Response to Anonymous Referee #1

First of all, we thank the referee for submitting their helpful and productive annotations, which lead to improvements and clarifications within the manuscript.

We prepared a revised manuscript that addresses the questions and comments of the referees. Furthermore, below we explicitly respond to each of the items raised in the comments of anonymous referee #1. 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 accepted the reviewers' suggestion and implemented it in the manuscript. The differences are also highlighted in separate PDFs using latexdiff. All line and page numbers refer to the ACPD manuscript version, not the revised manuscript.

Review of "Ice nucleating particle concentrations of the past: Insights from a 600 year old Greenland ice core" by Schrod et al.

General comments:

This reviewer supports publication of this manuscript in ACP. The research topic - researching INPs in the pristine past conditions - is an important addition to ACP for many reasons; e.g., providing a constraint to climate simulations/projections etc. In spite of many potential artifacts addressed throughout the manuscript, the authors conducted careful and dedicated offline lab experiments, and their findings warrant future follow up studies. Unfortunately, such care was not taken in the preparation of the manuscript (esp. after Sect. 2.2), with the manuscript containing a number of unusual word choices and non-intuitive statements. The reviewer has numerous revisions as listed below. Though most of them are minor, the reviewer would urge the authors of the manuscript to thoroughly proof read their manuscript for improving readability, as this list gets too long.

> We thank the reviewer for their careful reading of the manuscript. The long list of language edits, additional ideas and suggested literature are greatly appreciated. We agree that the suggested changes will improve the readability of the manuscript. We will go through the comments listed below one by one.

Specific and technical comments:

- *P1L13: The reviewer suggests the authors to specify dp is in a spherical diameter metric here.*

  > Okay.

- *P1L20-21: The reviewer appreciates the authors to be honest scientists extensively addressing some potential artifacts throughout the manuscript. However, the statement*

of "or some post-corning..." seems unnecessary to conclude the abstract. The reviewer suggests removing this part in the abstract.

Okay.

- *P2L6-7: Does the authors mean – "Unfortunately, heterogeneous ice nucleation, which is of primary importance of atmospheric ice formation, has not received..."?*

  The sentence was phrased this way to illustrate that although ice nucleation is very relevant to precipitation processes in the atmosphere and by extension to snow accumulation in the Arctic, ironically ice nucleation experiments on ice cores have not been sought out by researchers frequently. We rephrased the sentence to make this more clear:

  "Unfortunately, heterogeneous ice nucleation, which is of primary importance to atmospheric ice formation and therefore very relevant to Polar snow accumulation, has not received much attention in ice core sciences."

- *P2L7: As of today → Until now or To date (better word choice)*

  Okay.

- *P2L21: defines → constrains (this seems better fitting here)*

  Okay.

- *P2L27: Although...straightforward, → Evidently,*

  Okay.

- *P2L27: seen → implied*

  Okay.

- *P2L30: The reviewer finds the discussion of anthropogenic INP to be a very important part of the current manuscript and, therefore, wishes that the authors can extend the discussion a bit further? A suggestion for reading is Zhao et al. (2019, Nature Geosci.; https://www.nature.com/articles/s41561-019-0389-4?proof=trueMay) and references therein. Currently, the discussion of anthropogenic INPs is controversial, and the authors can help the community by including an extended discussion here. Doing such may reinforce the paper.*

  We agree with the referee about the importance of discussing the relevance of anthropogenic INPs for the manuscript. We thank the referee for pointing the suggested reading out to us. We added additional text to the manuscript to emphasize the discussion:

  "[...] Yet, the significance of anthropogenic pollution particles to atmospheric ice nucleation is still in question. Recently, Zhao et al. (2019) investigated the effects of pollution aerosol to the ice phase in moderate and strong convective systems in a top-down approach using a combination of satellite observations and model simulations. They present evidence that in the moderate convection case, where heterogeneous ice nucleation is more relevant, the ice particle effective radius is increased, indicating that continental pollution aerosol may in fact contain a considerable fraction of INPs. On the other hand, further experimental studies suggest that most anthropogenic aerosol particles are typically poor INPs. For example, Chen et al. (2018) found that the heavy air

pollution of Beijing did not affect the INP concentration in this urban setting in the investigated temperature range from -6 °C to -25 °C. Overall, there are still few studies available on the ice nucleation efficiency of anthropogenic aerosol and some of the presented evidence is conflicting. Although pure pollution aerosols are considered rather inactive INPs, this does not per se mean that the INP population as a whole has not changed at all over the last centuries. On the contrary, it seems rather likely that certain particles with ice nucleating potential may in fact be more abundant in today's atmosphere. [...]"

- *P2L34: Biomass burning aerosol is...least potential contributor to anthropogenic INP.*
  Okay.

- *P3L13: Indeed, soil dust, in part derived from agricultural systems/practices,... Is this what the authors meant? Feel free to modify it.*
  Yes, that's what we meant. Okay.

- *P3L17: ...global land area, of which approx. 9% were identifies...*
  Okay.

- *P3L20-23: Please clarify what "anthropogenic increase in mineral dust concentration" means. Also, a bit more discussion of aerosol particle episodes to Greenland would strengthen the paper.*
  There are several instances throughout the manuscript, where the dust transport patterns to Greenland are discussed. Therefore, we chose not to overly go into details in the introduction. We rephrased the sentence:
  "[...] however a recent increase in mineral dust concentration from these areas due to anthropogenic impacts is not documented."

- *P3L25-26: write out LINA and INDA? They appear once only, so it seems no abbreviations are necessary.*
  Okay. The line now reads:
  "[...] Leipzig Ice Nucleation Array (LINA, 90 x 1 µL) and Ice Nucleation Droplet Array (INDA, 96 x 50 µL)."

- *P3L30 INP analysis is...* → *Cumulative INP data is presented at temperatures of ...*
  Okay.

- *P3L30-31: Hartmann et al. (2019) observed...* → *The authors observed no alternation in the INP concentration over long-term period.*
  We chose to keep our phrasing, as it is more concise.

- *P3L31: Furthermore,* → *Instead,*
  We chose to keep our phrasing. "Instead" would indicate an opposite finding, but we feel the high short-term variability is an independent finding from the non-existing long-term trends.

- *P3L32: Please clarify what "dominate the total variability of the complete data set" means to the readers here. One may be able to guess, but the clarification would be appreciated.*

  We rephrased the sentence:
  "Furthermore, they found the "short-term" variability of INP concentrations from adjacent sub-year samples to be as large as or even larger than the total variability of the complete data set."

- *P3L32-35: this sentence runs too long. The reviewer suggests separating this sentence into two. For example - ... INP concentrations for the last few centuries. Their suggestion was to include...*

  Okay.

- *P4L3: write out FRIDGE.*

  Okay.

- *P4L22: B30. Complementary chemical profiles of...*

  Okay.

- *P4L25: Merge this sentence to the previous paragraph.*

  We are not quite sure what the reviewer meant by the comment.
  We deleted the line break.

- *P5L2-3: ... then split for the online chemical analysis and offline ion chromatography (IC) measurements, where discrete aliquots in vials were used (section 2.4).*

  We chose to keep our phrasing. In the suggested sentences the reader might think that IC measurements were performed right away, when in fact they were performed sometime later.

- *P5L4: thus covering → translating to*

  Okay.

- *P5L4: Further, depending on the exact...*

  Okay.

- *P5L5-6: Subsequently, the vials were refrozen and shipped to AWI to measure the concentration of major ions in order to complement the CFA measurements. Keep it simple!*

  Okay.

- *P5L7-8: Some of these samples → Some remained samples*

  We chose to keep our phrasing.

- *P5L11: The reviewer suggests deleting "Temperature variability ranged...15 hours."*

  Okay.

- *P5L13-14: ...were refrozen. (once again is repetitive of re:).*

  Okay.

- *P5L17: longitudinal pertains to vertical sections?*

  Yes.

  We deleted "longitudinal" for clarity.

- *P5L19: The reviewer suggests deleting "absolutely" – the sentence is good and makes sense without this accessory word, so not adding any value to the sentence. Perhaps let the readers decide on their own.*

  Okay.

- *P5L23: purpose-built*

  Okay.

- *P5L24-26: The reviewer suggests deleting "Additionally, trace-elemental...for this manuscript". If the data was not used in this study, then there is no need to report/mention in the reviewer's opinion.*

  Okay.

- *P5L31: are → were*

  Okay.

- *P6L2: are → were*

  Okay.

- *P6L2: The IC provides → In this study, the IC provided*

  Okay.

- *Comment: While the reviewer understands that everyone has their own style on how they use tenses in writing, the use of past/present etc. seems not consistent in this manuscript. The reviewer suggests the authors to improve the consistency on the tenses usage throughout the manuscript. Perhaps, the following site could help the authors: https://services.unimelb.edu.au/__data/assets/pdf_file/0009/471294/Using_tenses_in_scientific_writing_Update_051112.pdf*

  We thank the reviewer for noticing the inconsistencies in tense usage. We will carefully proof-read the manuscript again in this regard.

- *P6L6-: We placed a strong emphasis on having a data set with quasi-consistent time intervals for our samples (approximately decadal interval). Furthermore, our sample selection strategy was intended to consider the pre-industrial INP concentration vs. the INP concentration of the recent past (1960-1990).*

  Okay. "We considered these time intervals to be both meaningful and feasible." was added in between the suggested sentences.

- *P6L9:... in the latter time period to rightly match up sub-total sample numbers for each set.*

  We added the following clause to the sentence:
  "[...] in the latter time period to potentially enhance the statistical significance."

- *P6L12: ...as well as a couple of samples collected before and after it.*

  Okay.

- *P6L12-14: Please clarify what is meant by "Due to..." to the readers. Not intuitive to this reviewer.*

  We selected a subgroup of samples to be analyzed in FRIDGE according to the high-resolution CFA data. Some samples were selected, because there was a peak in the high-resolution dust or conductivity signal. However, the discrete INP samples are random means over several months around this peak, which means that a sample with a CFA peak does not necessarily have to have a high mean value. We will rephrase the sentence:

  "Due to the episodic nature of such an event and the fact that the INP samples were automatically collected as multi-month means, the sample containing the high-resolution peak signal does not necessarily need to have an extraordinarily high average value itself."

- *P6L15: ...were selected. These samples were typically...*

  We rephrase the sentence to:

  "Similarly, peak samples in the high-resolution signal of conductivity were selected. Large peaks in the electrolytic conductivity record are most often derived from high sulfuric acid deposition in the ice after volcanic eruptions."

- *P6L23-25: A majority (63%) of the analyzed samples averaged over a time period of 6 ± 2 months. The rest averaged over a shorter (26%) and longer (11%) time. Reads better this way?*

  We rephrase the sentence to:

  "The majority (63%) of the analyzed samples averaged over a time period of 6 ± 2 months. About a quarter of the samples (26%) averaged over a shorter time and 11% over a longer time."

- *P6L28: ...aerosol particles are activated to ice crystals by ...*

  Okay.

- *P6L30: The reviewer suggests the authors to briefly address the importance of droplet freezing. The question here is that - why was the droplet freezing mode selected and used rather than another? The readers would appreciate a justification.*

  We thank the reviewer for pointing this question out. First and foremost, immersion freezing is considered to be the most atmospherically relevant mechanism in heterogeneous ice nucleation for mixed-phase clouds (e.g Murray et al. (2012)). Moreover, using a droplet freezing assay (DFA) feels like the natural choice to study the ice nucleation ability of particles that are already immersed within ice core meltwater, especially considering that a DFA needs only few microliters of water. All other methods would require additional steps of particle generation (e.g. atomizer), which may introduce further contamination sources and would likely require more sample water. We added a few sentences to the manuscript:

  "We focused on the droplet freezing assay (DFA), because 1) immersion freezing is considered to be the most atmospherically relevant process in heterogeneous ice nucleation for mixed-phase clouds (e.g. Murray et al. 2012), 2) the use of a DFA seems to be the natural choice considering that the aerosol particles are already immersed within the ice core meltwater, 3) the technique requires only

a few mL of sample water, and 4) other methods would likely introduce further contamination sources through the particle generation setup (e.g. atomizer)."

- *P7L4-6: are → were (x3)*
  Okay.

- *P7L7: is decreased quickly → was quickly decreased*
  Okay.

- *P7L8: slowly lowered at...until all droplets were frozen.*
  Okay.

- *P7L10: is controlled → was measured (or was it really controlled?)*
  Okay.

- *P7L12: limit → minimize*
  Okay.

- *P7L13-16: Did the authors observe any half-or-less frozen droplets at given Ts? If so, how did the authors systematically judge the freezing moment/T?*
  The moment of freezing was registered automatically by the LabView software as freezing causes a significant change in brightness. However, sometimes the freezing of a droplet begins just as the images is saved. In these cases the software sometimes misses it, and would count the frozen droplet one image later. But we checked every image manually to account for this. However, it is possible that a droplet froze between two measurement images (which are 10 seconds apart). Therefore, the freezing temperature has an uncertainty of 1/6 °C at the freezing rate of 1 °C/min due to this effect.

- *P7L24-31: The reviewer thinks all future tenses should be changed to present.*
  Okay.

- *P7L30: Please provide an overall uncertainties in numeric terms, and discuss these here. The words "substantially" and "higher" seem too abstract.*
  We find it difficult to provide a general numeric uncertainty here, as individual freezing curves are substantially different from one another. Moreover, we cannot concisely predict how the cumulative INP concentration of a freezing curve would extrapolate at lower temperatures. Further, as stated in the text the underestimation is dependent on the temperature were the last droplet froze. For example, if sample a. was completely frozen at -24°C and sample b. at -28°C, the extrapolated cumulative INP concentration at -30°C would likely be much higher at sample a.

- *P8L2: The authors may want to recap the unique importance of 1977, 1680 & 1630 and provide the readers a brief justification of why they were picked for SEM analysis.*
  Unfortunately, as this was a novel measurement approach for us, the labor intensive SEM analysis was limited to a small number of samples in this study. We plan to increase the number of SEM samples future studies.

The three samples were selected for different reasons. The 1977 sample was selected as example for an IN active modern-era sample. The 1680 sample was selected, because it had an average INP concentration at -25°C and was in the middle of the time series. The 1630 sample was selected, because it showed extraordinarily high INP activity and we were interested to find out if we could identify the underlying reasons in the chemical aerosol signature of the sample.
We will add a short paragraph to the manuscript:
"The 1977 sample was selected exemplarily as an active modern-era sample. The 1680 sample was chosen for its average INP concentration at -25 °C, as well as being in the middle of the time series. The 1630 sample was analyzed with SEM, because it had an extraordinarily high INP activity at comparably warm temperatures."

- *Sect. 2.7: Briefly describe the operation conditions of SEM-EDX – beam intensity, WD, SS etc. Were these experimental variables all consistent for all analyses?*

  Yes, experimental variables were consistent for all three samples. We expanded the method description, which now reads:
  "For SEM-EDX analysis on each filter some 100 rectangular fields of about 100 µm x 100µm in the center of the filter were scanned and for all detected particles the size was determined and an EDX analysis (acceleration voltage: 20 kV, spot size: 4, acquisition time: 10 s, working distance: 10 mm) was performed. Using this procedure, particles down to approximately 250 nm were detected. Smaller particles will often be overlooked. This is also true for larger carbonaceous particles, because of their poor contrast on the polycarbonate filter."

- *P8L11-12: The reviewer suggests excluding "Smaller particles will...". Adding not much value to the section.*

  As reviewer 3 seemed to be confused which particles were collected for SEM analysis and which were likely lost during filtering, we think we should keep this sentence.

- *P8L16: review the state of the art of → reviewed several*

  Okay.

- *Sect. 2.8: In general, this section can be much more concise. Especially, P8L30-P9L2 seems containing repetitive information and, thus, could be excluded. Three most important sentences in this section are: P9L6 However,...; P9L7 Unfortunately,...; and P9L15 Therefore,... The reviewer suggests the authors to summarize the section by putting simple emphasis on these, and reduce the # of words. The reviewer defers to the point addressed in P9L18-21. No worries. The authors' method sounds.*

  We agree with the reviewer that the highlighted text passages are the most important sentences in this section. We understand that the presented approach of listing and addressing potential factors influencing the background signal is not strictly necessary and it may take the reader some time to find the most relevant bits concerning the actual measurement data. But we feel in that these uncertainties and measurement routines are often not clearly addressed in many publications. Also, this is the first first-author publication concerning the FRIDGE droplet freezing method, and we would like to be able to refer to this

method paper in our future publications. Therefore, we chose to be rather detail-oriented.

- *P10L19-20: This means as well... → This implies that INP concentrations may be higher in ice core samples than ambient INP concentrations at any given time. Or something similar?*

  We rephrase the sentence to:

  "This implies that INPs may be overrepresented in ice core samples compared to non-INPs or the ambient atmosphere at any given time."

- *P10L26-27: The reviewers agrees about INPs being preserved. The authors may add discussion of Beall et al. (Beall, C. M., Lucero, D., Hill, T. C., DeMott, P. J., Stokes, M. D., and Prather, K. A.: Best practices for precipitation sample storage for offline studies of ice nucleation, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-183, in review, 2020.).*

  We thank to reviewer for the suggested literature, which we were not aware of. Thanks to this note and a concern shared by reviewer 2 and 3, we now feel that we need to highlight possible losses of (high temperature) INP activity due to storage effects. Although we ensured a frozen storage at our laboratory, the samples needed to be melted and refrozen several times prior to the IN measurements, possibly deactivating warm INPs and thus lowering the cumulative INP concentration. Text passages were added to:

  Page 9, line 30: "However, recent studies indicate that sample storage (i.e. storage temperature) significantly affects the ice nucleation activity of fresh precipitation samples in the range of -7 C to -19 °C (Beall et al., 2020). For example, samples stored at room temperature lost on average 72% of their INPs compared to the freshly analyzed samples. An average INP loss of 25% was still observed, even when samples were stored at -20 °C. Storage time did only weakly affect the INP concentrations. Therefore, based on this study a loss of INP activity on the order of a factor of 2 – 5 is possible, if not likely for the ice core measurements presented here. Furthermore, it is likely that the warmer end of INPs were disproportionally affected by these disturbances, while cold-temperature INPs were likely more robust. However, as all the samples experienced the same sample history, relative changes within the ice core can still be interpreted."

  Page 10, line 27: "However, as previously stated, storage conditions may have affected the INP activation."

- *P10L31-34: Not adding much value to the section. The reviewer suggests removing this part from the manuscript.*

  We recognize that the quote does not add much to the manuscript. Although, we like the quote, we deleted the sentence from the manuscript.

- *P11L3: Where does this 'an order-mag.' come from? Please clarify in the text for the readers.*

  We think we give ample explanations in sections 2.9 and 2.10 why the conversion to atmospheric concentrations is uncertain. The phrase signaling that the conversion "should be interpreted only as an order-of-magnitude estimation", is added to the text to highlight these uncertainties and to sensitize other researchers, which might want to use the data for atmospheric modelling, etc.

- *P13L3: The reviewer accepts the idea of conversion. If the authors are confident it is only +/- 50%, the reviewer suggests massively cut # of texts/words in this section. In general, this section is hard to follow. Spending full 2 pages to derive seems a simple sub-conclusion (i.e., P13L6-7) seems overwhelming. You may list the typical value of each variable (A, v_dry, and epsilon) +/- 'reasonable' upper/lower ranges (that correspond to shape a blue square in Fig. 4) in a table format to reduce # of total words. For that matter, the reviewer wonders if Fig. 4 is really needed and meaningful. A different presentation (again, tabular format) may be considered.*

  We shortened the text substantially and moved the extended version into the Supplement. However, we feel that a visual representation of the conversion factor depending on dry and wet deposition gives the reader a good idea about the sensitivity of those uncertain parameters. Therefore, we prefer Fig. 4 to a table and chose to keep it.

- *P13L13-15: This paragraph seems not fitting here.*

  Okay.

- *P13L28: very steep freezing → local maximum in – or something similar*

  We chose to keep the phrasing.

- *P13L29-30: → We verified a reproducibility of our results by confirming two separate measurements agreed each other. This verification eliminated the contamination during our FRIDGE measurements. Does this what the authors mean?*

  We rephrased the sentence. The manuscript now reads:

  "We verified our results by reproducing the measurement of this sample. The separate second measurement confirmed the strikingly different freezing characteristics, thus eliminating a contamination during the FRIDGE measurements themselves."

- *P14L4: → they showed a frozen fraction of only 0.7% on average.*

  Okay.

- *P14L7: average in ice concentration → average* N_{INPice}

  Okay.

- *P14L9: Here → At this temperature,*

  Okay.

- *P14L10: From here onward,... → Next, our characterization of INPs at -25°C is specifically discussed.*

  We rephrased the sentence. The manuscript now reads:
  "Henceforth, the discussion of results is focused on the characterization of INPs at -25°C specifically."

- *P14L11-12: ...every single sample... → all samples showed some droplet freezing events at this T.*

  Okay.

- *P14L14: , so the reader can see → in order to clarify*

  We rephrased the sentence. The manuscript now reads:
  "[...] in order to illustrate the typical variation in the INP concentration, while still allowing for easy identification of differences in the absolute INP concentration level."

- *P14L15: The reviewer suggests deleting ", but is still..." – not much value added.*

  See above.

- *P14L16: arise from → can be inferred from*

  Okay.

- *P14L17: delete "somewhat" and specify/clarify what include "more recent samples" in the main text.*

  Okay. The manuscript now reads:
  "We find on average higher and more variable INP concentrations for the last couple of decades as compared to the rest of the time series."

- *P14L22: Yet, → Nevertheless,*

  Okay.

- *P14L30 moderate yet significant → notable*

  We kept the phrasing as is, because we think our phrasing is more precise.

- *P14L34-P15L2: Delete 'however' and re-write the sentence to clarify what the authors mean to the readers.*

  We rephrased the sentence and moved Tab. 2 to the Supplement:
  "When the data is grouped into subsets according to Fig. 1, we find that the correlation weakens for the 10 year samples and the modern day samples, but increases for special event and seasonal samples (Tab. S2)."

- *P15L6: We like to point out here → It is noteworthy*

  Okay.

- *P15L11: That being said, going forward → Regardless,*

  Okay.

- *P15L12-14: "The four..." – the reviewer could not understand what it meant. Please rephrase and clarify the sentence.*

  The sentence was added to clarify that samples with the years 1960, 1970, 1980 and 1990 originally belonged to both subgroups (10 years and 1960 to 1990), but in the analysis presented they are only included in the 1960 to 1990 subgroup. We now see that the sentence is redundant and confuses the reader more than it helps, so we removed the sentence from the manuscript.

- *P15L14—16: → The observed difference between pre- and post-1960 samples is based on Subramanian (2019), which defines the 20th century as the beginning of the Anthropocene. Keep it simple, and delete "Note, however,..." – not much adding in.*

  The above cited literature is an article from the News Feature from Nature and does only report about the scientific debate about when to start the Anthropocene. We now added a solid reference. The manuscript now reads:
  "The observed difference between pre- and post-1960 samples is based on Zalasiewicz et al. (2011), who propose to define the middle of the 20th century as the beginning of the Anthropocene."

- *P15L17-19: But, then, excluding it also biases the authors' data... It is an important outlier, correct? It can be still excluded, but the reviewer suggests the authors to provide a better (and more constructive) justification to exclude it in the text.*

  We changed the text to:
  "Furthermore, we excluded the sample from 1630 in most of the following analysis in favor of more consistent freezing spectra. The statistical outlier is certainly important, as it was the only sample that was completely frozen before reaching -22 °C. At this state, however, we cannot explain what caused its high IN activity (cf. section 3.2). Moreover, as stated previously a contamination prior to the INP analysis cannot be excluded completely for this sample. Including the outlier does not change the general results."

- *P16L1: delete "seem to"*

  Okay.

- *P16L7: Only 36 particles for 1977. Please provide a justification for this small #.*

  On this filter only a very small number of particles were detected during the SEM analysis in the analyzed filter center region. The border regions were not analyzed, because of a higher risk of artefacts. The reason for the very low number is not known.
  Page 16, line 15 now reads:
  "For example, only a very small number of particles were detected on the 1977 sample in the analyzed center region of the filter. Generally, the border regions were not analyzed due to a higher risk of artefacts."

- *P16L11-13: Please provide reference(s). "will be feldspars" sound awkward. Please rephrase it.*

  Although EDX analysis does not allow an unambiguously mineralogical phase assignment, the typical elemental ratios (e.g. Al/Si ratio) and the content of minor elements in the alumosilicate particles allow at least an appraisal of the present silicate classes. In this case most of the detected alumosilicates are most

likely feldspars (sodium and potassium feldspars), amphiboles and pyroxenes. Besides this, some quartz and clay minerals are also present. For a more profound and detailed phase classification Transmission electron microscopic (TEM) investigations could be performed. Such kind of investigations require a specific sample preparation and are very time intensive.
We rephrased the mentioned sentence to:
"[...] most of the detected alumosilicates are most likely feldspars (and here more sodium and potassium feldspars), amphiboles and pyroxenes."

- *P16L16: How did the authors define "fly ash" through SEM-EDX? Reference(s)?*
  The fly ash definition in SEM analysis is strictly following the morphological analysis. If "perfect" melting spheres of refractive particles are detected the particles will be classified as fly ashes. For particle types with high melting points (silicates, metal oxides) no other particles source (as the high temperature process producing fly ashes) is known producing "perfect" spherical particles beside volcanic activities. But even when particles from volcanic activity can also show "spherical-like" morphologies, they differ strongly in morphology and mixing-state from fly ashes.
  However, not all fly ashes are spherical, therefore not all fly ashes, but only the spherical ones, can be classified/identified in SEM analysis.

- *P16L17: No notable difference found here might be due to limited # of particles analyzed, correct? If so, it should be stated in the text.*
  Correct. The manuscript now reads:
  "Otherwise there was no obvious distinction between the modern-era sample and the other two samples with regards to their chemical composition, which might be due to the limited number of particles analyzed."

- *P16L27-28: does seem to follow → shows*
  Okay.

- *P16L33-P17L7: The reviewer suggests the authors to soften the tone regarding the annual cycle. Yes. It is nice to see the seasonal cycle exists in this subset of samples, but the authors might need to be careful on not generalizing it as a bold conclusion here. The authors need to make it clear in the text in this particular section that this applies to only what they have analyzed for. Otherwise, please provide a proper justification why the authors believe the seasonal cycle could persist for other eras.*
  Okay. The manuscript now reads:
  "These findings suggest that the INP concentration in this year was subject to the annual dust input in Greenland. As the seasonal variability in particulate dust number can be clearly detected throughout the entire core, we expect that such a seasonal INP variability will hold for the entire record. Future high-resolution studies will have to test this assumption. Bory et al. (2002) show that the main dust source in Northern Greenland is the Taklamakan Desert in Northern China. At the beginning of the monsoon season, the dust particles are transported to Greenland within a few days via the jet stream and cause the annual maximum dust input for Greenland in spring."

- *P17L1: How about an episode of dust along with Atlantic Monsoon? How about Iceland etc.? Suggested reading: Iceland is an episodic source of atmospheric ice-nucleating particles relevant for mixed-phase clouds, Sanchez-Marroquin, https://advances.sciencemag.org/content/6/26/eaba8137.abstract*

  We thank the reviewer for suggesting the interesting article. We agree that both Saharan and Icelandic dust are possible (episodic) contributors to atmospheric INPs reaching the Arctic. However, the literature consensus suggests that the listed East Asian deserts and the described mechanism are largely responsible for the dust input in Greenland. Nevertheless, we added a sentence to the manuscript:

  "Furthermore, episodic dust transport from the Sahara desert (Lupker et al., 2010) and Iceland (Sanchez-Marroquin et al., 2020) may have contributed as well."

- *P17L30-P18L4: The reviewer appreciates the authors being careful, honest scientists by these statements here and elsewhere in the manuscript. Nonetheless, this part (right before the conclusion!) may give a very negative impression about the authors' study to the readers. Scattered concern statements throughout the manuscript bothers this reviewer, at the least. The authors may compile their concerns here and there regarding all uncertainties in Sect. 2.9 in a brief manner. The readers would understand that the results come with uncertainties, and the authors do not need to be too sensitive to sound.*

  We did not want to overly interpret the data as there are a number of uncertainties. Therefore, we were cautious when stating and discussing the findings in the manuscript. However, we understand the point the reviewer is trying to make.

  Regarding the issue of a potential post-coring contamination, we now reanalyzed some existing Abakus (particle diameter >1 µm) and SPES (single particle extinction and scattering instrument, particle diameter <1 µm) ice core data, which we like to share. The Abakus was used on the B17 and the EGRIP S6 ice core. On the S6 core we also used the SPES instrument, which was not yet available, when B17 was measured:

  Abakus: Analyzing the Abakus data of those two independent ice cores we find an average twofold increase of the mean background concentration of particles larger than 1 µm (but also in Ca^{2+}) in the top 8 m, which roughly corresponds to the time interval of 1960 – 1990, compared to deeper / older data. Already at 20 – 30 m (around 1900), where the firn is still porous, we do not see such an increase. The seasonality in the top 8 m is not as clean as in deeper intervals, which is to be expected as there are lots of breaks and wicking effects within the top 8 m. However, the seasonality is still detectable and comparable in amplitude to intervals below 20 m or even below 60 m (solid ice). Obviously, we cannot rule out contamination effects with absolute certainty, but the existence of a distinct seasonal variation is a valid argument that the observed increase in the dust concentration may be atmospheric.
  We will try to resolve the seasonality of the porous firn in future INP studies.

SPES: The SPES data from the S6 ice core looks quite different. For particles smaller than 1 µm we observed the average background concentration to increase by a factor of 4 – 5 within the top meters of the ice core. Furthermore, there was no seasonal signal within the top 100 years of the S6 ice core. Therefore, we conclude that for these smaller particles the post-coring contamination of the porous firn is severe. At this state, we do not know what kind of particles they are, but the mean diameter of the number size distribution is about 0.6 µm.

In conclusion, the INP results seem to agree sufficiently to the observations made by the Abakus, which sees an average twofold increase for the 1960 – 1990 interval in B17 (and S6). One could cautiously argue that therefore the INPs seem to reflect the mineral dust input of particles larger than 1 µm. However, as we have seen by the SPES data (from another ice core), a contamination effect is likely for particles smaller than 1 µm (and cannot be excluded completely for larger particles). As we did not observe the 4 – 5 times increase in INP concentrations as the SPES did for particles smaller than 1 µm, we expect that these contamination particles are no particularly active INPs in the investigated temperature regime, either due to their size, which might be substantially lower than 1 µm, or their surface structure, morphology or chemical composition, etc.
Regardless, we carefully read the manuscript again and removed some repetitious sections that mentioned a possible post-coring contamination, while adding to other text passages. Below we now list each instance, where the possible effect was mentioned, and describe if the passage was kept, removed or changed.

Abstract: "[...] or some post-coring contamination of the topmost, very porous firn."
We removed the text passage from this section.

Section 2.9: The effect was not yet mentioned.
We now introduce the effect in Section 2.9 (Other uncertainties).
Page 10, Line 4 now reads: "Specifically, we like to emphasize that the topmost part of the ice core is made up of relatively porous firn, which is more prone to post-coring contamination of dust during storage as compared to the rest of the ice core. Preliminary results of two particle counters (Abakus: spherical diameter > 1 µm, SPES: spherical diameter < 1 µm) from the B17 ice core (only Abakus) and the EGRIP S6 ice core (Abakus and SPES, 75.62° N, 35.97° W, 2702 m asl, C. Zeppenfeld, personal communication) suggest that a contamination effect is likely for particles < 1 µm and rather unlikely for particles > 1 µm. However, post-coring contamination still cannot be fully excluded for the latter measurements."

The effect was introduced in Section: 3.1: "It is noteworthy that the topmost part of the ice core is made up of relatively porous firn, which is more prone to post-coring contamination of dust as compared to the rest of the ice core. Unfortunately, we cannot entirely exclude the possibility that differences emerged or are enhanced due to post-coring contamination of the firn, as the ice core was stored for some time, despite the CFA decontamination technique."

We now only mention the effect shortly here.
Page 15, Line 7 now reads: "Unfortunately, despite the CFA decontamination technique we cannot entirely exclude the possibility that differences emerged or are enhanced due to post-coring contamination of the porous firn, as the ice core was stored for some time. Preliminary measurements (cf. section 2.9) found a twofold increase of particles larger 1 µm in the top 8 m (roughly the time interval of 1960 – 1990) compared to older intervals, which does seem to match the results observed by the INP measurements. Further, a distinct seasonality could be established for the dust measurements of the top layers, which argues against a strong contamination effect."

Later in 3.1 (after the revision): "If there was, however, only little influence by post-coring contamination and the latter two listed effects, the findings suggest that certain particles that are ice nucleation active in a mid-supercooled temperature regime may be more abundant in today's atmosphere."
We kept the mention of the effect here to transition to the next topic (i.e. which INPs could be enhanced in today's atmosphere).

Section 3.4: "However, we cannot fully rule out post-coring contamination as the cause for the observed differences."
We removed the text passage from this section.

Conclusions: "Alternatively, differences may have been caused by post-coring contamination, which is likely more relevant for these samples as they stem from the more porous firn layer."
We removed the text passage from this section.

- *P17L23: Fig. 11 tells the reviewer that the diversity may derive from the concentration and size of dry & wet deposited particles rather than the listed differences? The variability due to composition is ruled out in Sect. 3.2, correct? Please clarify.*

  The stated line gives possible explanations why our range and means of absolute INP concentrations are different from what Hartmann et al. (2019) observed in two Arctic ice cores. After reading the comments of all reviewers, we added storage effects to the list. Then, as we understand the reviewer refers in the comment to the diversity of INP concentrations that is observed in our data set specifically, which is another matter. We agree that the concentration and size of deposited particles are likely a driver of the INP concentration in the ice core. However, we don't think that the results of section 3.2 definitely rule the chemical composition of particles out here. We explicitly say that "[...] due to the low number of analyzed particles, we were unable to determine significant differences in particulate composition of the particles and size distribution in the three samples." The sentence now reads:
  "This disparity may arise from experimental (droplet volume, etc.), methodological (e.g. sample storage conditions) and or geographical differences, which may affect the deposition mechanisms and efficiency"

- *P18L11: particularly → significantly or substantially?*

  We chose the keep the phrasing.

- *P18L12: group → selected subset*

  Okay.

- *P18L14: recap and specify "certain aerosol species" here for the readers.*

  Okay. The manuscript now reads:

  "Furthermore, we found significant correlations between concentrations of INPs and the insoluble particle concentration > 1.2 µm, Ca2+ concentration and the conductivity for a broad range of temperatures."

- *P18L20: Delete "several mechanisms can be considered by which". The sentence makes sense without it.*

  Okay.

- *P18L31: The reviewer strongly agrees ☺*

- *P19: Perhaps, one of top priorities for the future ice core INP research includes the assessment of particle size distributions in liquid samples by DLS etc. The authors may elaborate it as an outlook? Connecting INP properties to aerosol propensities may resolve some raised concerns?*

  We agree with the reviewer that the proposed method of dynamic light scattering to gain information about the particle size distribution in liquid aerosol samples is certainly very interesting and promising. However, as far as we know, this method has not yet been tested in ice core studies. Furthermore, the low number of particles might be challenging for the instrument. We added a sentence to the manuscript:

  "[...] Furthermore, we plan to expand our ice core analysis to include a more rigorous, systematic study analyzing the chemical and morphological composition of insoluble aerosol particles as well as their size distribution by scanning electron microscopy. Particle size distributions of liquid samples may also be attainable by the dynamic light scattering and the single particle scattering and extinction method."

- *Tables 1 & 2: Add "Temperature (°C)" as the first column header, and delete °C from the send row.*

  Okay.

- *Table 2: What are "dust, volcanic and seasonal"? Please clarify within the table caption.*

  We added a reference to Fig. 1 (and moved Tab. 2 to the Supplement).

- *Fig. 1 caption: → Time coverage of the samples selected for assessing IN properties.*

  Okay.

- *Fig. 1 caption: longer → long*

  Okay.

- *Fig. 2: Adding the least active spectrum from the core sample (P9L14) may increase the visual importance of this figure.*

  Okay.

- *Fig. 3: The authors may superpose the 1:1 ratio line on top of the fit line. Doing such reinforce the authors' point in a visible manner.*

  Okay.

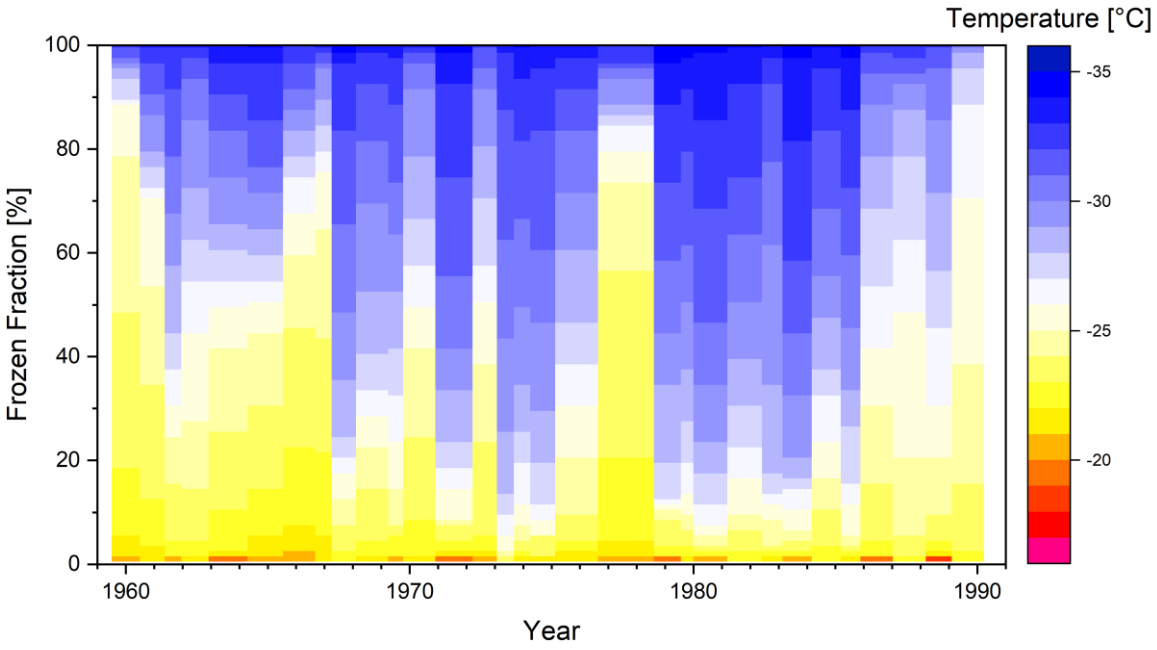
- *Fig. 6: INP /L ^-1_ atm/ or N_ INP_ atm? Perhaps, the authors may choose one way to improve the consistency throughout the manuscript.*

  Okay.

- *Fig. 7 caption: "However, data..." – the reviewer did not understand this. Please clarify.*

  The sentence points out that the underlying data (i.e. 31 samples that averaged over time periods of 0.24 years to 1.96 years, average 0.58 years, see Fig. 1b) was interpolated in time to create visually regular yearly columns of frozen fraction data, similar to what is presented in Fig. 5. After the interpolation 30 yearly columns are shown with the same width. We added the original (non-interpolated) data here and in the supplement (Fig S.2) for clarification. The caption of Figure 7 now reads:

  "However, data points are interpolated in time to generate yearly columns of regular width. The non-interpolated data is presented in Fig. S2."



- *Fig. 7 caption: Delete "Note, that".*

  Okay.

- *The reviewer enjoyed reading it. Hope some of suggestions/comments made here help the authors.*

  We are glad that the reviewer liked the manuscript. Again, we thank the reviewer for their valuable suggestions, which will definitely improve the paper.

Literature

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