

Interactive comment on “Annual variability of ice nucleating particle concentrations at different Arctic locations” by Heike Wex et al.

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Received and published: 30 January 2019

General Comments

This paper is a very nice to see effort to provide new ice nucleating particle concentration data for different Arctic locations, including long term coverage to establish seasonal and annual cycles. The use of samples collected for other compositional analyses provides additional utility. I will state upfront that I am involved in reporting of other measurements in this region, some of which are in review separately at this time, but have deemed that this does not color my review of this paper. I express some concerns about protocol and accounting for backgrounds, and I mention that I think that sections 3.2 and 3.3 can be combined as being about the same thing. In the discussions, I think

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that some comments about the need for coarse mode aerosol measurements could be useful. And the authors do not need to emphasize local sources so much as they do I think. It seems unlikely that local is the only influence, although I understand that the sites used limit saying much more. It is the case that specific sources are difficult to discern. This is the case now in many studies. Nevertheless, this paper lays a nice groundwork for future research, providing additional impetus for new measurements over broad regions of the Arctic.

Specific Comments

Abstract

Page 1, Lines 9 and 10: Seems out of place for an abstract. Activity at this temperature range alone is not a certain indication of biogenic INPs, or at least should not be stated where that cannot be accompanied by discussion, in my opinion.

Page 1, Line 11: Reported generally, or where and when? It would help if context were given.

Introduction

Page 2, Lines 8 and 9: Meaning pure supercooled water clouds? And is this referring to a certain season or an annual basis?

Page 2, Line 11: “For primary ice formation in clouds, ice nucleation has to occur...” Somewhat awkward phrase and sentence. This is the definition of primary ice nucleation. That is, primary ice formation requires a heterogeneous ice nucleating particles. Perhaps rephrase?

Page 3, paragraph ending line 26: The study of Irish et al. (2019) seems relevant as well here (<https://doi.org/10.5194/acp-19-1027-2019>), especially with regard to land versus marine sources.

Page 3, lines 28 and 29: Sometimes spectra are linear, but you emphasize some data

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that contradicts this (e.g., data more toward the upper bound of Petters and Wright, which is consistent with information in the book Microbiology of Aerosols, Chapter 3, Fig. 3.1.1). I understand where you are trying to go with this paragraph, but perhaps “typically”?

Page 3, lines 31-32: “But biogenic INP typically occur in low concentrations in the atmosphere.” Regardless, the population may be entirely biogenic (i.e., dominant) at times over oceans on the basis of lab and field studies (e.g., McCluskey et al., 2018a,b).

Page 3, lines 34 and 35: Do mineral dusts always occur in higher concentrations than biogenics, even at -20 °C. It would seem to depend on the scenario of measurement, as one could imagine few or no dust INPs over some locations at times.

Page 4, line 3-4: There are “particularly high fractions of supercooled water observed in Arctic stratiform clouds” compared to where and when? Is there an expectation that seasons may matter, since open and ice-covered seas are present at different times?

Page 4, line 10: Should immersion mode be immersion freezing mode to be more explicit?

Page 5, line 1: Can you explain the meaning of blanks being collected? For example, does it mean they were removed temporarily from foil/wrap, placed in sampling shelter, returned to foil/wrap? That is, were they otherwise handled the same? And regarding storage after sampling at room temperature, were all samples stored that way and shipped after a certain time (e.g., weekly? annually?) Same for blanks?

Page 5, lines 5-8: The order of sentences describing procedures implies that filter punches were immersed in water prior to transit. Is this so? If not, rewrite this to be clear that a portion of the filter was sent to TROPOS first. What kind of water was used? Was the water tested for freezing both with and without a blank filter piece? If the punches were immersed and frozen before shipment, was blank water with a blank filter piece shipped?

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Page 5, line 16: Same questions with regard to the Ny Ålesund filter samples. And another question might be if blank filters were also cut in the same manner with the same tools. These are just typical concerns regarding testing contamination due to processes used, and cutting with tools is a method not always used with filters.

Page 6, line 27: It is not clear from discussion here if the blank filter concentrations were used to adjust measurements or simply documented. Considering the statement on page 7, lines 16-17 that filters with 60 L volumes were not used due to background being too close to measurements, then it seems that many samples with 130 L volume (stated earlier) might also have sufficient background that a correction could be needed. This would seem to be important for establishing a clear lower bound to measurements.

Results

Page 8, lines 6-7: It seems not terribly surprising that over the limited T range assessed the curves do not intersect. There is no predicting what happens where measurements are not possible (e.g., lower T). The range measured is indeed the range where biogenic impacts generally and broadly occur, so I think this is expected, if that is the point.

Page 8, lines 11-12: While I can agree that the results suggest biogenic origins, it might be fair to point out that the studies referenced here conducted direct trials that more or less confirmed the nature of these sources as organic.

Page 8, Section 3.2: Although a statement is finally made about this later in the section (lines 32-33), one immediately wonders about the sufficiency of looking for correlations when the INPs represent an infinitesimal number fraction of all particles. It is a qualitative approach that many use, but it must only imply associations with certain air masses. I suggest to bring this point to the beginning of the section. This being clear, I wonder also about combining sections 3.2 and 3.3 under the general topic of associating INPs with air mass sources (e.g., source attribution of INP). The different

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information is really targeting finding the same information. I wondered after reading both sections if joint information on composition and trajectories might carry more information than either one alone.

Page 9. Line 28: Overall, the analyses in this section end up being not conclusive, even though it is a reasonable idea to ask about the time air spends in the boundary layer over different surfaces, and so it is worthwhile. But is there a reason that 100 m was chosen, as opposed to some typical marine boundary layer depth over which the surface may be coupled? In this regard, it is not clear why 500 m was used for Utqiagvik, even though I looked for this information later. Just for contrast?

Page 9, line 32: precipitation “is assumed to lead”?

Page 10-11, paragraph: This paragraph wanders from a start talking about sea bird colonies, but I think the focus is that coastal regions seem particularly important as influencing INPs.

Comparisons with literature

Page 11, lines 29-30: Point taken, but I note that Petters and Wright (2013) contain data mostly from over NH continents. Delort and Amato, Eds. (2018; cf. Fig. 3.1.1) show ocean-based samples also in this lower region, as derived from DeMott et al. (2016), but for regions both inside and outside of the Arctic. Additional data in marine air-affected regions in Mcluskey et al. (2018b,c) would fall in these lower INP regions. Hence, it may be the case that many regions have yet to be effectively sampled, and not only that the Arctic is unique as a region of especially low INP.

Page 11, Fig. 7: Are the bars on the figure, horizontal and vertical, intended to represent uncertainties? For which set of data? It is a little unsatisfying not to see uncertainties represented somehow. It could be useful to show some. I can say that I am not entirely sure what the uncertainties are for Rogers et al. (2001), but given that the values are based on 10 s data, they could be quite large from a statistical sampling

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standpoint. Uncertainties were characterized very well in the INP data set of Prenni et al., which are clearly measured at near the limit of detection of a CFDC. The confidence intervals on those data are reported in that paper, and they are quite large. In contrast, uncertainties on some of the immersion freezing data are relatively small. Is there any chance to represent some of these?

Page 12, last sentence: This statement may require a caveat that coupling and decoupling of the boundary layer from clouds, common at different times in the Arctic, will influence the conclusion.

Discussion

Page 13, lines 5-6: In speaking only about particle number concentrations, the possible need to consider size distributions is not discussed. And when size distributions are considered, it is apparent that most such data collected do not well capture the sizes of INPs that have recently been noted to be of most influence in the Arctic and coastal regions at the warmest activation temperatures (references already included). That is, most previous studies emphasize ultrafine and fine modes at sizes below 1 um, key for anthropogenic impacts, but ignoring the underappreciated coarse mode that may dominate INPs in this region. This seems an opportunity to mention this measurement need. You get to mentioning that ultrafine aerosols are not expected as INP sources, but these of course dominate total particle numbers and so should perhaps not be the focus of the start of discussion.

Page 14, line 19: Creamean et al. (2018) seems relevant to the suggestion about open leads.

Figure 7 caption: Measurements are referenced as ground based, but since some were over oceans, perhaps say “surface based”.

Supplemental

Figure S1. This figure highlights significant overlap of the range of background freez-

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ing spectra and sample data in some cases. What is done for correcting the sample freezing spectra in times like April/May or other periods? Thinking here of Vali's recent discussion of these things in a recent AMTD paper (Vali, 2018).

SI, Page 9, line 10: Polen et al. (2018) seems relevant to the statement here.

SI, Page 9, lines 11-13: I understand and appreciate the feedback given in this small section overall, but the statements here could merit a lot of discussion and seem out of place in any case, which I say with bias, but also some justification. Why mention this disconnect between the usual sampling regimes of different INP measurement methods, if you are also saying they can be used in a complementary manner? It is clear that they have their use for ambient measurements in different parts of the INP temperature spectrum (DeMott et al., 2017), but also that aerosol pre-concentration can improve this overlap (Tobo et al., 2013 and others).

SI, Page 9, line 16: You may wish to be a little more explicit in the last parenthetical note. Heat sensitivity testing is possible with certain filter materials, and even for the same sample used for INP measurements. That is, a single filter of the right material can suit both standard freezing spectra tests and tests for heat lability if the rinsed suspension is divided. Readers may not know that quartz filters are less suitable for such tests. Your point is well taken of course that second filters could help with additional compositional measurements.

Editorial Comments

Page 1, lines 8 and 10: "months"

Page 1, line 9: "known"

Page 2, line 27: "among others observations, measurements of NINP are needed"

Page 5, line 7: "from 4 to 13 days"

Page 5, lines 29-30: statement here somewhat repeats what is said earlier in the

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paragraph

Page 7, line 11: "From this limitation, it also follows that . . ."

Page 7, line 23: "Sec. 4 and 5"

Page 8, line 22: "there were anti-correlations"

Page 13, line 1: "introduced in this study"

SI, page 9, line 4: "Price et al."

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