EGUsphere, referee comment RC1
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RC: Reviewer comments on egusphere-2022-1242

Nadine Borduas-Dedekind (Referee)

Referee comment on "HUB: a method to model and extract the distribution of ice nucleation temperatures from drop-freezing experiments" by Ingrid de Almeida Ribeiro et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-1242-RC1, 2023

General comments:

The authors present open access Python code to estimate the subpopulations of potential ice nucleating substances from data obtained by drop freezing assays. They present codes that have the potential to be quite important in further discussing the ice-nucleating ability of ambient samples from mineral dust to organic aerosols. I command the others for this important detailed work and for their clear writing. I'd like to raise a few discussion points and point out a few minor issues to be addressed prior to publication.

I'd first like to highlight what I thought were the most important contributions within this paper.

- Clearly articulated problem to be addressed when using frozen fraction data (for example lines 15-16, 57-58, 75-80)
- The dilution discussion (section 3.1) is particularly valuable, and the authors can make specific recommendations for the community to move forward in their data analysis.
- The use of the HUB-forward code to estimate the presence of subpopulations.

Here are my recommendations for improvement:

I struggled a little with the chosen terminology of the code. Why use the term “HUB”? What does the “underlying-based” mean in atmospheric science and/or in statistics? The forward/backward terminology was also not intuitive to me, and it’s not clear why these terms pointing to a direction were used. Could there be better terms to be used such as “subpopulation determination” for HUB-forward? For example, the term could focus on the outcome of the code?

I’d like to challenge an assumption made in the manuscript (for example on lines 138-139) about the role of dilutions. I think the presented data analysis method is best applied to ice nucleating substances that are intact. For example, mineral dust and P. Syringae proteins. However, there is literature on organic matter and dilution series where dilutions can potentially change the shape, form and composition of ice-nucleating sites. For example, (Bogler and Borduas-Dedekind, 2020) showed that dilutions of the macromolecule lignin influences the mass-normalized ice nucleating ability of the material. I would recommend that the authors expand on the idea that this dilution method is for intact ice-nucleating ability. Alternatively, the authors could also use the open access lignin data and see how their code performs (that would be cool actually!).

I also wonder about the choice of Gaussian distributions (Eq3) for the freezing temperatures of populations of IN. Why not log normal? Lines 123-124 mention that other types of normalized distributions could be used, so it would be important to justify this choice. From my own understanding, ambient samples/datasets are typically log normal. See also (Andersson, 2021).

The manuscript is well written and well-motivated. The flow could be improved with more subsections to be able to find the information rapidly for the future reader. For instance, after reading the paragraph at lines 178-186 – I would have been interested to see this code applied in the following section. There could also be a Method section for the details of the math and then a Results and Discussion section with subsections for categories related to recommendations like dilutions series, subpopulations, etc. Subsections within pages 9-10-11 would also help the flow.

There are additional references that I would encourage the authors to consider and I've added them throughout my specific comments below.

Specific comments:

Title: The title might be improved by specifying the types of ice nuclei as well as either defining HUB or removing the acronym.

Lines 32-36 has a rather random assortment of references of some drop freezing assays. I can refer the authors to a < 2021 comprehensive table of reported techniques: Table 1 in (Miller et al., 2021)

Line 36: I would also comment that many drop freezing techniques are also used for ambient measurements with unknown concentrations and unknown surface area like sea surface samples and ambient aerosols. How would the authors use their code on these types of samples?

Lines 42-46 discuss the role of cooling rate which is important in data evaluation. I would encourage the authors to comment and reference (Wright et al., 2013). Also relevant to the discussion on lines 440-447.

Eq1b and differential freezing spectra have been discussed previously in (Creamean et al., 2019) and so this reference should be added and discussed.

Scheme 1: "I_u" is not defined. I also think this scheme could be improved by using graphics instead of terms. In other words, the authors could show a frozen fraction graph and show the type of graphs that may be generated based on their code. (especially since different research groups use different terms, a graphical visualization would be helpful – and could also serve as a TOC graphic)

Lines 102-109 could be omitted entirely as these sentences are redundant (more appropriate for a thesis rather than a manuscript)

The idea of Eq2 and the sum of all parts has been nicely discussed in (Steinke et al., 2020) and the authors should consider mentioning this work.

Figure 1 – PMF should also be defined in the text. It's also difficult to see the black line in figure 1. Perhaps making it bold would help?

Would it be worth relegating the tables to the SI? Some of the values could be added directly onto the graphs for instance.

Line 330-331 – there is much value in having code now to support this claim! Well done to the authors.

Lines 335-343 – excellent recommendations

Figure 6 – specify in the caption the difference between panels A, B, C and D.

Line 368-369: it would be worth describing how the choice of "2 subpopulations" was made. If I understood correctly, it was previously optimized? Or are the authors sourcing this information another way? It would be worth clarifying.

Figure 8 – there's an error on the panel labels in the caption. ABC should be ACD.

Line 386-387 – why were some points omitted from the optimization procedure?

Figure 9 Panel A is arguably an important graph and would benefit from being highlighted separately (perhaps moving the other panels to the SI?).

References:

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Steinke, I., Hiranuma, N., Funk, R., Höhler, K., Tüllmann, N., Umo, N. S., Weidler, P. G., Möhler, O., and Leisner, T.: Complex plant-derived organic aerosol as ice-nucleating particles – more than the sums of their parts?, Atmospheric Chemistry and Physics, 20, 11387–11397, https://doi.org/10.5194/acp-20-11387-2020, 2020.

Wright, T. P., Petters, M. D., Hader, J. D., Morton, T., and Holder, A. L.: Minimal cooling rate dependence of ice nuclei activity in the immersion mode, Journal of Geophysical Research: Atmospheres, 118, 10,535-10,543, https://doi.org/10.1002/jgrd.50810, 2013.