

Response to the interactive comment by Referee #2:

We thank referee #2 for his or her thoughtful comments and feedback. Please find below our responses and suggestions for the manuscript revision, with the referee comments in black, our answers in red, and suggested changes or additions to the manuscript in blue.

Schneider et al. present a well-written, succinct manuscript describing results from a full year of INP measurements in a boreal forest region. The study involves assessment of INP biogenic sources in addition to development of a new parameterization for boreal forest INPs. While I find the results and new parameterization valuable, there are a few issues with the manuscript that need to be addressed prior to publication.

While there are indeed very few year-long INP measurements at one location, there are several that report such measurements over an inter-seasonal scale (e.g. Šantl-Temkiv et al., 2019; Stopelli et al., 2015, 2016, and 2017). These studies are worth describing in the introduction. Additionally, it would be useful for the authors to report main findings from previous analogous studies to clearly demonstrate a comparison between those previous and the current results. The introduction is very short and could be beefed up by providing more details on these studies, including their limitations to promote the motivation for the current work.

Šantl-Temkiv, T., Lange, R., Beddows, D., Rauter, U., Pilgaard, S., Dall'Osto, M., Gunde-Cimerman, N., Massling, A., and Wex, H.: Biogenic Sources of Ice Nucleating Particles at the High Arctic Site Villum Research Station, *Environ Sci Technol*, 53, 10580-10590, 10.1021/acs.est.9b00991, 2019.

Stopelli, E., Conen, F., Morris, C. et al. Ice nucleation active particles are efficiently removed by precipitating clouds. *Sci Rep* 5, 16433 (2015). <https://doi.org/10.1038/srep16433>

Stopelli, E., Conen, F., Morris, C. E., Herrmann, E., Henne, S., Steinbacher, M., and Alewell, C.: Predicting abundance and variability of ice nucleating particles in precipitation at the high-altitude observatory Jungfraujoch, *Atmos. Chem. Phys.*, 16, 8341–8351, <https://doi.org/10.5194/acp-16-8341-2016>, 2016.

Stopelli, E., Conen, F., Guilbaud, C., Zopfi, J., Alewell, C., and Morris, C. E.: Ice nucleators, bacterial cells and *Pseudomonas syringae* in precipitation at Jungfraujoch, *Biogeosciences*, 14, 1189–1196, <https://doi.org/10.5194/bg-14-1189-2017>, 2017.

Thanks for this suggestion. We agree that the introduction should be more comprehensive and precise. Therefore, we described previous studies, which could be compared to our study, in more detail within the introduction focusing on the presented seasonal cycle, the considered time period, the continuity of measurements and the measurement locations. We also included the suggested publications Santl-Temkiv et al. (2019) and Stopelli et al. (2015, 2016), as those also show seasonal trends in their INP data sets. Stopelli et al. (2017) and Stopelli et al. (2016) have been added as references when discussing the relation between INP concentrations and precipitation (lines 44-45, revised manuscript).

We have included the following section to the introduction:

Hartmann et al. (2019) report INP concentrations from the past 500 years derived from ice core samples collected at two Arctic sites. These measurements do not show a long-term trend of INP concentrations over their multiyear period, but the variability within a year is observed to be large. They do suggest indications that biological INPs contribute to Arctic INP populations throughout the past centuries, for example, the general shape of the INP spectra and high INP concentrations at relatively high temperatures are typically associated with biological materials. Although they did not find a statistically significant seasonal variation, they assume that it is likely that the strength of local biological particle sources is enhanced during a particular time of the year influencing the INP

variability. However, due to the time resolution and dating uncertainty a seasonal relation could not be explicitly shown. Tobo et al. (2019) also report INP concentrations measured at an Arctic station in Svalbard in July 2016 and March 2017, which show seasonal changes with enhanced values in summertime. Tobo et al. (2019) link these enhanced concentrations to the emission of high-latitude dust from glacial outwash plains. Šantl-Temkiv et al. (2019) report INP measurements from the Arctic in spring 2015 and spring and summer 2016 and also show higher INP concentrations in summer than in spring, which they also associate with biological aerosol and biogenic compounds. In another study of pan-Arctic INP, Wex et al. (2019) report INP concentrations from four Arctic stations for different time periods and time resolutions measured between 2012 and 2016. At all locations, the highest observed INP concentrations are recorded in the summer months from June to September. The nature of the INPs was not explicitly determined, but high INP concentrations observed at high activation temperatures indicate a contribution from biogenic material. Wex et al. (2019) suggest potential INP source regions mainly on open land and open water. Stopelli et al. (2015) presents INP concentrations measured from snow samples collected at the Jungfraujoch on a few days per month in the period from December 2012 to September 2013. Here, INP concentrations are again higher in the summer months. Based on this dataset, a model to predict INP concentrations at the Jungfraujoch was established by Stopelli et al. (2016) and validated using several precipitation samples collected between May and October 2014. The dataset from 2014 shows a completely different seasonal pattern than the 2012/2013 dataset with the lowest values during summer and maxima in May and October. The authors suggest these maxima are related to a Saharan dust event and a cold front passage. Schrod et al. (2020) describe a global network of four INP sampling stations, at a range of northern hemisphere latitudes, where atmospheric aerosol samples were collected on substrates for two years from September 2014. The substrates were analysed for deposition and condensation mode INPs using the FRIDGE isothermal static diffusion chamber (Schrod et al., 2020). The Schrod et al. (2020) results do not yield a clear seasonality but instead show that short-term variability overwhelms long-term trends. However, that observation may not represent the full picture of INP in those locations. The short sampling times (low volumes, 1 hour per day) and/or colder activation temperatures may serve to maximize sampling variability and mask any potential biological signal (Schrod et al. 2020). To date these studies present the first observations and indications of the seasonal variability of INP concentrations, addressing the need for more long-term INP observations. None of these studies presents a comprehensive analysis of continuously recorded INP data for a full seasonal cycle at one location without interruptions. Moreover, the focus of most of the previous studies except for the Schrod et al. (2020) study was especially at Arctic INPs.

In regard to the very short introduction, perhaps more time could be spent on: (1) the motivation and objectives of the study itself (i.e. SMEAR II) and (2) more details on current parameterizations and modelling efforts for bioaerosols and biological INPs, which often are conflicting and based on a very limited subset of INP observations. This would inherently provide a clear motivator for developing the boreal INP parameterization.

Explanations of the objectives and motivation for this study have been added to the introduction. We have also extended the discussion of studies, which report about biological INPs and bioaerosols and included a discussion of the difficulties related to investigating and parameterizing these particles.

Text added to the introduction describing motivation:

The main objective of this study is to investigate and describe the variability and seasonal trends in INP concentrations and INP temperature spectra in a boreal forest environment. The absence of anthropogenic and/or dust aerosol sources in the boreal region motivates the additional investigation of biogenic ice nucleation activity and reveals the relevance of boreal forest areas as an important INP source. The comprehensive instrumentation provided at the measurement site at the

SMEARII station allows comparisons between INP measurements with simultaneous measurements of many meteorological variables. These measurements are complemented by measurements characterizing the sampled aerosol number concentrations, size distributions and chemical compositions in order to elucidate the potential origin and nature of the INPs. Heat treatments of the suspensions prior to INP analysis also help identifying the nature of INPs. We aim to improve the parameterizations describing atmospheric INP concentrations in the boreal forest by considering seasonal dependences in the formulations. Finally, this study provides motivation for further continuous long-term studies of INP in different environments across the globe.

Text added to the introduction discussing studies on bioaerosols and biological INP:

Several biogenic aerosol types have been shown to have atmospherically relevant ice-nucleating abilities (Augustin et al., 2013; Creamean et al., 2013; Hader et al., 2014; Möhler et al., 2007; Morris et al., 2004; O'Sullivan et al., 2015, 2018; Pratt et al., 2009; Schnell and Vali, 1973) especially at temperatures above -15°C (Christner et al., 2008; Murray et al., 2012). Although the contribution of biogenic INPs to the total global INP abundance is thought to be rather low (Hoose et al., 2010), biogenic aerosol may contribute substantially at regional scales where biological aerosol sources are important. For example, Tobo et al. (2013), Prenni et al. (2009) and O'Sullivan et al. (2018) have observed biogenic aerosol in the INP populations of the forested environments in Colorado, in the Amazon basin and in rural areas in Northern Europe. Furthermore, Pratt et al. (2009) and Creamean et al. (2013) showed that biological particles were frequently present in ice crystal and precipitation residues measured over the western United States and suggested that these particles play a key role in cloud ice formation. In a study about Swedish and Czech birch pollen, Augustin et al. (2013) reported the ice-nucleation activity of sampled macromolecules and formulated new parameterizations for the heterogeneous nucleation rates of two different ice-active macromolecules. However, in general, measuring and parameterizing the IN ability of biogenic particles has proven to be difficult for several reasons. For accurate biogenic INP model simulations, it is critical to understand the global distribution of biogenic INP, their source strength and their aerosolization and atmospheric transport mechanisms (O'Sullivan et al., 2018). It remains unresolved how the microphysical and chemical properties of biogenic aerosol may change during transport processes in the atmosphere. In field studies, which attempt to address these deficiencies, it remains difficult to identify biogenic aerosol particles and to separate them from non-biogenic particles (Möhler et al., 2007). Moreover, there are many biogenic species with a range of properties, which complicate comparisons and generalized parameterizations.

The snow cover information is useful and corroborates the INP concentration cycle, but what about the transition between melt and full growth of vegetation? Showing some sort of vegetation index and/or type information would be useful, particularly for the inter-seasonal transitions.

For our measurement site at SMEARII, the vegetation indexes NDVI (Normalized Difference Vegetation Index) and PRI (Photochemical Reflectance Index) are available, but only from 11 March 2018 to 11 September 2018. The NDVI and PRI data and the following interpretation is provided by Jon Atherton and Pasi Kolari from University of Helsinki (personal communication, October, 2020). For both vegetation indices, we calculated daily average values, which are averaged between 12:00 and 13:00 Finnish time, because the solar angle plays an important role for these indices. NDVI mainly tracks the development and loss of green leaf material including overstory trees and understory shrubs. We observe a trend in NDVI over the seasonal transition as the canopy slowly

greens from winter to summer. During summer, when the color of the canopy does not change significantly, the NDVI remains relatively constant. The increase in NDVI in the transition period from winter to summer is observed some days later than the increase in INP concentrations. The increase in INP concentrations is nicely represented by increasing air temperature and melting snow in early April 2018 (see Figure 4), whereas a strong increase in NDVI is observed in the second half of April. It has to be considered, that the NDVI is affected by snow resulting in smaller NDVI values. The PRI measures the abundance of photoprotective pigments called carotenoids, which help the tree to deal with excess light, which is potentially harmful. We observed a PRI increase in May, which indicates a carotenoid change of the trees. A relationship to the increasing INP concentrations in April is not observed.

We do not think that these comparisons are of great benefit to the story outlined within the manuscript and have decided not to include the comparison in the paper. However, we are glad it can be made available in this publicly accessible discussion and gratefully acknowledge the support of Jon Atherton and Pasi Kolari by providing and interpreting the NDVI and PRI data.

The methodology on the WIBS and L-ToF-AMS is incredibly limited. Because data from these methods are presented in the paper, the methods should include sufficient descriptions on each instrument, their operating parameters during SMEAR II, and data analysis and interpretation. Even though the L-ToF-AMS is presented in detail in Paramonov et al. (2020), there should still be a brief description of the instrument and data produced here.

Thanks for this comment. We agree that a more detailed explanation of these measurements is needed to support the results of this study. We have added a more detailed description of the WIBS instrument to Section 2.3. "Additional Instrumentation at SMEARII". As the other referees also asked further questions about the details on the WIBS and L-ToF-AMS, the added description addresses the comments of all the referees regarding this topic:

The WIBS-NEO (Droplet Measurement Technologies, Longmont, CO, USA) is a bioaerosol sensor that provides information on the fluorescence properties, size and asphericity ratio of individual aerosol particles. It operates with an inlet flow of 0.3 l min^{-1} and detects particles with diameters between 500 nm and 30 μm . From 11 March 2018 to 2 April 2018, the WIBS was located about 50m from the aerosol filter sampling line used for the INP analysis. There, it was attached to a total aerosol inlet, which is characterized in Vogel (2018). On 3 April 2018, the WIBS was moved and installed directly next to the filter sampling line and attached to a PM10 inlet, which is described in Schmale et al. (2017). For the WIBS data analysis, particles from 0.5 μm to 10 μm were considered. To analyse the fluorescence of the particles, the WIBS sensor utilizes two xenon flashlamps as excitation light sources (optically filtered at wavelengths of 280 nm and 370 nm) and two emission detection channels (wavelength bands 310 – 400 nm and 420 – 650 nm). Optical size information is acquired utilizing elastic scattering from a continuous wave laser with a wavelength of 635 nm and a photomultiplier tube located orthogonally with respect to the laser. The excitation pulses are fired into the sample volume at different times and both detection channels record the emission(s) from both excitations, leading to three distinguishable excitation-emission combinations (the 370 nm light saturates the 310 – 400 nm detection channel and therefore does not provide any information). Thus, the fluorescence can be divided into 7 unique fluorescence groups based on the excitation-emission wavelength pairs and their combinations, after Perring et al. (2015) and Savage et al. (2017): A (only FL1: excitation 280 nm, emission 310 – 400 nm), B (only FL2: excitation 280 nm, emission 420 – 650 nm), C (only FL3: excitation 370 nm, emission 420 – 650 nm), AB (FL1 + FL2), BC (FL2 + FL3), AC (FL1 + FL3) and ABC (FL1 + FL2 + FL3).

The WIBS performs an empty-chamber background signal check every 8 hours, during which the excitation pulses are fired into the optical chamber without any present particles. The background check collects a multitude of emission intensities that form a baseline for particle fluorescence. In this study, a particle is considered fluorescent, if the associated emission peak intensity is larger than $FT + 9\sigma$. FT is the mean value of the forced trigger intensities and σ is their standard deviation.

A more commonly used method would be to compare the emission peak intensity to $FT + 3\sigma$. However, some non-biological particle types such as wood smoke, African dust and black carbon are weakly fluorescent and therefore might satisfy the lower threshold value, leading to an overestimation of biological particle concentration. Furthermore, the stricter threshold only marginally affects the detection efficiency of biological particles, because they tend to have stronger fluorescence (Savage et al., 2017). More detailed descriptions on the WIBS are also available in Savage et al. (2017) and Perring et al. (2015).

We also added the following text to the description on Figure 4 (old Figure 3b) to Section 3.2 “Comparison to meteorology and aerosol properties” to explain the characteristic of the different excitation-emission wavelengths pairs:

The time series of the number concentration of particles with a fluorescence signal in other fluorescence groups is shown in the Appendix in Fig. A2. In this Figure, the strongest seasonal increase in the transition period from winter to summer is observed in the group ABC. Consequently, this fluorescent group correlates best with the measured INP concentrations (see Figure 4). The characteristics of each fluorescence group are comprehensively investigated and reported in Savage et al. (2017), who examined the fluorescence emissions of different types of pollen, fungi, bacteria, biofluorophores, dust, HULIS (humic-like substances), PAH (polycyclic aromatic hydrocarbons), soot and brown carbon. Using the $FT + 9\sigma$ threshold for defining a particle as fluorescent, nearly all dust and HULIS types show no fluorescence signal at all. Some of the soot and brown carbon types only show weak signals in A, and B, BC and A, respectively. Nearly all of the bacteria types show fluorescence only in group A. The fluorescence of fungal spores are also mainly detected in group A, but also in AB and ABC. The investigated biofluorophores show mainly fluorescence in the groups BC (Riboflavin, NAD), A (Pyridoxamine), AB (Tryptophan) and ABC (Ergosterol). PAHs show fluorescence mostly in groups ABC and A. Finally, most pollen types show fluorescence in groups ABC and AB. Some pollen types also show a fluorescence signal in groups A and B.

We also adjusted the discussion on the comparison of WIBS data to INP in the same Section as follows:

Such a correlation is also supported by the peaks of pollen and PBAP concentrations observed in snow-free periods in spring and in autumn, and by the increases of the organic aerosol mass concentration and fluorescent particle numbers of group ABC observed in spring. According to the study of Savage et al. (2017), we assume particles of fluorescence group ABC to be mainly pollen, particles containing PAH or Ergosterol, or fungal spores. ~~Fluorescent particles are expected to be primarily of biological origin except for a few percent, which could arise from non-biological materials (Pöhlker et al., 2012; Savage et al., 2017).~~

We also added a more detailed description on the L-ToF-AMS instrument to Section 2.3.” Additional Instrumentation at SMEARII”:

The size-resolved chemical composition of ambient aerosol was measured with the L-ToF-AMS. Its application in the same campaign has been described in Paramonov et al. (2020). It builds on the functionality and characteristics of the high-resolution ToF-AMS (DeCarlo et al., 2006). However, due to the longer time-of-flight chamber, the L-ToF-AMS, has a better resolution ($8000 \text{ M}/\Delta\text{M}$) than the

standard ToF-AMS (2000 M/ Δ M in V-mode). Detailed descriptions of the instrument, measurements and data processing are available in other publications (Canagaratna et al., 2007; DeCarlo et al., 2006). In general, the L-ToF-AMS measures the size-resolved, non-refractory composition of submicron aerosols, including organic, sulfate, nitrate, ammonium and chloride. The aerodynamic lens has a 100% transmission range of 75–650 nm (in vacuum aerodynamic diameter; Liu et al. (2007)) and focuses particles into a narrow beam that impacts the surface of a porous tungsten vaporizer heated to 600°C, followed by ionization by a 70eV electron source. Ions are detected by a long time-of-flight mass analyzer (ToFwerk AG). The sample flow of 0.09 l min⁻¹ is extracted from an extra suction flow (3 l min⁻¹) that is used to avoid aerosol losses in the inlet line. A PM2.5 cyclone mounted at the inlet removes large particles to avoid clogging the critical orifice (100 μ m), and before entering the L-ToF-AMS, the samples are dried by a Nafion dryer to keep the RH below 30%.

The L-ToF-AMS data were analyzed using standard ToF-AMS data analysis toolkits (Squirrel V1.61B and PIKA1.21B) using Igor Pro software (V6.37, WaveMetrics Inc.). To calculate mass concentrations an ionization efficiency (IE) was determined using 300 nm, size-selected, dry ammonium nitrate particles, and a relative ionization efficiency (RIE) for ammonium of 3.7 was determined. The default relative ionization efficiency (RIE) values of 1.1, 1.2, 1.3 and 1.4 for nitrate, sulfate, chloride and organics, respectively, were applied. A composition-dependent collection efficiency (CE) was applied based on the principles proposed by Middlebrook et al. (2012).

We also added the following text to the discussion on Figure 4 (old Figure 3b) to Section 3.2 “Comparison to meteorology and aerosol properties” to give more information about the measured organics:

The non-refractory organic components measured by the AMS include the commonly observed primary organic aerosol (POA) and oxygenated organic aerosol (OOA).

Figure 1: It would be useful to show an averaged spectrum per month overlaid on the data in each panel.

With monthly averaging, the variability of the spectra within one month and also the characteristic spectral shape is no longer visible. As this Figure is meant to show the daily/monthly variability and characteristic concentrations and spectral shapes throughout the full year of measurements, we have decided to keep the Figure as is.

Figure 2: The data in panel a are redundant from Figure 1. Suggest omitting and just keeping panel b.

We think it is reasonable to keep panel (a) in Figure 2, as the representation on a time axis allows easier peak identification in the INP concentrations and highlights the seasonal variability and trends.

For the “bulge” which is more pronounced in the heated versus unamended INP spectra for the summer samples, why would this be? There should be some discussion as to why this feature is more prominent when the samples were heated, and why this would occur only for samples collected during the summer.

This is a very interesting point. So far, the “bulge” is only an observation, for which we do not yet have a concrete explanation. We want to show what we observed, which may also be a basis for further studies and measurement campaigns that could directly be initiated with the objective to explain this observation.

Because n_s is shown earlier on than page 11, the calculation should be provided in the methods.

Thanks for this comment. We have added the following section to the Methods:

INAS densities were calculated as described in Eq. (2) in Ullrich et al. (2017), where ice number concentrations are normalized by the aerosol surface area concentration. Assuming that every INP triggers the formation of one ice crystal, the ice number concentrations are equal to the INP concentrations, which are determined by the INSEKT measurements. The aerosol surface area concentrations are derived from continuous size distribution measurements of the PM₁₀ atmospheric aerosol at SMEARII. Details on the size distribution measurements are given in the following section.

Like the introduction, the conclusions are brief and somewhat limited. The “bigger picture” should be reiterated for context of the measurements, and perhaps some discussion on what the authors recommend for the next step and future work.

We agree that the conclusion needed improvements – thanks for this comment. We now set our study in the context of the “bigger picture” and have added suggestions for further work:

As INPs strongly influence precipitation formation and cloud evolution, a description of INPs in weather forecast models is crucial. This study shows that the ice nucleation activity in the atmosphere is highly variable depending on the surrounding conditions. Therefore, it is important to investigate INP concentrations and INP types in different characteristic locations on Earth to establish an overall picture of the global INP abundance and variability. For investigating long-term variability, continuous long-term observations are needed to get a profound insight on the ice nucleation activity at a specific site with a good statistics accounting for the difficulties and uncertainties in INP measurements. With this study, we provide a first step to this overall picture by characterizing the INP population abundant in a remote location in the boreal forest. With continuous aerosol filter sampling for more than one year, we provide the first observation of a clear seasonal cycle, which seems to be dominated by the abundance biogenic aerosol. As in this remote environment, biogenic aerosols seem to play an important role, in other areas, the INP population might be dominated by other species. For further studies, we suggest to conduct further continuous long-term measurements of INPs at different locations on Earth, like anthropogenic influenced locations or deserts. Measurements with a higher time resolution might be useful to investigate relations to meteorological events like precipitation and frontal passages in more detail.

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