Anonymous Referee #1: Review of "Arctic marine ice nucleating aerosol: a laboratory study of microlayer samples and algal cultures" by Ickes et al., submitted to ACPD

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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).

We thank anonymous reviewer #1 for the positive review and the detailed comments on the manuscript. We have revised the manuscript accordingly (see track-changes in the manuscript). Our replies to your comments are given below in blue after the specific comment.

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.

In our study we analyse the ice nucleating potential of Arctic sea spray aerosol, which could have an influence on Arctic mixed-phase clouds and thus the climate. We will further emphasise the atmospheric relevance of our results in the abstract and conclusions of our paper by adding the following sentences: "In the Arctic, these INPs can influence water-ice partitioning in low-level clouds and thereby the cloud lifetime, with consequences for the surface energy budget, sea ice formation and melt, and climate." (abstract); "We discuss our results in the context of aerosol-cloud interactions in the Arctic with a focus on furthering our understanding of which INP types may be important in the Arctic atmosphere." (abstract) and "From our study it is difficult to answer the question whether Arctic regions may have local marine sources of INPs and how much they influence Arctic mixed-phase clouds. At temperature above 248 K the ice nucleation activity of the investigated samples was very diverse, with some samples reaching a quite high median freezing temperature of 262 K, thus potentially being able to trigger freezing in Arctic mixed-phase clouds. The measurements in the temperature regime below 248 K on the other hand did not show that the samples were particularly ice active, especially when compared to dust, despite the fact that the results show an upper limit for n_s. Both measurements differentiated in the way the samples were analysed (bulk vs. aerosol phase). This was most relevant for the cultured samples, giving some hint that aerosolisation of cell cultures may change the ice nucleation activity of these, a process that could be important in the environment as well." (conclusions).

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).

There is still a lot of uncertainty around which aerosol particles make good INP and why. We changed the sentence to: Despite increasing interest in INP (Szyrmer and Zawadzki, 1997; Hoose and Möhler, 2012; DeMott et al., 2010), it is still uncertain which types of aerosol particles that constitute good INP in the atmosphere (Kanji et al., 2017).

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).

We updated the references as recommended.

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.

Thanks for bringing our attention to this paper, we added it to Table 1 and also added it in the introduction.

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!

As stated above, we added a few more statements on that in the conclusions.

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.

We added the mentioned papers to the table (and a couple of other ones as well).

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."

We changed it to: "...sample is well mixed, so that particles are distributed randomly, and each droplet is representative of the sample as a whole, meaning each one has an approximately equal probability of containing an INP active at a given temperature."

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.

We removed the FF plot and changed the text accordingly.

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.

We agree that this comparison here is a bit far fetched since we compare the non-normalised results, which one probably should not do. We changed the sentence to: "The various SML samples show little variation at temperatures below 248 K when probed in the AIDA chamber, meaning that the SML samples all exhibited similar ice nucleation activity (n_s of 10^9 m^{-2} at temperatures between 240 - 244 K) and the individual $n_s(T)$-curves of the AIDA measurements form a rather compact block of data (Fig. 5)."

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.

The dry particle number size distribution consists of all particles (pure sea salt, mixtures of salt and organics, ...) and there is no discrimination based on particle composition. Thus we cannot derive *any information on composition from the size distributions. However, we can conclude from the measurements of the ice nucleation activity (AEGOR results vs. SML results) that the aerosol composition (organics/salt) has changed.*

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.

We added a sentence on the abundance of mineral dust vs. sea spray here: "In the (High) Arctic both transported dust and sea spray aerosol (transported or locally originated) can be present (see Willis et al. 2018 for a thorough review of literature). However, which source is dominant for ice nucleation might be locally very different. In regions dominated by sea spray aerosol the fraction of organic matter within the aerosol population is another uncertainty.".

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 back-ground 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.

We removed the sentence since it was misleading. We do not think it is the background influence in this case but rather an effect of run-to-run uncertainty. We have taken backgrounds into account when analysing 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.

We changed the sentence to: "As the investigated algae species show less ice activity in the temperature regime above 248 K compared to the natural field samples, we conclude that they, especially Melosira arctica*, cannot explain the freezing at the high temperatures.". And added: "A normalisation of the samples to the atmospheric algal content would be needed to quantify this observation.".*

Technical comments:

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

Thanks for pointing that out, we changed "freezing depression" consistently to "freezing point depression".

We use British English, where "aerosolisation" is the correct term.

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.

Thanks for pointing that out, we simplified the figure.

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.

Nutrients, but also their stoichiometric ratios, determine growth. In treatment 1, we had algae that were both dividing fast and had a large cell size. In treatment 2, the cell division rate was high, but the cell size was small (= phosphorus limitation). In treatment 3, both parameters were low (= nitrogen AND phosphorus limitation). We added this explanation to the manuscript as well.

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.

We changed the sentence to: "In the AIDA chamber, typically held at 250 K and a relative humidity of 78% during aerosol injection (see Sect. 2.4 for more details and an explanation on the low temperature), the aerosol particles were suspended as supercooled aqueous solution droplets.".

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

We changed this accordingly to: "A smaller 3.7 m^3-sized stainless steel vessel is located in the vicinity of AIDA. It is referred to as the APC (aerosol preparation and characterisation) chamber and can only be operated at ambient temperature.".

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.

This is true, the sample air flow is sheathed by prior dried synthetic air. We changed to wording to: “sample air flow is sheathed by particle free synthetic air (initially dry)”.

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.

We changed the sentence to: ”The temperature at which 50% of the droplets are frozen has been shown to decrease with increased cooling rate in Wright and Petters (2013); Herbert et al. (2014), also the dependence was shown to be rather small.”.

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

Thank you very much for this reference. We included it in the respective discussion.

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.)

We changed the blueish color of SM100c and hope it is better now (we did not want to add confusion by other symbols or change the consistency in between all plots).

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

It is correct that the SM100d appears twice: one is the pure/original sample, the other one is the nebulised sample as indicated in the legend and by different symbols. We added SM100b to the plot and added some text/discussion on this sample in section 3.1.

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.

SM100a and SM100c is coming from two different bags of the sample (bag 1: SM100a; bag 2: SM100b). SM100c is a subsample of SM100b (2. bag) after two months storage. To reduce the ambiguity we changed the text in the caption to: ”Two duplicate samples of SM100 (SM100a and SM100b) are reflecting the variability of the sample. SM100a and SM100b are from two bags collected from the same culture. SM100c is a sub-sample of SM100b after 2 months storage. SM100d, a sub-sample of SM100b, was in storage for 10 months, and was then nebulised and retested to determine the effect of the aerosolisation on the sample.”

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

Corrected.

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”.

Adapted.

line 539: Add “is” between “this” and “related”.

Corrected.

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.

”The whole section and sentence changed (due to other revisions) and now reads as: ”The aerosolisation technique might exert more of an influence on the cultured samples compared to the microlayer samples, where the INP are thought to be associated with submicron organic detritus, rather than intact cells.”.

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.

We changed the wording to: ”two diatom 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:"

Corrected.

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

Corrected.

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.

The freezing point depression in AIDA is negligible (< 0.1 K) because the seed aerosol particles are activated to > 10 μm sized cloud droplets with a remaining solute concentration of < 0.1 wt%, that is, a water activity close to 1 and has therefore not been accounted for.

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

The sentence was changed to: "This result indicates that the INPs active at the highest temperatures are not one of the two types of phytoplankton cells studied or their exudates.".

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!

Lines 629-630 refer to only one sample (SM100), which we did examine in terms of heat sensitivity. Line 636 refers to the general knowledge on marine INP - we added some citations now to emphasise this and rephrased it. We agree that this seems contradicting, but we did not want to derive too much from one single measurement. More measurements are needed for further conclusions.

Literature:

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