

Referee comments on "Using spectra characteristics to identify ice nucleating particle populations during winter storms in the Alps" by Creamean, Mignant, Bukowiecki and Conen.

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1 Summary

The two related goals of the paper are: in-cloud measurements of INPs and the identification of INP type or composition. The paper makes a good contribution to INP studies by providing a data set involving INP measurements in air (with size sorting), rime, and snow at the same time. They show evidence for the dependence of INP abundance depending of air mass trajectory.

- 5 Some inferences are drawn regarding local versus distant sources of INPs. The authors' conclusions are relatively simple and reasonably well supported by the data, but the analyses are limited without chemical and other supporting measurements. All that notwithstanding, an important revision is needed to take into account the points made in the Section 3 below.

2 General points

- 10 1) Stratification by storm type in Sections 3.1 and 3.2 focus only on air trajectory. It is surprising that precipitation during preceding days, cloud height, depth etc. are not considered. In some fashion those parameters are probably related to the air mass trajectories, but omission of attention to those factors makes the arguments presented sound incomplete and superficial. This sort of narrow focus in the treatment of the data is unsettling in various parts of the paper. Results in Section 3.2 and Fig. 6 are presented without separation of in-cloud and out-of-cloud samples. This sort of problem is not unique to this data set. It has also been there with the numerous papers reporting INP, and other, measurements at JFJ observatory.

- 15 2) The paper expands on other INP data from JFJ by determining INP abundance as a function of temperature, i.e. INP spectra. This is a useful step and avoids the somewhat simplistic comparisons that, unfortunately, arise from discussing and comparing INP concentrations without distinguishing between data obtained at -10°C or at -30°C . INP composition, their sources and their transport can be expected to be significantly different for widely different temperature regimes. The greater focus on this aspect of INP studies is a good contribution. Expanding further on the point, use of differential INP spectra for gaining
20 more information about INPs is a commendable goal as it goes even further in focusing on INP characteristics than the more frequently employed cumulative spectra. However, the manner this is done in this paper deserves closer examination and revision, as argued under the heading "Differential spectra" below.

3) As a general comment about interpretations of INP spectra, I'd express some caution. The paragraph in lines 267-285 about warm and cold-mode INPs is somewhat superficial. The temperature ranges and magnitudes of INP concentrations need to be considered quantitatively before deductions are made about possible sources of the INPs. Just looking at freezing temperatures (like onset and percentiles frozen) can be misleading since they depend on sample concentration (e.g. filter exposure time, dilution), number of drops tested and drop volume.

4) Related to the point raised in 1), one wonders what is underlying vision for comparing or correlating INP concentrations in air (via the filters) and in snow and in rime. These are three very different pathways for the INPs. Sources for each component can be different in space and time. With a broad separation such as NW versus SE airmasses perhaps these differences become unimportant but not necessarily so. It would be useful to read about the authors' perception on this issue. The paper glosses over such concerns thereby creating a degree of unease about the meaning of the results.

3 Differential spectra

5) What in this paper is called (Fig. 7, lines 26, 145, and other places) "normalized differential INP concentration" does not correspond to previously used definitions of that term. Earlier definitions are found in the references cited in the paper. Further references are Vali et al. (2015) (Atmos. Chem. Phys. 15, 10263-10270) and the recent Vali (2018) (Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2018-309>). To summarize, briefly, differential spectra, or differential INP concentrations, reflect the concentration of INPs or active sites per unit temperature interval. It also describes the probability of freezing for drops of given volume at given temperatures. As its name indicates, the differential spectrum corresponds to the differential of the cumulative spectra. The cumulative spectrum is denoted by $[INPs(T)]$ on line 147 of the paper. Other frequent designation of this cumulative spectrum¹ is $K(T)$, with $k(T)$ used for the differential spectrum. Both quantities are closely related to site density. Because the values are specific to each temperature, independently of what other INPs may be present in the sample, the differential spectra provide more acute diagnoses of INP characteristics than the more frequently employed cumulative spectra or the fraction frozen.

6) Line 152 states that differential values were obtained from the cumulative concentration. In fact, the results shown in Fig. 7 (right hand panels) and then used in Section 3.3 appear to represent some different quantity because differential spectra $k(T)$ usually exhibit a steady rise with decreasing temperatures at low temperatures. The large drop in the spectra at low temperatures seen in the right-hand panels of Fig. 7 suggest that what is shown in the graphs represent the differentials of the frequency of freezing events, $f(T)$, not of the cumulative INP concentration. Since fewer and fewer drops are left as the sample drops freeze and $f(T)$ approaches unity at the lowest temperatures in an experiment, the increase in $f(T)$ per temperature interval, $\delta f(T)/\delta T$, can be expected to drop off, just as it seen in these figures.

7) The differential $\delta f(T)/\delta T$ is a valid representation of the measurements but it lacks clear physical meaning. That differential is nearly the same as the 'freezing rate, R(T)' defined in Section 4.6 of Vali et al. (2015), but without the first ratio in

¹Symbols used in here follow Vali et al. (2015)

that definition. For $f(T) \ll 1$, i.e. at the higher (warm) end of the range $\delta f(T)/\delta T$ the first term in $R(T)$ is close to unity and $\delta f(T)/\delta T \simeq R(T)$ so that minor peaks are resolved in $\delta f(T)/\delta T$ without the distorting effect of reduced sample size. However, at smaller values of $f(T)$ that distortion becomes dominant, so that little significance can be attached to peaks such as those shown in Fig. 7 of the paper. That this is indeed the case can be demonstrated by changing the drop volumes. Such a change would shift the positions of the cold-mode peak. In contrast, $k(T)$ for different drop sizes overlap and form a continuous curve (e.g. Fig. 4 in Vali (1971)(J. Atmos. Sci., 28,402). Another illustration can be thought of with dilutions of the sample with INP-free water. The drop-off beyond the cold-mode peak of the original sample will not be reproduced with the diluted sample since large numbers of drops will be still available for substantial rates of freezing at that temperature. In this case too, $k(T)$ for successive dilutions overlap and form a continuous curve.

5 The ratio $\delta f(T)/\delta T$ has not been used in the past to describe experimental results, so the authors should define it clearly and explain why they chose that presentation. Renaming that quantity would be helpful in avoiding future misunderstandings. Alternatively, they could consider changing the analysis to using $k(T)$ or $R(T)$ to represent their data. The point is that $\delta f(T)/\delta T$ is an experiment-specific relative characterization of the results, while other, more objective quantities could be used.

10 An additional effect of the choice of analysis in terms of $\delta f(T)/\delta T$ is that freezing of some of the the drops at warmer temperatures decreases the number and, potentially, the gradient of the frequency at lower temperatures. Is that the reason why the so-called cold-mode for the SE samples seems to occur at warmer temperatures in Fig. 7 than for the NW samples?

4 Specific points

lines 33-34: Precipitation production is not limited, as implied, to clouds that contain both liquid and ice, Unfortunate phrasing. But the mention of mixed-phase clouds (MPC) in the subsequent several lines keep mixing the focus on the importance of INPs in general and the importance of MPCs for precipitation production.

20 line 37: There is jump here from the discussion of the INPs as important problems for weather and climate models to the practical problem of measuring INPs. Also, from none of the foregoing follows the argument of line 41 that in-cloud measurements are indispensable fo progress.

25 line 43: Another jump in logic. What?s said here had to be already taken for granted for the previous paragraph to make sense.

lines 43-76: These two paragraphs get into too much detail. The present study does not address many of the details described as possibly controversial or uncertain, so raising the issues is a diversion. Other review papers deal with what is known about the components of INPs. This paper is directed only to a broad classification of INP types by composition.

30 lines 104-105: To what extent did wind removal of snow from the pans hinder determinations of snowfall rates?

line 131: Drop size variability introduces errors due to INP content being proportional to volume, as also pointed out in Creamean et al. 2018b. How significant this error is depends on the range of variation of drop volumes. The way

this line is phrased is incorrect and subjective. The error introduced may be small or equal compared to other sources. The only concise way to test for this would be to do large numbers of repeated runs from the same sample. Recommend to change 'indeterminable' to 'undetermined' in line 130 and eliminate line 131.

5 line 136: Cooling-rate dependence is small but not non-existent as shown in the references cited. If the authors' tests showed no discernible dependency it must be because other variations hid the cooling-rate dependence.

line 142: The reader deserves to know what this 'custom software' was that connects visual detection with a recording system. Also, one wonders why is cooling rate considered a factor for each drop, when the previous paragraph states that there is no 'discernible' effect.

10 line 144: How were triplicate samples combined for analyses? Averages of fraction frozen at each temperature? How much variability was there among the three runs per sample?

line 153: Assigning significance to the fact that the data shown in the references cited extended to only about -20°C may indicate a misunderstanding. The -20°C limit was due to no fundamental limitation of the validity of definition of differential spectra. It was due to the background from distilled water and from the supporting surface becoming important at colder temperatures.

15 lines 160-163: Can the method used distinguish between clouds enveloping the observatory and clouds just a few hundred meters above it? Were there no in-situ instruments or visibility measurements available for detection of clouds?

line 173: Please define SDE.

Fig. 6 panel a: Are the lines shown averages for the each condition? If the curves in panel a) are differential concentrations, as the presence of a peak suggests, then the units of the ordinate are incorrect.

20 Fig. 6 panel b-h: The numbers of points doesn't seem to be the same in each panel. Some points may be missing for samples that had no freezing event at -10°C, but shouldn't the other panels contain the same numbers of points. It is hard to tell if that is the case. Perhaps due to overlap of points. Please check and explain if some additional selection has been made.

25 line 233->: It would be helpful if the authors stated what signals they consider significant in Fig. 6. The figure is complex and the data noisy. The jump into comparisons with the Stopelli data is hard to follow without first pointing out what deductions are extracted from Fig. 6.

lines 239-240: The phrasing could be improved. You mean cumulative concentrations at -15°C as the criterion used for the assertion?

30 line 243: Onset definition is based on single drops? Wording in parenthesis on lines 244-245 should be corrected and made more specific.

lines 265-266: This assertion about the role of hoar frost needs some explanation.

lines 267-285: This paragraph will need to be revisited after the authors examine the issue raised about the differential spectra, and also take into account the comment on line 153 above.

35 Section 3.3: Because of misgivings about the meaning of the cold-mode data, it is difficult to sort out what part of the conclusions is supported by the data. Focusing only on whether a minor peak was present or not and forgetting

about the cold-mode **W** entries in Fig. 8 the analysis may not change much since the position of the **C** peaks is almost uniform for all the days and contains little additional information.

line 348: Reference to 'differential INP spectra' need change in light of Section 3 of these comments.

line 351: The meaning of statements about the relative magnitudes of INP concentrations is unclear unless the temperature to which they refer is specified, and it is clear what quantity is used for the comparison ($f(T)$ or $k(T)$ or $K(T)$, or other).

line 352: Reference to bi-modal spectra may also need to be re-thought depending on what changes are made regarding the use of $\delta f(T)/\delta T$ or its replacement. The

line 354-359: The thoughts expressed here are basically sound but are somewhat overstated regarding how much information can be gained from cumulative versus differential spectra. The latter are more specific and make it easier to see differences among samples, but the information content is not different.