Review of "Condensation/immersion mode ice nucleating particles in a boreal environment"

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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.

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.

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.

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.

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.

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.

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.

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}.

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.

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 $sim1\text{x}10^{-9}$. 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 $\sim 2 \times 10^{-6}$. Furthermore, although this becomes a focal point of this section, there is little observational evidence that this BC is from residential heating. 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. 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.

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.

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; 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. 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 of pollen, soil dust, etc. 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.

3 Stylistic Concerns: Abstract Example

- Line 15: Parentheses around SMEAR II

- Line 17: Delete the phrase "found to be," it is wordy and slightly redundant
- 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.
- Line 21: You already made an abbreviation for INP number concentrations ([INP])–please use it here.
- 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.
- Line 24: Again you have already abbreviated INP number concentrations to [INP].
- 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.
- 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.

References

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