Vertical distribution of ice nucleating particles over the boreal forest of Hyytiälä, Finland

Zoé Brasseur et al.,

This manuscript describes ice nucleation measurements collected in three different atmospheric compartments; ground level, boundary layer, and free troposphere. Boundary layer and free troposphere measurements were made aboard a CESSNA aircraft. The duration of the sampling period ranged from the 20 April until 20 May 2018, and involved 19 separate flights. Ground based measurements took place at the SMEAR II station in Hyytiala. The aircraft was equipped with filter measurements (for INP analysis) and with SMPS and OPS measurements for aerosol size and number concentration

Several meteorological and remote sensing measurements were used to determine the development of the boundary layer each day. This information was later used to interpret the variability INP concentrations as a function of altitude.

Measurements are compared to previous studies at the same site over a longer period of time. These measurements and the subsequent aerosol measurements are used to develop an updated parameterization that is compared to existing parameterizations from a number of different studies.

There are few studies capable of providing measurements on the vertical profile of INP measurements and therefore this study presents a very unique and valuable data set. The manuscript is well written and the graphic are of very good quality.

Please find below some general comments and suggestions:

Data availability:

It would be appreciated to provide accessible data to the reviewers (as is stipulated in the policies of ACP).

Introduction:

The presentation and discussion of the parameterizations is an important part of this manuscript. A short discussion on the need for parameterizations in the introduction would be useful, and to present why having multiple variable parameterization's is useful compared to single parameterization's (currently used in cloud models). Can cloud models integrate these more complex parameterizations?

Methodology:

- Were any samples analyzed immediately after collection and again post freezing. What kind of impact is the freezing cycle expected to have on the ice nucleation activity of aerosol particles?
- Line 189: Can the authors rephrase this sentence. What exactly was passed through the filter and what was the purpose of using this filter. "First, the sampled aerosol particles were washed from the filter membrane into a solution using Milli-Q purified water (18.2 $M\Omega$.cm), which was passed through a 0.1 μ m Whatman syringe filter."
- What is the volume of liquid used in the sample wells of INSEKT, and how many wells are used? Was it possible to perform multiple runs on the same samples to determine the reproducibility of the filters? What impact of biological aerosol is thought to have on these samples? Schneider et al., performed a heat treatment of the samples, and observed a significant fraction of biological INPs. Do the authors suspect that this might also be the case in these samples, and how would this vary with altitude?

- Can the authors provide Fig. A1 in the same units (INP stdL) as the data presented elsewhere.
 It would be easier for the reader if Figure A1 and A2 are combined in the same figure a) and b).
- For the air mass backtrajectories, was the impact of airmass history (precipitation, and or cloud activation, from ECMWF calculations) considered in this analysis.

Results:

- Can aerosol mixing state be inferred from the size distribution measurements? What impact does aerosol mixing state have on INP properties?
- What causes the difference in the onset of freezing between Schneider et al., 2021 and the ground level samples (shown in figure 4). What are the differences between the Schneider sampling set up at SMEAR II and those in this work? The authors say that they were sampling behind a TSP inlet, however Schneider was sampling behind à PM10. What is the inlet height at the ground station?
- Figure 4b, would it not be more correct to label this normalized INP / particle number concentration. The axes being labeled Activated fraction, is misleading.
- Can the authors provide more details about the data points in Figure 4 (Median, percentiles etc..).
- Since measurements of the full size distribution were available why was the number of INP per aerosol surface area not calculated? This is a pertinent measurement of INP activity and would be possible to put it in context with a number of other studies.
- In the individual plots (shown in Figure 9), we observe very different distributions of INP concentrations as a function of temperature. For example at ground level on the 15th there was a first freezing mode until -18C then a sudden increase in freezing until -20C and then a gradual increase until the end. Whereas in the boundary layer the INP concentrations appear more homogenous with a consistent rise in INP as a function of temperature. Likewise in the FT measurements there appears to be a similar distribution as the BL. Are these changes in INP spectra a result of dilution steps (as discussed for 9c) or is there additional information that can be extracted? Are there different 'slopes', or freezing modes in the data.
- For the jumps in data points observed for the FT samples collected on the 17th, that the authors state is a result of a dilution step, should the reader focus on the trend in the FT sample after the -20°C of before the -20°C. Are measurements valid in both cases?
- The average INP plots over the full period were illustrated. As expected a high spread in the data points is noted. The authors showed that there was significant changes in weather (Figure 3) over all the flights.
 - Can the variation in the INP concentrations be explained by other variables than aerosol size, such as the temperature and pressure, and also aerosol chemistry (at least from ground based measurements).
 - Can aerosol chemistry (other than biogenic sources) be mentioned, at least for the ground based measurements (from measurements available at the SMEAR station), or those measurements already available in the Schneider paper. Was there any variability observed during the sampling period discussed in this manuscript.

- **Figure 9:** The authors mention that there were clouds present between 3000 and 4000 m on the flight of the 17th. Was the aircraft sampling inlet adapted to sample could droplets?
- **Figure 7:** For the Schneider parameterization (in both the BL and the FT), it seems that there are only a small number of points that are pulling the fit away from the 1:1 line.
 - Do these points correspond to a single flight? If so, is there something particular about this flight?
 - O Why do the authors not provide a test for the ground based measurements?
- In figure 7: Is it possible to apply this newly developed parameterization's to the ground based, boundary layer data, and also to Schneiders data. It would be interesting to determine if this parametrization can be used in other environments or if it is only suitable for FT measurements.
- In Table 1, it would be useful to also include the fitting values of the updated parameterization included here. Those that are listed at the end of the paragraph.
- In the development of the parametrization, what methods were used to find the optimum values for the coefficients? Were these calculated only using the FT samples?