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

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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.

Response: We thank the referee for their useful comments and suggestions which helped us improve the manuscript. We provide our responses below each individual comment.

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).

Response: The data will be made available as soon as possible with the following DOI: 10.5281/zenodo.10975295.

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?

Response: We agree with the referee that parameterizations could be mentioned in the introduction. We propose adding the following text after line 50:

"To overcome our lack of knowledge concerning INPs, there is a need for more observations of INPs worldwide. Such measurements are also needed to develop accurate parameterizations, which are an important tool used to constrain heterogeneous ice nucleation predictions in models (e.g., Fletcher et al., 1962; Meyers, 1992; DeMott et al., 2010)."

However, we believe that a discussion related to the different parameterization types and their implementations in models would be out of scope in this manuscript, especially since the comparison with existing parameterizations only represents one section of this manuscript and does not focus on producing new parameterization(s).

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?

  Response: The freezing of the suspensions of aerosol sampled at ground level at SMEAR II during the HyICE-2018 campaign did not have a significant impact on the measured ice nucleation activity (Kaufmann, 2019; see Fig. 3.13 extracted from the master thesis below). For some samples, we observed a slight reduction of the ice nucleation activity in the higher temperature regime (within the margin of error). It is therefore possible that the freezing of the filters sampled during the aircraft campaign might also have slightly altered the ice nucleation activity in the higher temperature regime. For this reason, the storage of the aerosol filters was kept as short as possible. Warm storage of filters was not an alternative, as it alters the activity especially for biological INPs (Stopelli et al., 2014).
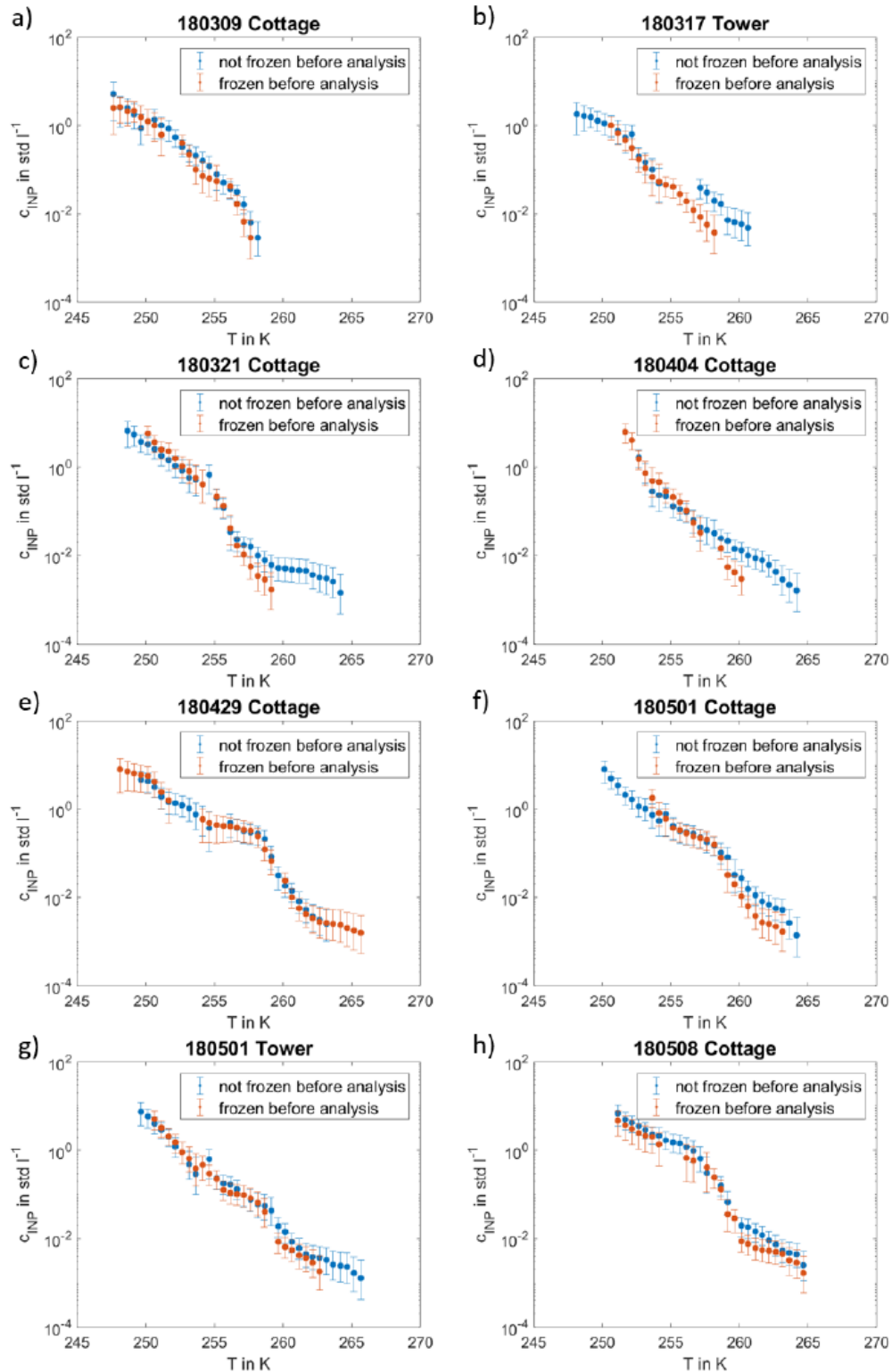
Figure 3.13: **Impact of freezing the aerosol suspensions.** For testing the impact of freezing of the aerosol suspension, several randomly chosen aerosol samples of the HyICE2018 campaign have been analysed directly after producing the aerosol suspension (blue) and a second time after the suspensions have been left in the freezer for several weeks (red).

- 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 MΩ.cm), which was passed through a 0.1 µm Whatman syringe filter."

  Response: We agree that the sentence needs to be clarified and propose rephrasing:

  "First, we used Milli-Q purified water (18.2 MΩ.cm), which was passed through a 0.1 µm Whatman syringe filter to remove possible remaining impurities, to wash the sampled aerosol particles from the filter membrane into a solution."

- 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?

  Response: Concerning the analysis with INSEKT, we propose adding the following information to the text line 189:

  "The resulting aerosol suspensions were then analyzed with the INSEKT by pipetting volumes of 50 µL into two 96-well polymerase chain reaction (PCR) plates. Typically, 32 wells were filled with nanopure water while the remaining 160 wells were used to analyze two samples at once. For each sample, 24 wells were filled with the undiluted suspension, 24 wells were filled with a 10 or 15-fold diluted suspension, and 32 wells were filled with a 100 or 225-fold diluted suspension (Schneider et al., 2021)."

  We did not perform multiple runs on the same samples to determine the reproducibility of the analysis.

  In lines 376-379, we make the hypothesis that similar INPs could have been sampled in our study and in Schneider et al. (2021). As mentioned, Schneider et al. (2021) found that the sampled INP population was dominated by heat-labile materials, and one could expect similar features from our ground-level and boundary layer samples. However, since we did not conduct heat tests on the samples collected during the aircraft campaign, we cannot investigate this hypothesis further. We propose adding the following text line 379:

  "It is therefore possible that biogenic particles represent an important fraction of the INPs sampled at ground level and in the boundary layer in this study. A recent study from Maki et al. (2023) showed that airborne microorganisms from forested areas could maintain similar concentrations from ground level up to 500 m under efficient vertical mixing conditions. Since the boundary layer was well-mixed during the aircraft campaign, we can expect that surface biogenic particles had a non-negligible impact on the INPs sampled in the boundary layer.

However, such a hypothesis cannot be confirmed with the data presented in this work, and more measurements such as heat treatment tests (e.g., Hill et al., 2016) would be needed to examine the presence of biogenic INPs in the samples."

- 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).

  Response: Since the background correction was done in INP concentration per ml of aerosol solution, we would like to keep this unit in Fig. A1. In addition, following Referee 2's comments on Fig. A1, the figure was modified to indicate which data points were not considered because they did not meet the criterion introduced (INP concentrations at least twice as high as the average background INP concentrations). We agree with the Referee that Fig. A1 and A2 should be combined into one figure with subplots. The updated figure is attached at the end of this document.

- For the air mass backtrajectories, was the impact of airmass history (precipitation, and or cloud activation, from ECMWF calculations) considered in this analysis.

  Response: Precipitation for wet deposition (both in cloud and below cloud scavenging) is considered in FLEXPART v10.4. However, since we used a passive air tracer and not aerosol/gases, precipitation does not impact the air mass backtrajectories presented in the manuscript. We propose adding the following information to the methods line 266:

  "The simulations were computed for a passive air tracer for which the wet and dry removal processes have no impact."

Results:

- Can aerosol mixing state be inferred from the size distribution measurements? What impact does aerosol mixing state have on INP properties?

  Response: Following the definition from Riemer et al. (2019), aerosol mixing state includes both chemical and physical properties of the overall particle population within an aerosol. As such, size distributions only give information on the physical property of the particle population and do not give information on the chemical mixing state of aerosols. Additional measurements of particle properties, such as chemical composition, morphology, optical properties, etc., could tell us more concerning the aerosol mixing state, but such measurements were not made during the aircraft measurements. Furthermore, as discussed in Riemer et al. (2019), the ice nucleation ability of a particle depends on its surface properties, which is in turn strongly tied to the aerosol mixing state. Thus, changes in the aerosol mixing state, for example through chemical aging or coating, can change the ability of the particles to form ice (e.g., Czisco et al., 2009).

  Based on this and Referee 2's comments, we have added the following text line 613:

"In addition, it is important to stress that, although the analysis of the particle number size distributions and concentrations gives valuable information on the vertical distribution and physical mixing state of the aerosol population, such information cannot necessarily be directly extended to the INPs, which represents a very small and highly variable fraction of the overall aerosol population (DeMott et al., 2010)."

- 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?

  Response: We suggest that the difference in the onset of freezing between the Schneider et al. (2021) and the ground-level samples is due to the shorter sampling times used for the ground-level samples, as explained lines 309-311. Indeed, the samples presented in Schneider et al. were sampled for 24 hours while the ground-level samples were collected for 3 hours approximately.

  Concerning the sampling set up at SMEAR II, we propose adding the following information lines 176-178:

  "At SMEAR II, the ground-level filter was sampled from a measurement container using a vertical sampling line connected to a total aerosol inlet. The inlet height was approximately 4 m a.g.l. and the average flow rate through the filter was 15 L min^{-1} with an average sampling time of 3 hours."

  In addition, we propose adding some information line 305:

  "Compared to the ground-level samples presented in this study, the samples used in Schneider et al. (2021) were collected from the aerosol cottage (approximately 20 m from the measurement container) using a PM_{10} inlet with an inlet height of approximately 4.6 m a.g.l."

- Figure 4b, would it not be more correct to label this normalized INP / particle number concentration. The axes being labeled Activated fraction, is misleading.

  Response: Here we defined the activated fraction as the ratio of INP concentration to the particle number concentration. A similar definition has been used in several previous studies (e.g., Porter et al., 2020; He et al., 2021 and 2023; Iwata et al., 2019) and therefore we believe that we can also use this definition here.

- Can the authors provide more details about the data points in Figure 4 (Median, percentiles etc..).

  Response: We propose adding more information line 305:

"In Fig. 4, the data is presented in the form of boxplots calculated for each activation temperature, where the line dividing the boxes in two represents the median value of the distribution, the lower and upper edges of the boxes represent the 25th and 75th percentiles, respectively, the lower and upper whiskers represent the minimum and maximum, respectively, and the outliers are represented as single point markers."

- 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.

   Response: We agree with the reviewer that including the INAS might prove useful to readers and future comparisons, and we therefore propose adding a figure presenting the INAS in the Appendix. We will also add the following text line 334:

   "The ice nucleation active surface site (INAS) densities, calculated as the ratio of the INP concentration to the surface area concentration of particles larger than 300 nm, are presented in Fig. A3."
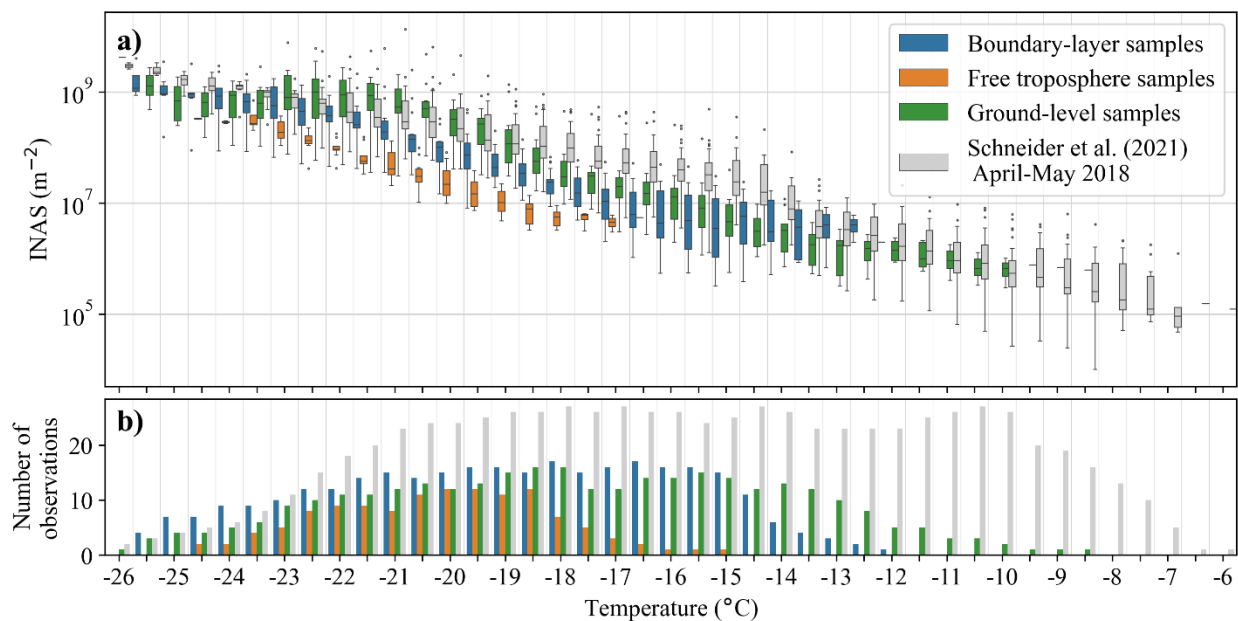

Figure A3. a) INAS densities as a function of the activation temperature for all the samples collected during the aircraft measurement campaign together with the ground-level data from Schneider et al. (2021) collected in Hyytiälä from 20 April to 19 May 2018. The INAS densities were calculated by normalizing the INP concentration by the aerosol surface area concentration following the method described in Ullrich et al. (2017) and assuming that each INP triggers the formation of one ice crystal. The aerosol surface area concentration was derived from the size distribution measurements of the particles larger than 300 nm obtained from the OPS and the combined DMPS-APS for the aircraft and the ground-level samples, respectively. b) Number of observations for each sample type as a function of temperature.

- 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.

Response: The changes in the shapes of the INP temperature spectra are not related to the dilution steps. As illustrated in the figure below, which is an enlargement of Fig. 9a, the changes in slopes do not occur specifically at the dilution steps and thus are not caused by the dilution(s).
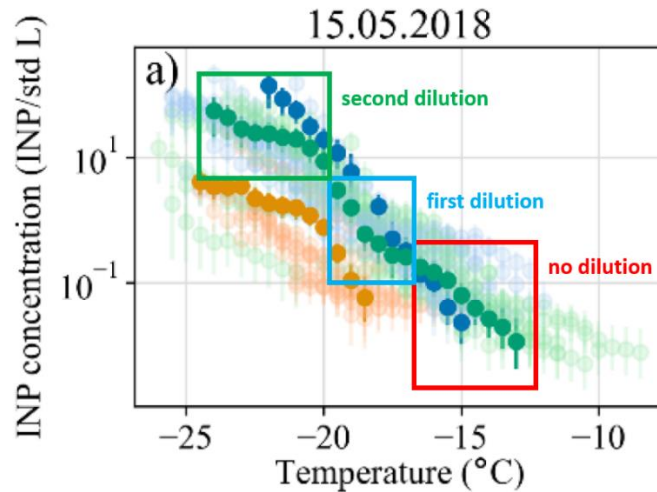
It is possible that these differences in the shapes of the INP temperature spectra indicate that different aerosol types dominate the INP populations in the different samples. Similar observations were made in Schneider et al. (2021), but at warmer temperatures (approximately -13 °C). However, without additional analysis of the samples (such as heat tests) or information concerning the chemical composition of the sampled particles, specific conclusion for the variations in the INP temperature spectra is unwarranted. We must leave it to future work to further investigate this matter.

- 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?

  Response: As we cannot fully explain the differences measured between the two dilutions, we decided to show both spectra instead of disregarding one of them without good reason. Therefore, we also cannot give guidance on which result is more valid.

- 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.

  Response: We would like to point out that, with the exception of the first two flights, all aircraft measurements took place in May 2018 during a period with relatively stable meteorological

conditions, as shown in Fig. 3. In addition, while there is some variability in INP concentrations in the samples, this spread is not unusually large (cf Fig. 8 and Fig. A2c) and less than the systematic difference between the free troposphere and the ground-level/boundary-layer samples.

- o 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).

  Response: All INP concentrations presented in this study were converted to standard conditions in order to eliminate their influence and thus make the samples collected at ground level and in the atmosphere aloft comparable. A direct effect of air temperature and pressure on the INP concentrations at the sampling location is not expected and, to the authors' knowledge, not reported in previous studies. Nevertheless, air temperature can be an indicator for the seasonal cycle and thus can be linked with the boreal forest's biological activity and the concentration of biological INPs, as described in Schneider et al. (2021). However, as mentioned previously, the meteorological conditions were stable in May 2018, hence it is unlikely that the variation in INP concentration is directly linked to meteorology.

  Regarding aerosol chemistry, no measurements of the aerosol chemical composition were conducted onboard the aircraft, and thus we cannot investigate the impact of such parameter on the INP concentrations measured in the boundary layer and in the free troposphere. However, we agree that aerosol chemistry should be investigated in future studies, as mentioned in the conclusions:

  "Future measurements should include additional analysis of the chemical composition and heat sensitivity of the sampled INPs, in a similar manner to what Sanchez-Marroquin et al. (2023), Hartmann et al. (2020) and Hill et al. (2016) have done."

  Concerning ground-level measurements, the link between aerosol chemistry and ground-level INP concentrations is discussed in detail in Schneider et al. (2021). As highlighted in Schneider et al. (2021), there was a clear seasonal transition from winter to spring between the end of March and the beginning of April 2018. However, the aircraft measurements presented here took place after this seasonal transition, when there was less variability in the concentration of biogenic aerosol. Additionally, the focus of this study is not on the ground-level INP measurements but on the aircraft measurements, for which we do not have any aerosol chemistry information.

- o 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.

  Response: Regarding the chemical composition of particles from sources other than biogenic, there is limited data to investigate potential links with the INP concentration

since the only instrument that gives aerosol chemical information is a long time-of-flight aerosol mass spectrometer (L-ToF-AMS) located at ground level in SMEAR II. However, this instrument measures in the size range 75-650 nm, which is generally not considered to be of the highest relevance for ice nucleation in the atmosphere, and only gives information in broad chemical categories including organic, sulfate, nitrate, ammonium, and chloride. Schneider et al. (2021) showed some slight correlation between the mass concentration of non-refractory organic compounds and INP concentrations and suggested that these were linked to the forest biological activity. However, such observations were made over long time scale to study the seasonal cycle of INP concentrations, and correlations might be less visible on shorter time scales such as those covered with the aircraft campaign. As mentioned previously, these ground-level measurements are discussed in Schneider et al. (2021) and since the study presented here focuses on the aircraft measurements conducted aloft (for which we do not have aerosol chemistry measurement), we refrained from investigating ground-level aerosol chemistry.

- **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?

  Response: The aircraft sampling inlet and setup in general were primarily developed for new particle formation studies (see for example Lampilahti et al., 2021). During the HyICE-2018 campaign, the setup was slightly modified to samples particles on filters in order to analyze their INP content subsequently. Measuring in-situ ice crystals and cloud droplets was not the objective of the campaign, and therefore the aircraft inlet was not adapted for such in-situ measurements.

- **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?

    Response: Assuming that the referee's comment concerns the few data points that are the further away from the grey shaded area (representing a range of a factor of 5 from the 1:1 line) in Fig. 7, where the observed INP concentration are higher than about 10 L^{-1} at temperatures around -20 °C, then no, these points do not correspond to a single flight. For the boundary-layer samples, these points come from three different flights on 14, 15, and 18 May 2018. These three flights had relatively high INP concentrations between -18 and -24 °C which were not reproduced by the Schneider et al. (2021) parameterization. However, the reason behind these elevated INP concentrations remains unclear and is not further explored. For the free troposphere samples, very few points are outside of the grey shaded area representing a range of a factor of 5 from the 1:1 line. The main deviation is observed for the sample collected on 16 May 2018, which corresponds to the highest INP concentrations measured in the free troposphere during the campaign and which is further discussed in section 3.7.

  - Why do the authors not provide a test for the ground based measurements?

Response: The focus of this manuscript is primarily on the aircraft measurements conducted at higher altitude, and the ground based measurements are used to bring context and additional information to the vertical profile of INP concentrations. Ground based measurements from HyICE-2018, including their representations by parameterizations, are described in length in the previous studies summarized in the introduction (Paramonov et al. 2020; Schneider et al. 2021; Vogel et al. 2024). Thus, we do not provide additional tests here to avoid repetition, and we prefer focusing on the aircraft INP measurements which were not presented previously.

We propose modifying the text starting line 433 to clarify:

"In Fig. 7a-f, the INP concentrations measured in the boundary layer and in the free troposphere are compared to INP concentrations predicted by three existing parameterizations from Schneider et al. (2021), DeMott et al. (2010), and Tobo et al. (2013), which are presented in Table 1. This section focuses on the aircraft INP measurements conducted at altitude, and a detailed comparison between parameterizations and INP concentrations from ground-based filter measurements similar to those presented here can be found in Schneider et al. (2021)."

- 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.

Response: The adjusted parameterization presented in Fig. 7g is not a newly developed parameterization, but simply the parameterization from Tobo et al. (2013) with adjusted coefficients. To avoid confusion, we modified the legend of Fig. 7g to "Adjusted Tobo et al. (2013)" and clarified the method used to adjust the parameterization (see comment below). Considering that the parameterization from Tobo et al. (2013) was adjusted to better fit the free troposphere data, we do not see value in testing it on ground or boundary layer measurements. In addition, as we stress in the manuscript lines 505-507, this adjusted parameterization should be used with caution as it was derived for a very limited number of observations (only 12 filters samples/119 data points), and we do not claim that it is suitable for all free troposphere measurements or other environments.

- 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.

Response: We propose modifying Table 1 to the following:

Reference	Temperature range	Equation	Input parameters
Schneider et al. (2021)	-25 to -12 °C	$n_{INP} = 0.1 \cdot exp(a1 \cdot T_{amb} + a2) \cdot exp(b1 \cdot T + b2)$ with $a1 = 0.074$ K^{-1}, $a2 = -18$, $b1 = -0.504$ K^{-1}, and $b2=127$	Ground-level ambient air temperature T_{amb} (K) Activation temperature T (K)
DeMott et al. (2010)	-35 to -9 °C	$n_{INP} = a(273.16 - T)^b(n_{AP,>0.5\ \mu m})^{(c(273.16-T)+d)}$ with $a = 0.0000594$, $b = 3.33$, $c = 0.0264$, and $d = 0.0033$	Number concentration of particles with diameters larger than 0.5 µm $n_{AP,>0.5\ \mu m}$ (cm^{-3}) Activation temperature T (K)
Tobo et al. (2013)	-34 to -9 °C	$n_{INP} = exp(\gamma(273.16 - T) + \delta)(n_{AP,>0.5\ \mu m})^{(\alpha(273.16-T)+\beta)}$ with $\gamma = 0.414$, $\delta = -6.671$, $\alpha = -0.074$, and $\beta = 3.8$	

Adjusted Tobo et al. (2013)	-34 to -9 °C	$n_{INP} = exp(\gamma(273.16 - T) + \delta)(n_{AP, >0.5\ \mu m})^{(\alpha(273.16 - T) + \beta)}$ with $\gamma = 0.7408$, $\delta = $ -16.0788, $\alpha = 0.2746$, and $\beta = $ -3.3184

- 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?

Response: The adjusted coefficients were indeed calculated using only the free troposphere samples, as mentioned lines 500-501. We propose rephrasing lines 499-501 and adding some information to clarify how such adjusted coefficients were obtained:

"To test this hypothesis, we adjusted the coefficients used in the parameterization from Tobo et al. (2013) to better fit our free troposphere data while keeping the same mathematical form. This was done using the in situ observations of number concentration of particles with diameters larger than 0.5 µm and INP concentrations measured in the free troposphere, and following the method described in the supporting information of DeMott et al. (2010). Each fitting was calculated using the Levenberg-Marquardt algorithm, and the following adjusted coefficients were obtained: […]"
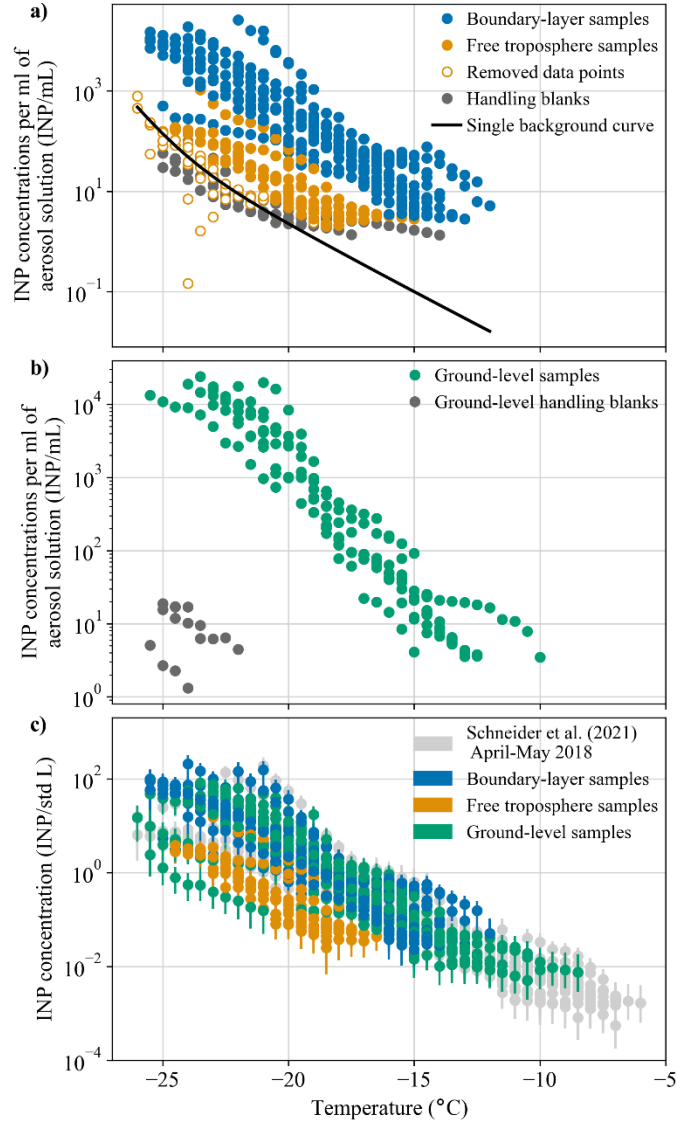
Updated figures:

Figure A2: a) **INP concentrations per ml of aerosol solution from the aircraft samples compared to the background signal derived from the handling blank filters collected onboard the aircraft. b) INP concentrations per ml of aerosol solution from the ground-level samples collected at SMEAR II at the same time as the aircraft samples compared to the ground-level handling blank filters. c) INP temperature spectra of all the samples collected during the aircraft measurement campaign together with the ground-level data from Schneider et al. (2021) collected in Hyytiälä during April and May 2018. The error bars represent the statistical as well as the systematic error of the INSEKT assay. More details related to the calculation of these error bars is given in Schneider et al. (2021).**

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