We thank referee 2 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 #2 Evaluations:
Scientific significance: Excellent
Scientific quality: Excellent
Presentation quality: Excellent

General Comments

- The paper does and can only answer the INP source question in a very limited way. Even though a lot of different measurements/analysis is used, the conclusions are a bit vague, which is probably due to the complex nature of the research question. They find indications that INP above -15◦C are dominantly biogenic, while at lower temperatures dominantly dust (this is not so surprising looking at previous field studies, even though not so many specific studies for the Arctic exist). They state that the sources could be marine, locally emitted for the INP above -15◦C, but only if the INP are enriched during aerosolisation by several orders of magnitude. It would be helpful to discuss more critically the limitation of the study, what can be taken from it for which conditions and what needs further investigation. The authors might need to break down the complex research question into subquestions etc..
We are aware that a campaign-based study such as ours has several limitations, which is why we specified the time of year and location in the title of the paper. To better highlight this restriction we now also explicitly mention this limitation directly after formulating our research questions: "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)

- Why are two different filter techniques (LINA and INDA) used and what are the advantages/disadvantages of both compared to each other? When is which technique used (figure captions are not always clear on that?)?
The main difference between LINA and INDA is that they are sensitive in different, but overlapping ranges of INP concentration (as described in section 2.3.5). In our eyes, the difference in their respective detectable range is neither an advantage or disadvantage but give a more complete dataset when combined.
The question which instrument was used was also asked by referee 1, therefore we repeat our answer given there: 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).

- It is not clear where the conclusions come from that the lower temperature INP are

dominated by dust (no clear indication from the trajectories?). Is that derived from the slope of the measurement and comparison to other field measurements?

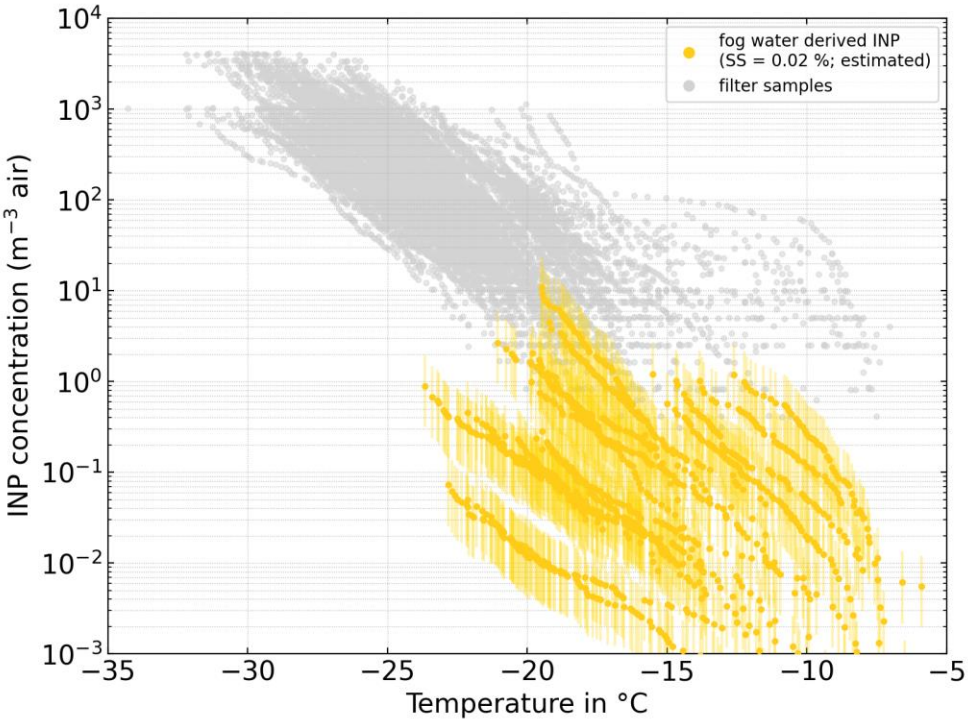
Please note that a referee 1 made a comment on the same topic, hence some parts of our answer here are identical to our answer given there (p. 7 there).

This stems primarily from the abundance and the temperature range of the INP we observed in comparison with other field measurements: To our knowledge, no study has yet found biologic INP to be present in such high numbers in that temperature range as shown in our Fig. 4. Also, only mineral dust is known to occur in such high atmospheric concentrations that it could explain the observed INP concentrations. The study by Welti et al. (2020) does confirm this assumption as they only observe similarly high INP concentrations at the same temperature, when they are in the outflow region of the Sahara, where the aerosol is known to be dominated by mineral dust. Additionally the study by Groot Zwaaftink et al. (2016) shows that mineral dust is produced within the Arctic, which is supported by the recent study by Sanchez-Marroquin et al. (2020), where Icelandic dust is identified as an important source for INP in the lower temperature regime. Then 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. These are of course only indications, but we also don't see any reason to assume that high concentrations of non-mineral INP that are ice active at low temperature should occur especially in the Arctic.

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_{INP} at these low temperatures is remarkable by itself as shown in Fig. 4. That figure shows N_{INP} 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_{INP} 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_{INP} at low temperatures during the campaign." (L311-327)

- Section 3.3.1: How large is the variation due to the assumptions made (SS etc.)? Could you add a section on commenting this and or add error ranges depending resulting from the assumptions made? Same for the variable N_CCN which might affect the result (and comes itself already with an uncertainty).

We have no measures of the actual SS during the fog events we encountered during the cruise. Therefore, the only way to assess the variation of the fog water derived INP concentrations is to vary N_{CCN} at different SS. As mentioned in the manuscript, the literature values for SS of fog range from 0.02% to 0.2%. Since no N_{CCN} below a SS of 0.1% were measured, we calculate an estimate for N_{CCN} at 0.02%SS as follows: We derived the factor by which N_{CCN} decreases from 0.3%SS to 0.2%SS (per 0.1% decrease in SS, N_{CCN} decreases by a factor of 0.81). With that factor we estimated N_{CCN} at 0.02%SS. Then, we calculated again the INP concentration as described in the main manuscript. A linear extrapolation to such low supersaturations has large uncertainties but gives an estimate for the lower boundary of the presented N_{INP} derivation. While the agreement between the scaled down fog water derived INP concentrations and the INP in the air (measured on filter samples) is not as good it was before, the majority of the fog water samples still overlap by one to almost two orders of magnitude. We see still a general agreement between the INP concentration in the air and the fog water



derived INP concentration, especially if the simplicity of this closure calculation is considered. We included a comment on this uncertainty in section 3.3.1 (L428-432): “In the SI (section S9), the fog water derived N_{INP} are shown for an extrapolated value of N_{CCN} at SS = 0.02%. With that value, the agreement between the filter and fog derived N_{INP} is reduced, nevertheless both still overlap by one to almost two orders of magnitude. A linear extrapolation to such low supersaturations has large uncertainties, hence it should be only seen as an estimate for the lower boundary of the presented derivation method of N_{INP} in air from fog water samples.”
Also the the calculation and figure presented above are added to the SI (section S9).

- How large are the uncertainties in the calculated trajectories? Please comment on limitations here as well.

The uncertainty for the 3-day back trajectories shown in Fig.13 are ca. 260 km. This uncertainty was derived by arranging an ensemble of 4 trajectories around the receptor site (i.e., the location of the vessel) with a distance of 0.05°. Then the average distance of the ensemble members to the center trajectory was calculated.
Additional uncertainty stems from the resolution of the meteorological input file given to the HYSPLIT model. We used the standard input for HYSPLIT (GFS1), which as 1° resolution (corresponding to

111km in latitude and 23km in longitude for the area around Svalbard). Hence this is the inherent uncertainty at any point along any trajectory we show. Kahl (1993) described high uncertainties (ca. 1000 km) for 5-day back trajectories in the Arctic. Therefore, we have refrained from interpreting back trajectories that extend beyond three days. We consider the trajectories as only general pointer towards a source region from which an air mass originates. Nothing added.

- Page 2, line 21 and 22: The sentence on the heat induced reduction... can be removed, this was stated already on page 1, line 9 ff.
Sentence was removed.

- Page 2, line 35-46: It would make sense to switch paragraph line 35-39 with the paragraph line 40-46.
Paragraphs were switched.

- Page 5, line 127: "within the ice pack" = open leads?
Yes, in the open leads within the ice pack. This is now specified in the manuscript: "[…] Seawater samples were taken from different environments, i.e., ice-free ocean, marginal ice zone (MIZ), open leads within the ice pack or from meltponds.[…] " (L126-L127)

- Page 9, line 246: What is a "vessel's Ferrybox system"? How does it work?
A Ferrybox is an online instrument to continuously measure oceanographic parameters in a flow-through system that was developed in the early 2000s and is commercially available. A Ferrybox system consists of a water inlet from which seawater is pumped into the measuring circuit containing multiple, modular sensors. Ferryboxes are deployed on research vessels as well as ferries, cruise, and cargo ships (see Petersen et al. 2007, 2011, for details). In the case of the RV Polarstern, the seawater inlet is located in a depth of around 11m, but due to the water flow around the body of the ship, also water from above can get in.

In the revised version the respective text passage now reads as (L260-263): "Chlorophyll-a (Chl-a) concentration was derived from the vessel's Ferrybox system (4H-FerryBox, Jena Engineering, Jena, Germany). A ferrybox is an autonomous online instrument with modular sensor assembly to continuously measure oceanographic parameters in a flow-through system (Petersen et al. 2011, Petersen et al. 2007). The data from the Chl-a sensor in the Ferrybox system were accessed via the DSHIP portal (https://dship.awi.de/) provided by the operator of the vessel."

- Page 12, line 290-291: Was the sampling time consistent for all environmental settings or how can that influence the statement?
The sampling time was set consistently (automatic sample changer) for 8h throughout the campaign except for four days, when the sampling period was reduced to 2h (see section 2.2.1). The statement is meant in a relative sense, i.e. that a higher fraction of the samples collected in the MIZ and ice-free ocean environment are show higher INP concentrations at the warmer temperatures compared to samples from within the ice pack environment. Since the statement does not refer to the absolute number of samples, but the relative number, the sampling time should not significantly influence the metric. In the revised version the statement is changed to:
"[…] 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. […]" (L304-305)

- Fig. 4: Is that figure needed in this paper?

In response to referee #1 additional discussion of the SPIN data was added in the revised version of the manuscript, which is why we would prefer to keep this figure in the manuscript. The Figure shows the surprisingly high INP concentration in the Arctic environment, concentrations which have previously only been observed within the Saharan dust outflow.

- Fig. 7: The labels are a bit small here.
Figure labels are larger in the revised manuscript.

- Page 15, line 340: Add: "...can be seen" due to filtration.
Suggestion added.

- Page 16, line 354-355: Is it legitimate to compare sea water and fog water directly?
Is there not a bias due to dillution effects?
We agree with the comment that this is not legitimate and removed the respective sentence.

- Fig. 11 a: The fit/slope at lower T is very difficult to spot in the graph.
The lines are now wider in the revised version.

- Fig. 12: Write variables in the caption.
Variables were added.

- Fig. 12: The arrows are very small to spot.
We assume the comment refers to panel b of that figure and enlarged the arrows there.

- Page 24, line 503: Be more specific what "similar in shape" means.
With “similar shape” we refer to the steep slope between ca. -7°C and -10°C, followed by an extended plateau region until ca. -21°C that leads into less steep slopes compared to warmer temperatures. A shortened version of this description is added to the bullet point to which the referee referred to: “The freezing spectra of atmospheric INPs are similar in shape, i.e. a steep slope at warm temperatures followed by an extended plateau region, followed by a less steep slope, indicating that during the case study consistent atmospheric INP populations were sampled.” (L540-542)

- Page 25, line 537-538: The most enriched samples featured the highest ice activity.
-> that refers to the SML samples?
Yes. This was made clearer in the revised version (L576-577): “The most enriched samples featured the highest ice activity in the SML samples.”

Technical corrections:
- Page 11, line 270: Remove brackets around the temperatures.
- Page 14, Caption Fig. 5: ...the samples were taken from.
- Page 14, Caption Fig. 5: Add a . at the end of the caption.
- Page 20, line 432: Remove the brackets around the citation.
All technical corrections were implemented in the revised manuscript.

References

Kahl, J. D. (1993). A cautionary note on the use of air trajectories in interpreting atmospheric chemistry measurements. *Atmospheric Environment. Part A. General Topics*, *27*(17-18), 3037-3038.

Petersen W et al. (2007) FerryBox: from online oceanographic observations to environmental information. In: Petersen W, Colijn F, Hydes D, Schroeder F (eds) EuroGOOS Publication No. 25. EuroGOOS Office, SHMI, 601 76 Norkoepping, Sweden ISBN978-91097828-4-4

Petersen, W., Schroeder, F. & Bockelmann, FD. FerryBox - Application of continuous water quality observations along transects in the North Sea. *Ocean Dynamics* **61,** 1541–1554 (2011). https://doi.org/10.1007/s10236-011-0445-0

Welti, A., Bigg, E. K., DeMott, P. J., Gong, X., Hartmann, M., Harvey, M., ... & Stratmann, F. (2020). Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern oceans. *Atmospheric Chemistry and Physics*, *20*(23), 15191-15206.