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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Arctic marine ice nucleating aerosol: a laboratory study of microlayer samples and algal cultures" *by* Luisa Ickes et al.

Anonymous Referee #2

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In their manuscript titled "Arctic marine ice nucleating aerosol: a laboratory study of microlayer samples and algal cultures", Luisa et al. describe findings from a series of ice nucleation measurements performed on sea surface microlayer (SML) samples collected from previous Arctic field campaigns and two culture phytoplankton species. This research topic is of current interest for the aerosol-cloud interaction community, particularly for remote regions and high latitudes. The introduction motivates the study and the descriptions of the approach and methods used in this study are detailed and well written, which is greatly appreciate. I have only one major concern, which relates to Section 3.3 (see general comment #5) and some specific minor comments. Overall, the manuscript is well written and these results do advance current knowledge related

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Discussion paper

to ice nucleating material in the marine environment. I recommend this manuscript for publication once these comments have been adequately addressed.

General Comments:

1. The title of the manuscript is pretty misleading – there are no measurements of Arctic marine ice nucleating particles, meaning these measurements were not made for aerosol collected in Arctic. I understand that the results may potentially have implications for the Arctic, but I recommend the authors consider changing the title to be more transparent about what this study entails.

2. Throughout the manuscript, it is difficult to know exactly what type of sample is being discussed: a bulk SML/culture sample, an nebulized aerosol sample, or a AEGOR aerosol sample. For example, it is not clear if Section 3.1 includes any measurements of aerosol samples or if it is strictly SML/culture samples. The section title is not very specific and Figure 1 has arrows pointed toward the "Droplet freezing experiments" picture from both the bulk sample and from the AEGOR, but I do not think there are measurements of AEGOR aerosol with the NIPI technique.

3. There are many instances where discussion on the interface between bulk seawater, SML, and aerosol is relevant for understanding the findings. The size-dependent aerosol composition is also relevant for interpreting the results from the two aerosol methods. I encourage the authors to consider a paragraph in the introduction that includes some of the literature on this topic and why studies such as this one are useful to address this knowledge gap in the context of ice nucleation research.

4. In calculating the ice nucleation site density, it's important to be clear and specific as to how the nebulizer will bias the ice nucleation site densities to higher values. Specifically, that the narrow size distribution with small particles that are likely more enriched in organic material compared to larger particles sizes will bias estimated ice nucleation site densities to higher values compared to natural aerosol and the AEGOR emissions.

5. My understanding is that Section 3.3. "Combined temperature regime – full ice nucleation spectra" aims to quantify the full ice nucleation temperature spectra from the different instruments, which includes measurements of both aerosol and bulk water/SML samples. To do so, the authors have estimated ice nucleation active site densities per mass of sea salt. I absolutely understand the experimental limitations that motivate this and I also think the authors include a thorough explanation as to why this approach may not be appropriate, which is very appreciated. This approach assumes that the ratio of the ice nucleation material to salt is equivalent in the bulk SML/culture samples and the aerosol. As the authors are aware, the transfer of organic material (likely responsible for the ice nucleation behavior) between the bulk water, SML, and aerosol phases is complex and varies depending on the solubility and surface active properties of the ice nucleation material. As such, I think presenting this analysis as "combined full ice nucleation spectra" is highly misleading to readers who may try to do the same in their own experiment without the careful consideration of discrepancies between bulk and aerosol composition or who may try to reuse the data. I do think these results are interesting and address the puzzling process of evaluating ice nucleation associated with the marine system (bulk, SML, and aerosol). Instead of presenting Section 3.3 as "full ice nucleation spectra", I suggest the authors to consider reframing these results as an approach for investigating the transfer of ice nucleation material from the bulk phase to the aerosol phase. Most of this discussion is already included, so it would require renaming the section and changing the order of text and therefore isn't really a major change.

Specific Minor Comments:

L15 – "we applied several aerosolisation techniques" – should this say "two aerosolisation techniques"?

L41 – the references listed for sea spray aerosol as an important INP source in remote regions includes only numerical modeling studies. This should be specified as such. There are also additional observational studies in remote regions that are cited in the

manuscript elsewhere and would also support this statement.

L57 –Why are these specific temperatures listed in reference to the DeMott et al. (2016) study? DeMott et al. (2016) evaluated INPs at a range of temperatures for their study -15 to -34 deg C for laboratory studies; -6 to -27 for ambient aerosol measurements (see Figure 1 from that manuscript).

L75 – ". . . suggested that absolute cell concentrations. . ." – is this referring to cell concentrations in air or in seawater?

L87 – "the ice nucleating potential of the aerosolised organic matter has not been examined in detail" – Do you specifically mean marine organic matter in the Arctic? Please be specific, as previous studies have investigated INPs associated with marine organic material and organic material in other settings.

Table 1 – Is this table necessary? Only a couple studies are mentioned in the introduction and several studies are missing if this is intended to be a full summary of marine INP studies. If you really want to include a table like this, I suggest including only the studies relevant for the Arctic region or laboratory studies since those are the focus of this paper. If it is decided that the authors want to include a table of all studies that have targeted ice nucleation observations of marine aerosol/SML/seawater, please take some time to be inclusive to all marine INP studies.

L112 – what is meant by "ex situ" ?

Table 3 – Are these for the bulk water samples or aerosol samples?

Table 3 - "we give in brackets how many mL of sample" – I think this should say parentheses, not brackets

L162 – Here, the order of the text suggests that these subsamples were with the ACCACIA campaign, but the table lists them as ASCOS. Is the sentence stating "The surface microlayer water was collected from . . .." out of order?

L178 – What does STN mean?

L190 – throughout the methods section, when referring to “samples”, the authors should be clear if they are talking about the bulk samples/mixtures or aerosol samples. Here, I think “the cell concentrations of algae in the experiment” is referring to bulk samples/mixtures (not aerosol), but this is not clear here nor in the Table 3 description (see general comment 2).

L200 – Why were the tank water temperatures changed for the different experiments? Was this intentional? Studies have demonstrated that aerosol production is sensitive to temperature (e.g., Zábori et al., 2012), so curious if there was a reason and if the authors can elaborate on this detail.

Zábori, J., MatisÄĄns, M., Krejci, R., Nilsson, E. D., & Ström, J. (2012). Artificial primary marine aerosol production: a laboratory study with varying water temperature, salinity, and succinic acid concentration. Atmospheric Chemistry and Physics, 12(22), 10709–10724. https://doi.org/10.5194/acp-12-10709-2012

L210 – This last statement suggests that you can account for differences between the two aerosol generation techniques just by applying a dilution factor. However, an important difference between the nebulizer and the plunging jet is the size distribution, which is shown in Figure 2, and the corresponding organic composition of the generated aerosol because of the size-dependent composition of nascent sea spray aerosol. This was demonstrated in numerous studies, such as O’Dowd et al., 2004 (field evidence) and Prather et al., 2013 (laboratory evidence).

O’Dowd, C. D., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., et al. (2004). Biogenically driven organic contribution to marine aerosol. Nature, 431(7009), 676–680. https://doi.org/10.1038/nature02959

Prather, K. A., Bertram, T. H., Grassian, V. H., Deane, G. B., Stokes, M. D., DeMott, P. J., et al. (2013). Bringing the ocean into the laboratory to probe the chemical complex-

ity of sea spray aerosol. Proceedings of the National Academy of Sciences, 110(19), 7550–7555. https://doi.org/10.1073/pnas.1300262110

L234 – This is a large range of variability (180 to 900 nm) that spans an important size range for sea spray aerosol composition (see referenced in previous comment). Could the median diameters and widths be included in Table 2 to aid in interpreting the figures and data that follow?

L304 – Are all sizes of particles transmitted to the to the INKA instrument?

L321 – "sample under investigation" – are these samples bulk samples or aerosol collected onto filters? Based on Figure 1, it looks like the cold stage technique is applied to the bulk sample and the AEGOR samples, but there are no details describing how the aerosol are collected from the AEGOR and then analyzed with the NIPI method.

Figure 3b – it is very difficult to see the difference between the SM100C, SM100d, and SM100d nebulized marker colors. Also, is SM100b missing?

Figure 3 – Is the artificial seawater missing from this? It is listed in Table 2 as having been analyzed with the NIPI method.

L411 – Another possible important difference in the ASCOS high mol. w. sample is the aerosol sizes, which were mentioned previously as the smallest sizes observed from the nebulizer method. The surface area normalizes the data, but it should also be mentioned that the composition (i.e., possible ice nucleating material) is strongly size dependent for sea spray aerosol. Thus, the ASCOS high mol. w. aerosol sample may include smaller particles with greater organic mass fractions compared to the other aerosol samples, which further supports your finding (see general comment 4).

Figure 5 – Are all of these data for aerosol generated from the nebulizer or the AEGOR? There is a triangle in the legend, but I only see one set of data points plotted with triangles and Table2 includes 5 samples and the artificial seawater that were aerosolized with the AEGOR.

Figure 5 - By including the full range from the DeMott study (including uncertainties), the impression provided by this figure is that ns for marine aerosol spans 3 orders of magnitude and dessert dust is perfectly known. I think this is a bit misleading and I suggest the authors may want to consider using the marine INP ns parameterization from McCluskey et al. (2018) or the parameterizations used in Huang et al., 2018.

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(11), 6196–6212. https://doi.org/10.1029/2017JD028033

Huang, W. T. K., Ickes, L., Tegen, I., Rinaldi, M., Ceburnis, D., & Lohmann, U. (2018). Global relevance of marine organic aerosol as ice nucleating particles. Atmospheric Chemistry and Physics, 18(15), 11423–11445. https://doi.org/10.5194/acp-18-11423-2018

L458 – The comparison between AIDA and IKNA data is interesting. I have no issues with what is included in this discussion, but wonder if the authors could comment on additional impacts associated specifically with the unique particle composition. That is, once the solution droplet/particle effloresces and re-deliquesces, will it have the same ice nucleation activity as the particle that enters the AIDA chamber? Additionally, what do these results suggest for naturally occurring aerosol-cloud interactions and which (AIDA or INKA) is more representative of natural sea spray aerosol production, transport, activation and nucleation?

Section 3.3 – See General Comment 5.

L552 – what is “mist”?

L574 – I do not think “direct comparison” is an appropriate description for the analysis completed here because the ice nucleation ability of aerosol and bulk samples are not directly comparable. I suggest that the authors change this language to “We have

normalized all of the measurements by the salt mass present in the bulk and aerosol samples to investigate the ability of ice nucleating material to transfer to the aerosol phase" or similar (see General Comment 5).

L611 – "We also tentatively show that nebulisation enhances the ice nucleating ability of some cell cultures. We suggest that the aerosolisation process might rupture individual cells allowing ice nucleating macro-molecules to be dispersed through the aerosol population" – Please specify that this result only has implications for laboratory studies, not reality, because the nebulizer is not a naturally occurring phenomena at the ocean surface.

L623 –This is a great discussion. While it should not and is not be expected that the authors know every paper on this topic, I do want to point out two others that are extremtely relevant to this discussion: McCluskey et al., 2018 identified two marine INP types during mesocosm experiments and also include a discussion on the timing/conditions of their emissions and a very recent paper by Wilbourn et al., 2020 describe additional phytoplankton species and may be interesting to include.

McCluskey, C. S., Hill, T. C. J., Sultana, C. M., Laskina, O., Trueblood, J., Santander, M. V., Beall, C. M., Michaud, J. M., Kreidenweis, S. M., Prather, K. A., Grassian, V., and DeMott, P. J.: A Mesocosm Double Feature: Insights into the Chemical Makeup of Marine Ice Nucleating Particles, J. Atmos. Sci., 75, 2405-2423, 10.1175/JAS-D-17-0155.1, 2018

Wilbourn, E. K., Thornton, D. C. O., Ott, C., Graff, J., Quinn, P. K., Bates, T. S., et al. (2020). Ice Nucleation by Marine Aerosols Over the North Atlantic Ocean in Late Spring. Journal of Geophysical Research: Atmospheres, 125(4). https://doi.org/10.1029/2019JD030913

L629 – The heat tests would be interesting to see, especially for inferring ice nucleation material type/properties and since it is later mentioned that "The fact that marine INP are very small and heat sensitive" – is this a fact based on the data (not shown) or a

hypothesis based on previous measurements?

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