

**Comments by Gabor Vali on the revised version of "Using freezing spectra ..... Alps" by Jessie M. Creamean, Claudia Mignani, Nicolas Bukowiecki and Franz Conen.**

March 29, 2019

The paper contains data from a field campaign of about one month duration at the Jungfraujoch observatory. Aerosol, rime and snow samples were taken for analyses of ice nucleating particles (INPs).

The main result is a difference between two wind regimes associated with air mass origins that differ in altitude and geography. These are sensible results, in reasonable accord with previous work and with what is known about INP source/activity relationships. Thus, the results here obtained reinforce general understanding but do not yield significant new insights regarding the sources and composition of INPs. What is new in the paper is the use of differential spectra of INP activity. While there are other good measures of INP activity, the use of differential spectra allows a clearer identification of the properties of INPs.

The basic result of the work emerges from the paper with sufficient clarity in spite of the somewhat excessive effort to provide explanations for every detail. Many rationalizations are speculative and qualitative. The connections explored provide some sense of reassurance that no unexpected phenomena need to be invoked, but do not rise to the level of proofs. This is not unusual for material dealing with the multitude of issues here involved: distant aerosol sources, in-cloud processes, precipitation fall-out, complex terrain and experimental difficulties. So, while readers may well find the paper overly burdened with detail, the authors' efforts can be respected.

Regarding the analyses of nucleus spectra, the inclusion of  $K(T)$  and  $k(T)$  is a clear improvement over the earlier version. As defined by the equations presented, these quantities provide absolute measures of INP concentration per volume of liquid (for rime and snow) and per volume of air (for the aerosol samples). However, a normalization is introduced (line 175) for reasons that are not clear. Perhaps it is done to compare the rime and snow samples with the aerosol samples on the same scale (line 314). That could be accomplished by plotting the two sets of data with different abscissa scales without the loss of absolute values of concentrations. As it is, the data from this paper cannot be compared to other results. Even on a procedural basis there is a problem: maximum values of  $k(T)$  are used as normalizing factor for each sample but these maximum values are obtained with very low values (usually 1 or 2) for both  $\Delta N$  and  $N(T)$  and so the ratio has large uncertainties and arbitrariness. The paper would gain from eliminating this normalization. If the authors insist on retaining it, the maximum values chosen should be presented.

Onset temperature and  $T_{10}$  and  $T_{50}$  are introduced as metrics for the INP measurements on lines 296-298 and are used in Fig. 6 (b) to (d) and in Section 3.3. This is understandable in view of the frequent use of these metrics in the past. Their use in this paper is regrettable as it goes against the goal stated in the title of the paper and elsewhere that spectra provide the focus of the analyses. The onset

temperature is statistically a weak metric,  $T_{10}$  and  $T_{50}$  are somewhat better, but both are experiment specific, most significantly because of volume and sample size dependence. They do not constitute independent measures of activity to reinforce what is indicated by  $k(T)$  and  $K(T)$  and they are easily substituted by more defensible measures. In this paper, the value of  $K(T)$  at  $-15^{\circ}\text{C}$ ,  $K_{-15}$ , or at some other nearby temperature would be a good way to distinguish between results for different air-flow regimes. To re-state, the results shown in Fig. 6 (b) to (d) are not wrong, but by-pass the stated goal to use spectra measures and are weaker metrics than what the spectra could provide. Thus, the paper loses effectiveness. Comparisons to other works are hindered.

Why  $df/dT$  is retained in the analyses is somewhat puzzling. This quantity provides no information different from  $k(T)$  for the warm part of temperature range, i.e. for low values of  $f(T)$ , and is clearly an artifact at the cold end of the data range. As argued in my previous comment (point #6)  $df/dT$  necessarily falls off when  $f(T)$  approaches unity and the location of the peak reflects primarily what fraction of the sample drops have already frozen at higher temperatures, not what is significant for the colder temperature region. Should the volume of the drops in the freezing tests been smaller, or should the water samples have been diluted with INP-free water, the  $df/dT$  peaks would have shifted to lower temperatures. As it is, the  $df/dT$  plots provide some comparison among the samples but are clearly misleading when used as a basis of interpreting the results in terms of INP types.

Phrasing the results in terms of warm mode and cold mode INPs (lines 261-265 and more) is a tempting but imprecise argument. While peaks near  $-10^{\circ}\text{C}$  are significant, the cold mode is an artifact. The most common pattern of INP concentration functions is a monotonic increase toward colder temperatures. The monotonic increase can arise for a single material or for mixtures of several types of INPs. In general, the interpretation of signatures in  $k(T)$  is not yet well studied. When a peak is found in the differential spectra, the peak it can be assumed to represent either the presence of a specific INP material or a type of site configuration with frequency above the general trend. However, decomposing differential spectra in terms of different contributions will require much further work. The association of peaks near  $-10^{\circ}\text{C}$  with biological INP sources, and activity observed below about  $-17^{\circ}\text{C}$  with mineral sources should be made with more recognition of its tentative basis than is done in this manuscript. This is specially the case, since no independent analyses of composition, particle surface properties or other potentially relevant parameters are presented in this paper.

Regarding different spectral features, it is remarkable that the slopes of the spectra below about  $-17^{\circ}\text{C}$  are very similar for the all snow and rime samples. While near  $-20^{\circ}\text{C}$  there is up to factor 100 range in concentration for the snow samples and not much less for the rime samples, the slopes are quite similar for all cases. Aerosol samples show more variability for the lower temperature range but still aren't far from the trend seen in the snow and rime samples. Such features are noteworthy but the underlying causes are not yet clear.

Minor points:

- line 58            Seems like the word 'While' is an error
- line 67            'intact' instead of 'in-tact'
- line 80            'intercomparison' doesn't express well the combined use of the data from the three different samples
- lines 100-101    There is a substantial difference in how the main goals and accomplishments of the work are stated on lines 80-81 and on these lines.
- line 109           'inherent time' ???
- line 117           'rime' instead of 'rimed'
- line 117           What is known about the collection efficiency as a function of particle size for the intake configuration?
- lines 145           The error introduced by variation in drop volume can be ascertained from the equations used for  $k(T)$  and  $K(T)$  -- it is of direct proportional magnitude. Stating this uncertainty as undetermined is incorrect.
- line 149           Was the variation in cooling rate due to slowing as the temperature lowered? Was it variable from one experiment to the other?
- line 162           ' average for each drop' ???
- lines 188, 195    Have BLI and SDE been defined?
- line 331           'not all samples contain a warm mode' would be better phrasing
- line 350           This is a problem with the use of  $df/dT$  for analysis. If all drops froze at temperatures above about  $-17^{\circ}\text{C}$  for this sample, the concentration of less active nuclei could have been determined by dilution of the sample with nuclei-free water.
- line 371           'differing' may be better instead of 'variable'
- Fig. 6a            A legend placed inside the diagram area would be better than the one along the right-side axis.
- line 373           At what temperature is this range of concentrations evaluated?