Response to the Editor Decision:

Dear authors,

Thank you very much for your revised version and your reply to the reviewer's comments. There are a few things to be corrected before your manuscript can be accepted for ACP:

1.) Reviewer 3 (and also reviewer 1) commented on the parameterization of INP number concentration via temperature. Although this is an interesting finding, I agree with the reviewers that the physical meaning is missing. The mathematical relationship is not really useful to be directly implemented into models. I asked an additional reviewer (with modelling expertise of INP) for an additional opinion and the reviewer agrees with reviewer 3 that the related text should be revised. It has to be clear that the found relationship is useful to constrain models (please also revise the abstract & conclusions accordingly) but it is not a direct parameterization. See detailed comments for reviewer 3 and 4.

2.) Reviewer 1 asked a question about the vegetation indices (LAI, NDVI). I found this to be a useful additional information to the manuscript. You could add this comparison at least to the supplement and add a sentence to the result section.

3.) Reviewer 2 had some more minor revisions.

Looking forward to your final revisions!

Many thanks and kind regards

Paul Zieger.

Dear Paul Zieger,

Thank you for your feedback! We really appreciate your effort to include an additional referee in this discussion. We found his/her comments very useful and helpful.

Regarding point 1): We agree, that we need to point out more clearly, that this is not a direct parameterization of INP concentrations in the atmosphere, like other common parameterizations that are functions of aerosol number or surface area concentration, but more a new way to formulate the annual variability of the INP concentration in a larger source region like the boreal forest in our case. There, the annual variation of the ambient INP concentration may be controlled or at least largely influenced by temperature dependent source processes. To account for this, we followed the suggestions of Referee #4 and modified the manuscript accordingly. Please see the response to Referee #4 for details.

Regarding point 2): We looked again into the data of NDVI and PRI and we still think that there is no benefit from this comparison to the story of the manuscript. We observe increases of NDVI and PRI during the transition period from winter to summer, but this does not happen in the same time period when we observe increasing INP concentrations. For now, we do not have a profound basis to relate our INP observations to the NDVI and PRI variability throughout the year. A thorough analysis and interpretation would raise and involve more questions, which are beyond the scope of this manuscript and need further detailed data analysis, which will be the subject of a follow-up study.

Regarding point 3): We followed the suggestions of Referee #2 and modified the manuscript accordingly. For details, please see the response to Referee #2.

Thanks again and best regards,

Julia Schneider, on behalf of all authors

Response to the referee report by Referee #2:

We thank referee #2 for his or her helpful 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.

The revised manuscript by Schneider et al. has significantly improved from the original version. I only have a few minor comments below that the authors should address prior to acceptance.

I appreciate that the authors took care in adding more sufficient background. Perhaps a bit long in some places. I suggest shortening the discussion on Hartmann et al. to a 2 – 3 sentences so it is similar in length to the others.

Thanks for this suggestion. We adjusted the discussion on the study of Hartman et al. (2019) as follows:

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~~ and 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.

The paragraph starting on page 1 is quite lengthy. The authors should consider splitting up into multiple, more focused paragraphs, e.g., one on previous work, one on boreal forest background, and one on biogenic INPs. That leads nicely into the second (last) paragraph that provides a synopsis of the current work.

We follow the referee's suggestion and split the respective paragraph into shorter parts: general introduction and previous work (first), boreal forest environment (second), biogenic INPs (third), information about this study (fourth).

Page 5, line 208: Why was only the one inlet heated? Would that not introduce variability that would affect the sizing results?

The heated inlet is a total aerosol inlet for the size distribution measurements, which are permanently installed at SMEARII. The heating of the inlet to about 40°C is important to also sample fog or cloud droplets when they are present at the site. Such droplets can be larger than 10 µm in diameter, and then do not pass the PM10 inlets, which we use for the INP filter sampling. In such a situation, we may underestimate the INP number concentration compared to a total aerosol inlet. At dry conditions without large droplets present, which was the case most of the time during this study in Hyytiälä, the difference between the inlets is negligible. Therefore, the dry aerosol measured at the heated total aerosol inlet is a good reference for our INP measurements at the PM10 inlet. In general, it is better to measure both INPs and aerosol parameters at the same inlet, but this was not possible during this study in Hyytiälä because of space and inlet flow limitations. The effect of different inlets, with different size cutoffs, and parallel INP sampling or measurements with size-selected aerosols are a topic of ongoing and upcoming studies.

Again, the paragraph starting at the end of page 5 is quite long. Consider breaking up into size distributions (DMPS+APS), fluorescence measurements (WIBS), and chemical measurements (LToF-AMS).

We split this section into three paragraphs, as suggested.

Page 6, line 239: Can eliminate the sentence starting with "Note" as that is redundant.

We removed this sentence from the manuscript.

Section 3.2: Another long paragraph. Consider splitting up to make more concise paragraphs.

We split Section 3.2 in four shorter paragraphs. In principle, three Figures (Fig. 3,4 and 5) are discussed in this Section. Therefore, we split the text in one paragraph for each Figure description (three paragraphs in total) and a fourth paragraph where the findings of the three Figures are contextualized.

Page 9, lines 293 – 303: This information belongs in the methods section.

We have shifted the information to the Methods Section "2.3 Additional instrumentation at SMEARII". We have also added a short description of the other instruments that measure meteorological variables at SMEARII and are used in this study.

We have added the following text:

Various meteorological parameters are continuously monitored at SMEARII. For this study, we used five basic variables including ambient air temperature, relative humidity, wind speed, snow depth and precipitation. The ambient air temperature was measured 4.2 m above ground with a Pt100 sensor inside a ventilated custom-made radiation shield. This 4.2 m temperature measurement is the closest to ground-level at SMEARII and thus we utilize this as the ground-level ambient air temperature in the following. The relative humidity was measured in 35 m height by a Rotronic MP102H RH sensor. For wind speed measurements, we used a Thies 2D Ultrasonic anemometer at 34 m above the ground by. The snow depth was measured by a Jenoptik SHM30 snow depth sensor, which is based on an opto-electronic laser distance sensor, in open field about 500 m southeast of the aerosol collection area of SMEARII. The precipitation, the liquid water equivalent, was measured by a Vaisala FD12P Weather Sensor in 18 m height.

Page 10, lines 341 – 343: Wouldn't this be expected given the wind measurements were above the canopy and the INP measurements were below?

During the intensive measurements period during the HyICE-2018 campaign from March to May 2018, additional INP measurements were conducted on a 35 m high tower to directly compare INP concentrations above and below the forest canopy. These measurements were not included in this study, as they were only available for a short period of the year and did not encompass a seasonal cycle. However, we observed no systematic difference between the INP concentrations below and above the canopy. Therefore, it was not implicitly expected that there is no relationship between INP concentrations below the canopy and the wind speed above the canopy observed.

More detailed analyses and additional comparisons of INP concentrations below and above the canopy will be included in future work, which will focus on the intensive measurement period of the HyICE-2018 campaign.

Page 16, line 452: What a remarkable reproduction of the data! Very great result.

Thank you!

References:

Hartmann, M., Blunier, T., Brügger, S. O., Schmale, J., Schwikowski, M., Vogel, A., Wex, H. and Stratmann, F.: Variation of Ice Nucleating Particles in the European Arctic Over the Last Centuries, Geophys. Res. Lett., 46(7), 4007–4016, doi:10.1029/2019GL082311, 2019.

Response to the referee report by Referee #3:

We thank referee #3 for his or her feedback. Please find our responses below 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.

The authors have identified a strong correlation between the ground temperature and the INP number concentrations, which they interpret as a link to seasonality. This correlation is then used to "develop a parameterization". In my original comment, I emphasize how, while the parameterization "works", it doesn't have a physical meaning and also wouldn't be something realistic to implement in a model. I think I would be ok if the authors want to present the analysis as a fit to data in supplemental material, but I don't think it belongs in the main text nor do I think it's a valid parameterization.

The authors respond "This formulation can still be useful as a technical basis for atmospheric models, which could help to improve the representation of atmospheric INP concentration by using the ground-level ambient temperature." - This is not a reasonable application, as this would require a model that includes tracers to determine the aerosol origin when calculating INP numbers at cloud level (this is not a common framework and is also very computationally expensive). I think the authors may not be familiar with how models estimated ice nucleation and INPs, as the models do not track INPs from various sources, but only represent the emissions of aerosols and the physical processes that act on aerosols (e.g., scavenging); Ice nucleation is then calculated based on a parameterization applied to a simulated aerosol parameter (e.g., abundance of biological particles, Hoose et al. 2010).

That is, the INP number concentration that is measured at the surface will not be the same as at cloud-level. Meaning, if using this ground-based-temperature "parameterization", one needs to have additional information on things like the number & type of particles to know how to scale the INP number concentrations at higher altitudes. That is, if you have an airmass that originates from this sampling location with e.g., ground temperature of 10 deg C and a corresponding number concentration of INP active at 257K (nINP) of 0.3 per liter, once that air mass reaches cloud level, the nINP will be altered due to 1) dilution, 2) scavenging, 3) mixing with other air masses from different location containing different aerosol.

Finally, the authors write "The models will not manage to describe the real physical processes behind aerosol particle behaviors as this is very complex and any model cannot resolve the involved time scales.", which is not correct. Aerosol models have become increasingly sophisticated and are able to represent number, size, and mass of aerosols from many sources. Model timescales are as capability of resolving aerosol processes as they are cloud processes. For the biological particles that appear dominate in the INP population in this study, for example, there are a many studies to illustrate growing modeling capabilities, for which I provide a couple:

Model-estimated bioaerosol evaluated against single particle mass spectrometer measurements

Zawadowicz, M. A., Froyd, K. D., Perring, A. E., Murphy, D. M., Spracklen, D. V., Heald, C. L., et al. (2019). Model-measurement consistency and limits of bioaerosol abundance over the continental United States. Atmos. Chem. Phys., 13.

Classical nucleation theory based approach for estimating IN, including biological particles:

Hoose, C., Kristjánsson, J. E., Chen, J.-P., & Hazra, A. (2010). A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model. Journal of the Atmospheric Sciences, 67(8), 2483–2503. https://doi.org/10.1175/2010JAS3425.1

All in all, I stand by my comment that the ground temperature based "parameterization" should not be included in the main text or presented as a parameterization.

We note the referee’s concerns and critical comments about the temperature-based parameterization. After discussion with the co-authors and consideration of the comments from other referees, we think there is justification for the presentation of the parameterization within the manuscript, given a few clarifications and modifications. We agree that the background for and applicability of this parameterization was not well explained and have therefore added explanatory statements. For example, as suggested by Referee#4, we explain that this is a mechanistic parameterization, which describes the near-surface INP concentrations as a function of the temperature, assuming that this concentration is dominated by temperature-dependent INP sources. In addition, we clarify within the text that this mechanistic parameterization only describes INP concentrations near the surface, and that it is only applicable in the boreal forest area. With this, we hope to clarify how this formulation can be used and applied in models.

We have changed the following passages, as shown below. For more details of the suggested changes to the manuscript, please see also the response to Referee #4.

Abstract: As current parameterizations do not reproduce this variability, we suggest a new mechanistic ~~parameterization~~ description for boreal forest environments, which considers the seasonal variation of INP concentrations. For this, we use the ~~ground-level~~ ambient air temperature measured close to the ground at 4.2 m height as a proxy for the season, which ~~affects~~appears to affect the source strength of biogenic emissions and thus~~by that~~ the INP abundance over the boreal forest ~~areas~~.

Methods: The ambient air temperature was measured 4.2 m above ground with a Pt100 sensor inside a ventilated custom-made radiation shield. This 4.2m temperature measurement is the closest to ground-level at SMEARII and thus we utilize this as the ground-level ambient air temperature in the following.

Description of Equ. (1) (Sec. 3.4): To account for seasonal dependencies in this formulation, the linear relation between the ~~ground-level~~ ambient air temperature T_{amb} in K measured close to the ground at 4.2 m height (called ground-level ambient air temperature) and the natural logarithm of the time series of INP concentrations […].

[…] with a1 = 0.074 ± 0.006, a2 = -18 ± 2, b1 = -0.504 ± 0.005, b2 = 127 ± 1 and with the activation temperature T and ground-level ambient air temperature T_{amb} in K (measured at 4.2 m height).

Discussion on parameterization (Sec. 3.4): ~~With this new approach, we do not directly describe the INP concentration in the atmosphere in a specific environment, as it was common in previous studies (DeMott et al., 2010; Tobo et al., 2013). Rather we have found a way to describe the boreal forest as an important INP emitting source. We suggest this formulation for application in atmospheric models to describe the source concentration of INPs abundant at ground level, which can then be further transported to cloud-relevant altitudes within the models. Here, it needs to be considered that~~ This new parameterization approach describes the annual variation of the near-surface INP concentration in the boreal forest, which provides a temperature dependent source of these INPs. We did not directly detect or quantify the INP source, but found a strong correlation of the measured INP concentration with the ground-level ambient air temperature. This leads to the assumption that the near-surface INP concentration in the boreal forest may be dominated by local or regional sources, and that this parameterization may be used in models to formulate the source strength or concentration of INPs in boreal forests near the surface. It should be noted that this is a mechanistic approach, which cannot necessarily be applied to regions other than boreal areas or to higher altitudes, where the INP spectrum may be influenced by other sources. It is further important to note that INPs might undergo changes in their size distribution and chemical composition, when they are transported from their sources to higher altitudes, which could affect their ice nucleation ability.

Conclusions: Thus, we suggest two new approaches for formulating and quantifying the annual ~~cycle~~ variability of INP concentrations over boreal forest areas. The first is a mechanistic approach, which considers the boreal forest as ~~an aerosol~~ a temperature dependent INP emission source~~, including INPs, which appears to vary strongly with seasonal changes~~ with a pronounced seasonal cycle.

Response to the referee report by Referee #4:

We thank referee #4 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.

I have been asked to comment on a specific point raised by Reviewer#3, but also mentioned by Reviewer#1 : the question of the feasibility of the temperature dependent parameterization.

This parameterization shows very impressive results when compared to observations. This suggests that the temperature close to the surface might represent the variability in measured INP concentrations better than any other meteorological parameter (such as humidity in the surface layer) or any index representative of the vegetation. I agree with Reviewer#3 that a physical explanation of this relationship is missing, but I wouldn't remove this temperature-dependent parameterization. The possible contribution of PBAPs to INP population is probably somewhat embedded in this relation. I still believe it is valuable for models because INP concentrations are generally very badly represented, or even not taken into account (many atmospheric models just use a temperature and supersaturation dependent parameterization). It could help to improve the representation of atmospheric INP concentrations and their seasonal variability. Hence, I think it should be kept in the paper. But, to avoid confusion for modelers who could be interested in this parameterization, it should be clearly stated that :

1/ what is called ambient temperature in the pre-exponential factor is the air temperature close to the surface. In models, it is generally represented by the 2m temperature;

2/ this is not a physical parameterization, but a mechanistic parameterization to represent sources near the ground level that may impact INP populations;

3/ this parameterization is only relevant over boreal forest areas, not elsewhere.

To 1): We did not use the air temperature directly measured at the surface, but in 4.2 m above the surface, which is the temperature measurement closest to the surface and continuously available at SMEARII. Therefore, we called this ground-level temperature here.

We made this clear point clearer in the manuscript by adding the information about real temperature measurement height of 4.2 m into the Methods section as well as in the description of equation (1), which gives the parameterization.

Methods: The ambient air temperature was measured 4.2 m above ground with a Pt100 sensor inside a ventilated custom-made radiation shield. This 4.2m temperature measurement is the closest to ground-level at SMEARII and thus we utilize this as the ground-level ambient air temperature in the following.

Description of Equ. (1) (Sec. 3.4): To account for seasonal dependencies in this formulation, the linear relation between the ~~ground-level~~ ambient air temperature T_{amb} in K measured close to the ground at 4.2 m height (called ground-level ambient air temperature) and the natural logarithm of the time series of INP concentrations […].

[…] with a1 = 0.074 ± 0.006, a2 = -18 ± 2, b1 = -0.504 ± 0.005, b2 = 127 ± 1 and with the activation temperature T and ground-level ambient air temperature T_{amb} in K (measured at 4.2 m height).

To 1), 2) and 3): We followed your suggestions and adjusted the corresponding passages in the text to clarify that we used the ambient air temperature measured close to the ground, that the parameterization is only valid in boreal environments and that the parameterization is only mechanistic, as follows:

Abstract: As current parameterizations do not reproduce this variability, we suggest a new mechanistic ~~parameterization~~ description for boreal forest environments, which considers the seasonal variation of INP concentrations. For this, we use the ~~ground-level~~ ambient air temperature measured close to the ground at 4.2 m height as a proxy for the season, which ~~affects~~appears to affect the source strength of biogenic emissions and thus~~by that~~ the INP abundance over the boreal forest ~~areas~~.

Discussion on parameterization (Sec. 3.4): ~~With this new approach, we do not directly describe the INP concentration in the atmosphere in a specific environment, as it was common in previous studies (DeMott et al., 2010; Tobo et al., 2013). Rather we have found a way to describe the boreal forest as an important INP emitting source. We suggest this formulation for application in atmospheric models to describe the source concentration of INPs abundant at ground level, which can then be further transported to cloud-relevant altitudes within the models. Here, it needs to be considered that~~ This new parameterization approach describes the annual variation of the near-surface INP concentration in the boreal forest, which provides a temperature dependent source of these INPs. We did not directly detect or quantify the INP source, but found a strong correlation of the measured INP concentration with the ground-level ambient air temperature. This leads to the assumption that the near-surface INP concentration in the boreal forest may be dominated by local or regional sources, and that this parameterization may be used in models to formulate the source strength or concentration of INPs in boreal forests near the surface. It should be noted that this is a mechanistic approach, which cannot necessarily be applied to regions other than boreal areas or to higher altitudes, where the INP spectrum may be influenced by other sources. It is further important to note that INPs might undergo changes in their size distribution and chemical composition, when they are transported from their sources to higher altitudes, which could affect their ice nucleation ability.

Conclusions: Thus, we suggest two new approaches for formulating and quantifying the annual ~~cycle~~ variability of INP concentrations over boreal forest areas. The first is a mechanistic approach, which considers the boreal forest as ~~an aerosol~~ a temperature dependent INP emission source~~, including INPs, which appears to vary strongly with seasonal changes~~ with a pronounced seasonal cycle.