

The manuscript by Brasseur et al focusses on the vertical distribution of INP over boreal forest in Finland and has been carried out by aircraft measurements during spring 2018 above Hyytiälä. The manuscript is well-written and includes new and important data. The topic is important for atmospheric scientists and might be published in ACP after some minor corrections.

Response: We thank the referee for their useful comments which helped improve the manuscript. We include our responses below in the context of individual comments.

Comments:

Introduction: The topic has been well introduced and the relevant new literature has been cited. However, the authors might mention their own paper Vogel et al. which is under discussion as well. Also, the older literature at similar locations deserves attention (e.g. Prenni 2013). The authors mention the ice nucleation temperature of desert dust being below -15°C but I miss a statement regarding the biological INPs and INMs which are below -2°C depending on the origin. In this regards the authors might also mention that dust could be a carrier, coated with bio INMs, which turns them into bio INPs.

Response: We propose adding a mention to the Vogel et al. (2024) paper line 112:

“Finally, Vogel et al. (2024) presented ground-based measurements conducted with the Portable Ice Nucleation Experiment (PINE) below -24°C and found moderate correlations between INP concentrations and concentrations of particles larger than $0.5\ \mu\text{m}$ as well as concentrations of fluorescent aerosol particles, hinting at a possible biological source of INPs active below -24°C .”

In addition, we propose mentioning previous studies conducted at similar locations (forested environments) line 93:

“These results agree well with previous studies conducted in similar forested environments. Prenni et al. (2009) for example showed that INP concentrations and abundance in a pristine rainforest of the Amazon basin could be partly explained by local emissions of biological particles. Huffman et al. (2013), who performed measurement in a semi-arid pine forest of North America, found a strong correlation between fluorescent biological particles and INPs during rain events. Similarly, results presented in Prenni et al. (2013) suggest that biological particles represent a significant portion of rain-generated INPs measured at a forested site in Colorado. Tobo et al. (2013) conducted measurements in a midlatitude ponderosa pine forest ecosystem in Colorado and found significant correlations between INP concentrations and the concentration of ambient fluorescent biological aerosol particles. Finally, Iwata et al. (2016) carried out measurements near forested mountain slopes in Japan and found that biological particles played an important role as INPs for temperatures warmer than -22°C , especially during rainfall events. However, all these observations were carried out at ground level and did not examine the transport of such INPs to higher altitudes.”

Concerning biological INPs, we propose modifying lines 47-50 to:

“Biological aerosols are considered another widely present type of INPs (O’Sullivan et al., 2015; O’Sullivan et al., 2018; Wex et al., 2019;). Although their global emissions are lower than dust, they can form ice at relatively warmer temperatures depending on the nature of the bioaerosols (Després et al., 2012). For example, the bacteria *Pseudomonas Syringae* is a very efficient INP at temperatures as warm as $-2\text{ }^{\circ}\text{C}$ (Maki et al., 1974; Joly et al., 2013). In addition, biological particles, including bacteria, have been found in dust aerosols, possibly enhancing their ice nucleation activity (Conen et al., 2011; Meola et al., 2015; Barr et al., 2023).”

Methods: The description is very detailed. However, I have remarks regarding the figures:

Figure 1a: The map is very pale and the scripts are small. I would recommend to add a general map where the location on the European continent is documented.

Response: Following the referees’ comments, Fig. 1a was updated to a 3D plot to highlight the flight tracks more clearly. A figure with the location of the measurement site with respect to the European continent was added in the Appendix. The updated figures are attached at the end of this document.

Figure 2: The color code is missing. The x-axis might be labeled “day time” or “position of the sun”

Response: We added a legend with the color code and a label for the x-axis. The updated figure is attached at the end of this document.

Results: Figure 3: Please explain the calculation of the TKE dissipation rate from the Doppler lidar in more detail.

Response: Thank you for your comment. We propose modifying the text lines 245-251 to the following:

“After the flights, data from a Halo Photonics Stream Line scanning Doppler lidar located at SMEAR II was used to estimate the limit between the boundary layer and the free troposphere, for comparison with the aircraft measurements. The Halo Doppler lidar was configured with vertically-pointing stare and conical scans (i.e., with velocity azimuth display (VAD) scans at 30° elevation angle) repeating every 30 minutes. Additional scans during the 30-minute scan cycle were not used in this study. The range resolution of the lidar is 30 m, with a minimum range of 90 m a.g.l.. More details on the Doppler lidar at SMEAR II can be found in e.g., Hellén et al. (2018). The data was post-processed following Vakkari et al. (2019): horizontal winds were retrieved from the VAD scans and the variance of vertical wind velocity was calculated from 12 consecutive vertical stare measurements. The instrumental noise contribution to the observed variance of vertical wind velocity was estimated from the post-processed signal-to-noise-ratio according to Pearson et al. (2009) and subtracted before calculating turbulent kinetic energy (TKE) dissipation rate profiles according to the method by O’Connor et al. (2010).”

Figure 5a: add ticks on the y-axis for every order of magnitude.

Response: We added ticks for every order of magnitude on the y-axis. We attach the updated figure at the end of this document.

Figure 6a: add a size bar, explain the color code in more detail

Response: We added a title to the legend and modified the figure caption to improve clarity regarding the color code. We attach the updated figure at the end of this document. However, we would rather not add a scale bar in Fig. 6a as it may be inaccurate due to the distortions that can occur with map projections at this scale. We believe that the gridlines representing the latitudes and longitudes should be enough to provide context to the map used.

Figure 7: add ticks on the x- and y-axes for every order of magnitude

Response: We added ticks for every order of magnitude on the x and y axes. We attach the updated figure at the end of this document.

Figure 11a and b: add size bar. Better explain PES.

Response: As in Fig. 6a, we would prefer not to add scale bars to Fig. 11a and b since adding a straight line to represent the scale would be inaccurate due to distortions in the projection. Concerning the PES, we propose adding the following text to the methods line 264:

“FLEXPART was used to calculate the potential emission sensitivity (PES) fields, where PES is the response function of a source-receptor relationship which estimates the potential source contributions for a given receptor (in this case the measurement site SMEAR II). The PES is therefore proportional to the residence time of the air mass in a specific grid cell, and it was calculated in units of seconds. High values of PES indicate source regions where emissions are likely to significantly impact the tracer concentrations at the receptor (Seibert and Frank, 2004; Stohl et al., 2005; Pisso et al., 2019).”

Conclusion: The conclusion reads more like a summary. I miss a real discussion and conclusion from the results along these questions: What do we learn regarding the emission of INP and their transport from the forest ecosystem, above canopy, into the free troposphere? Can you speculate about the transport mechanisms? How do wind and humidity influence these potential processes?

Response: Following the comments from Referees 1 and 2, we have shortened and focused the conclusions. Here is the updated text:

“In this study, we present the first aircraft measurements of INP concentrations above the Finnish boreal forest, and we shed new light on the vertical distribution of INPs above this environment. We found that local surface particles were transported and mixed within the boundary layer aloft through convective mixing, resulting in similar INP concentrations and activated fractions observed at ground level and in the boundary layer. INP concentrations and activated fractions measured in the boundary layer were within the same order of magnitude as those reported by Schneider et al. (2021) for the same period and were best predicted by the parameterization developed in the same study. This further suggested that INPs sampled in the boundary layer

primarily originated from the local boreal forest environment rather than long-range transported particles. Although the identity of the INPs sampled in the boundary layer, and whether or not they are dominated by biogenic aerosol similarly to what Schneider et al. (2021) found, has yet to be confirmed, our results suggest that the Finnish boreal forest is the main source of INPs observed in the boundary layer. 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 previously.

On the other hand, much lower INP concentrations were observed in the free troposphere. The distinct particle number size distributions observed there indicated that different aerosol and INP populations were encountered in the free troposphere and that local surface particles have a weaker influence at these altitudes. The free tropospheric INPs likely resulted from long-range transported particles from different sources, although the analysis of the air mass backward trajectories in the free troposphere did not yield conclusive results due to the limited number of observations. Additional measurements are needed to draw conclusions on the influence of the air mass origin(s) on the INP concentrations and identify the source(s) of INPs observed in the free troposphere.

However, one event was observed where INP concentrations and activated fractions measured in the free troposphere were higher and within the same order of magnitude as the concentrations observed at ground level and in the boundary layer. Analysis indicated that the air mass sampled in the free troposphere during this flight was influenced by the boundary layer. Although the exact transport mechanism remains unclear, it is possible that particles and INPs were transported to the free troposphere via boundary layer ventilation, likely caused by convection, turbulent mixing across the capping inversion, or upwards vertical motions of large scale weather systems (e.g., Donnell et al., 2001; Agustí-Panareda et al., 2005). Overall, this finding is of particular importance since INPs in the free troposphere can have longer lifetimes and travel farther, and can therefore impact cloud formation on a regional or global scale.”

Updated figures:

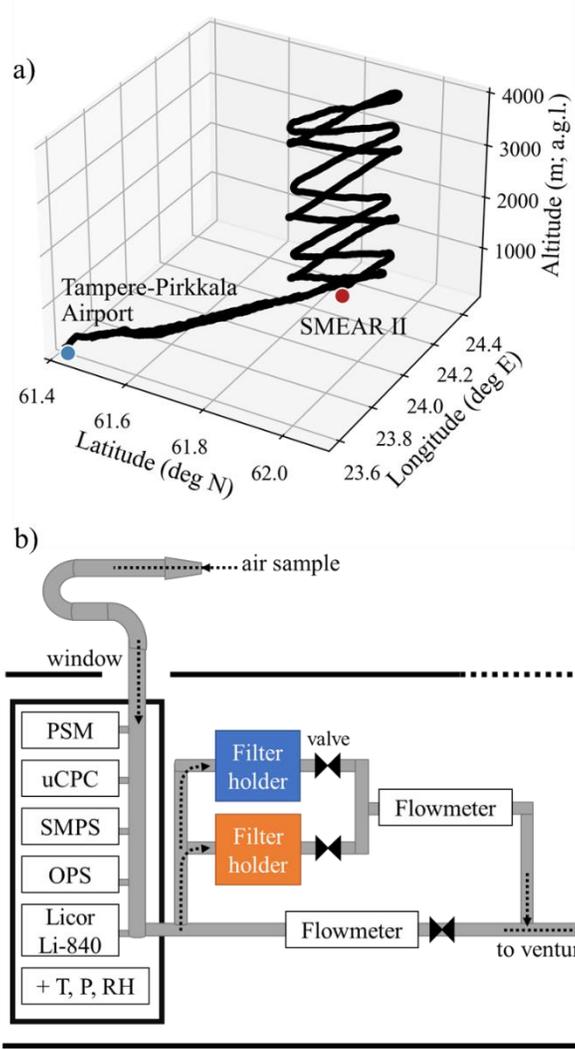


Figure 1: a) Example of a flight track from Tampere-Pirkkala airport to SMEAR II. The distance from the airport to the station is approximately 60 km. The location of SMEAR II with respect to Northern Europe is given in Fig. A1. b) Schematic of the instrumental setup viewed from above inside the Cessna 172, described in detail in Section 2.1.1.

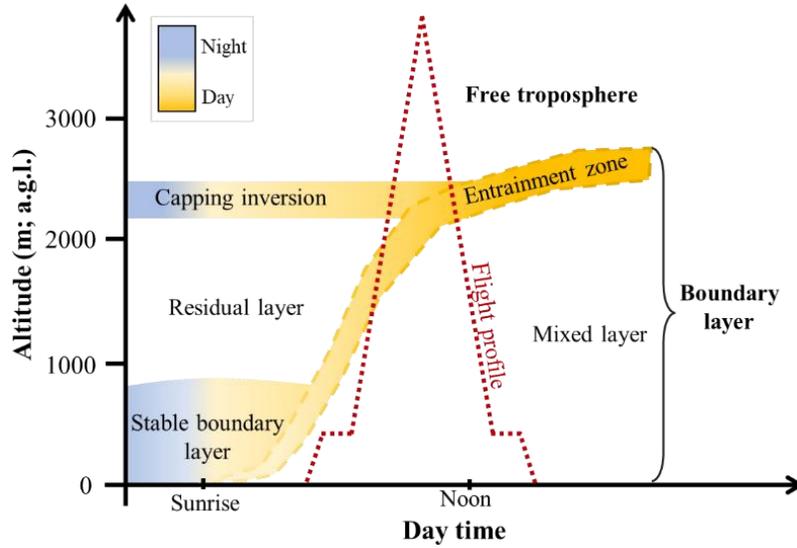


Figure 2: Schematic diagram of the boundary layer diurnal development adapted from Stull (2017) and Lampilahti et al. (2021), and overlaid with an example flight profile. The actual layer heights may vary from the values depicted on the vertical axis.

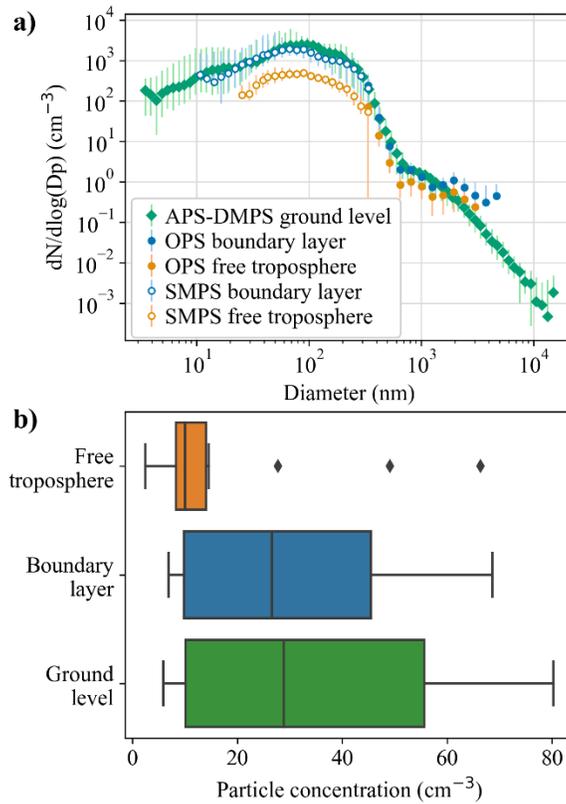


Figure 2: a) Median particle number size distributions calculated from ground-level measurements (SMEAR II APS and DMPS) as well as boundary layer and free troposphere measurements (aircraft SMPS and OPS) over the 19 flights of the campaign. The error bars represent the 25th and 75th percentiles. The size distribution shown with a linear scale can be seen in Fig. A3. b) Box plots of the concentration of particles > 300 nm measured at ground level, in the boundary layer and in the free troposphere calculated over all the flights.

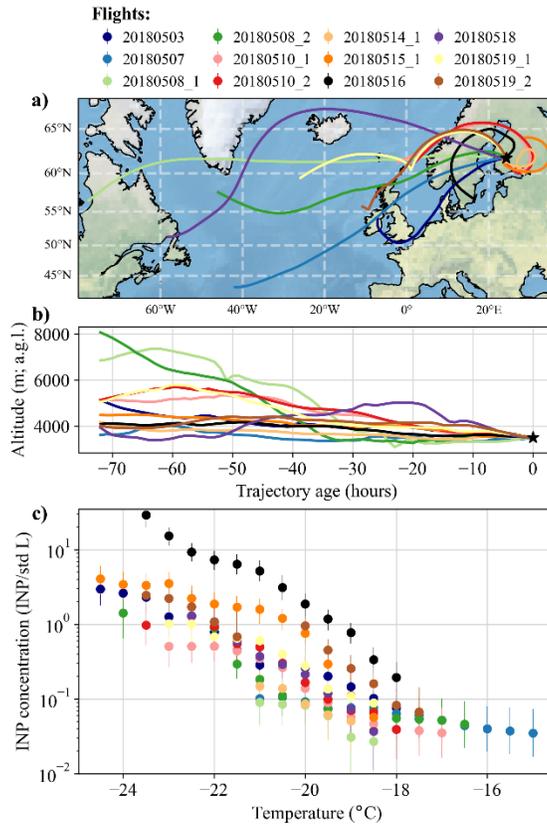


Figure 3. a) HYSPLIT 72-hour backward trajectories and b) altitude of the trajectories over time for the air masses arriving at 3500 m a.g.l. in the free troposphere above SMEAR II at the time when the measurement aircraft reached the free troposphere. c) INP temperature spectra of the free troposphere samples corresponding to each flight. In each plot, the color represents a specific flight, as indicated in the legend above panel a). Note that there were sometimes two flights per day, and each flight is identified by a number (_1 or _2) in the legend above panel a). In a) and b), the black star represents the measurement location in the free troposphere above SMEAR II, at 61°51' N, 24°17' E and 3500 m a.g.l.

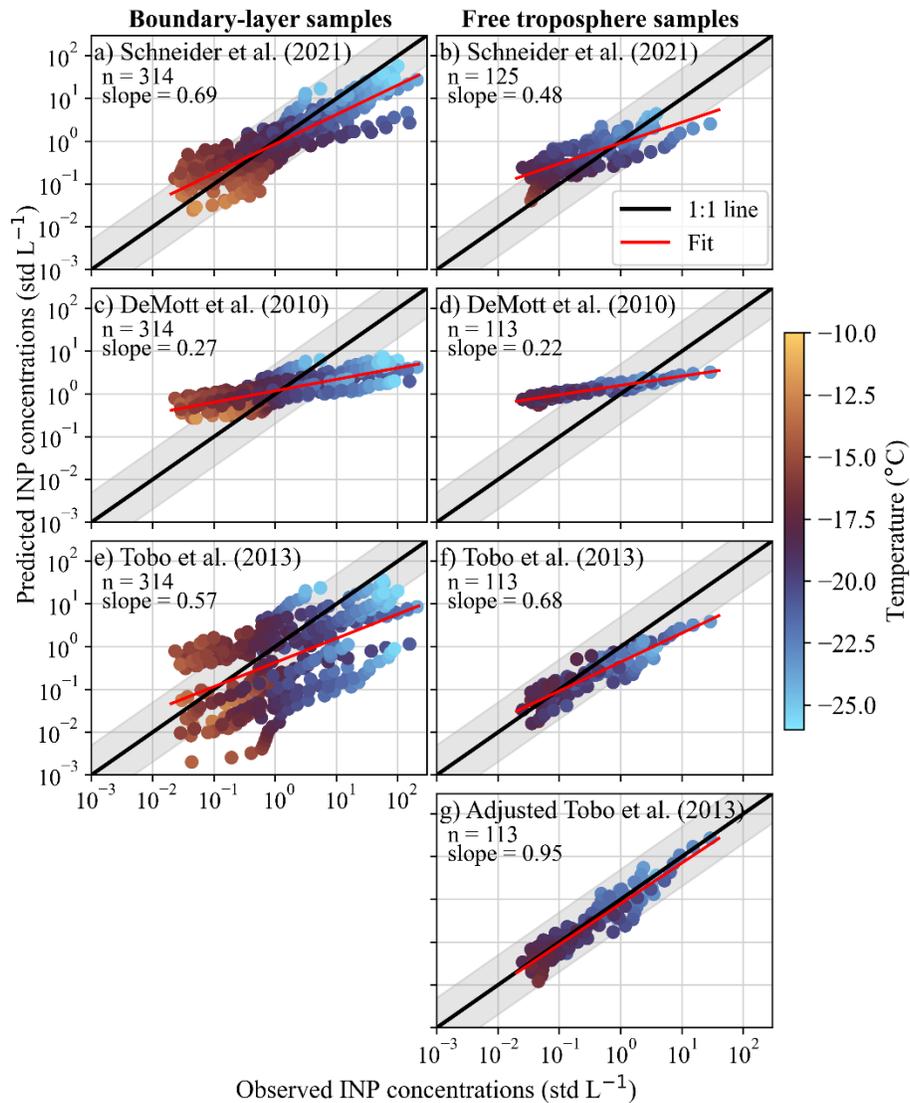


Figure 4. Comparison between the INP concentrations observed in the boundary layer (left side) and the free troposphere (right side) and the INP concentrations predicted using the parameterizations from a) and b) Schneider et al. (2021), c) and d) DeMott et al. (2010), and e) and f) Tobo et al. (2013). The black solid line represents the 1:1 line while the grey shaded area indicates a range of a factor of 5 from the 1:1 line. The red solid lines show a linear regression fit through the logarithmically transformed data points. The slope of the fit and the number of data points used is shown in each panel.

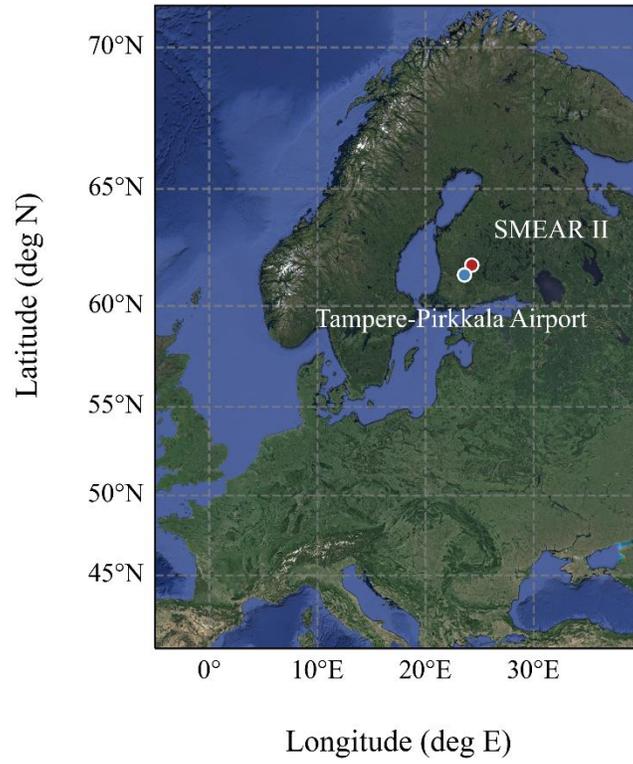


Figure A5: Location of the Tampere-Pirkkala airport and SMEAR II with respect to Northern Europe.

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