Response to Referee #2 – Paul DeMott

First of all, we thank Paul DeMott for submitting helpful and productive comments and annotations, which have led to improvements and clarifications within the revised manuscript we submit with this review response.

We have prepared a revised manuscript that addresses the questions and comments of all referees. Furthermore, below we explicitly respond to each of the items raised in the comments of Paul DeMott (reviewer 2). These comments are indicated in *italics,* whereas the author's response is presented in blue. Changes in the manuscript are given in green; changes to the supplement are given in purple. A response with "Okay." means we accept the reviewers' suggestion and have implemented it within the revised manuscript. The differences are also highlighted in separate PDFs using latexdiff. All line and page numbers refer to the ACPD manuscript (version 2), not the revised manuscript.

Interactive comment on "Long-term INP measurements from four stations across the globe" by Jann Schrod et al.

General comments:

This paper is excellent as a large compilation of INP data that has been processed in a consistent manner. The effort is to be commended for that reason alone. It is also a very well written manuscript, and with most of the details one would wish for, and the abstract highlights several key points: well mixed populations that do not vary greatly overall between northern and southern continental and marine sites, short-term variability dominating at all sites, certain site specific aerosol drivers of INPs, but no universal driving aerosol property driver, and no indication of anthropogenic influences. Nevertheless, as I read the paper as it is currently organized, I struggled in knowing how to relate the method and results from the standard FRIDGE method to drop freezing assays (or the immersion mode method sometimes applied using the FRIDGE device), which are possibly the most widely used present method. It seems to me that two things are required to assist readers in understanding the nature of the results, and potentially how to consider them in relation to immersion freezing data. First, the title should explicitly describe the basis for INP measurements. In other words, "Long-term deposition/condensation freezing INP measurements. . ." or something to that effect. When one sees the INP versus ice supersaturation data in this manuscript, there is no discontinuity that occurs at water saturation (as the authors readily note), and so it seems apparent that immersion mode freezing is indeed not represented at all. The authors provide a discussion of the dominant mechanisms at play in the data and the likely underestimate in comparison to immersion freezing mode operation of the FRIDGE only very late in the paper. This is critically important in understanding if the findings can be ascribed only to deposition and condensation-freezing mode INPs, or if the same is expected for immersion freezing populations. I suggest in the specific comments that the methods used may indeed limit assessment of strong local/regional impacts, at least for biomass burning. Of course, it will not be possible to make a conclusion about what was not measured, but it should be highlighted as a question for future inspection. This should all be made crystal clear. Hence, the second recommended change is to bring a discussion forward of what types of INPs the data describe, and what types the

generalized results may not describe. It will not detract from the great effort the authors have made to collect large quantities of ice nucleation data from multiple sites and discern answers to some of the key and enduring questions related to INP sources. However, I believe that it will better frame future needs.

We thank the referee his helpful feedback and review. After re-reading the paper and the reviews, we recognize now that it may indeed be difficult to understand for the reader what is measured here and what is not. As a matter of fact, we do absolutely think that we need to explore the differences in our own measurement methods, i.e. FRIDGE standard and droplet freezing mode, to a greater extent. As for the suggested implementations, we agree to the proposed changes. Adding the addressed nucleation mode in the title will immediately help the reader orient themselves. Also, we come to the same conclusion as the referee, that the discussion about the nucleation mode appears too late in the paper and can be better introduced in the methods section as the reviewer proposes in the specific comments.

Accordingly, we have changed the title of the manuscript to:
"Long-term deposition/condensation INP measurements from four stations across the globe".

Furthermore, we now introduce both operational modes of the FRIDGE instrument shortly in a new section 2.1, indicating clearly at the beginning of the methods chapter that only the standard mode has been used in this study:
"2.1 FRIDGE operational modes

The FRIDGE instrument was originally introduced by Bundke et al. (2008) and Klein et al. (2010), but was fundamentally reevaluated and updated by Schrod et al. (2016). Since this effort FRIDGE has participated in laboratory intercomparisons (Hiranuma et al., 2015; DeMott et al., 2018; Hiranuma et al., 2019) and field campaigns (Schrod et al., 2017; Thomson et al., 2018; Gute et al., 2019; Marinou et al., 2019). In its original design FRIDGE serves as an isothermal static diffusion chamber for offline analysis of ice nucleation. In this standard operation mode FRIDGE analyzes deposition and condensation freezing INPs on substrates that had been laden with atmospheric aerosol particles by electrostatic precipitation. To avoid confusion, we point out that the FRIDGE instrument can in fact be modified to serve as a cold stage for droplet freezing assay measurements as well, which was, however, not done for the results presented here."

The former sections 2.1 (2.2 in the revised manuscript) and 2.2. (2.3. in the revised manuscript) then follow, describing the typical procedure during sampling and measurements.

Finally, we have added section 2.3.1, which goes into more detail regarding what kind of INPs were actually measured in this study, i.e. deposition/condensation vs. immersion mode and how these might relate to each other:
"2.3.1 Freezing modes

It should be noted that we cannot predict how our deposition/condensation freezing measurements would translate to the immersion freezing mode in a situation given in the atmosphere. Some conclusions may however be drawn from previous parallel

measurements (unpublished) with the FRIDGE diffusion chamber and the FRIDGE droplet freezing assay in different environments during the FIN-03 (Storm Peak Laboratory, SPL, USA, 2015), GLACE (Jungfraujoch, JFJ, Switzerland, 2017) and PICNIC (Puy de Dome, PDD, France, 2018) campaigns. Daily average INP concentrations (i.e. one day sample and one night sample) covered three orders of magnitude at -25°C. When transforming the INP concentrations to log-space, we find that the two operational modes are well-correlated (R = 0.81, N = 44), with the immersion freezing INPs being on average a factor of 10 higher than deposition/condensation INPs. In fact, the INP concentrations measured in the droplet freezing assay were always higher. One may speculate that both species simply covary for the reason of having the same sources and sinks, or that deposition INPs may represent just a subset of immersion INPs, when observed by FRIDGE, or both. We will present the results of this comparison in more detail in a forthcoming publication, in which we will further investigate how exactly the nucleation modes of both methods are connected to each other.

Except when noted otherwise, the discussion presented in section 3 will focus on the highest ice supersaturation(s) RH_{ice} at each of the three examined activation temperatures (Tab. 2). At these highest saturation conditions, at or slightly above water saturation, we observe the highest INP concentrations. We expect the nucleation mechanism to be a mixture of deposition nucleation and condensation freezing. At lower supersaturations we qualitatively observe trends and variability in INPs that are similar, but at lower absolute concentration levels."

Specific comments:

1) *Introduction*

- *Page 2, Lines 8-9: Is there a reason to separate primary biological aerosol and marine biological aerosols? They are both primary biological aerosols, no? If referring to secondary marine aerosols, you might require evidence that those play any role as INPs.*

  This was rather unintentional. We have rephrased the sentence for more clarity (we included non-biological organics to the list as suggested by reviewer 1):
  "Known species of INPs include mineral dust, soil dust, primary biological aerosol particles of terrestrial and marine origin, as well as organics and glassy aerosols (Kanji et al., 2017)."

- *Page 2, Lines 30-32: I find this statement quickly becoming untrue, with many laboratories now involved in long-term measurements of immersion freezing (e.g., Schneider et al., 2020), some with agency support, and multiple online instruments are in development (or are already there) for automated or semi-automated operation.*

  We thank the referee for the interesting paper, which was not available at the time of submission. We certainly hope that the assessment of the referee proves to be correct, as we think that having more long-term INP measurements publicly available is a crucial step in understanding the spatio-temporal variation of INP concentrations worldwide. As for the sentences in question: We think that at least for the very recent past the phrasing is correct. We fully stand by the first sentence, stating that very few of the published INP

measurements cover multiple seasons or more. We edited the second sentence, adding the assessment of the reviewer:

"A further obstacle to INP monitoring was that many instruments were previously not suited for sustained, long-term monitoring tasks due to their complex and labor intensive operating principles. However, recent developments in INP instrumentation and a shift in sampling focus may lead to more long-term INP measurements becoming publicly available now and/or in the near future (e.g. Schneider et al., 2020)."

- *Page 3, line 30 to end of paragraph: With regard to anthropogenic influences, I do think that there is some literature on this topic. Some is recent, e.g., Levin et al. (2019) found no apparent influence of urban pollution on INPs in studies in CA, USA. Chen et al. (2019) and Bi et al. (2019) discuss urban pollution impacts in Beijing.*

  While we are aware that some literature exists on this topic, we wanted to emphasize in the paragraph that the anthropogenic influence on the INP abundance and efficiency is far from conclusive at this point. We added a sentence to the paragraph (P3L26):

  "Although some recent studies indicate that urban pollution aerosol do not make efficient INPs (e.g. Chen et al., 2018), the overall anthropogenic impact on the INP concentration is still rather inconclusive (see also Schrod et al., 2020)."

2) Methods

- *Page 4, lines 28-29: Have larger particle losses been quantified? This is important, as it is a weakness compared to an open-faced filter for example, and it is not clear as an advantage over the in situ instruments mentioned in the last sentence of the paragraph. For example, Schrod et al. (2016) report collection efficiencies only to 3 microns, which is not measurably much different that impactors used on some in situ devices. And larger particles might be imagined as the most efficient deposition nuclei. While collection of and a role for larger INPs is evident ultimately in Fig. 9 for the AZ site, one wonders if the drop off of INPs at sizes above 2 microns reflects the true contributions in these size classes or is influenced at all by collection efficiencies.*

  We recognize from both reviews that more care should have been taken when describing the inlet configuration. Unfortunately, only the particle losses at AZ have been quantitatively characterized (Moran-Zuloaga et al., 2018, see section 2.4.1). Regrettably, we don't think a thorough inlet characterization is feasible at this point as the sampling devices are no longer at the sampling sites. We will add a paragraph that mentions this shortcoming more clearly. P1L27:

  "No inlet size-cutoffs were used for the results presented here, and thus we expect to sample the complete particle spectrum, except for the usual particle losses that may occur for large particle sizes. The exact aerosol inlet configuration differed substantially between sites and was mainly predetermined by the local observatory facilities. Unfortunately, these inconsistencies may lead to some aerosol sampling artifacts with respect to the absolute particle losses. The individual sampling configurations are described in section 2.4 and Tab. 1."

Table 1. Main characteristics of the geographic sampling location and inlet configuration at the sites.

	AZ	MQ	TO	SB
Geograph. coordinates	2.144° S, 59.000° W	14.735° N, 61.147° W	50.221° N, 8.446° E	78.908° N, 11.881° E
Altitude [m AMSL]	130	487	825	474
Climate	tropical	(sub-)tropical	temperate	Arctic
Continental / marine	continental	marine	continental	marine
Mountain site	no	yes	yes	yes
Predominant vegetation	tropical rainforest	diverse (i.e. ranging from alpine to tropical rainforest)	coniferous forest	low-growing tundra (summer) / snow-covered (winter)
Anthropogenic impact	near pristine to polluted	remote to polluted	rural to polluted	near pristine to polluted
Inlet type	Total Suspended Particulate (Moran-Zuloaga et al., 2018)	1/4" tube, rain shield (no characterized inlet)	HORIBA ASS-370 type (ÖNORM, 2007)	Whole-air (Karlsson et al., 2020)
Inlet height [m AGL]	60	2	11	7.5
Isokinetic flow splitter	yes	no	yes	yes
Length of tubings to PEAC7 [m]	1.5	2	1	2

As we cannot retrospectively quantify the particle losses reliably for the inlet configurations at the stations we deleted the sentence about a possible advantage of not using size-cutoffs (P4L29-31).

Concerning Figure 9, as already mentioned the inlet configuration has been characterized by Moran-Zuloaga et al., 2018, as presented in the supplementary Fig. S1 of that manuscript. Here it can be seen that transmission efficiency from the inlet was calculated to be between 90 and 100 % at 2 µm. Depending on the particle density the transmission drops for larger particle sizes. For example, at 5 µm the transmission efficiency is still between 80 and 90 % for particle densities around 1 g cm^{-3}, but may be as low as about 60% for particle densities of 2 g cm^{-3}, e.g. mineral dust or sea salt. Taking these calculations into account, we believe that the presented INP composition vs. size likely represents the true contributions quite well, at least for the bins up to 5 µm. However, the last bins may in fact be influenced by particle losses to an unknown, but non-neglectable, degree. We added a sentence to P14L28:
"Note however, that the contribution to the larger size bins might be potentially underrepresented due to particle losses from the inlet configuration."

- *Page 5, line 13 paragraph: This description of the aerosol samples had me already wondering about sampler inlets and placement. You might state that this will be covered for each specific site. I do question the statement that 100 L samples provide for "well-resolved ice crystal numbers for a broad spectrum of temperatures..." INP concentrations can span several orders of magnitude from -5 to −35 C . This paper covers a 10C range for data presentation. Finally, is the statement on storage effects necessarily assured for biological INPs that might be exposed to dessicated and higher temperature conditions? This was qualified in Schrod et al. (2016).*

  First part: see previous response.
  Second, yes, although we think a span of 10 °C is still quite good, we agree to rephrase the statement:

"The sampled aerosol particles resulting from this 100 L of air were found to usually generate well-resolved ice crystal numbers in the investigated temperature regime using the FRIDGE analysis system."

Further, we cannot guarantee that biological INPs remained active during storage and transport. We expanded upon the paragraph:
"As a result, several weeks often passed between sample collection and analysis, which may introduce an aging effect. However, in a previous study no effect of storage time on ice nucleation activity was observed within the investigated temperature regime (Schrod et al., 2016). Since a frozen storage and transport could not be logistically guaranteed for all sites and for all times, samples were stored and transported at ambient temperatures, which may have affected the warm end of (biological) INPs."

- *Section 2.2: It is worth carefully explaining the valid activation modes for this work (should be deposition and condensation "freezing" mode on line 22), perhaps by reiterating a few points from Schrod et al. (2016). This first paragraph appears to be the clear place to expound on what is known about the potential underestimations compared to immersion freezing mode INP data as well. Instead, there is only a sentence, "In this context...", which is awkward and defensive considering that the FRIDGE instrument pre-dated many of the droplet freezing assays. The instrument is clearly a tool within the wider array of ice nucleation instrument types, and to my knowledge one of the few well-characterized and documented ones that allows for exploring the full temperature and ice relative humidity space (in the mixed-phase cloud regime) for single samples, in the same manner that droplet freezing assays allow for full temperature spectra. All of the advantages of the technique compared to more labor intensive diffusion chambers and drop freezing assays are well acknowledged. What is missing for this assessment of long-term records at multiple sites is a clear indication of the relation of the modes assessed to immersion freezing. What is known and what remains for future exploration, if the method could be meshed with additional immersion freezing measures?*

  Yes, we agree with the referee. See the response to the general comments section. Furthermore, we have removed the sentence starting with "In this context".

- *Page 5, line 24: The word meaningful seems unnecessary.*

  Okay.

- *Page 7, line 15: An additional question here is if there are any considered additional particle losses in the inlet entry to the sampling system. That is, is sampling from the main inlet isokinetic (or sub- or super-isokinetic) and are any additional large particle losses characterized for that last step in collection? Similarly on page 8, line 23, it says that the sampler and the OPS instruments were connected to a 2 m stainless steel line at OVSM. Were particle transmission efficiencies characterized/expected to be the same at this site? Given the outsized role of larger particles as INPs at some surface sites (e.g., Mason et al., 2016), it seems important to know if the relative collection efficiencies were the same, and what the upper limit might be. I also note no mention of sampling inlet protocol for either TO or SB sites.*

  The inlet sampling configuration of AZ is well-characterized in the supplementary information of Moran-Zuloaga et al., 2018 to which we refer. The

main sampling line is in fact connected to an isokinetic flow splitter that was connected to the PEAC7. The main particle loss mechanism considered is sedimentation of particles >0.5 µm. P7L15:
"An extensive inlet characterization can be found in the supplementary information of Moran-Zuloaga et al., 2018."

We do assume more or less similar particle loss scenarios at the other sites to what is stated in this section, although we, unfortunately, did not characterize the transmission efficiencies at the other sites. Also see previous response.
We now add information about the inlet configurations at TO and SB:

P9L14 (TO):
"Samples were collected from the upper level of Atmospheric Physics Laboratory at the hilltop. The aerosol inlet was at 11 m above ground. A main flow of ambient air was pumped through a Horiba ASS-370 type inlet (ÖNORM, 2007) with a 40 mm I.D. x 7 m length stainless steel tube into the laboratory. The PEAC7 collected aerosol isokinetically at 2 l min^{-1} from the main flow through a nozzle of 2.2 mm diameter."

P9L23 (SB):
"A whole air inlet was used for aerosol particle sampling according to the ACTRIS guideline for stations that are often embedded in clouds. The flow through the inlet was kept constant to ensure near isokinetic sampling conditions. A short description about the inlet characteristics of the Zeppelin Observatory can be found in Karlsson et al. (2020)."

- *Page 7, line 29: A minor note here that it would be interesting to know the vegetative differences in these sites. Images of the sampling sites could also be interesting, for supplemental information.*

  We have added one picture for each site to the supplement (Figs. S1 to S4). Further, we have added Tab. 1, which describes the most relevant site characteristics, including the predominant vegetation.

- *Page 9, line 20: What is meant by "direct influence of sea salt aerosol"? Is the Zeppelin site not within the boundary layer? This is important to know with regard to what influences are being measured there. Sea spray particles would seem as one key source.*

  Yes, sea spray is still expected to be a key aerosol source. Due to the elevated position of the observatory we expect lower absolute sea salt concentrations than what would have be measured at sea level (i.e. the other research stations in Ny-Alesund). The sentence now reads:
  "The mountain top Zeppelin Observatory was chosen for its elevated position, which likely limited the effects of locally produced pollution and of sea spray from the surf zone."

3) Results and Discussion

- *Page 10, lines 10-11: Considering the discussion above about INP mechanisms, this statement about deposition being considered relatively unimportant for mixed phase*

clouds is confusing. Is this not what is measured by the FRIDGE instrument? If the traces of INP versus ice supersaturation are continuous, how to know the difference between deposition and condensation freezing? Is not the highest RH value of processing used here so that the highest INP concentrations assessable are accessed? This is the only way to understand the following statement that "incomplete" condensation freezing is assessed. Again, this may be material to consolidate in the Methods section, where it can be pointed out that an emphasis will be placed on the highest RH values for inter-comparison of site data.

Yes, the results of the highest RH are shown to give the highest INP concentration. The intention here was to say that these concentrations are the closest we can come to immersion freezing in our instrument. We recognize that the phrasing adds more confusion than it actually helps. Therefore we have removed the sentence and moved the paragraph to the methods section (see response to the general comments).

- *Page 10, lines 31-32: It is great that the authors qualify the results regarding timing of the sampling, storage impacts, etc. However, I am not sure what this statement means about long-term trends being better captured by different sampling strategies. Can you expound? Does it mean spreading the sampling periods out across daily periods? Larger volume samples collected over longer time periods? Additional use of immersion freezing methods, as in that study, to investigate if that mode of ice nucleation also shows a lack of long-term trends at sites. Also, please note that the full publication on the noted results is now in press and under review in ACP (Schneider et al., 2020). That study does show trends linked to a regional source. One can imagine that regions close to mineral dust sources also show impacts of a strong regional source, where much higher INP concentrations are noted (e.g., Price et al., 2018). Likewise, higher latitude and polar regions, especially from ship campaigns in the Southern Hemisphere (McCluskey et al., 2018; Welti et al., 2020), appear to represent extraordinarily pristine INP environments. It is simply the case that for the sites selected for this paper and the methods applied, strong cycles are not noted and short-term variability dominated. The extent to which this can be generalized for tropical and mid-latitude regions remains to be seen.*

  Yes, we primarily meant longer sampling periods/larger sampling volumes. For example, Schneider et al. (2020) have used a time resolution of 24 to 144 h at 11 L min^{-1}. However, it would be rather difficult to adapt the FRIDGE standard technique to such long sampling times, as the number of resolvable ice crystals on a substrate is limited. All of the suggestions to implement a different sampling strategy are good ideas, i.e. spreading the total sampling time out over short increments of time throughout a day as well as complementing the standard mode with the FRIDGE droplet freezing mode and longer sampling times. The manuscript has been modified to read:

  "Comparisons with other recently published data sets suggest that long-term trends may be better captured using different sampling strategies (Schneider et al., 2020). The authors of that study observe a clear seasonal cycle of immersion INPs in a boreal forest using 24 –144 h filter sampling at 11 L min^{-1}, which is a much longer sampling period than has been used here. [...]"

We have not intended to overly generalize our results. To clarify we have added the freezing mode in more instances throughout the manuscript when discussing the "INP concentration". For example, P17L10 now reads:

"In spite of the great differences in basically all characteristics that are expected to define the aerosol concentration, composition and source apportionment, we observed fairly similar INP concentrations for all four stations for the methods and sampling strategy applied."

- *Page 11, lines 15-16 and elsewhere: I have a suggestion to consider for demonstrating the spectral differences between sites, and where they are distinguished for given sites. Currently, a temperature spectral plot is not included in the paper, with too much emphasis on ice supersaturation in my opinion. Figure 5 could be made differently or augmented with an additional panel. While sometimes a linear scale is preferable, in this case if you alternately (or additionally) put these data on the same log scale, one could see the temperature differences more clearly. For example, if the y-axis scaled from 0.01 to 10 on a log scale, the temperature spectra becomes evident for conditions near water saturation, which are arguably the most important for clouds.*

  The suggested change to Fig. 5 is appreciated. We have changed it accordingly:

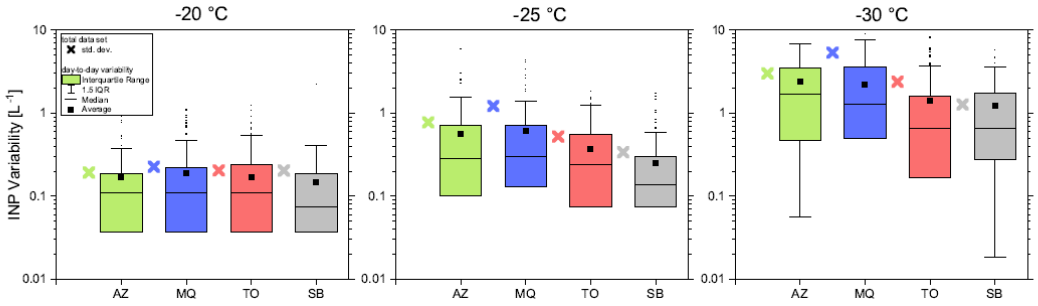

Figure 5. INP variability for $T = -20\,^{\circ}\mathrm{C}$ (left), $T = -25\,^{\circ}\mathrm{C}$ (middle) and $T = -30\,^{\circ}\mathrm{C}$ (right) at $RH_{water} = 101\,\%$. Crosses show the standard deviation of the total data set for each site. Box-plots show the distribution of sample-to-sample differences in the INP concentration of consecutive samples (i.e. day-to-day or every other day). Lower whiskers are not shown when the lower 1.5 IQR is at zero.

- *Page 12, lines 18-19: This comment harps back a little bit to the statement in Methods regarding the large dynamic range of measurements. While 100 L samples are more useful than the smaller sample volumes used in online instruments, the lack of resolution in the −20 C and warmer regime means that there is little or no access to the temperature range where one might expect most sites to be distinguished, considering for example the results shown in Petters and Wright (2015). This is also an important point to remember in the discussion here regarding whether any of the sites are distinguished by apparent biological particle influences. The measurements are just touching the regime of interest.*

  Yes, we agree. We have added a comment on P12L24, when mentioning the potential for biological INPs:

  "However, there is no strong evidence for such a signal in our data overall, possibly due to the comparably low sampling volume. As a result the temperature range of our measurements overlaps only very little with the regime where biological particles nucleate."

- *Page 13, discussion of Fig. 7: Figure 7 is a remarkable figure, and I find it astonishing that local sources do not come into play for either TO or AZ. I wonder if the authors might comment on whether INP removal is also a factor to consider, not only dilution/ mixing out from strong sources, as is inferred in the comment about "background" air masses?*

  It is possible that INPs are removed by either being activated to ice crystals or by deposition processes. However, it is difficult for us to assess how such effects may affect the distribution of INP concentrations found, and particularly if and how local sources are affected differently.

  Generally speaking, we also found the strong log-normal representations to be surprising. We believe that this is an area that deserves further attention within the community. In particular we would like to understand how skewness and departures from log-normality is affected by source function changes and atmospheric processing, but these questions go beyond what we can address in this manuscript.

- *Page 13, section 3.2.1: First, can you please clarify the timing of the "dry season" at AZ? It becomes obvious in Fig. 8, but it would be nice to see it stated in the discussion. And then one has to go back to figures to note the lack of an apparent influence of smoke. The reduction is AF is not really unexpected, right, in consideration of previous results regarding biomass burning INPs? Considering laboratory studies of surrogate and real combustion particles (Petters et al., 2009; Levin et al., 2016; Kanji et al., 2020) and field studies (Prenni et al, 2012; McCluskey et al., 2014; Schill et al., 2020)? Hence, the discussion could be clarified here, including the most recent references. One might even support that for realistic combustion particles, and not only black carbon isolated (Kanji et al., 2020) or contained in real biomass burning particles (Schill et al., 2020), water supersaturations and immersion freezing are required to see the influence of biomass burning on INP concentrations (e.g., Petters et al., 2009; Schill et al., 2020). That is, there are clear impacts of biomass burning on regional INP concentrations already demonstrated in the literature for other regions. I think this discussion needs more specifics than referencing a review paper and a single laboratory study on black carbon surrogates. Activity within the deposition and condensation freezing regime up to water saturation may be quite limited, so this may represent a case where the methods applied in this paper cannot resolve real influences on INPs, or it may indicate that fires are not sources at AZ. I think it is unresolved still.*

  The timing of the dry season is given in section 2.4.1: Dry season: August to November, wet season: February to May, transition periods in between. We will state this in 3.2.1 again now.

  We agree that the obtained results are not unexpected, as we wanted to indicate by including the Kanji et al. references. We will expand upon this point as the referee suggests, adding more references on this matter. We thank the referee for the suggested papers. P13L31 now reads:

  "The observed anti-correlation seems to suggest that aerosol particles from fires are relatively poor ice nuclei; an observation that agrees with previously published findings (Kanji et al., 2017, 2020). Considering the recent literature consensus regarding biomass burning INPs, these results are not unexpected. Biomass burning INPs have been studied in the laboratory investigating both surrogate and real combustion particles (Petters et al., 2009; Levin et al., 2016; Kanji et al., 2020), and in the field (Prenni et al., 2012; McCluskey et al., 2014;

Schill et al., 2020). Although at least a regional impact of biomass burning on INP abundance is reported, the nucleation temperatures are usually close to the homogeneous freezing limit. Some of these studies suggest that water supersaturation is a requirement for biomass burning aerosol to act as INPs (e.g. Petters et al., 2009; Schill et al., 2020). In this regard, our data may demonstrate the limits of what is explorable with FRIDGE. Either biomass burning aerosol is in fact a poor source for Amazonian INPs in the investigated temperature regime or the method simply cannot represent the freezing behavior of these particles accurately."

- *Page 14, lines 17-18: Following in the same line of comment, in fact the INP concentration results herein seem to be a factor of several lower compared to Prenni et al. (2009). It would be good to quantify what is stated presently as "on the low end".*

  Okay. The line now reads:

  "However, our observed concentrations are clustered at the low end of those presented by Prenni et al. (2009) (i.e. a factor of 5 lower at -30 °C), which is presumably due to the different nucleation modes addressed."

- *Page 15, lines 25-26: It is unclear if the conclusion here is that marine contributions to the INPs at MQ are represented in the lower range of values observed?*

  We removed the unclear sentence. The manuscript now reads:

  "[...] They measured INP concentrations of 0.06 L^{-1} at -20 °C and 0.3 L^{-1} at -24 °C, which agrees within a factor of two to our median INP concentrations at -20 °C and -25 °C."

- *Page 15, lines 32-33: Is this correlation with PM10 at TO shown anywhere? Can you at least state the r2 and p values?*

  In the next line we do present the Pearson correlation for -25°C (101%). The R-value is not impressively high, but is significant due to the large number of samples. We have added an indication of the p-value.

- *Page 16, SB section: As I read this section, I wondered about the issues brought forward at the end of the section with regard to signal to noise ratio, and how this influenced the lack of a seasonal cycle. For example, Hartmann et al. (2020) should also be referenced here. They also report winter values consistent with Tobo et al. (2019) and Wex et al. (2019). Hence, one wonders why no seasonal cycle is present in the data here. Is it just noise, or is the baseline potentially somehow even higher than you have estimated from blank data?*

  We have now added a sentence regarding the new measurements by Hartmann et al. (2020), and have added references to the measurements by Rinaldi et al. (2020) who do not find a seasonal shift in Ny-Alesund (suggested by referee 1). P16L23:

  "Very recent measurements from Greenland during March/April 2018 qualitatively agree very well to these concentration levels (Hartmann et al., 2020). However, a recent study by Rinaldi et al. (2020) did not observe a distinct seasonal signal in their INP measurements in the temperature range from -15 °C to -22 °C in the spring and summer of 2018 in Ny-Ålesund. Rinaldi et al. (2020) present INP concentrations from two separate methods, one of which is fairly

similar to FRIDGE, addressing the condensation freezing (DFPC) and immersion freezing (WT-CRAFT) modes."

Unfortunately, it is difficult to add more insight into why we did not observe a seasonal cycle in the SB data. Although some variation in the background concentration is present, we don't think that the baseline is higher overall than we have assumed. Rinaldi et al. (2020) conclude that the discrepancy of their time series with the clear spring-to-summer differences from other observations "likely indicates that the inter-annual variability of meteorological and biogeochemical conditions determining the INP atmospheric concentration over the Arctic is wider and more complex than previously assumed. For sure, the number of observations in the Arctic and their temporal coverage are still too limited to derive general conclusions on the INP concentration trends." In general, we agree with these statements, although it is still possible that we have missed a seasonal shift due to a poorly selected sampling strategy, as stated in the manuscript (see also above).

4) *Conclusions*

- *Page 17, lines 11-13: I find alluding to the Welti et al. paper results to not be a great comparison. In fact differences in the most remote locations were striking compared to mid-latitude and tropical locations in that paper and in other recent ship campaigns (McCluskey et al., 2018).*

  Okay. We have removed the reference.

- *Page 17, line 20: I think you should add "at all sites" when referring to the inability of single parameters to describe results. This is important, as influences were noted at some sites.*

  Okay.

- *Page 18, lines 3-6: This is the discussion point that needs to be introduced earlier in the paper, as I mentioned previously. One even wonders if the processing conditions emphasize certain INP types that are more well mixed in the atmosphere and contain few hygroscopic materials that would limit ice nucleation until strong condensation occurs at most of the temperatures investigated.*

  We agree and have moved the discussion to the methods section as described earlier.

  Second, unfortunately, we did not fully understand the arguments made regarding a potential bias towards more well-mixed INPs in our data, but we don't think that there is clear evidence for this hypothesis present.

5) *Outlook*

- *Page 18, lines 18-19: One wonders about varying sampling times over daily schedules to represent diurnal cycles. However, here, I wonder if it is necessarily true that longer sampling times would reduce short term variability? Would several hour samples reflect less differences than the short sample times used in this study? How do you know?*

  We do not know and this is a bit speculative. We have changed the wording:

"Longer sampling may effectively act as a low-pass filter and thereby reduce the considerable short-term variability in INPs that is observed everywhere."

- *Page 18, lines 21-22: Again I find myself disagreeing with this conclusion that automated and higher frequency sampling methods are too much of a technological challenge. It simply needs impetus and being made a priority, and I would judge that the time has already arrived.*

  We agree with the assessment of the referee and hope that soon such an effort will be made. We have changed the wording:

  "However, such an instrument and/or technique has not been available in the past and will likely present both technological and human resource challenges."

- *Page 18, lines 30-32: A reason that immersion freezing is considered so important is because clouds, and how they form, in many cases determine this result. Could immersion freezing measurements become an integral part of sampling and processing protocol for a device like the FRIDGE? Then all mechanisms except contact freezing would be assessed.*

  The sampling schedule of this manuscript began in 2014, when the immersion mode was not yet implemented. But in future FRIDGE measurements we will study the atmosphere with both the standard mode and the immersion mode setup whenever possible, just as the referee suggests.

- *Page 19, lines 9-10: I find the calling out of a single device to be inappropriate here, from a conference paper no less. Fortunately for this reference, the prime publication on the PINE came out the same day as this review (Möhler et al., 2020). However, automated CFDC instruments are already being built for surface sites (Bi et al., 2019) and under development for aircraft use. I do not understand the statement about a "vital intermediate step".*

  We thank the referee for bringing our attention to this paper (Bi et al., 2019), which we had missed. He makes a good point and we were not attempting to single out any particular device. In fact, since this time yet another instrument has also emerged (Brunner and Kanji, 2020), we reformulate to include both new references:

  "Although we are currently far from the best-case scenario of a (near) continuous automated global network of INP measurements, there are promising new developments (e.g. Bi et al., 2019; Brunner and Kanji, 2020; Möhler et al., 2020) that may provide a vital step towards long-term (semi-)automated measurements of immersion mode INPs in the near future."

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