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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 #1

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Review of "Arctic marine ice nucleating aerosol: a laboratory study of microlayer samples and algal cultures" by Ickes et al., submitted to ACPD

The manuscript is a very well written and quite exhaustive study about the ice nucleation ability of different samples related to Arctic sea water (algal cultures and sea surface microlayer samples), examined with different measurement approaches. Measurement approaches include the examination of liquid samples on a cold-stage, and measuring aerosolized samples with the expansion chamber AIDA and the in-situ instrument INKA (a continuous flow diffusion chamber). For aerosol generation, two different methods were deployed, aerosolization and a plunging jet sea spray chamber.

Measurements were all done with great care and with up to date knowledge. The work describes and summarizes all measurements well, observed results are interpreted broadly and with great care. I particularly liked section 3.3, in which all different methods are compared, based on a normalization wrt. the salt content of the samples (and again with sufficient care, in that caveats of using this normalization are mentioned as well).

Having said all these positive things, I did miss some final information about what was learned for the atmosphere from this study. It is clear that the contribution of "real world sea spray" to atmospheric aerosol and particularly to the INP fraction is a different topic. But the authors decided to submit their work to ACPD, and in that context it will benefit from some summarizing remarks about the applicability of results from this study for atmospheric implications.

Additionally, although I have no big concerns about this work, there is a rather longish number of remarks I give below. They are all not essential, but should be dealt with to make this already good work such that it then can be published in ACP.

Specific remarks:

line 37: "The types of aerosol particles that constitute good INP are uncertain (DeMott et al., 2010)." As the field of atmospheric ice nucleation research had a VERY strong revival in the last decade, this sentence is a) not really correct, and b) the citation is quite old. Since then, there were several review papers, and there is already an older one (Szyrmer & Zawadzki, 1997; Hoose & Möhler, 2012; Murray et al., 2012; Kanji et al., 2017). I don't want you to cite all of them, it's just to show you why I think this sentence needs revision (or deletion).

line 42: "sea spray aerosol could be an important source of INP" – again, it can't be expected that you give a complete review here, but as this is a focus of your work, I wanted to point out these papers on the topic: McCluskey et al., (2018a, b) and Creamean et al. (2019).

line 48: “three main groups” - There is actually a quite new paper in which SML and airborne concentrations were connected (including cloud water): Gong et al. (2020). For air masses that were continentally and/or mineral dust influenced (Cape Verde), INP concentrations in the SML did not explain atmospheric INP concentrations - that does not say anything about the remote oceans, but is a piece in the puzzle, nevertheless, which is worth mentioning, as this helps closing a gap between ocean and atmosphere.

lines 84/85 and line 95-97: You write: “Another goal of this study was to improve our understanding of whether Arctic marine regions may have local sources of marine INPs.” and “Through comparison of the ice nucleation activity of artificial seawater containing Melosira arctica with that of the SML samples we aim to shed light on how representative relevant algal cultures are for Arctic marine INP.” - Comments on that in the summary (that I proposed above) would be highly welcome, although it is clear that it is not straightforward to draw conclusions on atmospheric concentrations of sea spray aerosol from artificial lab work. But there are results you can summarize!

Table 1: As you try to give an overview here, including the papers I referred to above makes sense (McCluskey et al., 2018a,b; Creamean et al., 2019; Gong et al., 2020). And there is one more for coastal Mexico by Ladino et al., (2019) which may fit.

line 323: “sample is well mixed, so that particles are distributed uniformly, and each droplet is representative.” Actually, if the INP concentrations are so high that this is true, then each well freezes at the same time, yielding a super-steep freezing curve. When the sample is then diluted or is already more diluted to begin with (so that conditions are those for which measurements typically are made), a less steep increase of FF with decreasing temperature is observed. But then, strictly speaking, INP are Poisson-distributed and this here does not apply any more. Therefore, you could say "... so that particles are distributed randomly."

lines 385-389: Due to the dilution that was done, comparing FF does not make sense,

and the figure could have only been shown for INP concentrations (normalized) – at least a FF figure cannot be interpreted in this way. Please revise.

lines 408/409: This sentence here gives a wrong impression. It becomes clear in the next chapter (3.3), that there really is a lower variability for the AIDA data at the lower temperatures. But before normalizing data from the different instruments (as you do, based on the sea salt concentration below), you should refrain from any kind of comparison and discussion thereof.

lines 441-442: This conclusion confuses me a little. You used the dry(!) particle number size distribution to derive the surface area for both particle generation methods when you normalized. This basically means that the contribution of different aerosol particle types (such as particles consisting purely of salt, or of having a mix between salt and organics, ...) to the overall aerosol was similar, independent of the particle generation. And therefore, this conclusion does not hold.

line 449: And of course, for atmospheric relevancy, also the abundance of these different particle types has to be accounted for. Mentioning this here would be good.

lines 535-536: “the diluted sample having higher nm values compared to the undiluted sample in the same temperature regime.” This may be an indication that the background was hit, i.e., one already measures background, dilutes more and again only measured background, but normalizes to a higher dilution. Check if this is the case here, and if yes, omit the data.

lines 582-583: To be able to draw this conclusion, results would have to be normalized to "atmospheric algal content", which wasn’t done and (as you argue above) is difficult, even amongst the NIPI data. This should be mentioned.

Technical comments:

The word “freezing depression” is used consistently. “freezing point depression” is the more correct term, right? Same for “aerosolisation” -> “aerosolization”?

Figure 1: When initially looking at this figure, I wondered about the meaning of the arrow on the left (in the middle), going from the plunging jet tube to the droplet freezing experiment. It became clear later on. It's probably a matter of taste to leave it here or to delete it – I just wanted to point out my initial confusion.

lines 137-139: Doesn't the nutrient content determine the growth rate? - It puzzled me that you can somehow set things such that you get high growth with low nutrient content (middle case). Or is the growth determined separately, and you just already give these observations here? Then that should be made clear.

line 215ff: Upon reading this for the first time, I was confused about the influence this low temperature in AIDA during the preparation phase would have on the measurements. This is nicely explained later, and it would be good to point out that an explanation on the reason for choosing such a low temperature will follow.

line 247: "The smaller . . . vessel" could better be introduced here as "A smaller 3.7 m3-sized stainless steel vessel located in the vicinity of AIDA . . .".

line 306: "sample air flow is sheathed by dry particle free synthetic air" – that is only true initially - water vapor diffuses through the sheath air flows and the sample flow from one plate (the warmer) to the other. Consider reformulation.

line 371: Concerning a difference in ice nucleation temperature observed for different cooling rates, the papers you mention show that the influence of the cooling rate is rather small. Please add that explicitly! As you mention, this can rather not explain the differences you observe.

line 377: Concerning the loss of ice activity during storage, there is a paper on that for Snomax (Polen et al., 2016).

Figure 3: 1) The different shades of bluish green in b) are difficult to distinguish, engraved by the opacity changes. Maybe additionally also change the symbol styles between samples? Or use a broader range of colors? (Although it is nice how you use

consistent colors throughout the manuscript for the separate samples.)

Figure 3: 2) Also, check the legend in panel b): SM 100d appears twice, SM 100b not at all.

Figure 3: 3) "Two duplicate samples of SM100 (SM100a and SM100c)" - do you refer to the two bags in which the sample was delivered? In the text these are "a" and "b", while "c" is the one that was stored. Check and homogenize.

line 417: Delete "of" (at "contained of . . . gels") or replace "contained" by "consisted".

Fig. 5, 7, 8, and 9: Maybe use a separate legend explaining the different symbol types (AIDA, INKA, ...), as it was done for Fig. 6. - It's a bit confusing to see one entry in the legend saying "SMLx", and then another sample is "AIDA".

line 539: Add "is" between "this" and "related".

line 561: Shouldn't "since" rather be replaced by an "although"? If it's assumed that INP are small, and not intact cells, aerosolization might not do additional harm. In fact, I think this is why the data from AIDA and NIPI are not in completely different ball parks in the first place.

line 572: "two diatom species" does not sound right. This gives the impression that the diatoms themselves were aerosolized and examined, while you rather looked at the whole algal cultures.

lines 577-579: It might be better to split this in two sentences: "Our three main objectives were: first the comparison of the ice nucleating ability of two common phytoplankton species with Arctic microlayer samples, second examining the impact of the aerosolization technique on the results, and third deriving the sample variability over the entire mixed-phase cloud temperature range. Concerning these objectives, we can draw the following conclusions:"

line 581: "among" might be better than "within".

Fig. 7: Why is the freezing point depression not accounted for the AIDA data? In case this could not be made, it would make more sense to not do this correction at all.

line 584: Please exchange “triggered” by another term. The sentence is not clear to me, the way it is formulated now.

line 636: Above (lines 629-630), you mention “heat treatment test (not shown) . . . only a weak heat sensitivity”. Here you say they are heat sensitive!?!? Check / revise!

Literature:

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Murray, B. J., D. O’Sullivan, J. D. Atkinson, and M. E. Webb (2012), Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519-6554.

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