Anonymous Referee #2; Received and published: 29 May 2020

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 to ice nucleating material in the marine environment.

I recommend this manuscript for publication once these comments have been adequately addressed.

We thank anonymous reviewer #2 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.

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.

We changed the title to: "The ice nucleating activity of Arctic sea surface microlayer samples and marine algal cultures".

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.

Section 3.1 is only bulk SML/culture samples. We made this clearer in the manuscript by adding it to the section title. We also adapted Fig. 1 to reduce the confusion and adjusted all the labels or captions in Figure 3 and 4.

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.

Thanks for bringing this up. We added a paragraph in section 2.2 on what we expect from the two different aerosolisation methods and what we want to address with our study: "We expect that aerosolisation of the samples with the nebulizer results in an upper estimate of INP because the undiluted SML (or cultured) samples are aerosolised whereas AEGOR is aerosolising a dilution of the samples with artificial seawater, which could result in a lower estimate of INP. However, it is not only the dilution factor in the sea spray simulation chamber (see Table 3), which has to be accounted for. The aerosolisation process itself is different in AEGOR compared to the nebuliser. In the nebuliser the suspension is well mixed, while in AEGOR the aerosol particles are formed from an organic enriched surface microlayer at the top of the tank. That leads to different expectations depending on the sample type. For the SML samples we would not expect such a huge difference due to this aspect. Here, we aerosolise in one case the pure well mixed SML (nebuliser), while in the other case we aerosolise the SML that has formed in AEGOR, which should be similar to the original SML sample. For the cultured samples, however, we would expect a larger influence. In AEGOR the phytoplankton material is floating at the surface of the tank leading to organic enriched aerosol particles during the aerosolisation, while the nebuliser might produce less enriched aerosol particles due to the mixing of the sample. Note that this might depend on the algae culture as well. Another crucial aspect of the two different aerosolisation methods is the size distribution and the resulting chemical composition of the generated aerosol. It was demonstrated in the laboratory and as well measured in the field, that for sea spray aerosol the organic composition of the aerosol particles and the generated size distribution are related (O'Dowd, C. D. et al., 2004; Prather et al., 2013). One interesting aspect of our study is to see the influence of all the aspects mentioned above and to check if

the diluted samples aerosolised with AEGOR show a similar or a lower freezing signal compared to the aerosolised pure samples.".

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.

Both the nebulizer and AEGOR produce rather broad size distributions (see Fig. 2). There is therefore no reason to assume that the ice nucleation active site densities calculated from the nebulizer measurements are overestimated due to that. However, the nebulizer results are an upper estimate because the undiluted SML (or also cultured) samples are aerosolised whereas AEGOR is aerosolising a dilution of the samples with artificial seawater. One interesting aspect of our study was to check if the diluted samples aerosolised with AEGOR show a similar or a lower freezing signal compared to the aerosolised pure samples. We did measurements with both aerosolisation methods for five samples: SM100, SM10, MA100, SML5 and SML8. We did observe a similar freezing signal for SML5 and MA100 when aerosolised with AEGOR, despite the dilution. The measurements of SML8, SM100 and SM10 did not exhibit a detectable freezing signal above the background when aerosolised with AEGOR.
We added to section 3.2: "However, both the nebulizer and AEGOR are not producing very narrow size distributions (see Fig. 2).".
We also added some more discussion on the expected results from both aerosolisation techniques in section 2.2.

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.

We agree with the reviewer and shifted the focus of this section on the bulk- vs. aerosol measurements. We also changed the title accordingly. Both measurements methods are widely used and in our study we could investigate if both techniques complement each other or differ when normalised and brought into one context.

The new section reads now:

"**Combined bulk and aerosol phase measurements**

One of the central aims of this study was to analyse the ice nucleation behaviour of Arctic SML samples and two different algal cultures over the full temperature range relevant for freezing in mixed phase clouds. We also wanted to assess if the ice nucleation material is transferred from the bulk to the aerosol phase. The samples were measured with different instruments sensitive to different temperature regimes: AIDA and INKA below 248 K (aerosol phase) and µl-NIPI above 248 K (bulk). Here we attempt to directly compare the AIDA and µl-NIPI datasets. The INKA dataset is not included in the comparison since the AIDA dataset is more comprehensive and has a finer temperature resolution than the INKA data.
To enable comparison and answer the question if the ice nucleating material is transferred from the bulk to the aerosol phase, both datasets (AIDA and µl-NIPI) require normalisation so that the ice nucleation behaviour can be expressed with the same quantity as a function of temperature. We have chosen to normalise both sets of data to the mass of salt present in the solution droplets since this quantity can be estimated for both approaches. Thus, the ice nucleation behaviour is expressed as ice nucleation active site density per mass of salt (n_m; $[n_m] = g^{-1}$). It is more obvious how to treat and harmonise ice

nucleation data using materials like mineral dust which have a relatively well-defined surface area. The surface area of an aerosol dispersion can be used to derive n_s in much the same way as dust particles in bulk suspension. However, when the ice nucleating material in a sample is soluble or forms colloidal suspensions then it is less clear how to treat it. This is especially complex for the marine system, where the bulk sample can be very different from what is aerosolised into the atmosphere - one question that we want to investigate a bit further by comparing the AIDA and the µl-NIPI datasets. While we can, and have, derived n_s values for the AIDA and INKA data where the surface area is the surface area of the dry aerosol, we cannot do this for the bulk suspension measurements from the µl-NIPI instrument. Similarly, while we have a measure of organic mass for the bulk microlayer samples we do not have a measurement of the organic mass in the aerosol phase, hence we cannot normalise to organic mass. Solution volume cannot be used, since the volume of the solution of the aerosol changes as its concentration alters to come to equilibrium with the chamber conditions. Hence, we have chosen to normalise to the mass of salt, a quantity which can be readily estimated from both the bulk and aerosol experiments. When contrasting the resulting n_m values it should be borne in mind that the spread in activities is likely an indication of the range of concentrations of the ice active components as well as variability in the activity of those components. The objective of our work was to compare droplet freezing assay results with aerosolised measurements, rather than to derive a quantity which could be used to predict atmospheric INP. Ideally, we would quote active sites per unit mass of the nucleating component, but if the identity and mass of the nucleating component is unknown this is not possible (as in this case). However, this approach enables us to investigate if the bulk and the aerosolised samples behave similarly and if both ice nucleation techniques complement each other when normalised and brought into one context.

For the µl-NIPI data we derive the salt concentration for each sample in g/L using the measured water activity of the samples and the parameterisation linking the water activity and salt concentration of seawater presented by Tang et al. (1997). To calculate the ice nucleation active site density per mass of salt, the measured INP/L is simply divided by the salt concentration in g/L. For the samples where no water activity was measured as part of this study (see Table 3), the values from Wilson et al. (2015) (for the ACCAIA SML samples) or an average of all SML samples (for the NETCARE STN samples) was used. We added an additional uncertainty of 20% (arbitrary) to the error bars for the n_m values of the samples where the water activity was not directly measured. The ASCOS samples are not included in the unified dataset. Their water activity could not be directly measured because the remaining sample volume was too small. Furthermore, these samples were treated differently to the other microlayer samples so an average water activity might not be a good representation for theses samples.

For the AIDA data the measured FF was normalised with the measured mass concentration of dry particles (as obtained from the SMPS and APS measurements, see discussion in Sect. 2.2), instead of using the particles' surface area concentration for normalisation that yielded the n_s data shown in Figs. 5 and 6. The underlying assumption is that the dominating constituents in terms of mass is salt with a density of 2.017±0.006 g cm^{-3} [Sigma-Aldrich sea salt; Zieger et al. (2017)]. Considering the composition of marine aerosols as presented in Gantt and Meskhidze (2013) this assumption is fair for the typical sizes of aerosol particles aerosolised into AIDA.

How many INPs are transferred from the bulk to the aerosol phase?

The combined ice nucleation activity of the field samples is shown in Fig. 7. The combined temperature spectra for the ice nucleation activity of the algal samples is shown in Fig. 8 and Fig. 9; the samples were split in two figures for clarity.

We first turn to the comparison between the AIDA and µl-NIPI measurements for the algal and field samples focusing on the difference between aerosolised and bulk samples. A significant difference between the AIDA and µl-NIPI measurement is that one is derived from an aerosolised sample and one is derived directly from the pipetted culture medium. Comparison between µl-NIPI, AIDA and other instruments in a recent intercomparison was very good (DeMott et al., 2018). Inspection of the data in Fig. 7, Fig. 8 and Fig. 9 suggests that the data from the two techniques might be consistent, but n_m would have to be extremely steep at the intermediate temperatures. The discontinuity of the AIDA and the NIPI data, i.e. the shift of the AIDA data to higher n_m values might be related to a change of physical characteristics upon aerosolisation. Aerosolisation may alter the physical characteristics of the ice nucleating material compared to when it is in the culture medium through breaking up aggregates or disrupting cells. This was shown for Pseudomonas syringae *cells in the study of Alsved et al. (2018). Hence, it is feasible that the ice nucleation activities of the aerosolised samples in the AIDA experiments are higher than those in the µl-NIPI experiments. However, there is a recognisable difference between both types of samples. 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. For the SML samples, it is therefore reasonable to assume that the composition of the aerosolised solution droplets probed in the AIDA chamber is very similar to that of the corresponding bulk solutions used in the µl-NIPI measurements. Indeed, the n_m spectrum looks more uniform as compared to the algal cultures. Most samples feature a rather continuous slope in the temperature-dependent INP spectrum. One notable exception is the STN7 sample, which shows a pronounced, step wise change in the ice nucleation behaviour at about 263 K. For the algal cultures the assumption that the aerosolised and bulk samples are similar is not necessarily valid. In order to investigate if the process of nebulising influences the ice nucleating activity of cell suspensions, we nebulised a SM100 sample, collected the nebulised sample as a bulk liquid and retested its ice nucleating activity using the µl-NIPI. Nebulisation increased the activity of the sample (see Fig. 3). We suggest that this might be consistent with the break up or rupture of cells in the vigorous nebulisation process, which might then release macromolecular ice nucleating materials. Alternatively, there might be agglomerated cells or colloidal particles inside the sample. That means that ice active sites can be either inaccessible or simply concentrated in a few particles. These aggregates might remain relatively intact during pipetting, but may be disrupted on nebulisation. It would have the effect of dispersing the ice nucleating entities throughout the aqueous suspension, thus increasing the probability of freezing across the droplet distribution when nebulising the sample. However, nebulising MQ water (not shown) showed that some impurities can likely be introduced by the nebuliser itself. These hypotheses deserve further investigation in the future.

Further to this, we have the hypothesis that the aerosolised material entering AIDA was very different compared to the pure cultures. For example, first analysis of electron microscopic pictures of aerosol particles contained in AIDA (representative for particles aerosolised with a nebuliser into AIDA) during the experiments with Skeletonema marinoi *showed no cells or obvious cell fragments visible (see left picture of Fig. 10). This is consistent with the microlayer being dominantly composed of organic detritus and might be a result of biochemical processes within the microlayer. In contrast the right picture of Fig. 10, where SM100 droplets were pipetted directly from the solution, shows clearly cells, which are then also present in the droplets analysed with µl-NIPI. However, a more detailed analysis would be needed to give a final answer on the difference of the aerosol particles in AIDA compared with aerosol particles within pipetted droplets.*

Dilution tests bulk measurements

Figure 8 shows the $n_m(T)$ spectra for the SM100 culture and the variability including two SM100 samples (a and b for biological variability; c and d for storage effects) as discussed in section 3.1. The latter (Fig. 9) shows the spectra for MA100 and SM10. To bridge the gap in the ice nucleation spectra between the AIDA and the µl-NIPI data, we did additional dilution experiments with µl-NIPI to extend the temperature regime of the µl-NIPI data to lower temperature. Diluting the SM100 and MA100 sample has the effect of reducing the freezing temperature and increasing n_m. Thus the curves from the undiluted samples can be extended to lower temperatures. That works well for SM100 and partly also MA100. For MA100 the slope of the n_m curve continues to be steep throughout the dilutions. However, there are some points which may have been affected by the background signal, which are denoted by the larger lower error bar value. It is not clear why there is such a difference in the behaviour after dilution between the SM100 and MA100 samples, and further investigation into the differences in their composition and how this is related to their ice nucleating ability is necessary.

Temperature dependent difference in ice nucleation behaviour

As a striking result, there is much more variability in the ice nucleation activity of the samples when analysed with the µl-NIPI than with AIDA (approx. 15 K vs. 5 K). This larger variability in the high temperature range has been observed in other studies, too, e.g. for soil or agricultural dust (O'Sullivan et al., 2014; Schiebel, 2017; Suski et al., 2018). One explanation for this behaviour could be that there are multiple INP types in seawater, just like there are in terrestrial samples, leading to a high diversity of the INP spectra at high temperatures. At low temperature the ice nucleation activity is much less variable and low throughout all samples."

Specific Minor Comments:

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

Yes, corrected.

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.

We have added further literature to support our 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).

It refers to Fig. 2 in the DeMott et al. 2016 study, where the authors tried to relate the INP conc. at these temperatures to TOC or Chlorophyll a. However, we agree that this is confusing and removed the parantheses with the specific temperatures in the manuscript.

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

It is the cell concentrations of the phytoplankton - we added this in the manuscript. The sentence now reads: "It has been suggested that absolute cell concentrations of the phytoplankton are not the sole determining factor for aerosol flux and that aerosol size distribution can be affected by the growth conditions of the microorganisms.".

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.

Yes, that refers to the Arctic as this is the focus of our study. We have modified this sentence so it now reads: "..., the ice nucleating potential of the aerosolised organic matter has not been examined in detail for the Arctic region.".

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.

We did add further studies to this table to give a complete literature overview on marine INP studies. Note that we only mention articles that explicitly discuss marine sources of INP and focus on the mixed-phase cloud temperature regime.

L112 – what is meant by "ex situ" ?

It means that the aerosol particles have been sampled from the AIDA chamber and investigated with INKA, but not within the AIDA chamber using the expansion cooling experiment. To avoid confusion we removed the "ex situ".

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

These are for the bulk water samples - we specified it in the table caption, which now reads: "Characteristics of the bulk samples used during the study...".

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

Corrected.

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?

It is only the sampling catamaran which is the same as during the ACCACIA campaign. Otherwise it is clearly stated that this paragraph is about the ASCOS samples.

L178 – What does STN mean?

It stands for Station. This is not of relevance for the paper - we decided to keep the same labels as in the original papers but here they can just be seen as labels (the meaning does not matter).

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).

We adapted the text here: "The cell concentrations of algae in the AEGOR tank...".

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., MatisÄ Ans, M., Krejci, R., Nilsson, E. D., & Ström, J. (2012). Artificialprimary 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

We chose the temperatures to be the same as the temperature that was used for growing the cultures. For the SML samples we did choose a realistic temperature for the sea surface in the Arctic.

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

We agree that the sentence was a bit misleading formulated. We changed it to: "Given these differences, comparison of the ice activity of aerosol generated by these two techniques should enable us to determine whether INP material is preferentially aerosolised by bubble-bursting.". We also added an additional paragraph here emphasising more what the differences between the nebuliser and AEGOR are and what we expect for the different samples.

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?

We have added the median particle diameters as well as the width (geometric standard deviation) of the fitted size distribution for all the AIDA expansions to Table 2.

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

We assume that the particle loss in the sampling line is negligible, as shown in DeMott et al., The Fifth International Workshop on Ice Nucleation phase 2 (FIN-02): laboratory intercomparison of ice nucleation measurements, AMT, 11, 11, 6231–6257, 2018.

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.

We changed the sentence to: "To do so, the droplets of the sample under investigation (if not explicitly otherwise mentioned this is a bulk sample) are pipetted onto a silanised glass slide, which serves as a hydrophobic substrate.". We also added after the description of the different AEGOR samples for the µL-NIPI: "Note that all these samples are bulk samples." and changed the labels in Figure 4.

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

We adapted the color scheme since that was also criticised by reviewer #1. We also added sample SM100b and some discussion on this sample.

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

We added it to Figure 3.

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).

That could be an additional explanation for the difference in the AIDA measurements. We added: "The size distribution of the nebulised ASCOS high-molecular weight sample was the smallest compared to the other samples, which might have an influence on the ice nucleation activity as well since the

chemical composition of sea spray aerosol is highly size dependent. This sample might consist of smaller particles with a larger organic mass fraction compared to the other samples". However, this sample was independent of the size distribution more concentrated in organic mass due to the treatment. Note, that this sample was also the most effective in the μL-NIPI measurement.

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 Table 2 includes 5 samples and the artificial seawater that were aerosolized with the AEGOR.

There are five samples which were aerosolised with AEGOR (SM100, SM10, MA100, SML5, SML8) as mentioned in Table 2. Figure 5 shows two of these samples (SML5 and SML8), while Figure 6 shows the other three (SM100, SM10, MA100). For most of the samples the freezing signal from the AEGOR measurement was below the background, so that there are no datapoints to be shown. That is for SML8, SM100 and SM10, meaning that Figure 5 and 6 indeed each only includes one AEGOR measurement (SML5 in Figure 5 and MA100 in Figure 6). This is mentioned in the text at the beginning of section 3.2 ("With respect to the experiments where AEGOR was used for aerosol generation, some samples did not exhibit a detectable freezing signal above the background (SM100, SM10, and SML8) and are therefore not included.").

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

We added the McCluskey et al. (2018) parameterization to Figure 5.

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?

The question of whether efflorescence of the particles changed their INP activity is difficult to answer and we are not sure if the algae cultures are damaged while drying. This could be interesting to investigate in future. We added the following to the manuscript in section 3.2: "Note that efflorescence might as well change the INP activity of the aerosol particles.".
Regarding of the question of the comparability to naturally occurring aerosol-cloud interactions, the AIDA-experiments should be more comparable to natural processes.

Section 3.3 – See General Comment 5.

Adapted.

L552 – what is "mist"?

We replaced this with "nebulised sample".

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).

We changed it to "In order to compare the different approaches and the ability of the ice nucleating material to transfer to the aerosol phase we have normalised all of the measurements by the salt mass present in the bulk and aerosolised samples.".

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.

Changed to: "We suggest that the aerosolisation process using a nebuliser might rupture individual cells allowing ice nucleating macro-molecules to be dispersed through the aerosol population." We also added: "This might be unlikely to be relevant for environmental conditions.".

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

We agree and added this two references as well.

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?

It is based on literature, we changed the sentence (see answer to reviewer #1).

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