

Anonymous Review of *Sources and nature of
ice-nucleating particles in the free troposphere
at Jungfraujoch in winter 2017*

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1 Summary

In this work, Lacher et al., measured ice nucleating particle (INP) concentrations at -31 °C at the high-altitude research station atop Jungfraujoch (JFJ). INP concentrations were measured semi-continuously for one month (Jan. 22 - Feb 22., 2017) using the Horizontal Ice Nucleation Chamber (HINC). To increase large particle concentrations in the free troposphere, a Portable Fine Particle Concentrator was used. Finally, offline INP concentration measurements were taken to access warmer temperatures in the INP spectra. In addition to INP concentrations, the authors also measured aerosol size distributions, single-particle composition (LAAPTOF, ALABAMA), and bulk aerosol composition (AMS) in parallel. In addition to these aerosol measurements, size distributions and single-particle composition (ALABAMA) were measured behind an ice-selective counterflow virtual impactor (ice-CVI). Finally, back trajectory models were used to help define the sources of INP.

Overall, this work contains an impressive amount of information, which may be helpful to elucidate the role of aerosol size and composition on atmospheric ice nucleation. The HINC instrument is well-known to the ice nucleation community, and has published several "long-term measurement" publications." Much of the measurements are taken in the free troposphere, which is more relevant to cold-cloud formation than most ground-site operations. The paper itself is well-written, and only has a few technical corrections.

Most of the analysis is interesting, I particularly enjoyed the FLEXPART + single-particle mass spec. analyses, as well as the IPR measurements; however, several of the analyses may need further investigation. This are outlined in the general comment section. Most of the general comments are about the Spearman's rho analyses in Figure 5.

2 General Comments

- The comparison between these methods is not quite apples-to-apples. The size ranges and detection limits of all of these instruments are quite different. The authors do a good job of describing this problem in Section 2.3.1, but I feel the authors should add a Figure of "Detection Efficiency vs. Size," and add curves for at least HINC (with and without the PFPC), LAAPTOF, and ALABAMA.
- It is really hard to determine what the Spearman's rho values actually mean. While it's a well-known equation, it's main purpose is to detect if the relationship between two variables is monotonic. In this paper, it is used to define a correlation between INP concentrations and other aerosol measurements taken in parallel. It's a subtle distinction, and likely matters little if the rho values are very high (say > 0.8), but it becomes difficult to envision what a rho of 0.5 really means in this context. I suggest that the authors spend some time defining the equation and also plotting some rank correlations in the supplemental to help the reader envision how good / poor these rank correlations are.
- Likewise, I'm not sure that taking the mean \pm std. dev. of the rank correlations really defines what m/z have "significant" correlations. It makes the assumption that, in general, peaks in single-particle mass spec. are not correlated with INP concentrations. This assumption was not rigorously tested, at least in this paper, and I see no good reason why it would be true. For example, Mineral Dust spectra contain many relatively unique peaks; ostensibly, these peaks will correlate with INP concentrations.
- One of the most troubling aspects of this paper is that some of its results contradict themselves. For example, Figure 5 indicates that sea-spray aerosol (SSA) are correlated with INP; however, Figure 7 indicates that the opposite is true. One reason for this is that many peaks are not unique to a single particle type. Thus, trying to attribute a peak to single particle type as done in Figure 5 may give erroneous results. For example, m/z 60 is defined as mineral dust (SiO_2^+ for LAAPTOF, but elemental carbon (C_5^+) for ALABAMA. I suspect that it shows up in each particle type for both single-particle mass specs.

3 Minor Comments

- Some Figures are extremely hard to read in print format. I can not read the axes in Figure 4, some text and the "markers" in part C of Figure 5, and the legends in Figures 9 and 11.
- Line 155: A detailed description of how the concentration factor needs to be added here. To say that you increase aerosol $\zeta 100$ by a factor of 20 is greatly simplifying what is happening.

- Line 160: The caveats of converting ambient [INP] to n_s should be addressed here. By definition, n_s assumes that ice nucleation is deterministic, but it has been shown that this is not necessarily true for aerosol populations with a wide variety of ice-active site densities.
- Line 245: You are biased by the mass spec. detection efficiencies. Was there any attempt at normalization to the optical particle counters?
- Line 469: This statement is incorrect for two reasons. One—a large signal in one particle could greatly skew the average because the signal spans several orders of magnitude and a linear average was applied. That is one INP disproportionately affecting the signal average. Two—a single peak in a mass spectrum may qualitatively scale with abundance of that element or fragment, but it likely does not scale with the abundance of a "substance" or ice-active site.
- Line 537: I believe that dust particles fluoresce slightly in the WIBS. This is one reason why the FBAP thresholds are so strict. This should be mentioned here—or it should be mentioned why dust may not be suspected.
- Line 626: I'm not quite sure how you arrive at the conclusion that 70% of EC particles also contain mineral dust. From general aerosol knowledge, this seems like a vast overestimation. Thus, more details are need to support this statement.

4 Technical Comments

- Line 42: Are metallic particles and biological particles each 10% or are they 10% together?
- Line 60: I would not say that INP concentrations are entirely "unconstrained." Plenty of papers show a reasonable range of ambient INP concentrations depending on the air mass.
- Line 116: Is there an estimate of the transport time from the MBL to JFJ?
- Line 141: I think it should be "[INP] are naturally low ..."
- Line 141: This sentence could be split into two sentences. The phrase "but is generally higher ..." it difficult to follow because there are many potential subjects –e.g., nucleation temperature, detection limit, [INP], etc.
- Line 190: Should it be "into" instead of "onto?"
- Line 200: Should it be "decreases" instead of "decreased?"
- Line 206: Should it be "mJ / pulse?"

- Line 289: What is the typical size range of ice crystals at JFJ?
- Line 343: There is a lot of information in Figure 2, so it would be instructive to highlight these periods in Figure 2.
- Line 353: This reference to the Kammermann paper is not really supported by Figure 9.
- Line 483: Please move the legends in Figures 6 and 7, they are obstructing one of the high INP periods.
- Line 563: More secondary ice from your calculations, right? The way this sentence reads, it sounds like it's not an inferred measurement.