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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice nucleating particles in the marine boundary layer in the Canadian Arctic during summer 2014" *by* Victoria E. Irish et al.

Anonymous Referee #1

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General Comments

This paper represents a nice contribution to the literature, adding to the limited data on immersion freezing INP spectra in the Arctic region and emphasizing the dominant role that mineral dusts can play when overlain on pristine air masses that are otherwise representative of marine air. I was only curious about the use of a ratio calculation made on the basis of mineral and sea spray aerosol surface areas estimated by CCSEM, rather than actually calculating a surface-active site density on the basis of the aerosol distribution to support what composition is most responsible for the INP activity. While the ratio approach is one to take, it would be good to see actual surface area

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estimates in order to understand the consistency of the data with previous mineral dust parameterizations, rather than only assuming the validity a priori.

For example, only three samples were analyzed, understandable of course given the effort needed to perform the analyses of many particles for composition and size. Given this though, the unknown exact source of the dust, and the fact that what remains active after transport and any scavenging removal could differ from pure minerals tested in the laboratory, it could be interesting to know if the ice activity is truly consistent with dust parameterizations or is reduced and in a more competitive scenario with marine INPs. Otherwise, it is difficult to know if the inference of the dust dominance at all temperatures and loadings is as assumed. To know this, one might need to know mass fractions and surface areas very accurately. Hence, I suggest to add these actual values to the tables, and perhaps some discussion of alignment of data with the published or estimated parameterizations used to quantify assumed mineral influence. As for the estimates of marine INP contributions, the authors may know that an active site density parameterization is now in the literature (McCluskey et al., 2018), which seems to align quite well with the values they infer from published laboratory data.

I was a little curious about two factors in how the data were utilized. First, there is sparse data at -15 to -20 °C as emphasized temperatures. And I note on Page 9, lines 13-14, that even 36% of the samples had no INPs at -25 °C. Were "zero" or unresolvable data treated in some manner to come up with the average values plotted in Fig. 3? Also, a decision was made to not attempt to correct and discuss INPs for data at lower temperatures where background is present from the pure water. Was the correction simply too large in this region? Finally, I question whether time over marine areas would be expected to positively correlate with INP concentrations in any case. This is not obvious to me given the low strength of the marine source of INPs and the ready ability of terrestrial sources to dominate with any exposure to land emissions, which has been clear in some past studies.

Specific questions/comments for addressing before publication are listed below. The

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paper is otherwise very well written, so these extra comments are limited to the topics overviewed above.

Specific Comments

1) Page 3, lines 17-20: Could the meaning of the wind directions noted be clarified? Do these imply from open water? Or assumed free from stack contamination? This actually touches on the topic of using remote ocean data and assuming marine only influence of course, about which not much is said.

2) Page 5, lines 18-19: Can you justify not considering the freezing of drops not on a spot as another sort of background freezing spectrum? Is it because you cannot be certain that some small particles did not migrate under this position on the slide?

3) Page 7, lines 3-5: This statement about the footprint layer confused me because I could not clearly distinguish how this differed from the earlier statement that the terminal point was 0 to 60 m above sea level. Please add, if possible.

4) Page 7, lines 19-20: One sample with "low" INP was selected, but I see only 4 or 5 samples in Fig. 2 that even have data at -15 °C, so if three of these were used, then they are already not low INP samples I would judge. Is there a possible bias in the samples selected? After all, INP concentrations of 0.1 per liter at this temperature are fairly classical continental numbers in the first place. Your qualification about the conclusions being applicable only to the CCSEM cases on page 9 is duly noted. I wonder if you might comment about the influence of selection of samples for microscopy.

5) Page 7, lines 23-24: Please clarify if fully quantitative EDS analysis is performed to determine atomic percentages. That is the counts are interpreted quantitatively or qualitatively? Figure S4 shows actual concentration values as a means of interpreting mixtures as sea salt or dust, but there are no mixtures. It would be interesting to know how many of the dusts were salty, as processing may potentially alter their assumed behaviors (i.e., compared to parameterized dust).

6) Page 7, lines 27-28: Please also tabulate surface areas attributed to mineral dust and sea salt. It seems that one could also derive n_s for each of these. This is critically important to the assumption that these can simply be applied in existing parameterizations that will then be turned into a ratio according to your Eq. 3.

7) Page 8, lines 18-21: Following from what I just said, if you have surface area, can't you compute n_s and compare to parameterizations on the assumptions that particles are all SSA or all minerals, or use the mineral ratio to parse it out? Are the results consistent in any way with the parameterization or does the dust act differently than expected? Otherwise, by applying Niemand et al., you are making assumptions about the activity of the dust in these samples that may or may not be true. The same could be said about the marine spectrum, but the dust appears to dominate here, so is the most critical assumption.

8) Page 9: The correlation analyses are not especially impressive on first glance. Nevertheless, it seems to make sense that residence time over land would have a positive impact on INP concentrations, while time over water would not be expected to matter. This is the natural result when the land surface is a so much more powerful source by the amount that has been noted in past studies. The marine source would only be expected to show up when there is little or no land influence. Both sources would seem to depend more on wind conditions.

9) Page 10, lines 11-12: As noted, there is field evidence for marine INP ice active surface site densities in line with what is assumed for the exercise here.

10) Page 10, line 14: The comment about biological INPs is kind of a hanging thought. Are you inferring that the results are influenced by these? Your references suggest a range of source types including microbes or macromolecules it seems. While one might agree with the sentiment, it is not clear how it relates to the subject of this paper and why it only appears in the conclusions.

11) Page 10, lines 26-27: While the correlation analyses and dispersion modeling

support the role of mineral dust especially in case of higher loading (e.g., a few to 10% of surface area represented is a good amount of mineral dust), this does not indicate at what level marine INPs become important for lower INP concentrations does it? In a number of cases, INPs were apparently below detection limits, possibly consistent with typical surface areas and marine INP n_s, as well as with limited sample volumes assessed (limited warm temperature assessment). And again, any time spent over land would be expected to influence INPs strongly, while time spent over what might be a more constant and low INP source like the ocean would not be expected to correlate with INP concentrations. Those numbers may be relatively uniform independent of time spent over the marine source, but instead more correlated with marine conditions that influence emission rates. The fractional marine influence does not seem fully evaluated, as it would have required assessing more cases at the lower end of the spectrum of measurements I suspect.

References

McCluskey, C. S., Ovadnevaite, J., Rinaldi, M., Atkinson, J., Belosi, F., Ceburnis, D., et al. (2018). Marine and terrestrial organic ice-nucleating particles in pristine marine to continentally influenced Northeast Atlantic air masses. Journal of Geophysical Research: Atmospheres, 123. https://doi.org/10.1029/2017JD028033

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