

Response to Anonymous Referee #1

First of all, we thank the referee 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 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 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.

Review of “Long-term INP measurements from four stations across the globe” by Schrod et al.

General comments:

The authors made enormous amount of efforts tackling the current challenges of the INP research community – wide spatiotemporal coverage of ambient INP measurements. This reviewer is impressed with a comprehensiveness of this work (for 1212 samples) as well as persistence and articulation of the authors, and supports publication of this manuscript in ACP. The results and discussions provided in this manuscript tightly fit in the scope of ACP. The reviewer has only technical (some are minor) suggestions to make (see below). But, the reviewer noticed different writing styles/tones involved over different sections (before/after Sect. 2.3.). Consistency in writing will improve the readability as well as importance of this paper even more.

We are grateful for the positive feedback of the reviewer. We hope to improve the readability (and substance) of the manuscript by implementing the suggestions of the reviewer.

Specific and technical comments:

- *P1L4: → Unfortunately, only a few ...*
Okay.
- *P1L14-15: This statement introduces a multitude of perspectives – one may consider physicochemical properties have negligible impact on INP abundance/propensity, thereby ambient INP estimation could be rather simple than ‘complex’. This may be true and somehow supported by what the authors found (i.e., P1L9-11 & P1L18-19; great statements, by the way). Perhaps, incorporating this counter-thought (on top of*

what already exists) in an abstract and other parts in the main text would increase the readability/flexibility to both authors and readers.

Admittedly, one could argue as the reviewer proposes here. We argue for a “complex” and unresolved interplay of factors determining the INP concentration as we did not find an individual parameter (i.e. particle number concentration $> 0.5\mu\text{m}$, PM_{10} , etc.), which managed to predict the number of INPs to a satisfyingly high degree at any one site let alone all sites. Hence, we think that the observed high short-term variability is a clear sign that we do not sufficiently know all processes involved, or at least that the supporting physical and chemical parameters at hand did not cover all relevant aspects of the ice nucleation process.

- *P2L6-8: Depending on ... - the reviewer is not sure if this statement is adding any meaningful aspects in this paper. The authors may consider removing this statement. The CCN is not discussed in tandem with INP much in this manuscript.*

Okay.

- *P2L9: Non-biological organics are deemed to be overlooked here. The authors may review Knopf et al. & Kanji et al.?*

We now include non-biological organics in the list. The sentence now reads:

“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).”

- *P2L19: Vertical distribution – very good point. This is somehow one of the things INP community has been missing for a long time in the reviewer’s opinion. This should be pointed out in the outlook section?*

We agree with the referee that vertical distributions of INP need to be explored more by the community, as INP concentrations at heights where clouds form may differ significantly from those at ground level (e.g. Schrod et al., 2017). We added a paragraph to the outlook (see later).

- *P2L21: ...in identifying globally relevant INP... → ...in identifying some or potentially atmospheric-related INP...*

We changed the phrasing to:

“...in identifying some of the INP species of global relevance...”

- *P3L1: The reviewer totally agrees with this statement. This statement is a nice complement to previous studies. Nice writing.*

Thank you.

- *P3L2-17: Perhaps summarizing the examined temperature and the n_{INP} ranges from these previous studies in a tabular format with minimum explanation instead of prolonged texts would increase the readability of this section.*

We understand the argument for a better readability, yet we feel that the paragraph and the entailed efforts made in the early years of ice nucleation research deserve some space in a manuscript, highlighting the importance of long-term INP measurements.

- *P3L20-21: Yes. This is a very good motivation statement. Good job.*

Thank you.

- *P4L6-8: Seeing long/lat coordinates for these locations in their first appearances would be nice. The reviewer is aware these coordinates appear later on. This is just a suggestion from the reader's perspective. The authors can decide what to do.*

We have added the coordinates within the introduction as well.

- *P4L11: Please clarify what "semi-automated" really means. Please also clarify how the samples were stored while transporting here. Frozen at a certain temperature all the way? The reviewer is aware that the authors mention an insignificance of storage method on their INP characterization in P5L18-19. Perhaps, transportation and storage discussion can be combined here or P5?*

We think there is no need to add a very detailed description of these sampling related specifics here in the introduction. Much of these questions are answered (i.e. "semi-automated" sampling) within the following section (see section 2.2 Aerosol sampling). We will, however, add to this section, addressing the items raised by the referee. P5L19:

"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."

- *P4L17: factors could include local dynamics, thermodynamics, large scale meteorology, and/or a combination of any?*

All these factors surely influence ice nucleation in the atmosphere, yet we largely did not consider these in the analysis as we feel they are outside the scope of this manuscript. When formulating this sentence we were mainly thinking about aerosol species and sources.

- *P4L26-27: So is this correction incorporated/applied in relevant INP # in this study? Please state it if so.*

Yes, it is. The sentence now reads:

"The PEAC7 collection efficiency has been found to be about 60%, independent of particle size (Schrod et al., 2016). Accordingly, a correction factor of 0.6 has been applied to the data."

- *P4L27-: It would be meaningful to have a discussion of all inlets configuration and properties (e.g., length, flow rate – if any, cut-size – if an impactor was used in part, transmission efficiency, transmitted aerosol particle size range etc.) from individual sites here (rather than in Sect. 2.3). Maybe, the authors can use a table summarizing the inlet config. characterization (if done/any). Also, listing previous INP research done at the sites would be meaningful info for the readers.*

We recognize from both reviews that more care should have been taken when describing the inlet configurations. 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. P4L27:

“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

We added to 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.”

We added to 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).”

As for previous INP research at the sites: This part is discussed in the respective sections 3.2.1, 3.2.2, 3.2.3 and 3.2.4 when available.

- *P5L2: “for use within an INP monitoring network” seems misleading – sounds like a strong promotion. The reviewer suggests altering this to → to collect aerosol particles*

at multiple field sites for subsequent offline INP analysis. This way, the tone would be reduced, and the point can be made for the concurrent work.

Okay.

- *P5L12-13: Please elaborate the difficulties a bit further.*

The sample substrates need to be thoroughly cleaned before use as contaminant particles may introduce significant background freezing. This is a problem observed especially at lower temperatures (i.e. ≤ -30 °C). During the stated time frame we struggled to meet the workload associated with the cleaning procedure and INP analysis. As a result we were only able to guarantee clean (low background) substrates for temperatures ≥ -25 °C. The manuscript now reads:

“Between October 2015 and February 2016 some unexplained contamination in the process of wafer cleaning prevented to clean substrates to below the desired background level of INP at the lowest temperature. As a consequence no data below -25°C are available for this period.”

- *P5L13: Representativeness of local noon & short sampling time is questionable (the reviewer is aware that the discussion is given later on). On the other hand, the reviewer supports the best practice of pursuing consistency with this strategy employed by the authors for this study. Perhaps, such should be mentioned here to justify the strategy. The readers will understand.*

We added the following to the description of the sampling strategy:

“However, the level of representativeness of the deployed sampling strategy is difficult to assess (see discussion). Yet, the pursued sampling protocol ensured a consistent data base.”

- *P6 Sect. 2.3.: Very informative and detailed. But, this section seemingly better fits as SI in the reviewer’s opinion. Especially, P6L20-P7L10 & P7L23-P8L20 seem not relevant to the main focus of this study. Putting a subset in SI at the least would even increase the readability – the reviewer’s suggestion is based on the readers’ perspective.*

We understand that the site descriptions are unusually long in comparison to other studies, but we feel it is important to this manuscript to emphasize the contrasting features of the measurement stations by including a rather thorough characterization here. Especially, as one of the main findings emerging from this study is that the deposition INPs do not seem to differ all that much from site to site, despite these differences.

- *P10L15: Delete (incomplete).*

Okay. Note that we try to address the matter of nucleation mechanisms addressed more clearly in the revised manuscript in response to the feedback of reviewer 2.

- *P10L10-11, 16-12, and 27-29: The reviewer is impressed with these statements. Congratulations on finding these.*

Thank you.

- *P11L1: Besides storage effects, inconsistency in inlet configurations and IN mechanisms can also play a role in the reviewer’s opinion. If a proper inlet is not used for aerosol particles sampling, sampling efficiency of the sampler could be affected by local*

turbulence and other dynamic/thermodynamic conditions (e.g., sampler port get frozen/clogged). These points should be incorporated, otherwise the readers might be misled.

We added the following to P11L1:

“Furthermore, differences between the inlet configurations of the individual sites may have influenced the particle sampling process (see section 2.2).”

- *P11L7-8: Add reference(s) for bio-INPs that the authors are mentioning here or elaborate it.*

We have added to this text passage:

“For example, O’Sullivan et al. (2018) found that immersion INP concentrations at -20 °C at a northwestern European site were reduced by more than a factor of 2 in 59 % of the cases when samples were heated to 100 °C. For warmer temperatures the reduction was found to be significantly higher.”

- *P11L9-16: So what is the implication of such a strong IS dependence? Are the authors trying to point out the condensation/droplet freezing is more predominant as compared to deposition?*

As reviewer 2 (Paul DeMott) points out, an INP activation spectrum that is not dependent on temperature implies that immersion mode INPs were not represented in the data. We will try to address this point more clearly in the revised manuscript (see the revised text in section 2.1 and 2.3.1, and the responses to reviewer 2).

- *P11L19-22: This part is speculative. The reviewer sees lots of “may” words. But, it does justify that the sentence can remain speculative. Please introduce some references/citations to support the authors’ idea at the least.*

We agree that the part is speculative and we believe the chosen phrasing makes this clear to the reader. Section 2.4 lists some references that indicate that long-range transport of mineral dust is a regular feature of the AZ and MQ sites.

We added a sentence (P11L22):

“Our view of a generally higher abundance of mineral dust at the low latitude sites MQ and AZ as compared to the high latitudes of SB and TO is supported by dust observations from surface stations (Prospero et al., 1996), remote sensing (Kaufmann et al., 2005) and models (Zender et al., 2003; Lee et al., 2009).”

- *P11L28-29: Yes. The reviewer agrees.*

Good.

- *P12L8: That said, -> However (too informal for a scientific journal).*

Okay.

- *P12L8-9: The source of INPs is important, but how aerosol particles are sampled at the sampling location through what sort of inlets is also an important source of potential data variation. See the reviewer’s comment regarding an inlet above.*

We agree. See previous responses.

- P12L16: which one is bimodal? Please clarify this in the text.*

There is no clear indication of a bimodal frequency distribution, yet, as stated, some of the data hint at it (e.g. SB).
- P12L19-19: distribution analysis with higher sensitivity at high Ts would be a good future work (may be incorporated in depth in an outlook section?).*

We agree with the reviewer that future works could focus on the frequency distribution of the INP concentrations for warm temperatures. Furthermore, we would like to point the reviewer to a publication by Welti et al. (2018) that shows a similar figure for INP measurements in the subtropical marine boundary layer at temperatures up to -8 °C. We have added a paragraph to the outlook (see later).
- P12L27-29: This sentence is running too long, diluting an important message. The reviewer suggest breaking it down and carefully reformulate this sentence.*

We rephrased the sentence:

“Therefore, local species of plants or bacteria may be less likely to have evolved traits that induce freezing. It has previously been posited that some microbiology (e.g. bacteria like *Pseudomonas syringae*) gain an evolutionary advantage by being able to induce freezing (Morris et al., 2014).”
- P13L9-10: background air masses mean local ambient T and RH etc.? The authors may want to add “More discussion of insignificant role of local sources is provided in the next section” or something similar to smoothly guide the readers to e.g., P13L31.*

We are not sure what is meant by reviewer’s first comment. We believe that the data supports the idea that the measured deposition INP concentrations are largely determined by large-scale background air mass movements. The ambient conditions (T and RH) define if and how many INPs will be activated to ice crystals.

We have added a short sentence to guide the reader to section 3.2:

“More discussion of the site specific local sources and characteristic features is provided in the following section.”
- P14L1-13: Though the reviewer finds this part (bio aerosol - INP - precipitation interactions) very interesting, some parts sound speculative simply due to the lack of sufficient data – e.g., rain intensity, wind/gust condition, rain duration etc. etc. What is discussed in this sub-section seems supplementary, not the main point of this study. The reviewer suggests either elaborate it rigorously or eliminate it completely.*

We believe that although we cannot present sufficient evidence for the importance of biology-precipitation interactions in our data, the discussion would lack a potentially substantial INP feedback for AZ, if we completely removed the discussion. We have shortened some of the text passages. The manuscript now reads:

“[...] Another way to interpret the anti-correlation of AF and biomass burning markers is by coupling the metric to precipitation rates. There are several intricate interactions of note here. On one hand more precipitation leads to higher aerosol particle (and INP) removal by wet deposition. Moreover, enhanced precipitation during the wet season can largely prevent wild fires and the accompanied particle emissions in the first place. On the other hand, it has

been postulated previously that precipitation may be a driver of biological INPs (Huffman et al., 2013), and large tropical rainforests like the Amazon have been highlighted in that regard (Morris et al., 2014). However, the processes responsible for the release of the biological particles have not yet been deciphered in detail.”

- *P14L18: likely → presumably*
Okay.

- *P14L24: Then, the local source seems important... This seems contradicting to the point made in P13L9-10. Please clarify.*

On one hand we observed that during a distinct LRT episode INP concentrations were significantly correlated to mineral dust particles. However, even when no clear dust transport was registered (e.g. by back-trajectory analysis and particle measurements), electron microscopy analysis of six samples indicates that mineral dust is responsible for about half of the INPs at AZ. Therefore, we argue that there seems to be a well-mixed and diluted background concentration of mineral dust INPs at all times present at AZ. We have rephrased the text to make our argument more clear:

“However, mineral dust may be a relevant INP in this region even in the absence of distinct LRT events: An analysis of the average composition of INPs of six samples (4 in April 2016, 2 in December 2016) using scanning electron microscopy (SEM, Figure 9), identified that nearly half of the particles that activated to ice crystals in FRDIGE were mineral dust. This finding suggests that there seems to be a well-mixed and diluted background concentration of mineral dust INPs at all times present at AZ. The diameter of most of the INPs investigated by SEM in this study was between one and a couple of micrometers (Figure 9b).”

- *P14L29-31: Very good statement.*
Thank you.

- *P16L5: Given → Due to*
Okay.

- *P16L5: ...atmosphere, the Arctic...(comma)*
Okay.

- *P16L13-27: The authors may consider mentioning about a more recent study by Rinaldi et al. (2020 - <https://acp.copernicus.org/preprints/acp-2020-605/>). The reviewer believes that findings of Rinaldi et al. (Ny-Alesund, Gruebadet station through a semilaminar flow TSP inlet during 2018) are consistent with what is presented in this study (2015-2017). Another place to potentially add Rinaldi et al. is on P17L11 in addition to Welti et al. (2020).*

We thank the referee for the suggested reading. We have added a paragraph:

P16L23: “However, a recent study by Rinaldi et al. (2020) did not observe a distinct seasonal signal in their INP measurements between -15 °C and -22 °C in the spring and summer of 2018 in Ny-Alesund. 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.”

P16L27: “Rinaldi et al. (2020) present evidence that Arctic INP concentrations are influenced by sources of marine biological INPs by providing a spatio-temporal correlation analysis between Chlorophyll-a fields from satellite data and a trajectory model.”

- P16L32: → ... anthropogenic Arctic Haze phenomenon during our study period. The reviewer supports the authors’ view, but the authors may want to reduce the tone. Otherwise, it may sound like a personal attack even without an intention. Just a suggestion to be fair on everyone in our community.

We meant to achieve quite the opposite effect here. In fact, we are rather a little concerned with the quality of our data due to the lack of a seasonal feature, as is frequently reported by others. We had hoped to get this message across by the last sentences of the paragraph (see P16L33 and following).

We have added the word “concerning” in P16L34:

“The concerning lack of meaningful correlations and/or seasonal trends may be in part related to a relatively poor signal-to-noise ratio in our SB measurements.”

- P17L4-9 & P17L22: Very good summary – the reviewer’s additional hope is a consistency in an inlet sampling system.

We now list inconsistencies in the inlet system as one cause of uncertainty in P17L32:

“However, when using the presented data one should be aware of the substantial limitations of the conceptual aspect of the approach and the uncertainties that are inherent in the aerosol sampling and INP measurements themselves.”

- P18L17-18: The reviewer disagrees. The finer time resolution of INP measurements for prolonged period of time with a reasonable detection - perhaps by semi-autonomous technique as mentioned towards the end of this section by the authors - is an ultimate goal/outlook for ambient INP measurements in the reviewer’s opinion. With a long(er) sampling time, researchers would overlook subtle change in INP episodes or local dynamic condition that has certain roles on INP propensity.

We agree with the reviewer and point him/her to the very next lines (P18L19 and following).

There are quite more important things to be listed as more specific future study ideas out of this study (e.g., inlet consistency, P2L19, P12L19-19 etc.). These could be addressed in this section.

Other general outlook can be made, but the authors may look through Murray et al. (2020 -<https://acp.copernicus.org/preprints/acp-2020-852/>), and adapt the authors’ ideas on top? Just a suggestion.

We thank the reviewer once again for sharing this excellent suggested reading. We feel it is beyond the scope of our manuscript even to attempt to fully and satisfyingly include all the listed needs of future INP research as done by Murray et al. (2020). However, we now refer to the paper to direct the interested reader

to the more extensive list. We have added a paragraph to the end of the manuscript:

“In addition to the goal of establishing more long-term global observations of continuous INP concentrations there are certainly other important areas for future research to address. For example, as most measurements are conducted at ground level, we believe there is a need to systematically study the vertical distribution of INPs – for example at heights where INPs are transported over long-ranges and/or where cloud formation occurs. Moreover, more extensive data sets from long-term INP monitoring might shed light on what mechanisms result in the observed log-normal INP frequency distributions (and departures from ideality etc.) as presented here and, for example, by Welts et al. (2018). Murray et al. (2020) has recently enumerated many crucial areas into which future INP research should delve. First and foremost, the authors emphasize the need to accurately implement ice nucleation related cloud-phase interactions in climate models in order to predict future climate scenarios correctly. We gladly refer the interested reader to Murray et al. (2020) for a more extensive list of future ice nucleation related research questions, as is presented in this study.”

- *P19L8: Möhler et al. may become publicly available soon. The authors may keep an eye on it, or touch base with Dr. Möhler.*

We have updated the reference from the Lacher et al. (2019) conference abstract to the newly available Möhler et al. (2020) paper.

- *The reviewer enjoyed reading this paper. Hope some of suggestions/comments made here help the authors (and future readers).*

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

Literature

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