Review of "Condensation/immersion mode ice nucleating particles in a boreal environment"
Anonymous Reviewer
October 10, 2019

1. Summary
In this work, Paramonov et al. report the first measured, condensation/immersion mode INP concentrations from a boreal forest. Measurements were taken from 19 February to 2 April 2018 in Hyytiälä, Finland. INP concentrations were measured using the PINC instrument; on March 2nd, an L-TOF-AMS co-sampled with PINC, and a WIBS was added to both PINC and the L-TOF-AMS on March 11. In addition, total aerosol concentrations (CPC), size distributions (DMPS, APS), and black carbon (MAAP) were measured at the nearby SMEAR II station.

These measurements are important, as they provide constraints to INP concentrations from models in a region where few INP measurements exist. They also provide some insight into the natural variability of INP at a single site over several months.

Authors' response: The authors would like to thank the reviewer for a detailed and insightful review. The line numbers below refer to the version without track changes.

That being said, the authors are extremely liberal in their interpretations. This is especially true in Section 3.2. The authors are INP experts, thus this reviewer is surprised that Pearson correlation coefficients are being used to chemically speciate the measured INP. As the authors note several times in the paper, INP are a rare subset of all aerosol particles. Thus, correlation certainly does not imply causation, and much of the paper beyond Section 3.1.2 suffers from this questionable-cause logical fallacy.

Authors' response: The authors acknowledge the limitations of using the Pearson correlation coefficients to interpret the data. The instrumentation setup during the field campaign did not allow for the single-particle analysis of ice crystal residuals, and this is addressed on lines 537-541 (page 17). Additionally, the fact that correlation *does not* imply causation is caveated throughout the manuscript on several occasions, e.g. lines 413-417 (page 13-14). This manuscript is not the first attempt at linearly correlating [INP] with various parameters in order to be able to say something about the physical and chemical properties of ambient INPs. There is a multitude of studies that have derived some use from this approach (Tobo et al., 2013; Mason et al., 2015; Wright et al., 2015; Welti et al., 2018).

The authors further note that Pearson correlation coefficients are not used to chemically speciate the measured INP. They are used to infer the predicting capacity of [INP], i.e. to explore if a parameter (and which one) can be used to predict the ambient INP number concentration. Predicting [INP] is one of the biggest challenges facing the IN communities, and attempts to find such parameter have been proposed before (Richardson et al., 2007; DeMott et al., 2010; Tobo et al., 2013). However, we add the following clarifications to address the reviewer concerns regarding correlation and causality.

- Lines 444-448 (pages 14-15), added text “It should also be noted here that correlation of [INP] with fluorescent particle concentration does not imply that INPs are necessarily fluorescent. Similar to the previous case, fluorescent particles may simply be a feature of the present airmass or a tracer for some other IN-active particle species. However, it is possible that fluorescent particles do contribute to the INP population given previous studies that show INP residuals sampled from the atmosphere to show an increased fluorescent fraction compared to background measurements (Boose et al., 2016).”.
- Lines 510-512 (page 16), added text “Third, and also similar to other special cases examined, a significant correlation between [INP] and the concentration of sub-0.1 μm particles does not imply that sub-0.1 μm particles are INPs; instead, they may be a tracer of some IN-active particle species or an airmass feature.”.
- Line 530-531 (page 17), sentence modified to read “On the day with the highest [INP], sub-0.1 μm particles, most likely nanoscale biological fragments such as INMs, were found to exhibit a significant correlation with the elevated INP number concentrations.”

I have outlined my primary concerns in the general comments section.

In addition, the language in the manuscript is generally wordy and often imprecise. Although this is a stylistic concern, it is prevalent enough throughout the manuscript that it detracts from its clarity. It is the responsibility of all authors on the manuscript to ensure that the manuscript language is clear, and the results are evident. As an example, I will provide minor/technical comments on the abstract paragraph; however, similar corrections need to be applied to the entire manuscript.

Authors’ response: We acknowledge the stylistic concerns of the reviewer. However, some redundancy is intentional to in order to remind and guide the reader through the manuscript. However, the language of the manuscript has undergone some modification (see tracked changes version) to account for reviewer’s comments.

2. General comments

1. **Section 2: As it is written as one, large chunk of text, the methods section is hard to digest. It would be helpful to the reader if the authors split this section into more subsections, (e.g., PINC, instruments co-located with PINC, instruments located at SMEAR II station, back trajectory analysis, etc.). I also could not find much detail about the back trajectory analysis in the methods section.**
   Authors’ response: Section 2.2 is now split into several subsections as per reviewer’s suggestion. A new subsection 2.2.6 is added to describe the trajectory analysis, and the related text moved from section 3.1.2 to section 2.2.6.

2. **Figure 1: Figure 1 is missing the L-TOF-AMS. It is also unclear from the figure that the DMPS, CPC, and APS were not at the same location as PINC, WIBS, and the L-TOF-AMS.**
   Authors’ response: Figure 1 has been updated to include all instrumentation that was used, and the figure caption updated to read: “Figure 1: Instrumental setup.”.

3. **Line 286: I am not sure why the authors believe that the back trajectories render a "potential marine source of the Norwegian Sea and the boreal forest source NE of Hyytiälä as improbable." To show surface influence, it would be more correct to see how many back trajectories were below the boundary layer height. Presumably, even during NH, high-latitude winter/spring, aerosol are well-mixed within the boundary layer. I strongly suggest that the back trajectory analysis be changed from <100 m to the <boundary layer height.**
   Authors' response:
   - We choose the trajectory arrival height of 100 m because we are interested in the potential influence of the Earth's surface and its emissions.
   - The definition of the boundary layer implies a well-mixed layer that is influenced by the surface. So if the BL height is 1 km, the trajectory arrival height should not matter so long as it is within the 1 km layer. This height should be well-representative of the BL conditions at SMEAR II. Only if the BL height was below 100 m would the trajectory arrival height of 100 m be inappropriate for studying the effects of the surface. But this is unlikely since most of the INP measurements presented took place during the day when the BL, as implied by the reviewer comments, is > 100 m.

4. **Figure 3: It is odd that a low fraction of trajectories at the SMEAR II site do not encounter the surface. Is this a known feature of Hyytiälä in the NH winter/spring? Or is this a result of your back trajectory arrival height? If the latter, did the authors test how sensitive the back trajectory results are to arrival height? Without such a sensitivity analysis-it would be even more difficult to exclude local INP sources from marine regions and the nearby boreal forests.**
   Authors' response:
   - Actually, Figure 3 shows the contrary. With a few exceptions, a very low fraction of trajectories is in contact with the surface. This is signified by the prevalence of blue colours in Figure 3C. While it is not possible to say if this is a known feature at SMEAR II, we did look at the results when the trajectory arrival height was set to 200 m and 500 m. In both of these cases, the likelihood of contact with the ground was even less, and the potential sources of INPs became even more inconclusive. This is consistent with expectations. If the airmasses arriving at a height of 100 m come from above 100 m, it is not very likely that airmasses arriving at a height of 500 m would be coming from below 100 m.
   - Since we are interested in the surface effects, the lowest possible arrival height of 100 m seems most appropriate. No changes made.

5. **Section 3.1.3: I do not see the value of this section; the range of INP concentrations in Hyytiälä at one temperature spanned almost 3 orders of magnitude. As CFDCs have lower detection limits around 1 L^{-1}, then this indicates that these measurements are similar to any site where the [INP] spans ~1-1000 L^{-1}.**
   Authors' response:
   - Our measurements and results are not stand-alone; they are part of an overall endeavour by the IN community to characterise ambient INPs. It is, therefore, necessary to provide context, to see how our results fit in the bigger picture and

how they compare to previous studies. Thus, though the value of this section may not be apparent, we keep this section in for the above reason and good scientific practice of comparing new results to those published previously on a similar topic.

- The reviewer is correct in saying that "the range of INP concentrations in Hyytiälä at one temperature spanned almost 3 orders of magnitude". The reviewer is also correct in saying that "these measurements are similar to any site where the [INP] spans ~1-1000 L-1", which is basically any site around the world without strong local sources. This section is meant to explicitly highlight that the range of ambient [INP] seems to be same around the globe and, given this, to raise the question of whether similar ambient INP measurements are necessary in the future. See lines 543-547 (page 17-18). No changes made.

6. **Section 3.2: As stated in the summary section, I find no reason why high correlations with the 22 measurements in this manuscript suggests anything about the composition of the actual INP. Thus, I do not believe that these correlations alone implicate BC, large biological particles, or small, biological nanoparticles as the INPs measured by PINC.**
   Authors' response: This issue is addressed above in the summary section. Each of the 3.2 sub-sections includes an explanation that the particle types correlating with [INP] may not actually be the particles acting as INPs. This can be seen on lines 413-417 (page 13-14), lines 444-446 (page 14), lines 469-470 (page 15) and lines 510-512 (page 16).

7. **Section 3.2.1: There is very little evidence in the literature that BC acts as an immersion-mode INP at activated fractions relevant to this work. This is true for both fossil fuel emissions (Schill et al., 2016) and for biomass burning surrogates (Levin et al., 2016), whose BC INP activated fractions are sim1x10^{-9}. It has also been shown that photochemical aging does not increase the INP efficiency of BC (Schill et al., 2016), in contradiction to the statement made on line 390. Thus, the INPs responsible are likely not BC, but some INP co-emitted with BC and present in activated fractions of ~2x10^{-6}.**
   Authors' response: Sentence in lines 405-406 (page 13) has been modified to read "Despite the short daylength, the cloudless conditions may result in the freshly emitted BC being subject to photochemical oxidation, leading to an increase in its ability to act as CCN (Li et al., 2018)." See further responses below.

   Furthermore, although this becomes a focal point of this section, there is little observational evidence that this BC is from residential heating.
   Authors' response:
   - It has been reported in previously published literature that the concentration of BC at SMEAR II is highest in February and March (Virkkula et al., 2011). The study used the light absorption technique to come to this conclusion. At the same time, Lewis et al. (2008) provided evidence that high light absorption coefficients are typically associated with different types of biomass burning. It, therefore, becomes quite likely that the source of BC at SMEAR II in February and March is biomass burning.

- Sentence added in lines 403-405 (page 13): "This notion has been indirectly supported by Lewis et al. (2008) and Virkkula et al. (2011) who reported that the high aerosol light absorption at SMEAR II in February and March is likely associated with biomass burning."

Finally, BC is not the only measurement that correlated with INP. INP are positively correlated with almost all of the aerosol indicators >100 nm, suggesting that there is something special about the air mass, not the BC, that is supplying INPs.
Authors' response: Yes, that's correct. It is well-known that large particles are more likely to act as INPs than smaller ones (Hoose and Möhler, 2012; Pruppacher and Klett, 1997). However, size alone does not tell us anything about what these INPs could be. We have found an interesting correlation with BC and we investigated further using existing literature. Whether BC is acting as an INP or is simply a tracer/airmass feature remains open for further research as mentioned in the manuscript (lines 413-417, pages 13-14).

One last specific note-the reference to the Prenni et al. (2012) paper is incorrect. They find INP in biomass burning emissions, but they do not attribute them to BC specifically. A follow-up paper by McCluskey et al. (2014) does show that BC can be found in INP residuals while sampling from prescribed burns, but the BC INP activated fractions were not reported.
Authors' response: Sentence in lines 407-410 (page 13) modified to read: "However, biomass burning has been shown to produce INPs active above water saturation at 243 K (Prenni et al. 2012) and warmer temperatures (McCluskey et al., 2014), supporting the notion of IN-active biomass burning particles originating from wood burning and heating during the examined time period.".

8. **Section 3.2.2: Similar to BC in Section 3.2.1, This section does not implicate biological particles as INP. Again, the authors ignore that INPs are correlated with all particles >100 nm, and focus only on a subset of their observations. Thus, again, the correlations indicate that a certain air mass type is correlated with [INP], not that a certain type of aerosol are INP.**
Authors' response:
   - Lines 444-448 (pages 14-15), added text "It should also be noted here that correlation of [INP] with fluorescent particle concentration does not imply that INPs are necessarily fluorescent. Similar to the previous case, fluorescent particles may simply be a feature of the present airmass or a tracer for some other IN-active particle species. However, it is possible that fluorescent particles do contribute to the INP population given previous studies that show INP residuals sampled from the atmosphere to show an increased fluorescent fraction compared to background measurements (Boose et al., 2016).".
   - We agree with the reviewer in that the correlation in this subsection indicates that a certain air mass type is correlated with [INP]; however, this implies that the INP correlate with aerosol in the air mass, given INP are a subset of condensed phase aerosols and not gas phase species. It is possible that the gas phase species in an air mass serve to modify the aerosol species causing an enhancement or

suppression of [INP]; however, the correlation should still be with the aerosol properties within an air mass type.

9. **Section 3.2.3: I am not sure why the authors chose to show the time series here instead of a figure similar to Figures 5 and 6;**
Authors' response: We wanted the reader to see the evolution of [INP] on what we think is the most interesting day of our measurements, as well as to have a visual cue as to what was happening on that day with respect to the weather and other parameters. Figures 5 and 6 do not provide such information.

however, even without the Pearson correlation coefficients, I agree with the authors that the correlation with sub-100-nm particles is striking. The authors suggest that the INP must also be sub-100-nm-this, however, is not supported by any observations.
Authors' response: Addressed above and in lines 413-417 (page 13-14), 444-448 (pages 14-15) and 510-512 (page 16). Also, see response below regarding sub-100-nm particles.

The authors hypothesize that these sub-100-nm INP biological nanoparticles, likely because most other known INP lose their ice nucleation activity below 100 nm (Marcolli et al., 2007). To support this hypothesis, the authors note that biological nanoparticles have previously been implicated as INP (Pummer et al., 2012; Fröhlich-Nowoisky et al., 2015; O'Sullivan et al., 2014); however, these biological nanoparticles are found in the ambient atmosphere attached to carrier particles >100 nm such as pollen, fungal spores, and soil dust. The ice nucleating entities were determined to be <100 nm by rinsing the ice nucleating entities off pollen, soil dust, etc.
Authors' response: That's correct. Previous studies have found IN-active biological nanoparticles attached to larger, carrier particles. However, if pollen particles during the warmest day of the measurement campaign are to rupture (either due to high humidity or mechanical rupture from turbulent wind conditions), sub pollen particles (SPPs), which are in the Aitken mode, are released and thus suspended in air on their own. It is reported that up to 70% of pollen grains rupture in the troposphere and release SPPs which get dispersed into the troposphere (Wozniak et al., 2018 and references therein). It is not necessary for biological fragments to be appended by carrier dust or soil particles. Previous studies have shown that biological molecules can be transported on soil or desert dust to long distances, but this is not the only form of bio-molecules existing in the atmosphere, and certainly not for the boundary layer, relevant to our measurement site. Thus, we cannot neglect the likelihood of these particles existing independently at least for some non-trivial amount of time, especially close to the emission source.

To the reviewer's knowledge, no study has observed unattached, ice nucleation active, sub-100-nm biological nanoparticles in ambient aerosol samples. Thus, attributing biological nanoparticles as the INP responsible for the high-[INP] event on 25 March 2018 is speculative at best.
Authors' response: Yes, we agree. However, the fact that SPPs have been observed to be active CCN even in the Aitken mode (Steiner et al., 2015 GRL) suggests that it is very

likely these particles can act as immersion INPs. Furthermore, most particle detection techniques used to identify ice crystal residuals are limited to observing populations of aerosols > 100 nm (cite Cziczo et al., 2017, AMS monographs), which could be a reason for this. However, the correlation here, albeit not a causation, can demonstrate the importance and need to consider potential identities of sub-100 nm INPs.

3. Stylistic Concerns: Abstract Example

- **Line 15: Parentheses around SMEAR II**
  corrected here and in line 103

- **Line 17: Delete the phrase "found to be," it is wordy and slightly redundant**
  corrected here and in line 515

- **Line 18: The INPs are not necessarily "a result of" dilution and long range transport. This suggests that dilution and long-range transport create INPs. The INPs are a result of long-range transport and dilution of INPs sourced far from the measurement site. This needs to be clear.**
  corrected here and in lines 518-520 to say "…INPs at SMEAR II are a result of long-range transport and dilution of INPs sourced far from the measurement site".

- **Line 21: You already made an abbreviation for INP number concentrations ([INP])- please use it here.**
  corrected

- **Line 23: The phrase "any of the examined relevant parameters," is vague here. If parameters do not correlate, then are they relevant? Furthermore, since you are not using these parameters to define INPs (or any system), they are not parameters. They should be called measurements or observations.**
  The world "relevant" removed here and in lines 453, 522, and 909.

- **Line 24: Again you have already abbreviated INP number concentrations to [INP].**
  corrected

- **Line 25: You use the subordinating conjunction "although," which suggests that you should omit the comma beforehand. In fact, "although" is connecting two independent ideas-thus, it would be clearer for the reader if you split this sentence in two.**
  The conjunction "although" is used here instead of "but" or "however", and it is meant to split two independent clauses. Using "Although" at the beginning of the sentence would make a fragment sentence. No changes made.

- **Line 28: You should not connect "correlated" with "found in," because they are not the same thing. The former is true, you did find a correlation; the latter is not, you did not find anything in the INP.**
  corrected here and in lines 527-528. "Signatures of" also removed in both instances.

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