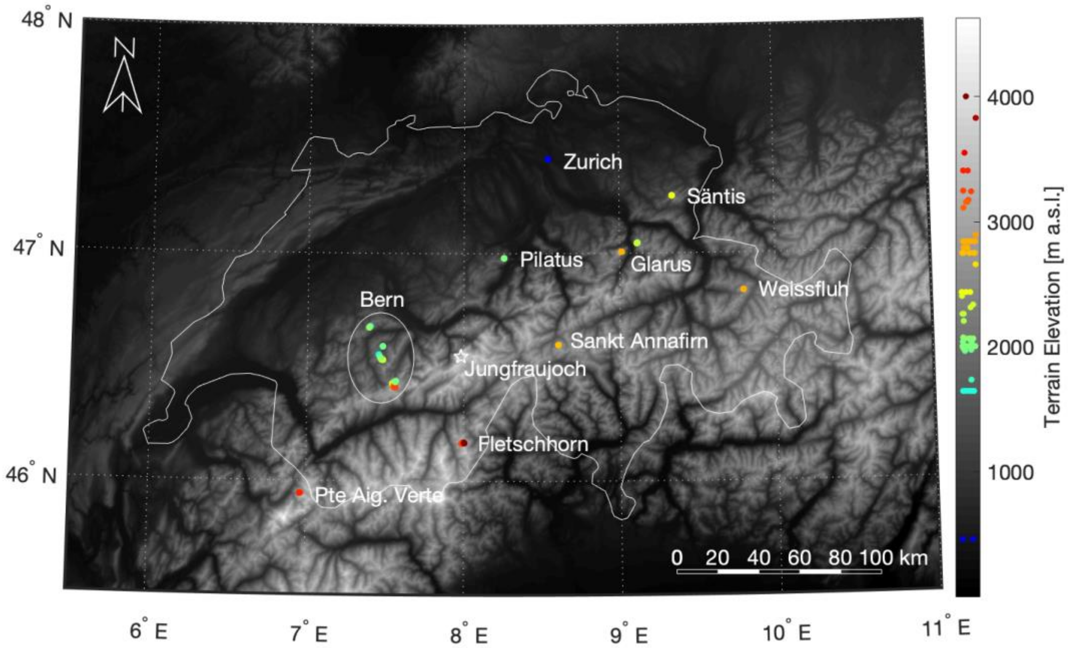
These samples were collected where no evidence (as far as we can tell) of snow making was observed during the ski day at those resorts. We clarified our statement which now reads,

"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.

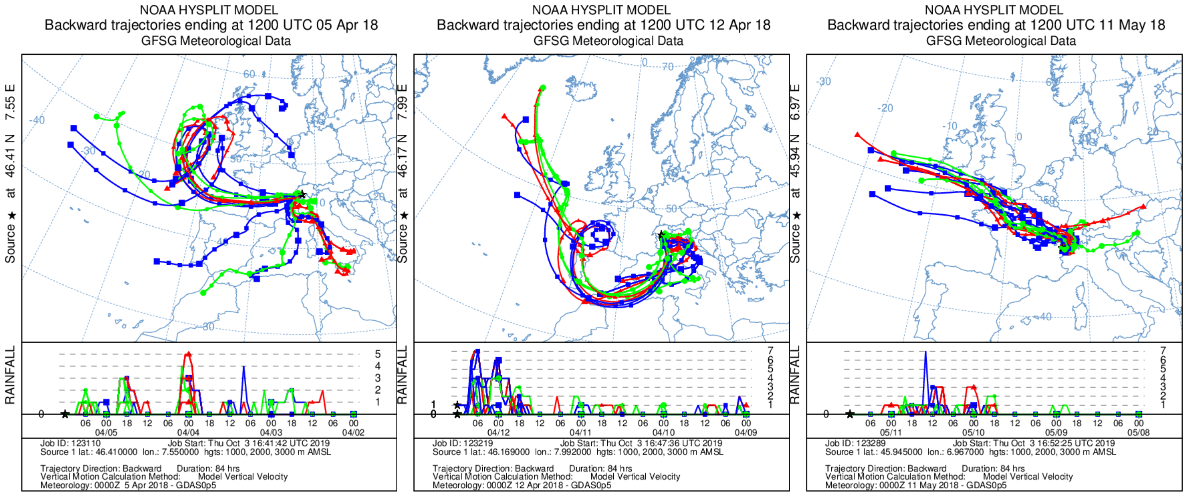


Figure RC1: HYSPLIT backward trajectories were generated at the last snowfall at the site using GDAS at 0.5 degrees resolution. The model parameters employed included model vertical velocity for the year, month, day and hour of sampling at Gorssstrubel (14.04.2019), Fletschhorn (22.04.2019), and Pointe 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.

Spatial and temporal variability in the ice nucleating ability of alpine snowmelt and extension to cloud frozen fraction

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Abstract.
Ice nucleating particles (INPs) produce ice from supercooled water droplets through heterogeneous freezing in the atmosphere. INPs have often been collected at the Jungfraujoch research station (at 3500 m a.s.l.) in central Switzerland; yet spatially diverse data on INP occurrence in the Swiss Alps are scarce and remain uncharacterized. We address this scarcity through our Swiss alpine snow sample study which took place during the winter of 2018. We collected a total of 88 fallen snow samples across the Alps at 17 different locations and investigated the impact of altitude, terrain, time since last snowfall and depths on freezing temperatures. The INP concentrations were measured using the homebuilt DRoplet Ice Nuclei Counter Zurich (DRINCZ) and were then compared to spatial, temporal and physiochemical parameters. 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, undiluted samples had cumulative INP concentrations ranging between 1 and 100 INP mL^{-1} of snowmelt over a temperature range of −5 to −19 °C. From this field-collected data set, we parameterize the cumulative INP concentrations per m^{-3} of air as a function of temperature with the following equation $c^*_{air}(\boldsymbol{T}) = \boldsymbol{e}^{-0.7T-7.05}$, comparing well with previously reported precipitation data presented in Petters and Wright, 2015.

When assuming (1) a snow precipitation origin of the INPs, (2) a cloud water content of 0.4 g m^{-3} and (3) a critical INP concentration for glaciation of 10 m^{-3}, the majority of the snow precipitated from clouds with glaciation temperatures between −5 and −20 °C. Based on the observed variability in INP concentrations, we conclude that studies conducted at the high-altitude research station Jungfraujoch are representative for INP measurements in the Swiss Alps. Furthermore, the INP concentration estimates in precipitation allow us to extrapolate the concentrations to a frozen cloud fraction. Indeed, this approach for estimating the liquid water to ice ratio in mixed phase clouds compares well with aircraft measurements, ground-based lidar and satellite retrievals of frozen cloud fractions. In all, the generated parameterization for INP concentrations in snowmelt could help estimate cloud glaciation temperatures.

1. Introduction

The supercooled liquid water to ice ratio significantly alters the radiative and dynamic properties of mixed-phase clouds. The abundance of ice crystals in a cloud can influence its optical transmittivity and reflectivity, as well as affect the formation of precipitation (Matus and L'Ecuyer, 2017; Mülmenstädt et al., 2015; Prenni et al., 2007; Vergara-Temprado et al., 2018). Since homogenous freezing of cloud droplets only occurs at temperatures below −38 °C (Koop, 2004; Pruppacher and Klett, 2010; Stöckel et al., 2005), heterogeneous nucleation which involves the presence of ice nucleating particles (INPs) can occur at warmer temperatures (Fletcher, 1962; Pruppacher and Klett, 2010; Vali et al., 2015). Aerosol-cloud interactions, including INP-cloud interactions, currently contribute to the largest uncertainty in total anthropogenic radiative forcing in global climate models (Boucher and Quaas, 2013) and hence constraining INP concentrations and better predicting the liquid water to ice ratio could help reduce this

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Spatial and temporal variability in the ice nucleating ability of Swiss alpine snow meltwater and extension to cloud glaciation temperatures

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Deleted: is required for ice formation uncertainty. Indeed, a growing number of INP parameterizations are helping to better constrain and predict INP concentrations as a function of temperature (DeMott et al., 2010; Petters and Wright, 2015; Phillips et al., 2008, 2012), surface area (Niemand et al., 2012) or organic carbon (Borduas-Dedekind et al., 2019; Wilson et al., 2015).

Mixed-phase clouds are responsible for the majority of precipitation over land (Hande and Hoose, 2017; Henneberg et al., 2017; Mülmenstädt et al., 2015; Murray et al., 2012) through immersion freezing, defined by the freezing of cloud droplets due to the presence of an INP (de Boer et al., 2011). As the majority of ice precipitation in mixed-phase clouds is initiated via immersion freezing (de Boer et al, 2011), the composition, abundance and sources of INPs responsible for the precipitation can be obtained through the analysis of precipitation samples (Fan et al., 2018; Morris et al., 2014; Petters and Wright, 2015; Stopelli et al., 2015; Wright et al., 2014). Indeed, INP concentrations have been measured in precipitation samples such as rain, snow, hail and sleet and have been recently revisited to obtain a representative INP concentration range for different temperatures (Petters and Wright, 2015). Examples of precipitation samples with ice nucleating activity have been collected in the United States (Failor et al., 2017; Hader et al., 2014; Hill et al., 2014; Petters and Wright, 2015; Wright et al., 2014), Canada (Rangel-Alvarado et al., 2019; Vali, 1966, 1971a), France (Joly et al., 2014), Switzerland (Creamean et al., 2019; Stopelli et al., 2014, 2017) and Israel (Zipori et al., 2018). Furthermore, geographically diverse precipitation samples have shown consistent immersion freezing activity due to biological material (Christner et al., 2008a, 2008b). These field studies highlight the ubiquitous presence of INPs in precipitation while simultaneously emphasising high variability in INP concentrations.

Meanwhile, studies conducted in the laboratory to further understand heterogeneous ice nucleation have found mineral dust, metallic oxides, and biological material to be efficient INPs (Hoose and Moehler, 2012; Kanji et al., 2017). 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). In addition, organic matter may also act as INP in the immersion freezing mode (Borduas-Dedekind et al., 2019; Hill et al., 2016; Hiranuma et al., 2015; Irish et al., 2019; Knackstedt et al., 2018; Knopf et al., 2018; Wilson et al., 2015).

Field measurements of INPs in the Swiss Alps are largely bound to the high-altitude research station Jungfraujoch (Boose et al., 2016; Chou et al., 2011; Conen et al., 2012, 2017; Ehrman et al., 2001; Farrington et al., 2016; Hammer et al., 2018; Lacher et al., 2017, 2018a, 2018b; Lloyd et al., 2015; Meola et al., 2015; Mertes et al., 2007; Nillius et al., 2013; Stopelli et al., 2016, 2017). Due to its common free tropospheric conditions, measurements of INPs at Jungfraujoch can be representative of INP concentrations globally. However, the impact of local sources and INPs present in the boundary layer cannot be ascertained. Therefore, to determine the representativeness of free tropospheric INP measurements across the Swiss Alps, geographically diverse sampling sites are required.

Thus, the first purpose of this study was to determine the heterogeneity of INP occurrence in the Swiss Alps and to investigate its correlation with various physicochemical properties and sampling site characteristics. 88 snow samples were collected using sterile Teflon tubes in the winter of 2018 across 17 locations on 15 different days spanning an approximate area of 35,752 km^2 in the Swiss Alps. INPs in the snowmelt were measured in the immersion freezing mode using the Droplet Ice Nuclei Counter Zurich (DRINCZ) (David et al., 2019). The ability of the snowmelt to freeze is reported as 96 freezing temperatures in one-dimensional boxplots. Furthermore, this study investigates INP occurrence by location, time, altitude, snow depth, snow age, total organic carbon, conductivity and size. We further propose a parameterization of INP as a function of temperature to complement the data set from Petters and Wright, 2015.

The second purpose of this study was to extrapolate our measured INP concentrations to frozen cloud fractions within mixed-phase clouds over the Swiss Alps. The first assumption is that the INPs

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quantified originated solely from precipitation without contributions from dry deposition. We then assumed a cloud water content of 0.4 g m^{-3} (Petters and Wright, 2015) to determine the cumulative concentration of INPs in air and subsequently assumed a critical number of INPs for cloud glaciation, ranging between 10 and 1000 m^{-3}. From this information, glaciation temperatures of mixed-phase clouds and supercooled liquid water to ice ratio were estimated. Finally, we discuss the limitations of our approach for arriving at frozen cloud fractions as well as compare our results with aircraft measurements, ground-based lidar measurements, satellite retrievals and global circulation models (McCoy et al., 2016a).

2. Methods

2.1. Sampling sites

Snow sampling locations were selected to provide a breadth of elevation, terrain type, distances from Jungfraujoch and snow age during one winter season to represent conditions in the Swiss alpine area during the winter of 2018. The campaign took place between February and May 2018 and the sites were accessed by ski or by alpinism. All samples were taken from undisturbed top layer snow, unless it was a depth profile. Specifically, 17 sites throughout the Swiss Alps were chosen (Table 1 and Table S1). 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. The coordinates of the sampling sites were measured using the GPS on a smartphone, and span an area of 218 km from east to west and 164 km from north to south to cover an approximate area of 35 752 km^2. The altitudes of the sampling sites ranged from 440 to 3981 m above sea level (a.s.l.) with a median value of 2294 m a.s.l. (Figure 1), and therefore cover a wide range of altitudes within the Alps according to the Bundesamt für Landestopografie swisstopo, 2018. Based on the date and time of sampling, snow age representing the timespan between the snowfall event and the sampling instance was determined by using the snowpack database (Institut für Schnee- und Lawinenforschung SLF, 2018). More specifically, we used the snowfall history at the SLF measurement station closest to the sampling site, typically within a few kilometers and at most within 10 km. Exceptionally for Pilatus, the closest SLF station was within 25 km due to fewer stations in the Luzern region.

Table 1: List of dates, sites (latitude and longitude), altitude, location including the Swiss canton and number (n) of samples collected, during this Swiss alpine snow sample study. Additional details with recorded parameters for each sample are available in the supplementary information. See Figure S5 for selected images of the different sites.

Date	lat [°N]	lon [°E]	Altitude [m a.s.l.]	Location/Canton	n	notes
04.02.18	46.68	7.40	1715 - 2033	Schibe/Bern	2	
13.02.18	46.53	7.48	2053 - 2316	Rauflihore/Bern	2	
24.02.18	47.04	9.11	2242 - 2294	Schilt/Glarus	4	
01.03.18	47.42	8.54	440	Wahlenpark/Zurich	2	
04.03.18	46.59	7.48	1945	Meniggrat/Bern	1	
17.03.18	46.55	7.46	1625	Alpetli/Bern	8	Depth profile
02.04.18	46.54	7.47	2045	Chalberhöri/Bern	9	Depth profile
14.04.18	46.44	7.57	1954 - 1967	Engstligenalp/Bern	6	
14.04.18	46.41	7.55	2185 - 3222	Grossstrubel/Bern	7	
14.04.18	46.44	7.56	1983	Engstligenalp/Bern	14	Depth profile
15.04.18	46.84	9.79	2822	Weissfluh/ Graubünden	9	10 m homogeneity
19.04.18	47.25	9.34	2416	Säntis/Appenzell	4	
22.04.18	46.17	7.99	3227 - 3981	Fletschhorn/Valais	3	
22.04.18	46.60	8.60	2729	Sankt Annafirn/Uri	6	10 m homogeneity
25.04.18	46.98	8.26	2020	Pilatus/Obwalden	2	
12.05.18	45.94	6.96	3137 - 3388	Pte Aig. Verte/France	4	

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21.05.18	47.01	9.01	2772	Vrenelisgärtli/Glarus	4	
15 days of sampling	**45.94 to 47.42 (164 km)**	**6.96 to 9.79 (218 km)**	**440-3981 (median at 2294 m)**	**17 locations**	**88 samples**	**3 depth profiles**

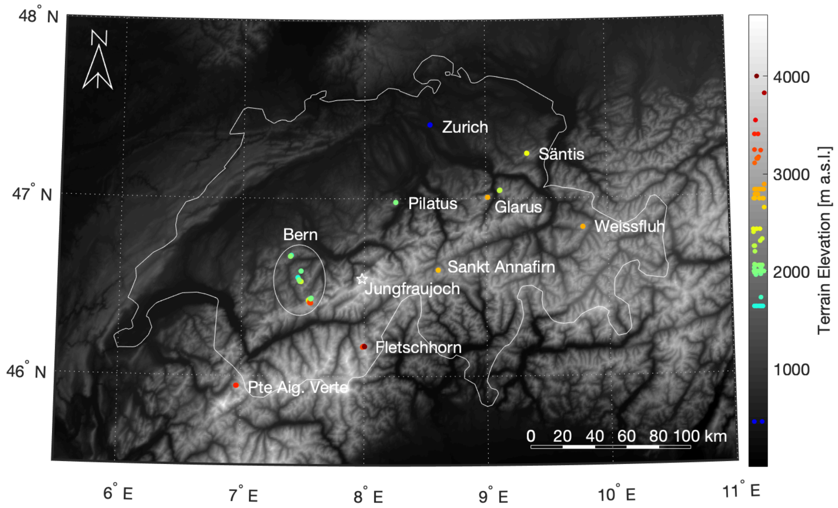

Figure 1: Map of all sampling sites (coloured dots) within the Swiss border (white outline). The circled sites are within the canton of Bern and include Schibe, Rauflihore, Meniggrat, Alpetli, Chalberhöri, Engstligenalp, and Grossstrubel. The high-altitude alpine research station Jungfraujoch is identified with a star for reference, although no snowmelt was collected at that site. The respective altitudes of the sampling sites are shown on the colour bar to the right, scattered in the x-direction to avoid overlapping of points. The terrain elevation is greyscale shaded according to a digital elevation model produced using Copernicus data and information funded by the European Union – EU-DEM layers (European Environment Agency, 2018). See Figure S5 for selected images of the sites.

2.2. Sampling procedure

Previously reported snow collection procedures for INP studies have involved a Teflon-coated tray put in place before snowfall (Stopelli et al., 2017). This method is convenient and adequate for alpine research stations where appropriate accommodation is available. However, to assess the heterogeneity of INPs in the Alps, we needed to access remote locations. Collecting snow samples from the top of the snowpack allows sampling in multiple locations compared to other precipitation collection procedures, such as rainfall collection, which require sampling site infrastructure. Through ski-touring and alpine mountaineering, we accessed 17 different locations for one-time snow sampling. Indeed, no infrastructure was necessary and remote locations as high as 3981 m a.s.l. could be uniquely accessed.

Snow samples were collected in sterile 50 mL polypropylene conical centrifuge tubes (Techno Plastic Products AG, Switzerland) without pre-treatment. 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 exact collection procedure for gathering snow samples is described step

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Deleted: , methanol and acetonitrile as well as 0.1 M HCl by step in the supplementary information. Briefly, the collection was done by a skier or an alpinist wearing nitrile gloves and they ensured that only the tubes came in contact with the snow. The snow was inertially compacted and the tubes were sealed. After collection, the snow samples were kept as cold as possible until the return to the laboratory, during which some of the snow did melt. Nonetheless, refreezing experiments suggested little difference in frozen fraction (see Figure 3), allowing us to conclude minimal sample degradation during transport. 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.

Snow depth profiles were dug with an aluminium avalanche rescue shovel and were dug until the underlaying dirt or rock surface was reached (see Figure 10 for a site photo). 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. The depths of the samples collected within the snowpack were determined with a graduated avalanche rescue probe (Mammut AG, Switzerland). Sample depths were distributed throughout the snowpack depending on the number of sampling tubes available on the specific trip.

2.3. Immersion freezing experiments and data analysis with DRINCZ

The DRoplet Ice Nuclei Counter Zurich (DRINCZ) technique used in this study, as well as the data processing and analysis are described in-depth by David et al. (2019). Additional information on the immersion freezing experimental details and on the frozen fraction (*FF*) analysis and can also be found in the supplemental information.

The instrument DRINCZ yields the *FF* from the number of wells frozen at a given temperature ($n_{frz}(T)$) and the total number of wells ($n_{tot} = 96$). A *FF* curve is a monotonic function of temperature and is derived according to Eq. (1). In other words, the frozen fraction stays the same or rises with decreasing temperatures.

$$FF(T) = \frac{n_{frz}(T)}{n_{tot}} \qquad (1)$$

The DRINCZ technique is limited to snow samples that freeze at temperatures above −22.5 °C, since 50% of the wells of the water background freeze at that temperature (T_{50} = −22.5 °C).

In addition to showing *FF* versus temperature in a two-dimensional line graph (blue line in Figure 2), we show all 96 raw data points as freezing temperatures in a boxplot (bottom half of Figure 2). 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 (Figure 2). 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).

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) (Figure 2). 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

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

where $n_{mw}(T)$ is the cumulative INP concentration per mL of snow meltwater as a function of temperature and V_d is the droplet volume in mL (0.05ml) (Figure 2).

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concentrations. Thus, the first two wells to freeze out of the 96 wells were omitted for calculating cumulative INP concentrations.

Furthermore, triplicates showed good reproducibility; T_{50} values of sample triplicates fell within 1°C (Figure 3) and standard deviations did not depend on average freezing temperature, consistent with Wright et al. (2013). Refreezing results showed a similar spread in T_{50} values as the triplicates, which suggest that the IN activity of the samples is only minimally affected by freezing (Figure 3). Consistent with the observed spread in triplicate freezing temperatures, the reported temperature uncertainty of DRINCZ is ± 0.9 ˚C (David et al., 2019). 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).

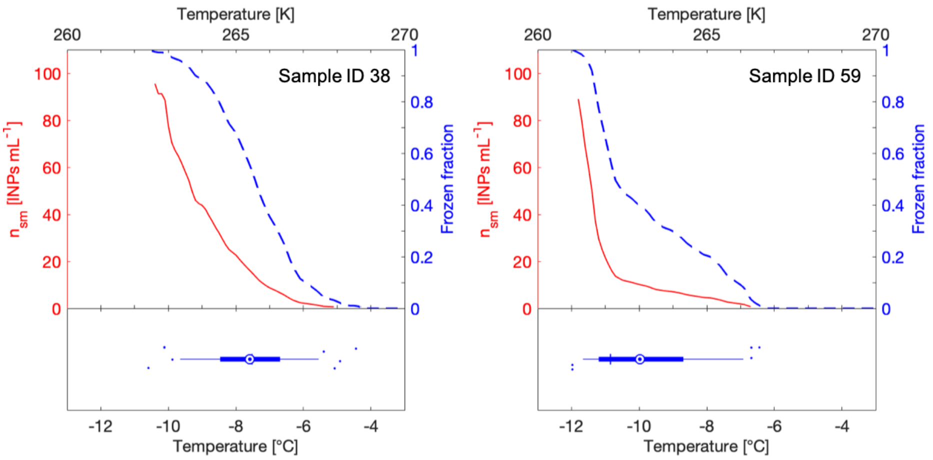

Figure 2: Examples of *FF* curves (blue y-axis), freezing temperature boxplots (bottom of the graphs) and INP concentrations (n_{sm}) (red y-axis) generated for two snowmelt samples (from sample #38 and from sample #59, see Table S1). On the boxplot, the thin blue vertical line shows the median and is equal to T_{50}, the mean is shown as a blue circle with a concentric dot. The blue box ranges from the 25th to the 75th percentile, whereas the whiskers extend from the 5th to the 95th percentile. Outliers are drawn as blue dots and scattered vertically to avoid overlapping. The figure on the left shows a linear FF curve with overlapping T_{50} and median values on the boxplot, whereas the figure on the right shows a FF curve with a kink with T_{50} and median values differing by at least 1 °C.

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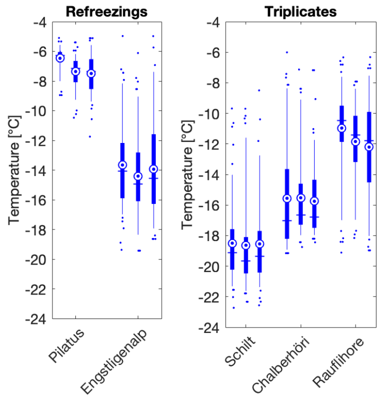

Figure 3: Refreezing and triplicate data at different locations show that the variability is within the instrument error of 0.9 °C. The standard deviation of the refreezings (± 0.47 °C) is comparable to the standard deviation of the triplicates (± 0.28 °C). On the boxplots, the blue vertical line shows 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.

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2.4. Physicochemical analyses

The Swiss alpine snow samples were submitted to chemical analyses in an attempt to correlate parameters to INP concentrations. In particular, total organic carbon (TOC), pH and conductivity measurements were made for all samples, whereas filtering procedures were conducted for a subset of 17 samples.

The total organic carbon was quantified as the non-purgeable organic carbon (NPOC) in solution using a total organic carbon (TOC) analyser (TOC-L CSH, Shimadzu, Japan). This instrument uses a 680 °C combustion catalytic oxidation method, while providing quantification in water samples above 0.1 mg C L^{-1}. A detection limit of 4 µg C L^{-1} is achieved through the use of a nondispersive infrared (NDIR) detector. Method sparge time and gas flow were 1.5 mins and 80 mL, respectively. For the NPOC calibration, two calibration solutions were prepared with concentrations of 20 ± 0.2 mg C L^{-1} and 2 ± 0.02 mg C L^{-1} from a potassium phthalate TOC standard solution of 1000 mg C L^{-1} (Sigma-Aldrich).

pH values were measured with a Metrohm pH glass electrode and the values obtained were consistent across all samples, ranging between 5.2 and 6.2. Considering the error on the pH measurements from the unbuffered snowmelt solutions, the pH of all the samples were within error and consequently was not ascribed to any INP variability.

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The conductivity of the snowmelt was measured using a handheld conductivity meter (LAQUAtwin COND, Horiba, Japan). It uses small sample volume (less than 0.5 mL) and has an accuracy of ± 1 µS cm^{-1} for conductivity measurements between 0 and 1999 µS cm^{-1}. The instrument was calibrated with a 1413 µS cm^{-1} standard solution. Before the measurement was obtained, the sensor was washed three times with nanopure Milli-Q water and then 1 mL of the sample was flushed over the sensor for conditioning.

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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,

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BGB-Analytik, USA) yielded lower freezing temperatures than with the cellulose acetate membrane filters and were thus not used further for this study.

3. Results and discussion

This study investigated the freezing temperatures of 88 snow samples collected over 15 days and 17 different locations during the winter of 2018 in the Swiss Alps. The T_{50} values and freezing temperatures are presented in three sections: physiochemical properties, times series and sampling site characteristics. T_{50} values from the entire dataset ranged between −5.3 °C and −21.6 °C with a mean value of −12.5 ± 4.0 °C (Table S1), all above the DRINCZ instrument's detection limit. We also report correlation analysis using T_{50} as the temperature where $FF = 0.5$ (Table 2). We first use T_{50} values to compare samples to different physicochemical properties of the snowmelt, such as TOC, conductivity and particle size, as well as to collection time within the winter season. We then use freezing temperature boxplots to compare the freezing behavior of samples based on terrain, altitude and snow age. Next, INP concentrations in precipitation were estimated and a parameterization is derived based on a temperature dependence. Finally, we extrapolate the INP concentrations to frozen cloud fractions to infer glaciation temperature and liquid water to ice ratio of mixed-phase clouds over the Swiss Alps in the winter of 2018.

Table 2: Correlation analyses between T_{50} values and different physicochemical and terrain parameters. R^2 values characterize the linear relationship between T_{50} and the parameter; the p-value identifies whether the correlation is statistically significant; n represents the number of snowmelt samples used in the correlation.

Parameter	R^2	p-value	n
Conductivity	0.0015	0.722	88
TOC	0.0085	0.192	86
Snow age	0.0623	0.057	59
Altitude	0.0015	0.722	88
Date	0.0005	0.833	89
Depth	0.0028	0.624	88
Distance	0.2140	0.174	88

3.1. Physicochemical properties

3.1.1. Chemical properties

To study the INP concentration dependence on chemical parameters, T_{50} values were compared to total organic carbon (TOC) and conductivity measurements (Figure 4). Organic carbon values ranged between 0.3 and 5.2 mg C L^{-1}, with a median of 0.64 mg C L^{-1} and an average of 0.9 ± 0.7 mg C L^{-1}, similar to a TOC range of 1.3 to 4.0 mg C L^{-1} previously measured in snow samples in North America (Rangel-Alvarado et al., 2015). The sample with the highest TOC value of 5.2 mg C L^{-1} was collected at Grossstrubel in Valais at 2638 m a.s.l (Figure 4 and Table S1). Its T_{50} value was −14.2 °C, and nothing was remarkably unique about this location. Furthermore, 13 snow samples were collected the same day with TOC values closer to the average concentration. Low TOC concentrations were found to span the entire range of T_{50} values, indicating no correlation between TOC and freezing temperatures of the snowmelt (Table 2). Wilson et al. (2015) propose an INP parameterization for sea surface microlayer samples based on TOC, however our snow samples suggest no correlation with organic carbon content.

Conductivity is a measurement of ionic strength of the snowmelt and salt can lower the freezing temperatures of a sample. However, conductivity values of the snowmelt ranged from 0 to 14 µS cm^{-1} with a median at 3 µS cm^{-1} and an average of 4.1 ± 2.5 µS cm^{-1}. The three samples with the highest conductivity measurements were samples 88, 96 and 73 and were from Fletschhorn, Pointe Aiguille

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Verte and Weissfluh, respectively (Figure 4). These samples had TOC concentrations close to 1 mg C L^{-1} and uncorrelated T_{50} values. The conductivity values of this dataset are small, and close to the limit of detection of the conductivity meter, approximately representing salt concentrations below 400 µM of for example NaCl.

The pH values obtained were consistent across all snowmelt samples, ranging between 5.2 and 6.2. This result is consistent with a pH of unbuffered pure water in equilibrium with atmospheric CO_2. The pH variability has no clear effect on IN ability and lies within the uncertainty of the measurement.

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Note that the chemical properties of the snowmelt are representative of all types of aerosols present within the collected snow sample. Yet, only a subset of these aerosols are INPs and can thus be related to the freezing temperatures. Since total aerosol concentrations are several orders of magnitude greater than INP concentrations (Kanji et al., 2017), the chemical signature of an INP is possibly lost among total aerosol chemistry, even if the INPs are expected to be concentrated in precipitation samples. Therefore, it is not possible to develop a meaningful parameterization for INP concentrations based on TOC or conductivity from the snowmelt samples collected in this study (Figure 4).

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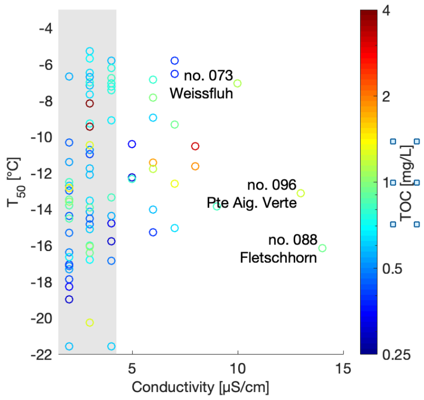

Figure 4 Scatterplot of T_{50} versus conductivity. TOC is shown in colour and the three most conductive samples are labelled with the sample number and sampling site. The range of the conductivity instrument background is shaded in grey.

3.1.2. Snowmelt filtration experiments

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Determining the size of INPs may be useful in identifying their sources, transport and sedimentation pathways in the atmosphere. The snowmelt was thus filtered to three sizes: 0.45, 0.20 and 0.02 µm (Figure 5). Filtrates from 0.45 µm and 0.20 µm filters retained the ice activity of the unfiltered sample. Three exceptions were observed from Figure 5; three of the highest T_{50} values from the unfiltered snowmelt lost between half to all their ice nucleating ability. On the other hand, filtrates from 0.02 µm filtrations completely removed the ice nucleating activity of the original snowmelt . We wondered whether salts from the filter could have been introduced into our sample, leading to a freezing point depression. However, the conductivity of molecular biology reagent water (89079-460, Sigma Aldrich, USA) filtered through a 0.02 µm filter was measured at 4 µS cm^{-1}, a value too small to lead to a noticeable freezing point depression. We conclude that the low freezing fractions observed with the 0.02 µm filtrates of the snowmelt was indeed due to loss of INPs in solution. We further conclude that most INPs in the collected snow samples were between 0.20 and 0.02 µm in size.

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This sizing test suggests that most of the INPs were found in the accumulation mode and smaller, and consequently only a few, albeit the most active, INPs were present in the coarse mode (0.5-5 µm) and larger. Sizes relevant for dry deposition are therefore only sparsely represented in our samples,

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suggesting that the majority of INPs found in the snowmelt were deposited in the snowpack through wet deposition by snow precipitation. This observation is expected and consistent with INPs preferentially removed by precipitation by initiating it (Stopelli et al., 2015). In addition, we make the same conclusion by plotting T_{10} and T_{50} which give the same trend in the size dependence of INPs (Figure S3). Although T_{10} has been used in previous studies (Irish et al., 2017), T_{50} was chosen here, as it was used in the rest of our analysis and as the same conclusions can be drawn from both values.

The majority of INPs found within our samples were smaller than 0.20 µm, consistent with findings of existing studies on organic matter (Irish et al., 2017, 2019; Wilson et al., 2015). In contrast, Mason et al., 2016 found that the majority of INP particles had aerodynamic diameters in the coarse mode when sampled from ambient air. However, it is important to note that it is not possible to determine the change in particle morphology when particles are immersed in water and therefore the same INPs may be responsible for the observed ice nucleation activity in precipitation and air samples. Regardless, the size classification of INPs from this study provides further support for the limited role of bacteria in ice nucleation over the Swiss Alps, since bacteria are larger than 0.20 µm. 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).

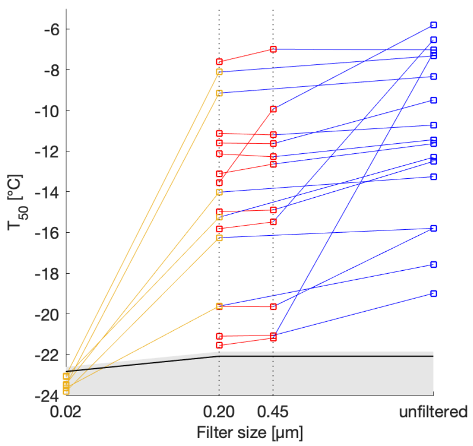

Figure 5 T_{50} as a function of filter pore size for selected samples. The T_{50} values of the background molecular biology reagent water are shown in black. The grey shaded area above the line represents one standard deviation. The samples were filtered through 0.02, 0.20 and 0.45 µm and the lines connect the different filtrates of the same sample. The T_{10} version of this graph can be found in the supplementary information for further comparison with Irish et al., 2017 and with Wilson et al., 2015.

3.2. Time series of T_{50} values

The variability of T_{50} values was observed throughout the measurement period and there was no trend over time (Figure 6). Noticeably, all samples taken in May 2018 had TOC values above the dataset average, and specifically above 1.2 mg C L^{-1}, however the time since their last snowfall was only 1-2 days and so these samples were still relatively fresh (Table S1). Nonetheless, the scatter of T_{50} values indicate no trend overtime of the freezing temperatures of INPs, consistent with a lack of seasonality in INP concentrations measured by a continuous flow diffusion chamber at Jungfraujoch (Lacher et al., 2018a). The absence of time dependency on the INP occurrence within the campaign timeframe indicates that the variability is consistently large throughout the entire timespan investigated.

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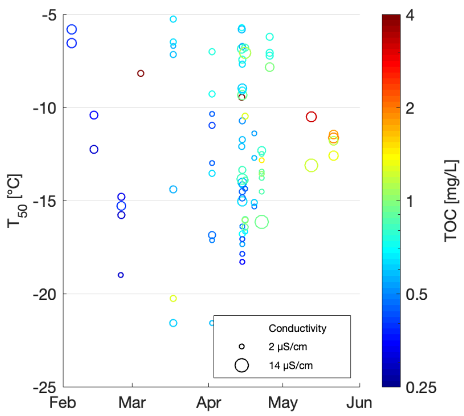

Figure 6 Time series distribution of the T_{50} values during the entire field campaign, displayed as months in 2018. TOC values of the samples are shown on a colour scale and conductivity values are shown on a marker size scale.

3.3. Spatial, altitudinal, snow age and depth variability of freezing temperatures

3.3.1. Spatial heterogeneity of freezing temperatures

To assess the local variability of INP concentrations spatially, samples from the same site, sampled on the same day were compared based on their distance from each other (Figure 7). The sampling site at Schilt showed consistent freezing temperatures for samples separated by only 5 m, but showed a difference in T_{50} of 4 °C for the sample located 300 m away, within a similar environment. Furthermore, the Sankt Annafirn site also displayed little variability of the median and mean freezing temperatures within a 5 m trajectory on top of the snowpack (Figure 7). Thus, we find that for two sites, snowmelt samples displayed similar INP concentrations within a radius of approximately 5 m. However, 300 m was enough to observe differences in INP concentrations, as was observed at Schilt.

However, for two other sites, Engstligenalp and Weissfluh, the T_{50} values of snowmelt varied by up to 8 °C and 10 °C, respectively. We studied the snowmelt at Engstligenalp due to its location in a flat flood plain of approximately 1 × 2 km in size (Figure S4). Because of the topographical homogeneity of the Engstligenalp site, similar local sources of INPs were expected. However, within the plain of less than 2 km, a difference in T_{50} of 8 °C was observed (Figure 7). Additionally, samples taken on the side of a snow hill at the Weissfluh site, showed a wider spread in freezing temperatures within a distance of only 8 m. Both the Engstligenalp and Weissfluh sites displayed visual evidence of snowdrift, which could explain the large variability observed compared to the Schilt and Sankt Annafirn sites (Figure 7). In fact, wind drift can lead to an extremely heterogeneous snowpack (Gauer, 2001), which could lead to locally heterogenous INP concentrations. Specifically, at Weissfluh, the origin sample and the samples at distances of 2 and somewhat at 4 m had significantly warmer T_{50} temperatures and a narrower spread in freezing temperatures than the snowmelt samples at other distances. 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 (Figure 7).

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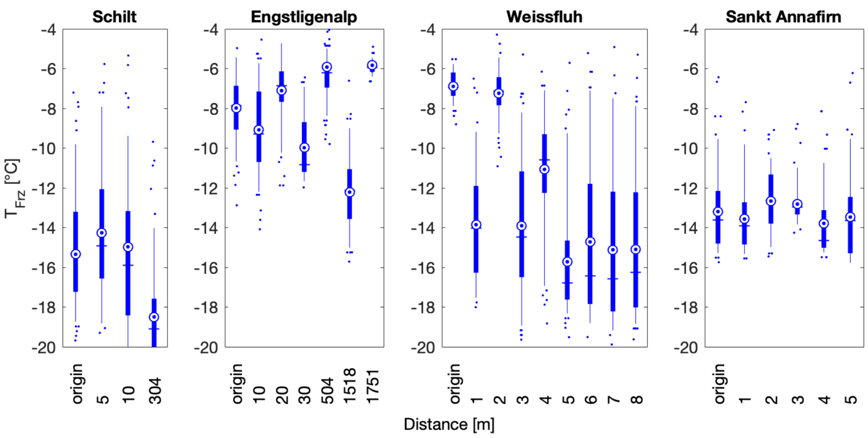

Figure 7: Spatial distribution of freezing temperatures of snow samples collected at four different flat sites, at Schilt on February 24, 2018, at Engstligenalp on April 14, 2018, at Weissfluh on April 15, 2018, and at Sankt Annafirn on April 22, 2018. Weissfluh and Sankt Annafirn are particularly striking since the snow samples collected at these sites show heterogeneity within 1 m distances. All the samples at the same location were collected on the same day. On the boxplots, 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, and the whiskers extend from the 5th to the 95th percentile.

3.3.2. Altitude dependency of freezing temperatures

To investigate the influence of boundary layer and local sources of INPs on snowfall across the Swiss Alps, sampling sites were chosen to cover a broad range in altitude. The sampling sites ranged in altitude between 440 and 3981 m a.s.l. with a median altitude of 2294 m (Table 1), thereby covering a range of sites affected by rare snow events (in Zurich at 440 m a.s.l.) and by eternal snowpack (at Fletschhorn at 3981 m a.s.l.). When investigating the role of altitude on INP occurrence in the same sampling region there is a qualitative decrease in T_{50} at higher altitudes for sites Fletschhorn and Pointe Aiguille Verte (Figure 8). The reasons are potentially two-fold. First, the more active INP nucleate ice at warmer temperatures and are therefore removed earlier and lower in the clouds (Stopelli et al, 2015). Second, INPs in the boundary layer are advected into the cloud due to orographic lifting or turbulence at the top of the boundary layer. Indeed, Lacher et al. (2018b) saw an increase in INP concentrations during periods of boundary layer air at Jungfraujoch, albeit at much colder sampling conditions than measured in this study.

In contrast to an altitude dependence observed at Fletschhorn and Pointe Aiguille Verte, Grosstrubel had no clear dependence on median and mean freezing temperatures with altitude, although samples at altitudes lower than 2500 m had significantly narrower spreads of the 25th and 75th percentiles (Figure 8). Considering the observed spatial variability when sampling at distances over 5 m, it is difficult to disentangle the effect of altitude and of spatial variability in INP occurrence. This dataset is therefore limited in its ability to compare free tropospheric snowfall and snowfall influenced by the boundary layer to assess the importance of boundary layer aerosols on ice formation in clouds.

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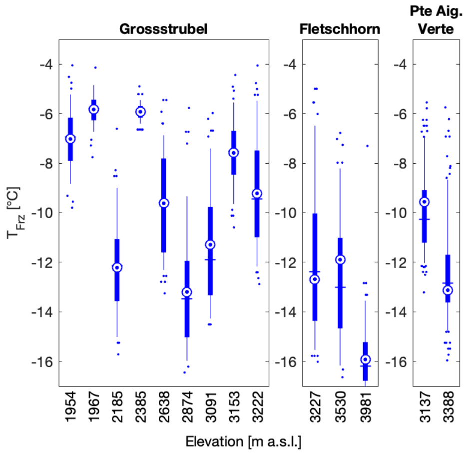

Figure 8: **Snow samples were collected at different altitudes at Grossstrubel on April 14, 2018, at Fletschhorn on April 22, 2018, and at Pointe Aiguille Verte on May 12, 2018. On the boxplots, the blue vertical line shows the median and is equal to *T_{50}*, the mean is shown as a blue circle with a concentric dot and the box ranges from the 25th to the 75th percentile, and the whiskers extend from the 5th to the 95th percentile. Note that the two Pointe Aiguille Verte boxplots contain 192 data points each as two individual snow samples were collected at the same location and thus combine for this figure.**

3.3.3. Snow age through time and depth

The effect of snow age on the TOC, conductivity and freezing temperatures were evaluated using two types of snow sampling and data analysis. The first method involved classifying the T_{50} temperatures of the snowmelt as a function of the day of the last snowfall reported at the nearest SLF station. The second method involved digging through the snowpack to the ground and collecting samples along the depth profile (see photo in Figure 10).

Comparing different snowpack ages, which we define as the number of days between the most recent snowfall at the nearest SLF site and the sampling day, allows the assessment of variables affecting snow properties since precipitation, including; (1) dry scavenging (Zhang et al., 2001), (2) microbial, plant and mineral effects from underlaying ground, (3) photochemical and ozone aging (Borduas-Dedekind et al., 2019; Gute and Abbatt, 2018; Kanji et al., 2013) (4), melt-process influence (Colbeck, 1981; Kuhn, 2001), (5) rainfall and (6) sublimation and deposition within the snowpack and between the snowpack and the atmosphere (Birkeland et al., 1998). The correlation between snow age and T_{50} was however found to be insignificant (Figure 9). However, the highest TOC value of 5.2 mg C L^{-1} was observed for snowmelt sampled on April 4, 2018, nine days after the last snowfall which could have allowed the surface snow to concentrate (Figure 6, Figure 9). However, this hypothesis does not hold across the rest of the samples. Based on this result, we conclude that INP concentrations are not preferentially found in fresh snow samples nor are they concentrated over time nor is there significant dry deposition. This result is consistent with Hartmann et al., (2019) who found similar variability and concentrations of INPs in ice cores, which have much longer ageing times. However, it is important to note that we did not investigate the meteorological conditions between snowfall and sampling, nor did we measure the INP concentration at the same location on different days without any snowfall in between the days. Thus, weathering, which has been shown to affect the INP concentration in collected snow samples (Stopelli et al, 2014), and dry deposition could have occurred to some extent. Nonetheless, we can conclude that the role of ageing does not change the INP activity beyond the natural variability of the samples.

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Sampling depth profiles in the snowpack permitted a comparison of snow that precipitated at different times in the same location without the logistical burden of returning to the site several times during the field campaign. Of note, the first widespread snowfalls that reached altitudes bellow 2000 m a.s.l. were registered on November 6, 2017 and thus, the oldest snow and the deepest depth samples would have precipitated no earlier than this date. The depth samples in all three locations were very diverse, with T_{50} values ranging from –20 to –5 °C (Figure 10). The depth profile sites of Alpetli and Chalberhöri were separated by only 1.5 km and were sampled 16 days apart, so one could have expected a similar pattern in the depth profile if INP concentration were driven on the synoptic scale. However, no similarity nor trend were found as a function of depth at the three sites (Figure 10).

Furthermore, the Alpetli site had consistently warmer T_{50} values at depths of more than 0.8 m from the surface of the snowpack in comparison to the other two sampling sites (Figure 10). Between 0 and 0.6 m beneath the snow at Alpetli however there was a wide spread in INP occurrence, and the freezing temperatures was similar to the boxplots of the other sites (Figure 10). The high activity of the INPs at Alpetli might be dominated by its particular proximity to trees shedding biogenic particles, known to efficiently nucleate ice (Morris et al., 2014). In contrast, the profiles at Engstligenalp and Chalberhöri had less variability in INP activity with depth and had a mean T_{50} value of –13.1 and –12.7 °C, respectively. Since the lower half of depth samples at the Alpetli site contained more efficient INPs in higher concentrations, we considered whether this high efficiency was due to contamination from the underlying soil or any organic matter within the soil, which has been shown to act as efficient INP in the immersion freezing mode (Hill et al., 2016; O'Sullivan et al., 2014; Suski et al., 2018; Tobo et al., 2014). As the depth of efficient INP extends well above the bottom of the profile (Figure 10) and as the other two depth profiles do not display this behaviour, we conclude that contamination from the underlying soil is likely not responsible for the high T_{50} values observed in the lower half of the Alpetli profile.

Although the age and exposure of the individual snow samples are unknown, there is no systematic decrease in INP activity with depth, consistent with our snow age observations and with literature ice core measurements (Hartmann et al, 2019). Therefore, we argue that snow depth profile measurements are an appropriate method to analyze the concentration of INPs over a winter season. However, a study comparing the concentration of INPs in freshly fallen snow to that of the same corresponding snow in a depth profile is warranted. Ultimately, such a study would help to understand the representativeness of the chronology within a snow depth profile, the influence of ageing on INPs and the impact of dry deposition on the concentration of INPs on a snow surface.

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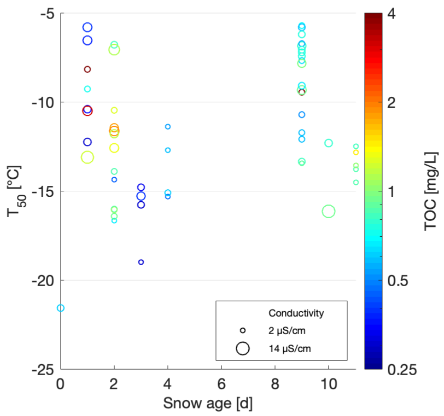

Figure 9: Scatterplot of T_{50} versus minimum snow age. TOC values of the samples are shown on a colour scale and the marker surface area represent the conductivity.

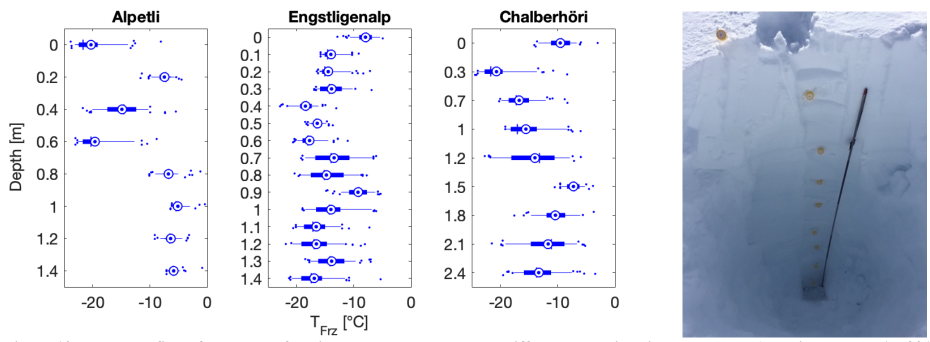

Figure 10: Depth profiles of snowmelt freezing temperatures at three different sampling sites, namely at Alpetli on March 17, 2018, at Engstligenalp on April 14, 2018 and at Chalberhöri on April 2, 2018 (Table 1). Right inset: a 2.7 m deep snow profile was dug with an aluminium avalanche rescue shovel at the sampling site Engstligenalp. The probe depicted is an avalanche rescue probe and was used to measure the snow depth. The fresh layer snow was 9 days old and the first snowfall at that site was recorded on November 23, 2017. On the boxplots, the blue concentric dot is the mean, the blue horizontal line is the median, the box is the 25th to the 75th percentile, and the whiskers extend from the 5th to the 95th percentile.

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3.4. INP concentration parameterization

Next, the freezing temperatures were converted into cumulative INP concentrations within the snowmelt (n_{sm}) following Eq. 2. The n_{sm} values were extrapolated to cumulative INP concentrations in air (c_{air}) by assuming a cloud water content (CWC) of 0.4 g m^{-3} (Petters and Wright, 2015) and the density of water of 1 g mL^{-1} (ρ_w), according to Eq. 3. This calculation assumes no dry deposition contribution (see section on snow age) and no scavenging of INPs during snow precipitation (Petters and Wright, 2015). Nonetheless, we appreciate that scavenging of microbial ice nucleators during rain events can occur (Hanlon et al., 2017).

$$c_{air} = \frac{n_{mw} \times CWC}{\rho_w} \quad (3)$$

A two-dimensional histogram of all INP concentrations could be obtained by binning the INP concentrations to a 1 °C bin-width laterally and a logarithmically constant bin-width vertically (Figure 11). The probability scale represents the probability of observing INPs within a given temperature and concentration bin based on all of our 88 observations, including the depth profiles. The measured

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temperatures for a given INP concentration fell nicely within values found in previously published studies (Petters and Wright, 2015), and furthermore within field-studies conducted at Jungfraujoch (Conen et al., 2012; Stopelli et al., 2014, 2016, 2017) (Figure 11).

The large variability in temperatures and INP concentrations made it difficult to find a clear influencing factor on INP occurrence, consistent with other field studies (Garimella et al., 2017; Lacher et al., 2018a; Welti et al., 2018). Diverse sampling sites in multiple regions with varying environmental factors and their accompanying uncertainties complicated the determination of INP concentration predictive factors. Yet, Stopelli et al. (2016) found that upstream precipitation and wind speed accounted for 75% of the observed INP variability at the Jungfraujoch site. Unfortunately, these meteorological parameters could not be assessed in this study. Nonetheless, this study's dataset was used to generate a parameterization for potential applications in simulating INP concentrations in as a function of temperature (Figure 11, Table 3). The parameterizations are given as $n^*_{nw}(T) = e^{-0.7T-6.02}$ for snowmelt and $c^*_{air}(T) = e^{-0.7T-7.05}$ for air, where $n^*_{nw}(T)$ and $c^*_{air}(T)$ are defined as the cumulative concentrations of INPs per mL of snowmelt and per m^3 of air, respectively, as a function of temperature (Table 3). Note that the asterisk is used to denote the parameterization values, and to distinguish them from the measured values from our study. In particular, the $c^*_{air}(T)$ parameterization can be directly applied to general circulation models in order to predict INP concentrations as a function of temperature.

The large variability throughout the dataset (Figure 6) suggests that differing synoptic conditions influenced the snow at different timespans. Although contributing factors to INP occurrence are likely due to differing source regions and microphysical pathways upstream of the sampling locations, they could not be matched with physical or chemical properties (Table 2). Additionally, high variability of snowmelt within close proximity of each other on the same day suggest that a site-specific contributing factor would be responsible. However, another explanation for local variability in INP occurrence is the wind drift which might have transported snow during or after a precipitation event or might have uncovered an older layer, which was then later sampled next to a freshly precipitated layer. These complicating effects yield a heterogeneous snowpack (Gauer, 2001), consistent with the measured heterogenous INP concentrations in this study. Nonetheless, the total measured INP concentrations fall well within the range of previously published values (Figure 11). Due to the fact that INP occurrence in the measured snow samples have been determined as non-site specific, the studies conducted at Jungfraujoch, measuring the free troposphere are representative for INP assessments across the Swiss Alps.

Table 3: Coefficients of the INP concentration parameterization, based on values between 0.2 and 90 INPs per mL and 0.08 and 36 m^{-3}.

Function: e^{a*T+b}	a	$b = n^*_{nw}(T)$	$b = c^*_{air}(T)$
Mean	−0.70	−6.02	-7.05
Lower bounds (mean-1 SD)	−0.60	−6.81	-7.84
Upper bounds (mean+1 SD)	−0.96	−5.84	-6.87

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This study's dataset was used to generate a parameterization for potential applications in simulating INP concentrations in precipitation ($n^*_{nw}(T)$) and in air ($c^*_{air}(T)$) (Figure 11, Table 3). The parameterizations are given as $n^*_{nw}(T) = e^{-0.7T-6.02}$ for meltwater and $c^*_{air}(T) = e^{-0.7T-7.05}$ for air, where $n^*_{nw}(T)$ and $c^*_{air}(T)$ are defined as the cumulative concentrations of INPs per mL of meltwater and per m^3 of air, respectively, as a function of temperature (Table 3). In particular, the $c^*_{air}(T)$ parameterization can be directly applied to general circulation models in order to predict INP concentrations as a function of temperature.¶

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Influence on cloud cover and precipitation through Saharan dust for example poses difficulties to numerical weather models in Europe (Knippertz and Todd, 2012), of which some portion is linked to uncertainty in INP activity.

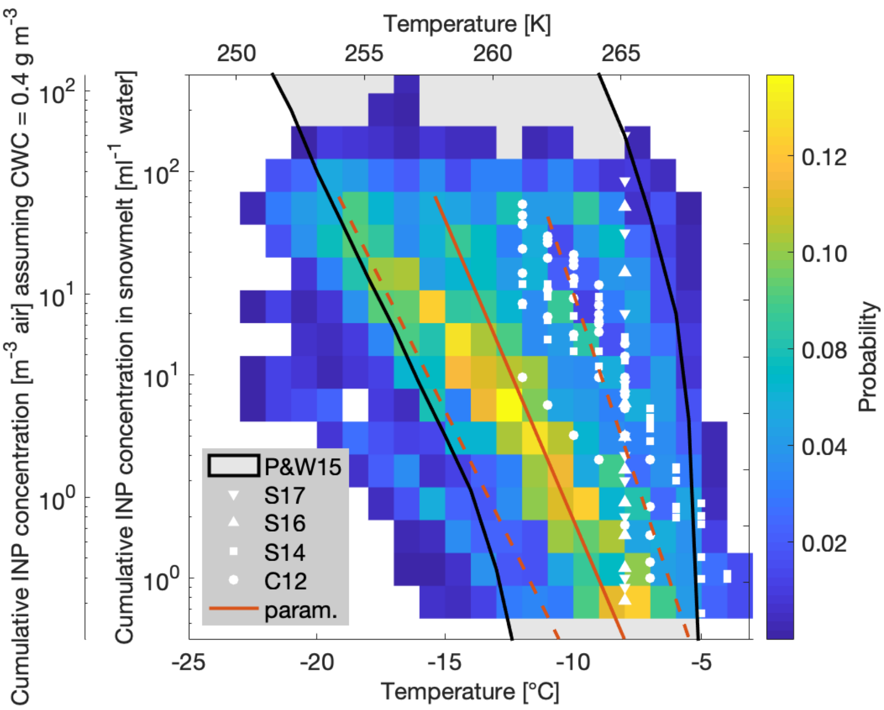

Figure 11: Observations in INP concentrations in snowmelt (n_{sm}) and in air (c_{air}) for all of the measurements (88 samples) as a function of temperature. The observations are shown as probability in a two-dimensional histogram. The grey area with black borders shows the concentrations found in the literature (Petters and Wright, 2015). The white symbols correspond to other field-studies in the Swiss alps (Conen et al., 2012; Stopelli et al., 2014, 2016, 2017). With the parameterization $n^*_{mw}(T)$ and $c^*_{air}(T)$ in red (solid) and 1σ standard deviations thereof (dashed).

4. Atmospheric implications for mixed phase cloud glaciation

An accurate onset temperature of ice nucleation in supercooled clouds and the subsequent temperature transition to mixed-phase and eventually to glaciated clouds are important factors for determining cloud lifetime and thus radiative forcing. Yet, these onset and transition temperatures are variable and lead to large uncertainties in weather and climate models (Boucher and Quaas, 2013; Prenni et al., 2007; Vergara-Temprado et al., 2018). The glaciation temperature of a cloud further depends on the supersaturation within the cloud and is highly sensitive to updraft velocity (Korolev et al., 2017; Korolev and Isaac, 2003; Korolev, 2008). As ice crystal concentrations in clouds can be several orders of magnitude higher than INP concentrations (Wex et al., 2010), secondary ice processes can play an important role in the evolution of supercooled clouds (Beck et al., 2018; Hallett and Mossop, 1974; Lauber et al., 2018; Mignani et al., 2019; Petters and Wright, 2015). However, the process of ice multiplication is not fully understood, with multiplication factors ranging from one to multiple orders of magnitude (Mignani et al., 2019; Wang, 2013). Nevertheless, it has been proposed that complete glaciation of supercooled clouds can be initiated through ice multiplication with less than 10 INPs per m^{-3} (c_{air}) (Crawford et al., 2012; Mason, 1996) by the process of riming and ice splintering (Hallett and Mossop, 1974; Mossop, 1978). Therefore, by choosing a threshold for the number of INPs required to glaciate a cloud, say 10 m^{-3}, we can estimate the frozen cloud fractions based on our cumulative INP concentrations (Figure 12).

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Specifically, we identified the freezing temperature from all 88 of our samples which corresponded to a calculated c_{air} value of 10 m^{-3} (also equivalent to 25 ml^{-1} of snowmelt (n_{sm}). We then sorted these 88 temperatures from warmest to coldest and plotted them in a linear space from 0 to 1, corresponding to a frozen cloud fraction (Figure 12). We repeated this analysis with assumptions of c_{air} values of 20 m^{-3} and of 40 m^{-3} required for cloud glaciation. However, the highest INP concentration from this study did not exceed ~ 200 m^{-3}, because of the -22.5 °C limit of detection of DRINCZ. This temperature thus limits the extrapolation of frozen cloud fractions to lower temperatures. Yet, since other studies suggest that hundreds of INPs per m^{-3} can be necessary for cloud glaciation to occur (McCoy et al., 2016), in contrast to the lower limit of 10 m^{-3} by Crawford et al. (2012), we use our parameterization to estimate the frozen cloud fraction from an order of magnitude higher INP concentrations. The change in temperature required for c^*_{air} to increase by an order of magnitude can be calculated as $\Delta T = \frac{ln(10c^*_{air})}{0.7} - \frac{ln(c^*_{air})}{0.7}$ and is equal to –3.3 °C. Thus, to extend the frozen cloud fraction using 400 m^{-3} as the INP concentration necessary for cloud glaciation, the frozen cloud fraction at 40 m^{-3} was translated to colder temperatures by –3.3 °C (Figure 12).

Our 50 % frozen cloud fractions fall within the range reported from airborne, ground based, and satellite measurements as summarized in McCoy et al (2016) (Figure 12). Furthermore, these results are remarkably consistent with the typically observed transition zones between supercooled liquid and ice clouds in models and observations (Costa et al., 2017; Henneberg et al., 2017; Lohmann et al., 2016; McCoy et al., 2016; Pithan et al., 2014). Indeed, global circulation models partition liquid and ice in a given atmospheric volume as a monotonic function of temperature, but precipitation and freezing and melting cycles affect the liquid water to ice ratio, thereby modifying the cloud phase transition in often poorly constrained ways (Cesana et al., 2015; McCoy et al., 2015). Thus, temperature remains the dominant effect on influencing liquid cloud fraction (Tan et al., 2014).

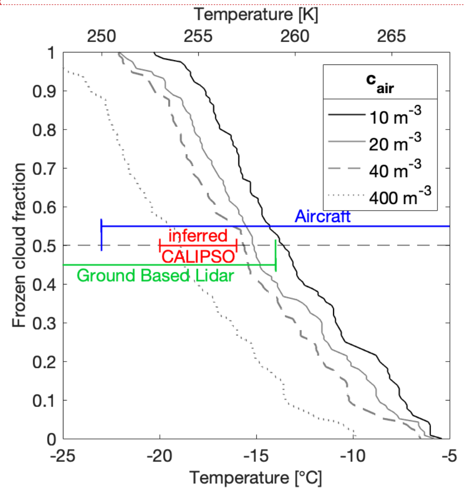

Figure 12. Frozen cloud fraction as a function of temperature derived from all 88 samples from this study at four different critical INP concentrations necessary for cloud glaciation, namely 10 m^{-3}, 20 m^{-3}, 40 m^{-3}. The frozen cloud fraction was further estimated using the c^*_{air} parameterization to 400 m^{-3} (see text). The aircraft (blue), inferred CALIPSO (red) and ground based lidar (green) temperature ranges for 50% frozen cloud fractions are adapted from (McCoy et al., 2016a).

Code/Data availability

Freezing temperature data for all 88 snowmelt samples are available in the supplementary information. Raw data and MATLAB data analysis code are available upon request.

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$T_{glac.} = -(ln(n^*_{nw}) + 6.02)/0.7$ (4)

By reorganizing Eq. 4, the change in temperature required for c^*_{air} to increase by an order of magnitude can be calculated as $\Delta T = ln(10^{om})/0.7$ where om is the order of magnitude in $n^*_{nw}(T)$. When $om = 1$, the change in temperature is equal to –3.3 °C. To extend our glaciation temperatures beyond 40 m^{-3} and thus beyond the measured INP concentrations in this study, we can estimate c_{air} at 400 m^{-3} (Figure 13).

Moved up [3]: By rearranging Eq. 3, the INP concentration parameterization in Table 3, the temperature required for a certain concentration of INPs can be calculated as:

$T_{glac.} = -(ln(n^*_{nw}) + 6.02)/0.7$ (4)

By reorganizing Eq. 4, the change in temperature required for c^*_{air} to

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Competing interests

The authors declare that they have no conflict of interests.

Author contributions

ROD and NBD designed the field study with contributions from KPB. KPB and NBD collected the samples. KPB conducted the experiments and analysed the data with contributions from ROD and NBD. The manuscript was written by all authors.

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