We thank referee 1 for reviewing our manuscript and for giving comments and suggestions. Our answers are given in blue, below, while the original text of the review was kept in black.

Referee #1 Evaluations: Scientific significance: Good Scientific quality: Excellent Presentation quality: Good

Major comments:

I appreciate how the authors introduced scientific questions that cover crucial gaps in Arctic INP research. However, they are indeed quite broad and cannot solely be answered by one study in one region during one time period. They are indeed a fantastic objective, but the authors should be clear that their results provide insight into these questions, i.e., specifically for the European Arctic in the summer. These findings are likely not consistent with other locations and certainly, not other seasons (e.g., the spring Arctic haze season when long-range transport is prominent or winter polar night where open water and biological productivity are at their minima).

We agree that a single, two months field campaign cannot answer all open questions regarding Arctic INP. We think that already the title puts into perspective that we show results for a specific season and region. In the revised version of the manuscript we added an additional sentence to highlight that our findings are constrained by season and region: "[...] Our findings are constrained to the season and region in in which measurements took place. They will nevertheless contribute to a better understanding concerning Arctic INPs and their potential effects on Arctic clouds.[...]" (L81 – L82)

The combination of LINA and INDA is very useful, however, it is not clear why: (1) both techniques are not presented per sample even though the methods imply sample prep for both was executed and (2) one technique was presented over the other in the figures instead of both. For example, are Fig 2 and 8 INDA or LINA data? Why were INDA data not shown for Fig 3 and LINA data not shown for Fig 5? Perhaps this issue would be resolved if the authors articulated which offline technique was conducted for which samples and why.

Which instrument was used was constrained by instrument availability. While the filter samples were scheduled to be analyzed, INDA was not continuously available, hence a full dataset for these exists only from LINA. For the sea water samples LINA and INDA measurements are available, and are now also shown in Fig, 5. In general, INDA measurements are more sensitive to low INP concentrations at warm temperatures that are relevant for the biological INP and also provide better signal to background ratio (sample in comparison to pure MilliQ water). Hence, data measured with INDA was preferred over LINA were possible.

In the revised version of the manuscript, we now indicate which instrument was used in the figures showing freezing spectra (Fig. 2, 3, 5, 6, 7, 8, 10 and 11).

Were the blank spectra subtracted from the filter sample data? The only place I see a blank spectrum is in Fig 11a, but is this one filter or an average of all the blanks collected as indicated in the methods? And is this blank shown from the filter in ultrapure water or ultrapure water alone? Because the background for INPs can widely vary for the type of ultrapure water used, a bit more detail on the source of the ultrapure water used should be provided (e.g., Milli-Q, DI, etc.). Blanks for both plain ultrapure water and the blank filters in ultrapure water should be shown to demonstrate the reliability of the ultrapure water and filters used. Additionally, DI blanks can be all over the board depending on the container used for sampling things like seawater. Was a blank conducted for putting ultrapure water in similar sampling containers as the SML, BSW, and fog water were collected

in? If so, these blanks should probably be shown in the SML, BSW, and fog water spectra as well (at least, the first occurrence of each).

We did not subtract blanks from real samples. As ultrapure water we used Milli-Q. This information is added in the revised manuscript ("[...] ultrapure water (Type 1; Direct-Q3 Water Purification System, Merck Millipore, Darmstadt, Germany) [...]"; L154-L155). The blank in figure 11 is the average of the field blank filters taken during the campaign. That information was added to the capture of Fig 11: "The black dots depict the mean freezing spectrum of the field blanks scaled to atmospheric concentrations with the mean sampled air volume of the 8h filter samples."

Prior to each measurement day, we typically perform a test of the Milli-Q water that is used on that day, in order to notice possible problems. For the data shown it can be excluded that contamination of the Milli-Q water (above the "usual level") is an issue. Also contamination of the containers that were used can be excluded: In the field, SML, BSW and fog water samples were collected into Nalgene bottles pre-cleaned with HCl solution (10% v/v) and then divided into two aliquots for the saccharide analysis of Zeppenfeld et al. (2019) and the INP analysis of this manuscript. The aliquot for the INP analysis was stored in a sterile centrifuge tube (50 mL, Cellstar, Greiner Bio-One, Kremsmünster, Austria), which is the same kind of container in which the washing of the filters was performed. These centrifuge tubes are used for all INP measurements of filter samples in our laboratory and so far contamination by the container was not observed. Therefore, we are confident that our typical background always is valid. Where applicable freezing spectra of pure MilliQ water and field blanks are added to figures (Fig. 3, 5, 8).

Frankly, it is a bit difficult to discern the generalized difference in the offline INP spectra from icefree ocean, ice pack, and the MIZ for the aerosol (e.g., in Fig 3, perhaps this is somewhat clear > -15C, but not the bulk of the spectra) and those + melt ponds for the SML and BSW (e.g., Fig 5 has highest spectra from the ice pack and MIZ for SML; and ice-free ocean, melt pond, and ice pack for BSW, depending on the temperature). To support the authors' claims regarding which regions have the highest INPs, perhaps a figure summarizing the data would be useful. For example, a figure showing: (1) aerosol, (2) SML, and (3) BSW INP concentrations at select temperatures (e.g., =10, -15, -20, etc.). Then, this would clearly demonstrate which indeed had higher concentrations and at which freezing temperatures.

We are not sure if we understand the referee's suggestion for a figure correctly, but in order to improve the discernibility we changed/added two things in the revised manuscript:

1. we adjusted the color-coded background in Fig. 2, the time series of  $N_{INP}$ , to match the categorization of the samples.

2. we created a box plot of the data shown in Fig. 3 for selected temperatures (-10°C, -15°C, -20°C and -25°C). The box plot is shown below.



Regarding the boxplot, the way we treated values outside the detectable range has to be kept in mind: In offline INP analysis, N<sub>INP</sub> cannot be derived for  $f_{ice} = 0$  and  $f_{ice} = 1$ , i.e., when no or all droplets of an experiment are frozen respectively. Therfore, the lower and upper limit of detection (LOD) is given by N<sub>INP</sub>( $f_{ice} = 1/n_{total}$ ) and N<sub>INP</sub>( $f_{ice} = (n_{total} - 1)/n_{total}$ ) respectively (with  $n_{total} =$  the number of droplets used in the experiment). How the LOD is treated, when calculating statistics such as mean, standard deviation, etc. directly influences if the used statistic is over- or underestimated. For example: If the lower LOD is (i) treated as the absolute value, the mean would be overestimated; (ii) treated as zero, the mean would be underestimated; (iii) excluded from the data set, the mean would be overestimated; (iv) substituting the LOD with a value between zero and the LOD, may lead to over- or underestimation of the mean. To our knowledge, for offline INP measurements no established way for the treatment of the LOD in such cases exist. Hence, we adapt the way of treatment which is widely used in environmental chemistry (U.S. EPA, 2020) and substitute the lower LOD with LOD/2 and the upper LOD with LOD\*2 in order to calculate the statistics which are also shown in the box plot. In the same manner box plots for the SML and BSW data were created:



We also want to emphasize the point that we did not claim in the manuscript, that one of the environmental settings has generally higher INP concentrations, but rather that the most ice active samples (at warmer temperatures) are more often associated with the MIZ than the icepack (for filter and SML samples). Hence, in terms of the boxplot, not only the box itself, which represents the quartiles, but also the fliers have to be considered.

The box plots shown above were added to the SI with the following description: "Fig. S10 shows a box plot of the filter derived N<sub>INP</sub> shown in Fig. 3 of the main manuscript. The horizontal line represents the median and the green triangle the mean. The whiskers have the length of  $1.5 \times IQR$  and data points outside the range of the whiskers is shown with diamond markers. It should be noted that for samples whose value was outside the detectable range at the selected temperature, the value was substituted in order to minimize the over- and underestimation of the summary statistics needed to create the box plot. If at the selected temperature a sample had an N<sub>INP</sub> value below the detectable range, the value was substituted as LLOD/2 (LLOD = lower limit of detection). Analogous values above the detectable range were substituted with  $ULOD \approx 2$  (ULOD = upper limit of detection) Box plots for the SML and BSW samples were created in the same manner as described before (Fig. S11 and S12; same data as in Fig. 5 in the main manuscript)." (L41-48)

We refer to these figures in the main manuscript: "Additionally, Fig. S10 in the SI shows a box plot of the very same filter samples, in order to emphasize the general differences between the environments." (L284-285) and "Additionally, Fig. S11 and S12 in the SI show box plots of the very same SML and BSW samples in order to emphasize the general differences between the environments." (L332-333)

Where were the SML and BSW samples collected from in the ice pack, if not from melt ponds? Were these samples collected in leads I assume?

Yes, in the open leads within the ice pack. This is now specified in the manuscript: "[...] Seawater samples were taken from different environments: ice-free ocean, marginal ice zone (MIZ), open leads within the ice pack or from meltponds.[...] " (L126-L127)

The authors state several times that the heat tests suggest the presence of biogenic INPs, but this is generalized without any discussion on how this statement came to be.

The heat test is an established method to test for biogenic, proteinacaeous INP and used widely in the literature: Conen et al., 2011, 2012, 2017; Conen and Yakutin, 2018; Felgitsch et al., 2018; Hara et al., 2016a; Joly et al., 2014; Moffett et al., 2018; Hill et al., 2016; Huang et al., 2021; McCluskey et al., 2018; Kunert et al., 2019; Pouleur et al., 1992. The test is based on the heat sensitivity of biogenic, proteinacaeous INP which are denaturated at 95 °C, while most other INP are heat resistant. In the revised version the paragraph now reads as follows: "The test for heat-labile INPs (Fig. S6 and S7 in the SI) demonstrates that ice activity of the samples is reduced when heated for1 h at 95°C. Especially INPs that nucleated ice at temperatures above ca. -16°C are gone after the heating. This is widely seen as an indicator for the presence of biogenic, proteinaceous INPs as those become denatured during the heating, which reduces their ice activity (Conen et al., 2011, 2012, 2017; Conen and Yakutin, 2018; Felgitsch et al., 2018; Hara et al., 2016; Joly et al., 2014; Moffett et al., 2018; Hill et al., 2016; Huang et al., 2021; McCluskey et al., 2018; Kunert et al., 2019; Pouleur et al., 1992)." (L305-310)

In looking at the SI figure, indeed some spectra show a decrease in INPs after treatment, but it is difficult to tell if this is the case for all samples at all temperatures.

We agree. In L306 of the manuscript and L22-23 of the SI, we state that this is not the case for all temperatures. We observe the main decrease at temperatures above  $-16^{\circ}$ C (see Fig. S6 in the SI, showing that no heated sample nucleated ice above  $-16^{\circ}$ C). We argue that the main change is to be expected above  $-16^{\circ}$ C, as biological INP are typically ice active at these higher temperatures. At lower temperatures only a minor decrease occurs. Also in response to the next referee comment below, we created a figure, that shows better how much N<sub>INP</sub> was decreased after heating at different temperatures (see our response to the next comment below for details).

In the results and discussion when first mentioned, the authors should elaborate briefly, meaning providing a more quantitative assessment of the decrease (how much of a decrease exactly?), if this was consistent for all samples and temperatures (was it?), and why or why not. For the figure below, we quantified the effect of the of the heat treatment as a decrease in N<sub>INP</sub> in percent: If N<sub>INP</sub> of the unheated sample at the selected temperature would be 100 L<sup>-1</sup> and if after the heat treatment N<sub>INP</sub> would have been 20 L<sup>-1</sup>, this would be counted as an 80% decrease. Therefore, samples that showed some freezing at the selected temperature before the heat treatment, but no freezing after the heat treatment, are counted as a 100% decrease in N<sub>INP</sub>. This decrease is shown as a boxplot for all samples at selected temperatures. It can be seen that the decrease is most pronounced for the warmer temperatures, but is present throughout the whole temperature range. For temperatures of  $-16^{\circ}$ C and above, the decrease almost always 100%, which indicates that all samples contained heatlabile INP at these temperatures. At temperatures of  $-18^{\circ}$ C and below, the variability in the decrease becomes higher, which indicates that at the lower temperatures some samples still contain mostly heatlabile INP, while in other samples also more heat-stable INP are present.

The figure of the decrease in INP was added to the SI, along with the description: "Fig. S7 shows a box plot of the decrease in N<sub>INP</sub> after the heat treatment at selected temperatures of all samples that were heated. Boxes represent the 25% and 75% quartile. The horizontal line represents the median and the green triangle the mean. The whiskers have the length of 1.5 \* IQR (interquartile range) and data points outside the range of the whiskers is shown with diamond markers. It can be seen that the decrease is most pronounced for the warmer temperatures, but is present throughout the whole temperature range. For temperatures of  $-16^{\circ}C$  and above, the decrease almost always 100%, which indicates that all samples contained heat-labile INP at these temperatures. At temperatures of  $-18^{\circ}C$  and below, the variability in the decrease becomes higher, which indicates that at the lower temperatures

some samples still contain mostly heat-labile INP, while in other samples also more heat-stable INP are present." (L24-31)



I assume this is because it would overcrowd the figures, but why are uncertainty bars not shown on any of the spectra, aside from the fog water derived INPs? Is there a way to possibly show uncertainty to corroborate the statements regarding differences between the different sample types? In the revised manuscript we added exemplary errorbars to data points in Fig. 3 and 5 to show the range of uncertainty without overcrowding the figures. We also added a paragraph on how the errorbars were derived: "The uncertainty in N<sub>INP</sub> was calculated with a formula by Agresti & Coull (1998). Agresti & Coull (1998) published an approximation for binomial sampling intervals, which was applied to N<sub>INP</sub> measurements by e.g., Gong et al., (2020), McCluskey et al, (2018) and Hill et al., (2016). Following their approach the confidence intervals for f<sub>ice</sub> are calculated by:

$$\left(f_{\rm ice} + \frac{z_{a/2}^2}{2n} \pm z_{a/2} \sqrt{[f_{\rm ice}(1 - f_{\rm ice}) + z_{a/2}^2/(4n)]/n}\right) / (1 + z_{a/2}^2/n),$$

where *n* is the droplet number, and  $z_{a/2}$  is the standard score at a confidence level a/2, which for a 95% confidence interval is 1.96." (L208-213)

The case study is indeed interesting but could be described in more detail beyond the one SML37 sample. For instance, what did the BSW spectra look like during this time period? If there was some level of blooming happening, that should be evident in the bulk seawater more so than the SML since phytoplankton reside in the upper trophic levels of the ocean. From the limited body of work on blooms and linkages to INPs, we would not expect chl-a to correlate directly with INPs (e.g., work by Creamean et al., Irish et al., McCluskey et al., and Zeppenfeld). It is more likely that INPs increase following a bloom due to enhancement in biogenic byproducts and bacterial growth following the

peak of the bloom, versus INPs originating from the phytoplankton, especially considering phytoplankton have been shown to serve as only moderately effective INPs at colder temperatures. We mentioned that chl-a does not necessarily have to be related to INP in the manuscript L507-511 (before revision L470-475), citing the work by Zeppenfeld et al. (2019). Typically, we observed either no enrichment or an enrichment from BSW towards SML. Therefore, we refrain from discussing the BSW samples in this context. The spectra of the BSW sample corresponding to SML37 can be seen in the figure associated with our next answer below (lower panel, orange dots). It can be seen, that the high ice activity during the case study period is constrained to the SML. The BSW sample is like the majority of the BSW samples rather unremarkable.

Looking at Fig 12, particularly at temps > -15C, it is interesting how LV195 had the highest warmtemperature "bump" of INPs followed by LV196 and LV197 towards the end of and following the spike in chl-a, while LV194 was also relatively high into the first half of the spike. It certainly would be interesting to highlight and discuss any SML samples following this period in addition to BSW samples before, during, and after, for comparison.

We agree that it would be interesting to investigate SML samples for the period following the case study, but unfortunately July 15 was the last day when SML samples could be collected and no following samples exist. However, for the sake of completeness, please find below a figure where we



highlighted the SML/BSW samples from the case study (orange), and the SML/BSW samples that were taken before the cases study (blue; collected ca. 24 hours prior to SM37/BSW37)

Also, it looks like some of the relatively warmer temperature SPIN data were high at the beginning of this event (-26 to -32C), but it is difficult to tell from Fig 2. To provide a nice, holistic wrap up by presenting all the data during the case study, showing and discussing the SPIN data would be useful as well.

SPIN measurements have been added to Fig.12. For convenience we converted the SPIN data to cm<sup>-3</sup>, which allows easy comparison with N<sub>CCN</sub> and N<sub>total</sub> which are also shown in Fig. 12. The SPIN data shows only little variability for a certain temperature and also no trend or correlation with one of the other parameters. The following was added to the manuscript: "Lastly, panel f) in Fig. 12 shows N<sub>INP</sub> measured with SPIN. Similar to the filter N<sub>INP</sub>, also the INP measurements with SPIN remain fairly constant during the period of the case study and no correlation with the other parameters shown in Fig. 12 can be seen." (L511-513)

Along these lines, the comparison of the slopes is a bit tenuous, given the slope changes could be cause by numerous factors including but not limited to sample-to-sample variability, influences from other airborne sources as indicated, etc. etc. In my opinion, the slope analysis does not add value to the results and are not as convincing as, say, the relationship between bloom time and INPs described in my previous comment. I suggest eliminating the slope discussion and focusing more on comparing the increases and decreases in the INP concentrations between the water and air in the context of the supporting measurements (chl-a, NC, NCCN, etc.). These are what provide empirical evidence of INP sources instead of arbitrarily defining slopes for only select spectra.

We agree that the slope is not an unambiguous indicator. However, from L490 to L496, we discuss the slope comparison thoroughly and point out the speculative nature of that comparison. The central findings regarding the supporting measurements (chl-a, NC, NCCN, etc.) and the INP concentrations is that the supporting measurements change, while the INP remain the same. In that sense we do not see the benefit of discussing these parameters in more detail.

The authors might consider reordering the sections in the results and discussion. As it stands, this section does not flow as well as it could, and it is not clear why the case study is section 4 when it is still a part of the results and discussion (section 3). This inherently causes some of the more important findings are buried. It makes more sense to order in the following as a combined section 3 of results and discussion (note that the titles do not need to be exactly these, but something along the lines). First, describe the sources: 3.1 Atmospheric INP sources over the entire campaign (this should include low temperature as a paragraph and not a subsection) and 3.2 INPs in the SML and BSW over the entire campaign. Next, describe linkages between the sources and airborne INPs: 3.3. connecting to sea spray. Then, possible effects on fog, which includes measured and derived: 3.4 Measured and derived fog INPs. Last, focusing on a unique case: 3.5 INPs from a phytoplankton bloom. Following the reviewer's suggestion, we restructured some parts of the manuscript.

The conclusion that dust represents the cold temperature INPs solely based on the transect in Fig 4 is very tenuous. Dust is also present at temperatures warmer than -32C, so why is this generalization based on only one temperature? How can the authors be sure what they measured were not algal or diatom material INPs, which do glaciate at the same range as dust?

The assumption that mineral dust dominates cold temperature INP was derived from its abundance and temperature range of activity, based on laboratory experiments. To our knowledge, no study has yet found biogenic INP to be present in such high concentrations as shown in our Fig. 4., the only INP known to occur in such high atmospheric concentrations that it could explain the observed INP concentrations, is mineral dust. Additional confirmation comes from the study by Welti et al. (2020) reporting that they only observed similarly high INP concentrations at -32°C, when they were in the outflow region of the Sahara, where the aerosol is known to be dominated by mineral dust.

However, the referee is of course correct with the comment that also silica-algae like diatoms might contribute to the INP population at the lower temperatures. Therefore we rephrased the paragraph on the cold temperature INP: We still present the argumentation for mineral dust, but also mention diatoms as a source for biogenic, mineral INP. We also now conclude the section with the more general term "mineral INP" rather mineral dust.

The whole paragraph now reads as follows: "In the previous section we described that at warmer temperatures for example at -10°C, samples with high INP concentrations are found more often in the MIZ and less frequently within the ice pack. In comparison, at the lower temperatures measured with SPIN (cross markers in Fig. 4) no correlation with the environmental setting is found. However, in global context the level of N<sub>INP</sub> at these low temperatures is remarkable by itself as shown in Fig. 4. That figure shows N<sub>INP</sub> in the Arctic at -32°C measured with SPIN during PS106, but also SPIN data by Welti et al. (2020) of a transect from Bremerhaven (Germany) to Cape Town (South Africa) along the western coast of Africa. It is striking that at these low temperatures N<sub>INP</sub> in the Arctic are in the same order of magnitude as in the outflow region of mineral dust from the Saharan desert. While we have no means of proofing the presence of mineral dust at these colder temperatures during PASCAL, to our knowledge there are also no other known sources of INP that can produce such high concentrations throughout whole time period of the campaign. Also, it was recently shown by Sanchez-Marroquin et al. (2020) that Iceland can be a strong Arctic dust source. Also Irish et al. (2019a) suggested that observed INP were mineral dust particles originating in the Arctic (Hudson Bay, eastern Greenland, northwest continental Canada), rather than particles originating from sea spray. And global model transport simulations done by Groot Zwaaftink et al. (2016) show that mineral dust is not only transported into the Arctic from remote regions but also, possibly increasingly, generated in the region itself. However, it is also possible also other sources of mineral INP contribute to the INP population at these temperatures. E.g., diatoms represent a biogenic, but mineral source of INP, as they have a cell wall made of silica (Xi et al., 2021) Therefore, it is likely that mineral INP, possibly mineral dust, contribute to N<sub>INP</sub> at low temperatures during the campaign." (L311-327)

What kind of drop in concentrations occurred in the heat treatments at these low temperatures? The LINA measurements do not extend to these low temperatures, therefore no additional conclusions can be drawn in this regard.

Can the authors link the air mass transport pathways to any major dust source like the Sahara? It does not look like it in Fig 13, especially considering how much time the air masses spent over water as compared to land.

Extending the back trajectories much further back in time is not meaningful in our view. Kahn (1993) showed that 5-day back trajectories have uncertainties of ca. 1000km. We would also expect dust sources in high latitudes to be more significant than the Saharan desert. It was recently shown by Sanchez-Marroquin et al. (2020) that Iceland can be a strong Arctic dust source. In addition, Irish et al. (2019) suggested that observed INP were mineral dust particles originating in the Arctic (Hudson Bay, eastern Greenland, northwest continental Canada), rather than particles originating from sea spray. Global transport model simulations done by Groot Zwaaftink et al. (2016) show that mineral dust is not only transported into the Arctic from distant regions but also, possibly increasingly, generated in the region itself.

The "dust extinction" (aerosol optical thickness at 550nm) provided by NASA's GEOS-5, shows little variation throughout the whole campaign and no transport from mid- and low latitude dust sources can be seen. For the period of the case study, only on the 14.7.2017 some dust appears to be transported from Novaya Zemlya to the general region north-east of Svalbard. Nevertheless, the dust extinction near the location of the vessel does not change significantly. In general, this is in alignment with our

observations shown in Fig. 4, where  $N_{INP}$  is relatively high, but does not vary extensively throughout the campaign.

Certainly, aerosolized redistributed dust from the ocean surface could have been a possible source, but there is no evidence of this specific to this study region. From the evidence provided, the authors should not rule out other possible INP sources at these low temperatures, especially without proof of air masses interacting with dust sources along their transect, which would entail a more detailed analysis that might include remote sensing data.

We agree that we do not have observational proof that mineral dust is certainly the INP at the low temperatures, but to our knowledge no other source could produce high enough concentrations of INP at these temperatures. It also must be kept in mind, that during most of the campaign, the ship was closer than 200km to Svalbard. It seems reasonable to assume that in such close proximity, Svalbard provides an ubiquitous background concentration of mineral dust. This aligns with what we described in the previous answer, where we mentioned that not much variation in the "dust extinction" can be found throughout the campaign.

## Minor comments:

The manuscript presents interesting new data on INP concentrations and collocated observations during late winter in the High Arctic. Material and methods were appropriate to produce robust results. Results are clearly presented and discussed. I enjoyed reading the paper.Lines 21-22: The statement on heat is redundant from earlier in the abstract; should remove.

Statement was removed.

Lines 111-112: How long were samples stored until analysis? The samples arrived in October 2017 at Tropos and where then analyzed over the course of ca <sup>3</sup>/<sub>4</sub> of a year.

Lines 229-234: Since the online measurements are a primary focus of the manuscript (i.e., not supporting), they should maintain their own section in the methods, perhaps before or after the offline INP measurement section.

The paragraph describing SPIN was extended. In the revised manuscript the paragraph now reads as :" In addition to the off-line INPs analysis of the filter samples, also the SPectrometer for Ice Nuclei (SPIN; Droplet Mea-surements Techniques, Boulder, CO, USA) was deployed to measure N<sub>INP</sub> in immersion mode on-line. SPIN is a continuous flow diffusion chamber (CFDC) with a parallel plate geometry and the measurement principle of SPIN in immersion mode can be briefly described as follows: aerosol particles are activated to cloud droplets and then exposed to conditions where ice can form. The number of formed ice crystals is then optically detected. SPIN is described in detail in Garimella et al. (2016). SPIN was placed within a measurement container. Together with the other aerosol instrumentation was the aerosol was fed to SPIN through one main inlet, but with additional subsequent drying of the aerosol. SPIN sampled in half-hourly intervals of constant temperature and relative humidity and each sampling condition was repeated three times within 24 h. The SPIN dataset is also part of the overview of global ship-borne INP measurements by Welti et al. (2020)" (L236-244)

Fig 2: Why are SPIN data missing from the very beginning of the campaign? If these data were not obtained or missing, that should be noted in the caption or figure itself. SPIN was not operational in the beginning of the campaign. We added to the caption of Fig. 2: "[...] Note that SPIN measurements were only obtained beginning with May 31." Lines 277-278: Which "others"? This statement is a bit vague. It is specified that with "others" we mean the spectra that go up to 1e3 m<sup>-3</sup>.

Lines 281-283: This is not clear given the 8-h and 2-h samples are not visually distinguished in Fig 3. Perhaps use of different markers for each would help?

Due to the large number of samples it is difficult to distinguish the spectra by different markers. Instead, we added a figure to the SI (Fig. S7) where the 2h samples are distinguished by color from the 8 h samples.

Fig 6: What does it mean by "stands out in terms of ice activity"? Was this qualitatively or quantitatively determined? Either way, there should be some description of how this was defined. This was a qualitative categorization that meant that the samples belong to one of the clusters described in the text. However, with the addition of the LINA measurements to Fig. 5 it was not possible anymore to indicate the individual clusters without overcrowding the figure. In the revised version we divide the area with the majority of very similar samples and those that we consider standing out by line

Fig 11: Clarify in the caption that the grey spectra are those from the entire study. Added to the caption.

## References

Conen, F., Eckhardt, S., Gundersen, H., Stohl, A., Yttri, K.E., 2017. Rainfall drives atmospheric ice-nucleating particles in the coastal climate of southern Norway. Atmos. Chem. Phys. 17, 11065–11073. https://doi.org/10.5194/acp-17-11065- 2017.

Conen, F., Henne, S., Morris, C.E., Alewell, C., 2012. Atmospheric ice nucleators active  $\geq$ -12 °C can be quantified on PM10 filters. Atmos. Meas. Tech. 5, 321–327. https://doi.org/10.5194/amt-5-321-2012.

Conen, F., Yakutin, M.V., 2018. Soils rich in biological ice-nucleating particles abound in ice-nucleating macromolecules likely produced by fungi. Biogeosciences 15 (14), 4381–4385. <u>https://doi.org/10.5194/bg-15-4381-2018</u>

Conen, F., Morris, C.E., Leifeld, J., Yakutin, M.V., Alewell, C., 2011. Biological residues define the ice nucleation properties of soil dust. Atmos. Chem. Phys. 11, 9643–9648. https://doi.org/10.5194/acp-11-9643-2011.

Felgitsch, L., Baloh, P., Burkart, J., Mayr, M., Momken, M.E., Seifried, T.M., Winkler, P., Schmale Iii, D.G., Grothe, H., 2018. Birch leaves and branches as a source of ice- nucleating macromolecules. Atmos. Chem. Phys. 18, 16063–16079. https://doi.org/ 10.5194/acp-18-16063-2018.

Hara, K., Maki, T., Kakikawa, M., Kobayashi, F., Matsuki, A., 2016a. Effects of different temperature treatments on biological ice nuclei in snow samples. Atmos. Environ. 140, 415–419. https://doi.org/10.1016/j.atmosenv.2016.06.011.

Hill, T. C. J., DeMott, P. J., Tobo, Y., Fröhlich-Nowoisky, J., Moffett, B. F., Franc, G. D., and Kreidenweis, S. M.: Sources of organic ice nucleating particles in soils, Atmos. Chem. Phys., 16, 7195–7211, https://doi.org/10.5194/acp-16-7195-2016, 2016.

Huang, S., Hu, W., Chen, J., Wu, Z., Zhang, D. and Fu, P., 2021. Overview of biological ice nucleating particles in the atmosphere. Environment International, 146. <u>https://doi.org/10.1016/j.envint.2020.106197</u>.

Irish, V. E., Hanna, S. J., Willis, M. D., China, S., Thomas, J. L., Wentzell, J. J. B., Cirisan, A., Si, M., Leaitch, W. R., Murphy, J. G., Abbatt, J. P. D., Laskin, A., Girard, E., and Bertram, A. K.: Ice nucleating particles in the marine boundary layer in the Canadian Arctic during summer 2014, Atmos. Chem. Phys., 19, 1027–1039, https://doi.org/10.5194/acp-19-1027-2019, 2019.

Joly, M., Amato, P., Deguillaume, L., Monier, M., Hoose, C., Delort, A.M., 2014. Quantification of ice nuclei active at near 0°C temperatures in low-altitude clouds at the Puy de Dôme atmospheric station. Atmos. Chem. Phys. 14, 8185–8195. https:// doi.org/10.5194/acp-14-8185-2014.

Kunert, A.T., Pöhlker, M.L., Tang, K., Krevert, C.S., Wieder, C., Speth, K.R., Hanson, L.E., Morris, C.E., Schmale Iii, D.G., Pöschl, U., Fröhlich-Nowoisky, J., 2019. Macromolecular fungal ice nuclei in Fusarium: effects of physical and chemical processing. Biogeosciences 16, 4647–4659. https://doi.org/10.5194/bg-16-4647-2019.

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., DeMott, P. J., 2018. A Mesocosm Double Feature: Insights into the Chemical Makeup of Marine Ice Nucleating Particles. J. Atmos. Sci. 75, 2405–2423. https://doi.org/10.1175/ JAS-D-17-0155.1.

Moffett, B., Hill, T., DeMott, P., 2018. Abundance of Biological Ice Nucleating Particles in the Mississippi and Its Major Tributaries. Atmosphere 9, 307. https://doi.org/ 10.3390/atmos9080307.

Pouleur, S., Richard, C., Martin, J. G., & Antoun, H. (1992). Ice nucleation activity in Fusarium acuminatum and Fusarium avenaceum. Applied and environmental microbiology, 58(9), 2960-2964.

Sanchez-Marroquin, A., Arnalds, O., Baustian-Dorsi, K. J., Browse, J., Dagsson-Waldhauserova, P., Harrison, A. D., Maters, E. C., Pringle, K. J., Vergara-Temprado, J., Burke, I. T., McQuaid, J. B., Carslaw, K. S., and Murray, B. J.: Iceland is an episodic source of atmospheric ice-nucleating particles relevant for mixed-phase clouds, Science Advances, 6, https://doi.org/10.1126/sciadv.aba8137, 2020

U.S. EPA, Guidance for data quality assessment. Practical methods for data analysis. (Office of Environmental Information, Washington DC, 2000).

Welti, A., Bigg, E. K., DeMott, P. J., Gong, X., Hartmann, M., Harvey, M., Henning, S., Herenz, P., Hill, T. C. J., Hornblow, B., Leck, C., Löffler, M., McCluskey, C. S., Rauker, A. M., Schmale, J., Tatzelt, C., van Pinxteren, M., and Stratmann, F.: Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern oceans, Atmos. Chem. Phys., 20, 15191–15206, https://doi.org/10.5194/acp-20-15191-2020, 2020.

Zeppenfeld, S., van Pinxteren, M., Hartmann, M., Bracher, A., Stratmann, F., and Herrmann, H.: Glucose as a potential chemical marker for ice nucleating activity in Arctic seawater and melt pond samples, Environmental Science & Technology, p. acs.est.9b01469,https://doi.org/10.1021/acs.est.9b01469, 2019

Zwaaftink, C. D. G., Grythe, H., Skov, H., and Stohl, A.: Substantial contribution of northern high-latitude sources to mineral dust in the Arctic, J. Geophys. Res.-Atmos., 121, 13678–13697, https://doi.org/10.1002/2016jd025482, 2016.