

Referee comments 2

We thank referee 2 for the valuable feedback on our manuscript acp-2022-21. In response to the questions and suggestions, please find our answers and corrections listed below. **referee's comments are reproduced in bold** and author responses in the normal font; *extracts from the original manuscript are presented in red italic*, and *from the revised manuscript in blue italic*.

Review of "Predicting atmospheric background number concentrations..." by Li et al.

The authors report INP measurements at Ny-Alesund (Svalbard) at temperatures between 0 and -30 °C for 12 weeks (Oct-Nov and March-April). They did not see a significant difference in INP concentrations between Oct-Nov and March-April. In addition, the results fall within the range of INP concentrations previously reported for the Arctic. Also, they show that parameterizations developed for mineral dust, sea salt aerosol, and bioaerosols over pine forests do not predict the measurements well. Also, other parameterizations developed from measurements in other regions do not describe the measurements well. In addition, they fit their data as a function of temperature and suggest that this parameterization could be used to describe background number concentrations in the Arctic.

The measurements at Svalbard are important, and I congratulate the authors for generating an important dataset. I think it is also useful to show that these measurements cannot be reproduced with previous parameterizations, even though the disagreement is expected since the parameterizations were developed for other regions or for specific types of aerosols. I support the publication of this part of the manuscript, although publication as a Measurement Report may be more appropriate than publication as a Research Article.

We thank the reviewer for their positive evaluation of our manuscript.

Like Referee #1, I have major concerns about the section that describes the new parameterization of the authors' data. The authors are suggesting that their measurements from one location in the Arctic and during only 12 weeks can be used to predict background concentrations for the entire Arctic. To me, it does not make a lot of sense to use measurements from one location in the Arctic and during only 12 weeks to develop a parameterization for the entire Arctic, especially when some studies have shown a seasonal dependence on INP. If one wanted to make a parameterization for the entire Arctic, it would make more sense to use all the currently available INP data collected in the Arctic. I am not suggesting doing this, since INP concentrations are expected to change with season and location in the Arctic (at least in some cases), and one parameterization using all the previous measurements would miss this variability.

We agree with Reviewer 2 in this regard and have toned down the statements regarding the general applicability of our parameterization for the Arctic. Rather, we specify that our data fills in a gap in the literature for INP concentrations during the transition seasons (fall and spring) where we believe our data and parameterization are most useful. This point also explains the absence of seasonality observed in our data set compared to those reported in the literature, which are mostly comparing summer to winter seasons.

Below are specific comments:

Figure 7 shows that the parameterization developed by the authors is off by approximately 2 orders of magnitude in some cases. To claim the new parameterization is doing a good job for the entire Arctic seems to overstate the power of the new parameterization. Furthermore, if you fit all the previous Arctic data, you would get a different parameterization and one that would be likely more applicable to the whole Arctic, since it would be based on measurements for different locations and many different times of the year. Why develop a parameterization from just one location and for a very limited time?

We agree with the reviewer here that to develop a parameterization valid for all Arctic regions and all seasons, it would make sense to use all available data. To tackle this, we now specify in the conclusions section (see lines 314-316 in the revised manuscript) and the abstract (lines 10-15 in the revised manuscript) of the revised manuscript that our parameterization is useful for the transition seasons and explain that this could be one of the reasons why we do not observe a seasonal effect in INP concentrations. Furthermore, to justify the use of the simplistic T-dependent parameterization as we have presented in our paper, we would also require showing that all other INP concentrations from the literature in the Arctic are also not well predicted by their respective aerosol and meteorological parameters. However, this would be beyond the scope of the current paper, as it would require also collecting the raw high time resolution aerosol and meteorological data from all published studies.

Page 1, line 10. Consider changing “not feasible” to “not successful”.

We agree with the reviewer and have changed the wording accordingly (See line 12 in the revised version).

Page 4, lines 61-63. “This parameterization will help evaluate the role of cloud phase interactions in Arctic MPCs, and contribute to the progress on accurately estimating cloud influenced climate predictions in the Arctic”. Like Referee #1, I think the authors are overselling their work here. They have done measurements at only one location for 12 weeks, and then they used this data to develop a parameterization for the whole Arctic. A more reasonable approach would be to take all the previous measurements in the Arctic and then generate a parameterization based on this combined data set. Even then, I do not think this is a very useful approach since previous work has shown that concentrations can vary with season and location in the Arctic (at least in some cases).

We agree with the reviewer and refer them to the response above in part. We agree that higher INP concentrations were observed previously, especially in summer when the surface is free of ice and snow cover, thus pronounced local sources (e.g., dust, biological INPs) were expected. Despite the importance due to augmented local sources in summer, only a few Arctic INP measurement campaigns (less than 9 % of total reference data) focused on transition seasons, highlighting the need for more measurements during the transition season. In this context, seasonal variation was not observed from our measurements. We have now acknowledged the limitation of our parameterization in the paper (see responses above). In addition, we note that the distribution-based parameterization provides useful information on a general level and range of INP concentrations in the Arctic environment, which could be applied to regional climate models for long-term predictions. In this regard, several sentences are added to discuss the discrepancy in line 216 (revised manuscript line 261ff): *“However, a few studies (e.g., Wex et al., 2019, Tobo et al., 2019, Creamean et al., 2018) observed consistently higher INP concentrations in the summer, indicating enhanced sources of local emissions due to the decreased ice cover. We note that the parameterization herein was derived from the measurements during transition seasons (autumn and spring), aiming to predict the general level and range of INP concentrations in the Arctic. Therefore, applying it to generate INP concentrations in specific seasons, particularly the Arctic summer, could introduce a low bias.”*

Page 3. Line 84. The Coriolis impinger has a cut-off of 0.5 μm . Does this mean the technique is missing a large fraction of INPs? Could this explain the lack of agreement with the parameterizations based on particles above 0.5 μm ? Please discuss. Did the HINC have a similar cut-off?

The cut-off size of 0.5 μm represents the lower bound for the Coriolis impinger, i.e., the impinger efficiently collects particles with aerodynamic diameters larger than 0.5 μm . *“(lower limit)”* is added in the text for clarification. Therefore, it is adequate to link the INP parameterization to the particles above 0.5 μm . So, this would not explain why there is a lack of agreement with other parameterizations based on particles above 0.5 μm (see Fig. S2 in the supplementary information). However, HINC has a different cut-off with an upper bound D_{50} of approximately 2.5 μm (Lacher et al., 2017), i.e., it efficiently samples particles smaller than 2.5 μm . In this regard, we note that the concentration of particles above 2.5 μm is negligible.

Page 13. Line 259-261. “Our INP parameterization promotes future modeling studies via a more realistic microphysical representation in the Arctic MPCs, especially the vertical profile of primary ice distribution

(Hawker et al., 2021), thus, improving the predictions for the future Arctic climate.” Again, similar to Referee #1, I think the authors are overselling their results here.

We agree with the reviewer and have modified this sentence completely to see lines 321-324 in the revised manuscript. The new sentence reads *“We hope our INP parameterization promotes future modeling studies via a more simplistic prediction of INP concentrations in the Arctic environment as a function of temperature, particularly during the transition seasons of fall and spring thus improving the representation of MPCs and Arctic climate.”*

Page 13, lines 251-253. “Note that the presented INP parameterization is specified for the Arctic environment, where the atmosphere is well-mixed and transient effects average out.” What is the evidence for a well-mixed atmosphere in the Arctic? I think there is a lot of field data that shows that the Arctic is not a well-mixed system. Please correct me if I am wrong.

Thanks. We agree with the referee. We have modified the statements regarding the well-mixed to specify that the INP concentration is not dominated by a single aerosol species. See lines 314-316 in the revised manuscript.

Page 13, lines 254-256. “The presence of well-mixed INP air masses is exhibited by the absence of a relationship with aerosol properties and further by the inability of previous aerosol-based INP parameterizations to reproduce the observations from this study.” I do not understand the authors’ logic here. The absence of a relationship with aerosol properties may have nothing to do with a well-mixed INP air mass. Furthermore, the inability of previous aerosol-based INP parameterizations to reproduce the observations from this study may have nothing to do with a well-mixed INP air mass. Please correct me if I am wrong.

We agree with the reviewer here and similar to the response above, we have modified the text in the manuscript to reflect that the absence of INP dependence on aerosol parameters could be caused by the absence of a major dominating aerosol species in the air mass (see lines 316ff in the revised manuscript).