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.

Specific and technical comments:
P1L13: The reviewer suggests the authors to specify dp is in a spherical diameter metric here.

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.

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

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

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

P2L27: Although…straightforward, → Evidently,

P2L27: seen → implied

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.

P2L34: Biomass burning aerosol is…least potential contributor to anthropogenic INP.

P3L13: Indeed, soil dust, in part derived from agricultural systems/practices,… Is this what the authors meant? Feel free to modify it.

P3L17: …global land area, of which approx. 9% were identifies…

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.

P3L25-26: write out LINA and INDA? They appear once only, so it seems no abbreviations are necessary.

P3L30 INP analysis is… → Cumulative INP data is presented at temperatures of …

P3L30-:Hartmann et al. (2019) observed… → The authors observed no alternation in the INP concentration over long-term period.

P3L31: Furthermore, → Instead,

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.

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…

P4L3: write out FRIDGE.

P4L22: B30. Complementary chemical profiles of…

P4L25: Merge this sentence to the previous paragraph.

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).

P5L4: thus covering → translating to

P5L4: Further, depending on the exact…

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!

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

P5L11: The reviewer suggests deleting “Temperature variability ranged…15 hours.”

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

P5L17: longitudinal pertains to vertical sections?

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.

P5L23: purpose-built

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.

P5L31: are → were

P6L2: are → were

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

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

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).

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

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

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

P6L15: …were selected. These samples were typically…

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?

P6L28: …aerosol particles are activated to ice crystals by …

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.

P7L4-6: are → were (x3)

P7L7: is decreased quickly → was quickly decreased

P7L8: slowly lowered at…until all droplets were frozen.

P7L10: is controlled → was measured (or was it really controlled?)

P7L12: limit → minimize

P7L13-16: Did the authors observe any half-or-less frozen droplets at given *T*s? If so, how did the authors systematically judge the freezing moment/T?

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

P7L30: Please provide an overall uncertainties in numeric terms, and discuss these here. The words "substantially" and "higher" seem too abstract.

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.

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?

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

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

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.

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?

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.).

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

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

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.

P13L13-15: This paragraph seems not fitting here.

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

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?

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

P14L7: average in ice concentration --> average N_{INPice}

P14L9: Here → At this temperature,

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

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

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

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

P14L16: arise from → can be inferred from

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

P14L22: Yet, → Nevertheless,

P14L30 moderate yet significant → notable

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

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

P15L11: That being said, going forward → Regardless,

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

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.

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.

P16L1: delete "seem to"

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

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

P16L16: How did the authors define “fly ash” through SEM-EDX? Reference(s)?

P16L17: No notable difference found here might be due to limited # of particles analyzed, correct? If so, it should be stated in the text.

P16L27:-28: does seem to follow → shows

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.

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
A. Sanchez-Marroquin
https://advances.sciencemag.org/content/6/26/eaba8137.abstract

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.

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.

P18L11: particularly → significantly or substantially?

P18L12: group → selected subset

P18L14: recap and specify “certain aerosol species” here for the readers.

P18L20: Delete “several mechanisms can be considered by which”. The sentence makes sense without it.

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?

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

Table 2: What are “dust, volcanic and seasonal”? Please clarify within the table caption.

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

Fig. 1 caption: longer → long

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

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.

Fig. 6: INP [L^-1_atm] or N_INP_atm? Perhaps, the authors may choose one way to improve the consistency throughout the manuscript.

Fig. 7 caption: “However, data…” – the reviewer did not understand this. Please clarify.

Fig. 7 caption: Delete “Note, that”.

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