Re-review of "Condensation/immersion mode ice nucleating particles in a boreal environment"

Anonymous Reviewer

February 28, 2020

1 General Comments

The reviewer would first like to reiterate that the measurements in this work are novel, and should be included in the literature; however, the reviewer still believes that the authors' interpretation of the data, especially the Pearson correlation coefficient analysis in Section 3.2, is incorrect. In their response to this reviewer's main comment, the authors explicitly state "that the Pearson correlation coefficients are not used to chemically speciate the measured INP." Instead, the authors insist that "they are used to infer the predicting capacity of [INP]." This sentiment, however, is not evident from the text in the manuscript. Instead the authors have only added several statements that correlation does not necessarily imply chemical speciation of the INPs; these statements are added as caveats after large sections that imply that correlation does indeed mean causation. With ice nucleation, the sentiment that correlation implies a potential role as an INP is *not* a caveat. It is generally false, and any instance should be scrubbed from the paper. If these sections are allowed to stay in the paper, then other people in the atmospheric chemistry and physics community can cite this paper and incorrectly assume that, for example, 10 to 100-nm biological nanofragments are INPs in boreal forests. This is not supported by the evidence collected in this work. The following are several examples of the authors attempting to use correlation to imply a role as an INP (all line numbers correspond to v4 of the manuscript).

- L26: "On shorter time scales, several particle species correlated well with [INP] implying their potential role as INPs"
- L27/28: "... sub-0.1 μm particles, most likely nanoscale biological fragments such as ice nucleating macromolecules (INMs), have been found in the INP signal."
- L98: "the investigation of INP physical and chemical properties"
- L332: "In order to probe the identity of the measured INPs ..."

- L411/12: "It seems as though the potential role of BC as INPs active under mixed-phase cloud regime during these particular weather conditions cannot be excluded."
- L417: "Reasons why the signature of BC as an INP shows up only during this time period and not others remain unknown."
- L430: "This supports the general notion that larger, supermicron particles are better INPs"
- L436: "It can, therefore, be said that significant linear correlations of [INP] with supermicron and fluorescent particles, as well as with organics, accompanied by the very high ambient RH likely indicate the importance of these biological particles released by the surrounding forest as INPs"
- L446/7: "However, it is possible that fluorescent particles do contribute to the INP population"
- L465: "What this might mean is that INPs on this day are below 0.1 μm in diameter, potentially even below 0.01 μm in diameter."
- L470: "these sub-0.1 μm particles that are most likely acting as INPs on this particular day."
- L488: "One possible identity of these highly IN-active sub-0.1 μm particles could be that of nanoscale biological fragments"
- L499/500: "Given the available data, it seems as though the highest measured [INP] during the campaign can be attributed to highly IN-active nanoscale biological fragments originating from surrounding vegetation."
- L527: "On shorter time scales, several particle species correlated well with [INP] implying their potential role as INPs"
- L530-4: "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. Reasons for why certain particle types act as INPs during certain conditions and not during others and why none of the particle species mentioned above correlate with [INP] across the entire campaign remain unknown."

As the authors note in their–these short term correlations are more likely indicators of specific air masses. Indeed, the authors do a nice job in Section 3.2.1 of stating that the correlation with BC and the back trajectories indicate that this air mass contains INP from biomass burning; however, the extra step in saying that there is a "the signature of BC as an INP" should be avoided. This type of discussion (and avoidance) should be developed in each subsection of section 3.2

2 General Comments

- L60: There is a logical fallacy here–black carbon is called a "well known" INP, but their INP activity is then called into question in L69 and L407.
- L63/64: DeMott 2010 only stipulates that n500 is correlated with INP, not that INP are larger. The Mason 2016 reference, however, is correct.
- L70: This mistake was made previously for a reference by Prenni et al., but Petters et al., 2009 only shows that biomass burning can be a source of INP, not BC. In fact, BC are shown *not* to be correlated with INP during the FLAME-2 study.
- L206: Several reviewer comments were centered around the choice of 100 m a.g.l. for the back trajectories. These comments were answered in the author responses to reviewer comments. This should be outlined here–especially the notion that 100 m is the lowest available level in the back trajectory, how well 100 m corresponds to the well-mixed boundary layer, and also the 200 m and 500 m sensitivity studies.
- L388: Interesting that the activated fractions and absolute [INP] are both low here. Does this suggests that biomass burning is not an efficient source of INP (compared to median values at SMEAR II), but could be important regionally?
- L406 and L443: These two lines in this paper correspond to increased CCN activity, however, the CFDC RH is 105%; is there any evidence that these particles do not activate at 5% supersaturation prior to photo-oxidation/sulfate accumulation?