

Dear Reviewer!

We thank you for doing this review and for your suggestions that helped to improve our manuscript. Below, please find your original comments in blue and our responses in black. As your suggestions often concern larger parts of the text that were mostly moved and only changed a little, we typically did not paste the new text into our answers below but instead point to where they can be found in the new version of the manuscript. This new version of the manuscript is attached at the end of this answer, with text that is going to be deleted printed in yellow and new text in blue, so that you can easily find the changes we made.

### General comments

The manuscript presents longer term observations of INP concentrations at four Arctic stations. These are then discussed in the context of chemical compounds, back trajectories, and previously published data. The results are interesting, approach and methods are sound. The text is mostly clear but should be more concise. In particular the Introduction section has some lengths.

We thank you for the positive remarks. Following your suggestions below, some reorganization of the text was done. We agree that the Introduction section is detailed, but did not want to miss aspects related to the topic. The second reviewer (Paul DeMott) suggested, on a few occasions throughout the text, to even add citations or some more detail. We still feel that the text is organized in a better way, now, and hope you can agree with the changes we made.

I also wonder what the storage and transport histories of the samples from the field stations to the laboratory are meant to tell us. Atmospheric INP that could perish at room temperature would probably do so within a short time, perhaps already during a sampling period of several days. I can not imagine why it would help to freeze them again at a later stage. Possibly, many INP found in the atmosphere, even biogenic INP, can well be stored when dry at room temperature for many years (see Vasebi et al., 2019, <https://www.biogeosciences-discuss.net/bg-2018-496/#discussion>).

It is correct that the sample examined in the here cited discussion paper retained most of its ice nucleation ability (we have observed this too, in our lab, with a sample from Russel Schnell), but other biogenic INP are not so stable. There is e.g., a discussion around *Pseudomonas syringae*, which even when frozen has sometimes (but not always) been observed to lose its ice activity (Polen et al., 2016) - and from our own experience we know that it loses its ice activity quickly (in a matter of days to weeks) when it is kept in a refrigerator (> 0°C) or unfrozen. To the best of our knowledge, the full scale of changes that occur to ice activity of atmospheric samples during storage is not known in the community right now, but will certainly (have to) be addressed in the future. Therefore, freezing samples and reporting exactly what was done seems needed.

But as the transport of samples to TROPOS was similar in all cases, this part was summarized and is now only described once (in the first paragraph of Sec. 2 (now called “Methods”) on the top of page 5), instead of repeating the almost similar text four times. We also added the reason given above for taking the precautions concerning cooling the samples.

The comparison of INP concentrations with chemical compounds in the same of air masses revealed no correlation. The discussion of that finding does not go much beyond current understanding, so it could be shortened substantially.

Following one of the second reviewer's remarks, we changed the order in this section and first mentioned the fact that generally no correlation is to be expected. Some correlations between INP and mineral dust particles in general were reported for Arctic samples in literature, and these are now mentioned, too.

The description of our results was shortened so that it reports in only one sentence that there is mostly no correlation. The remaining passage describes the results for POC+CC (pyrolyzed organic carbon and carbonate carbon) which we feel is justified, as this at least gives some indication of the nature of the INP.

For changes, see Sec. 3.2.1, starting on page 10, line 11.

The back trajectory analysis to illuminate possible changes in source regions to explain observed increases in INP during spring or summer is original. I wonder how much the results of this analysis depend on the fact that a trajectory model was employed and not a particle dispersion model, in particular the results related to Utqiagvik.

We used both, trajectories (combined with a potential source contribution function) and a particle dispersion model for tracking of sources of particles and cloud condensation nuclei for a study about data from the Antarctic recently (Herenz et al., 2019) and obtained similar results from both methods. Interestingly, back then, during the review process, it was rather deemed unnecessary by a reviewer to use both approaches. We cannot be certain that the same results would also be found for this new study discussed here for a dispersion model, but adding this second method here currently is beyond the scope of what can be done in the framework of this review.

The first part of section 3.3 (Determination of possible source regions; page 9, lines 5-33) should be moved into the Methods section.

We followed your recommendation, moving the first three paragraphs of Section 3.3 to the end of Sec. 2, which is now called "Methods" (instead of "Measurements"). Required slight changes were made in the text. For that please refer to the new version of the manuscript, Sec. 2.7 on page 8, line 20.

Section 4 (comparison with literature) could be reduced in order to avoid repetition of what has been said already in the Introduction (or vice versa).

There is, admittedly, some redundancy in the Introduction and Sec. 4. But while in the Introduction the literature was shortly introduced with respect to its content and scientific

outcome, in Sec. 4 it is mostly only discussed in terms of the INP concentrations or in connection to possible sources of INP. This section is one of the location where we were asked to include some more detail by the second reviewer. Therefore, for the time being, no deletions of text were done.

The Conclusion section would benefit from a couple of sentences relating back to the potential influence of INP on Arctic clouds stated in the Introduction. Do concentrations observed at any of the four stations fluctuate between concentrations that would have different effects on clouds?

We added the following paragraph at the end of the discussion section:

“Concerning Arctic mixed-phase clouds at temperatures above  $-20^{\circ}\text{C}$ , recently Norgren et al. (2018) observed a depression of cloud ice in these clouds when they are polluted, as observed during Arctic haze. Under these conditions, they have a lower amount of cloud ice mass for a given amount of condensed liquid mass. Based on the results of the here presented study, this can be explained by the lower concentrations of INP during times of Arctic haze - a reasoning that sheds light on the importance of the here reported findings which could not yet have been discussed in Norgren et al. (2018). With respect to recent modeling results, Solomon et al. (2018) did large-eddy simulations (LES) of Arctic mixed-phase stratocumulus clouds and find indications that changes in INP in the Arctic aerosol dominate over changes in cloud condensation nuclei (CCN). This shows the potential importance of determining INP and their changes due to climate change in the Arctic. However, Taylor et al. (2018) examined the Arctic annual cycle in cloud amounts from 24 different models and found significant disagreements. They conclude that the parameterization of the ice microphysics in the models contribute to the observed differences. Overall, the field of Arctic mixed-phase clouds and related ice nucleation currently is one of intense research, and the herein presented results contribute to our understanding and lay ground for improved descriptions of INP in modeling in the future.”

Based on what is described in this new paragraph, and particularly the results by Taylor et al. (2018), not enough is known, yet, to allow for more details on e.g., the concentrations of INP that are needed to influence these Arctic mixed-phase clouds.

### Specific comments

Page 2, lines 13-15: “Not many measurements . . . exist . . .” Either say that “measurements ...were done” or that “data . . . exist”).

Done.

Site description: Please indicate the altitude of all four stations.

Done.

Page 5, line 6: “...the volume of air sampled per 1 mm piece of filter was roughly 270 or 540 L.” Does this mean that you are not sure about the sample volume?

We changed the formulation to: "... the volume of air sampled per 1 mm piece of filter differed for the different samples and varied from roughly 270 to 540."

Page 8, lines 29-31: "...and therefore might be indicative of biogenic material. This might point towards a direction in which more detailed studies could be undertaken in the future." I wonder what substance is left of a thought after qualifying it with two "might" and one "could".

This passage deals with POC (pyrolyzed organic carbon) which is rarely measured. But you are correct that the formulation here was overly cautious, and we changed it to: "This points towards a direction in which more detailed studies should be undertaken in the future."

Page 11, lines 29-30: "...N(INP) in the Arctic may, at times, be lower than at other locations." This statement probably applies to any location on Earth, except, if there is a location where N(INP) is always the global minimum N(INP).

You are totally right! We changed it to: "lowest Arctic  $N_{\text{INP}}$ , as those we observed in the winter months, might be below the lowest values observed on continents in mid-latitudes."

Page 12, lines 30-31: "PNSD measured on ground and in heights up 1200 m generally agreed, . . ." Agreed on what? Or, do you mean "were similar"?

Oups, a non-native-speaker issue, "were similar" is what we meant. Replacement done.

Literature mentioned in our answers:

Herenz, P., H. Wex, A. Mangold, Q. Laffineur, I. V. Gorodetskaya, Z. L. Flemming, M. Panagi, and F. Stratmann (2019), CCN measurements at the Princess Elisabeth Antarctica Research Station during three austral summers, *Atmos. Chem. Phys.*, 19, 275–294, doi:10.5194/acp-19-275-2019.

Polen, M., E. Lawlis, and R. C. Sullivan (2016), The unstable ice nucleation properties of Snomax (R) bacterial particles, *J. Geophys. Res.-Atmos.*, 121(19), 11666-11678, doi:10.1002/2016jd025251.



Dear Paul!

Thanks a lot for doing this review, for your thorough reading of our manuscript and for your encouraging words. The points you raised were mindful and certainly helped improving this manuscript. Below, please find your original comments in blue and our responses in black. Concerning the literature cited in this answer to your review, we ask you to refer to the attached new version of the manuscript (with tracked changes, showing text that is going to be deleted printed in yellow and new text in blue). Similarly, the new version of the supplemental information is also attached at the end.

## 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 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.

In our answers to your comments below, you will find our answers and the related changes we made concerning the points you raise there. On the issue of local sources, which did not come up in your specific comments again, we wonder where you feel that we stress them too much. Should you still want us to remove some mentioning of that, we would ask you to be more specific on where this should be done.

## 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.

The sentence might have been misleading as it was not meant to say that we showed that the highly ice active INP are certainly biogenic in origin. We changed it such that this should be clear now:

“Although the nature of these highly ice active INP could not be determined in this study, it often has been described in literature that ice activity observed at such high temperatures originates from the presence of ice active material of biogenic origin.”

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

This was meant to refer to the results from the section “Comparison with literature”. It was changed to:

“Spectra observed at the lowest temperatures, i.e., those derived for winter months, were on the lower end of respective values from literature on Arctic INP and INP from mid-latitude continental sites, to which a comparison is presented herein.”

## Introduction

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

This was a try of a short summary of studies on different basis. To insert the requested information, this had to be extended. It now is:

“In the range of temperatures (T) down to  $-20^{\circ}\text{C}$ , fractions of supercooled liquid clouds were reported to be well above 50%, based on annual mean data for Europe and North America (both including the Arctic) from satellite remote sensing (Choi et al., 2010). For a multi-year analysis of all clouds, based on ground-based remote sensing at two western Arctic locations (Eureka and Utqiagvik), clouds containing only liquid water occurred at least 20% of the time in all months with a maximum of 56% in September (Shupe, 2011). Also during two Arctic aircraft campaigns operating out of Inuvik, each in April and May of two different years, based on in-situ measurements, at least 60% of the clouds observed down to  $-18^{\circ}\text{C}$  were characterized as mostly liquid (Costa et al., 2017).”

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?

Done, it now is: “Ice nucleation forms primary ice in clouds ... “

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.

We added: “Irish et al. (2019) derived  $N_{\text{INP}}$  during a ship cruise in the Canadian Arctic marine boundary layer in summer. They suggest that mineral dust contributed more strongly to the observed INP than sea spray, with mineral dust particles likely originating in the Arctic (Hudson Bay, eastern Greenland, north-west continental Canada).”

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

The sentence in question was amended with: “, although at higher  $T$  steep increases may be observed, followed by a weaker increase or even a plateau region down to roughly  $-20^{\circ}\text{C}$  (e.g., Petters & Wright, 2015; O’Sullivan et al., 2018; Creamean et al., 2019).”

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).

You are right, and we extended this part as follows:

“Biogenic INP typically occur in low concentrations in the atmosphere, but nevertheless, at remote marine locations as the Southern Ocean, where  $N_{\text{INP}}$  is generally low, marine biogenic INP might make up a large fraction or even the entire INP population (Burrows et al., 2013; McCluskey et al., 2018). At less remote locations, the majority of atmospheric INP consists of mineral dust particles originating from deserts or soils.”

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

Indeed, there are scenarios imaginable where there may be more biogenic INP than mineral dust INP that are ice active at  $-20^{\circ}\text{C}$ , close to sources for biogenic INP or far away from sources for mineral dust particles (e.g., remote marine locations, see your comment above). We added “with the above mentioned exception of remote marine locations, typically”.

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?

The sentence is more specific now: “The existence of particularly high fractions of supercooled water observed in Arctic stratiform clouds, as e.g., observed in Costa et al. (2017) in the temperature range above  $-20^{\circ}\text{C}$  in comparison to more convective clouds in midlatitudes and the tropics, could be expected to be linked to a lack of biogenic INP in the Arctic, due to sparse biological activity.”

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

Done (here and in the following sentence).

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?

We added the following clarifying information: “These field blanks were treated similar to the other filters, i.e., inserted into the sampler, however, without an airflow through them. They were also stored similarly to the sampled filters at all times.”

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?

The shipment is now only described once at the beginning of Sec. 3, as it was similar for samples from all four stations. In all cases, the filters were shipped, and punching and immersion in water was done in Leipzig directly prior to the measurements. We added at / moved to the end of the first paragraph in Sec. 2:

“Transport from the four institutes where the samples had been kept to TROPOS was done in a insulated boxes, together with cooling elements. The shipment was organized such that transport was fast (one to three days) and that upon arrival at TROPOS the temperature in the boxes was still below 0°C. At TROPOS, samples were again stored at -1°C until the measurements were done. These measures during storage and transport are precautions, as for biogenic INP, storage at temperatures above 0°C or even storage in general has been described to reduce their ice activity (Wex et al., 2015; Polen et al., 2016, respectively). In this study, until mentioned otherwise, all samples from all stations (including also field blanks) were treated similar during all procedures. In the next sections, peculiarities of the separate four stations are described, followed by details on the measurements and its evaluation.”

The section you refer to here has been reworded for clarity:

“From these filters, a circular piece with 47 mm in diameter was shipped to Leipzig for this study.

The total sampling area on the filters was 17.8 cm x 22.8 cm. For the measurements at TROPOS, described in detail in Sec. 2.5 below, circles with 1 mm diameter were punched out from the samples using sterile biopsy punches, and immersed in ultra-pure water separately. The volume of air sampled per 1 mm piece of filter was differed for the different samples and varied from roughly 270 to 540 L.”

As now said in the text, additional information on topics such as the water is (and had been) given in Sec. 2.5. See also our comment to your next to next remark.

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.

Your comment here relates to topics we clarified above and we hope that you feel that all you are referring to here is sufficiently clear now.

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.

This comment, combined with your comment on the treatment of the background wrt. the Supplemental Information (SI) was tackled by inserting a more detailed analysis on the treatment of the background to the SI. As this resulted in almost one additional page of text and an extra figure, we ask you to review the SI directly (attached at the end, following the revised version of the manuscript).

At the location you mention here, we additionally inserted the following:

“A subtraction of the signals of the field blank from those of the measurements was not done. This is justified in a detailed discussion in the SI, and the interpretation of the results from the filters presented in this study is the same for both uncorrected and background-corrected samples.”

## 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.

This passage justifies that results can be discussed based on measurements done at a single temperature, instead of having to discuss the entire measured INP spectra. But indeed, it is correct that there is no predicting what happens where measurements are not possible, and we reworded the first sentence in this paragraph to:

“It is worth noticing that once a sample has a comparably high concentration at one  $T$  this is generally observed at all  $T$  at which measurements are available, and vice versa, i.e., curves do not intersect much (see Fig. 2). Therefore, the curves shown in Fig. 3 can be used to discuss observed trends for INP that are ice active at high  $T$ .”

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.

The sentences were changed to:

“These highly ice active INP will be in the focus of this study in the next two sections (Sec. 3.2 and 3.3). Similarly highly ice active INP have been suggested to be biogenic in origin based on

tests such as heat treatment (Hill et al., 2016; O’Sullivan et al., 2018), tests which due to the limited available amount of filter material could not be done in the present study.”

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 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.

The last paragraph from the former Section 3.2 was moved to the beginning of this section, as you suggested. The following was added:

“At a lower  $T$  of  $-25^{\circ}\text{C}$ , recently Si et al. (2019) reported that mineral dust tracers correlated with INP, which suggests that mineral dust was a major contributor to the INP population at that  $T$ . Si et al. (2018) found that for three coastal sites in Canada a model based on K-feldspar as the only INP calculated INP concentrations that fit measurements well at  $-25^{\circ}\text{C}$  while at  $-15^{\circ}\text{C}$  measurements were under-predicted, suggesting a missing source of INP that are active at higher  $T$ . In the following it will be examined if there is a correlation between the INP that are ice active at high  $T$  detected in the present study with chemical composition.”

Former sections 3.2 and 3.3. were combined in the sense that a new Sec. 3.2 was formed (“Sources of INP”) with these former sections being subsections in it. A further combination of the two topics was not done, as the correlation with chemical composition (as could be expected) hardly gave any insights, making a combination irrelevant.

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?

Our analysis focuses on finding locations where emissions from open ground or open water could have taken place, and for this, the marine boundary layer depths does not play a role, as we are not looking into real clouds. As said:” An altitude restriction was used as we were trying to geographically locate INP sources on the surface.” (This was in the part that now was moved to the new Sec. 2.7.) During the analysis for this study, we had looked at a number of different altitudes from 100m to 1000m and got quite similar results, independent of the altitude. Choosing 100m seems reasonable, as this allows for some uncertainty in the back-trajectories.

The extension to a larger altitude (and trajectories that reach further back) for Utqiagvik was done, as for this site, no apparent influence was found, independent of the altitude or length of the trajectories that were used (up to 10 days). This was to show that the altitude restrictions

and the choice of 5 day back-trajectories did not influence the analysis for this site. A passage of the text was reformulated to explain that better (new text in bold, here):

“Fig. 5 shows the number of time steps when air masses were over open land or open water for the separate filter samples. Additionally, gray bars in the background indicate the percentage of time the air masses collected on one filter were below 100 m for Alert, VRS and Ny Ålesund. It can already be seen, that the presence of highly ice active INP on a filter is related to air masses that fulfill the above given criteria, i.e., that traveled over open land or open water at a low altitude. It also can be seen that this was not found for Utqiagvik. **Initially, no open land and hardly any open water had been found for this site when 5-day back-trajectories were used, together with an altitude restriction of 100 m, i.e., air masses did not travel over open land or open water at altitudes below 100 m. To check if the length of the back-trajectory or the chosen maximum altitude influenced our results for Utqiagvik, an analysis was also done using 10-day back-trajectories and 500 m as the altitude limit, presented in Fig. 5. This extension** simply only resulted in larger percentages of time for which the air masses were below this altitude limit. But still not a large number of time steps was found for which air masses traveled over open land or open water for Utqiagvik. We will get back to this again below.”

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

Done. Please note that this is in the part that now was moved to the new Sec. 2.7.

Page 10-11, last 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.

The text in this section was rearranged and reformulated:

“The above analysis shows that coastal regions may be particularly important as source for highly ice active INP, including open waters close to coasts. Indeed, highly ice active biogenic INP were found in Arctic surface waters before (e.g., Wilson et al., 2015; Irish et al., 2017). For the highly ice active samples collected on Ny Ålesund on June 12 and June 28, 2012, the surrounding of the measurement station was completely snow free during the times when these samples were collected, whereas for all other cases there was at least partial or total snow cover around the stations. In other words, local terrestrial sources close to the measurement station may also contribute as sources for highly ice active INP, as already discussed in Creamean et al. (2018). Also Irish et al. (2019) describe Arctic land masses to be the source for observed Arctic INP (ice active at  $-15^{\circ}\text{C}$ ,  $-20^{\circ}\text{C}$  and  $-25^{\circ}\text{C}$ ), and these INP were suggested to be mineral dust. On Svalbard, Tobo et al. (2019) found higher atmospheric  $N_{\text{INP}}$  in July than in March, and they additionally described glacial outwash sediments in Svalbard to be highly ice active. This ice activity was assumed to be connected to small amounts of organic (likely biogenic) material. Based on these findings, Tobo et al. (2019) suggest the higher  $N_{\text{INP}}$  in summer to be connected to organic (biogenic) components in glacially sourced dust. Some coastal regions in the Arctic, e.g., the west coast of Greenland together with the region around Baffin Bay and the Canadian Arctic Archipelago as well as the area around the Bering Strait and also Svalbard are known for their abundance of sea bird colonies (Croft et al., 2016). These regions partially coincide with regions highlighted as possible INP sources in Fig. 6. These



regions are known to emit ammonia, which plays a role in new particle formation in the Arctic (Croft et al., 2016). But clearly, newly formed particles are not expected to contribute to atmospheric INP at the temperatures examined in this study, and INP are likely simply also emitted from regions with high biological activity. In Sec. 5 we will discuss possible INP sources in more detail.”

## 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 Mcliskey 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.

This passage was revised and now is:

“At the lowest temperatures at which we detected INP spectra,  $N_{\text{INP}}$  are lower than data from Petters and Wright (2015), i.e., lowest Arctic  $N_{\text{INP}}$ , as those we observed in the winter months, might be below the lowest values observed on continents in mid-latitudes. It is worthwhile adding that still lower concentrations were observed in marine remote locations in the Southern Ocean (McCluskey et al., 2018a) and for clean marine air in the North East Atlantic (McCluskey et al., 2018b).”

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 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?

The blue vertical bar showed an average value and was removed as the separate data points from Rogers et al. (2001) are shown, too. The horizontal brown bar shows the range of literature data cited by Rogers et al. (2001), which is now explicitly mentioned in the figure caption. Error bars from Prenni et al. (2007) are now shown in Fig. 7 and the following text was added to the manuscript:

“For the data taken from (Prenni et al., 2007), the reported measurement uncertainty is shown, representative for typical uncertainties for the type of instrumentation used in Rogers et al. (2001) and Prenni et al. (2007). Error bars indicate one standard deviation at the higher end. The lower end is indicative of the detection limit, and for a substantial fraction of measurements no INP were detected in Prenni et al. (2007).”



As typical uncertainties for the immersion freezing instrumentation used in this study were shown in Fig. 3 and 6 already, they were not added again in Fig. 7, to not overcrowd the latter.

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.

The sentence was complemented and now is:

“Hence, INP detected by ground based measurements may well be able to influence ice formation in clouds, at least during times when the cloud layers are coupled to the surface.”

## 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  $\mu\text{m}$ , 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.

The opening paragraph of the Discussion section now cites the recent literature concerning this matter to stress the point you make here:

“The annual cycle observed for  $N_{\text{INP}}$  in this study is not in tune with what is known for particle number concentrations and size distributions occurring across the Arctic (Tunved et al., 2013; Nguyen et al., 2016; Freud et al., 2017). This is not too surprising: Recent studies, including Arctic locations, found that a large fraction of the number of INP are super-micron in size (Mason et al., 2016; Si et al., 2018; Creamean et al., 2018), while the majority of the number of particles are in the sub-micron size range.”

The paragraph dealing with new particle formation was shortened by one sentence.

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

Done: “It should be added that Creamean et al. (2018) found a similarly large increase in highly ice active INP during May at Oliktok Point in northern Alaska, only roughly 300 km east of Utqiagvik. This increase was related to INP from tundra surfaces and open water, particularly the marginal ice zone, while polynyas 700 km away from the measurement site were not found to contribute, likely due to settling of particles during the long traveling times in the slow moving air masses that were observed.”

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

Done.

## Supplemental

Figure S1. This figure highlights significant overlap of the range of background freezing 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).

We went back to check this very carefully. This is treated in much more detail now, in the first section of the SI (adding ~ one page of text and a figure), and we refer you to that to check if you can agree with our treatment of the background and the reasoning behind it.

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

We incorporated this citation.

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).

The point under discussion here was intended to say exactly what you say in this remark, i.e., that in-situ devices and filter-offline techniques operate in different sampling regimes (based on INP concentrations), and that, in this sense, they are complementary. It is formulated now in a positive way, including the possibility of pre-concentration:

“However, they need comparably high  $N_{\text{INP}}$  to overcome their detection limits and hence typically contribute values at lower  $T$  where  $N_{\text{INP}}$  is higher. Concentration of the aerosol prior to the in-situ sampling, done e.g., in Tobo et al. (2013) can help to increase that  $T$  up to which measurements can be made, increasing the range of  $T$  for which overlap between in-situ and off-line techniques can be obtained.”

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.

The text here was extended:

“It should be added that also the choice of filter material is important. Polycarbonate filters can be washed off and different analysis, including heat treatment, can be done on the suspensions. Teflon filters were found to not work well for washing off in Chen et al. (2018) and also could not be punched. When considering heat treatment, parts of sampled quartz fiber filters could be heated prior to punching filter pieces for the analysis, while suspensions cannot be made as INP are retained by the fibers (Conen et al., 2012).”

## Editorial Comments

Please note: when these comments only required the addition of a letter or a word, these changes were not always highlighted in the new version of the manuscript.

Page 1, lines 8 and 10: “months” Done.

Page 1, line 9: “known” Done.

Page 2, line 27: “among others observations, measurements of NINP<sup>~</sup>are needed” Done.

Page 5, line 7: “from 4 to 13 days”

Sorry, but it is not clear what you mean. This line talks about the duration of the transport from the institutes which originally collected and stored the samples to TROPOS, and this never took longer than 3 days (even much shorter for the samples from Europe). (Special transport was arranged for these shipments.) 4 to 13 days were the sampling times at Utqiagvik.

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

Information on the transport to TROPOS, which was repeated again separately four times, was summarized and moved to the end of the first paragraph of section 2.

Page 7, line 11: “From this limitation, it also follows that. . .” Done.

Page 7, line 23: “Sec. 4 and 5” Done.

Page 8, line 22: “there were anti-correlations” Done.

Page 13, line 1: “introduced in this study” Done.

SI, page 9, line 4: “Price et al.” Done.

# Annual variability of ice nucleating particle concentrations at different Arctic locations

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**Abstract.** Number concentrations of ice nucleating particles ( $N_{\text{INP}}$ ) in the Arctic were derived from ground-based filter samples. Examined samples had been collected in Alert (Nunavut, Northern Canadian Archipelago on Ellesmere Island), Utqiagvik, formerly known as Barrow (Alaska), Ny Ålesund (Svalbard) and at the Villum Research Station (VRS, Northern Greenland). For the former two stations, examined filters span a full yearly cycle. For VRS, 10 weekly samples, mostly from different months of one year, were included. Samples from Ny Ålesund were collected during the months from March until September of one year. At all four stations, highest concentrations were found in the summer months from roughly June to September. For those stations with sufficient data coverage, an annual cycle can be seen. The spectra of  $N_{\text{INP}}$  observed at the highest temperatures, i.e., those obtained for summer months, showed the presence of INP that nucleate ice up to  $-5^{\circ}\text{C}$ . It is known from literature that ice activity observed at such high temperatures indicates originates from the presence of ice active material of biogenic origin. Spectra observed at the lowest temperatures, i.e., those derived for winter months, were on the lower end of respective values reported in literature on Arctic INP and INP from mid-latitude continental sites, to which a comparison is presented herein. An analysis concerning the origin of INP that were ice active at high temperatures was carried out, using back-trajectories and satellite information. Both, terrestrial locations in the Arctic and the adjacent sea were found to be possible source areas for highly active INP.

## 1 Introduction

The Arctic warms faster than any other region on Earth, a phenomenon which is known as Arctic amplification (Serreze and Barry, 2011; Cohen et al., 2014; IPCC, 2013). Many different processes, of which some are heavily interconnected, contribute to this (Pithan and Mauritsen, 2014). However, not all of these processes and feedbacks are fully understood, and some might even still be unknown. Clouds in the Arctic are special in that they often form extended, persistent, low-level stratiform cloud layers, which are kept stable for days by different feedback processes (Shupe et al., 2006; Morrison et al., 2012; Shupe et al., 2013). These clouds influence the energy budget and generally warm the surface, compared to clear skies (Intrieri et al., 2002). They often contain supercooled liquid water. In the range of temperatures ( $T$ ) down to  $-20^{\circ}\text{C}$ , fractions of supercooled liquid clouds were reported to be well above 50%, based on annual mean data for Europe and North America (both including the Arctic) from satellite remote sensing (Choi et al., 2010). For a multi-year analysis of all clouds, based on ground-based remote sensing at two western Arctic locations (Eureka and Utqiagvik), clouds containing only liquid water occurred at least 20% of the time in all months with a maximum of 56% in September (Shupe, 2011). Also during two Arctic aircraft campaigns operating out of Inuvik, each in April and May of two different years, based on in-situ measurements, at least 60% of the clouds observed down to  $-18^{\circ}\text{C}$  were characterized as mostly liquid (Costa et al., 2017). ground-based and satellite remote sensing (Shupe, 2011; Choi et al., 2010) as well as on aircraft measurements (Costa et al., 2017).

For primary ice formation in clouds, ice nucleation has to occur. Ice nucleation forms primary ice in clouds, and for  $T$  from  $0^{\circ}\text{C}$  to roughly  $-38^{\circ}\text{C}$ , ice nucleating particles (INP) are needed to induce this nucleation process. Not many measurements on number concentrations of INP ( $N_{\text{INP}}$ ) in the Arctic exist were done up to now, however, these particles play an important role in the lifetime and radiative effects of Arctic stratiform clouds. A number of effects of ice in clouds are described in Prenni et al. (2007), among them that ice clouds are optically thinner than supercooled liquid water clouds so that the former emit less longwave radiation towards the surface. A modeling study showed that an increase in  $N_{\text{INP}}$  may cause a faster dissipation of these stratiform clouds, (Loewe et al., 2017), which in turn will influence the surface energy budget. But it should also be mentioned that  $N_{\text{INP}}$  of  $> 1 \text{ L}^{-1}$  were needed to obtain an effect in this modeling study, and such concentrations were observed for mid-latitude regions only for  $T$  below  $\approx -15^{\circ}\text{C}$  (Petters and Wright, 2015; O’Sullivan et al., 2018). Recycling of INP was assumed to be possible in Arctic clouds, again based on a modeling study (Solomon et al., 2015), i.e., ice crystals falling from a cloud could sublime and re-entrain into clouds from below, which might potentially enhance the effect of changes in  $N_{\text{INP}}$ . It also has been shown with large-eddy simulations that ice crystal number concentrations significantly influence cloud structure and evolution of Arctic mixed-phase clouds (Ovchinnikov et al., 2014). Overall, the cloud phase (i.e., supercooled water versus ice) is important for the radiation budget and hence the effect of Arctic stratiform clouds on climate. Kalesse et al. (2016) examined in detail a mixed-phase stratiform Arctic cloud and its phase transitions for roughly 1.5 days. They used a large set of measurements and applied modelling to gain further understanding. Observed changes in the cloud were related to changes in air mass, but it was also said explicitly that for a better understanding of cloud phase transitions, among other observations also measurements of  $N_{\text{INP}}$  are needed. All of this highlights the importance of insight on the abundance of INP in the Arctic and on their sources and sinks.

In the past, some studies on Arctic INP were done. However, most studies only included samples collected during short term deployments. A comparison of results of the present study with some of those from literature will be made further down in Sec. 4, while the main outcomes of these previous studies are already described in the following. In general, it can still be said that data on Arctic  $N_{\text{INP}}$  is scarce, which is particularly true for data at high  $T$ . Borys (1983) and Borys (1989) derived  $N_{\text{INP}}$  based on ground-based and aircraft measurements, respectively. It was suggested that mid-latitude pollution did not contribute INP to the Arctic aerosol, as  $N_{\text{INP}}$  were found to be lowest in winter, when Arctic haze, originating from anthropogenic pollution, was present. Bigg (1996) measured INP during a ship cruise in the Arctic in the months from August to October. It was concluded that INP were oceanic in origin while land was only a weak source, and that the upper troposphere was deficient in INP. Bigg and Leck (2001) derived  $N_{\text{INP}}$  also during an Arctic ship cruise in July to September, and found a decline in  $N_{\text{INP}}$  during that phase. At least for some of the detected INP, the most likely sources were assumed to be bacteria and fragments from marine biota emitted via bubble bursting from the open sea. Rogers et al. (2001) detected INP during aircraft measurements in the Arctic during the month of May. They reported strongly varying concentrations and found some INP that contained Si that were likely mineral dust particles, while other INP seemed to consist of low-molecular-weight components. In Prenni et al. (2007), also aircraft measurements of  $N_{\text{INP}}$  were made in the vicinity of Arctic clouds during fall. They found, reporting lower values than Rogers et al. (2001) had obtained in spring. and theyIt was concluded that typical Arctic values for  $N_{\text{INP}}$  might be overestimated by current parameterizations. Mason et al. (2016) derived  $N_{\text{INP}}$  for size segregated aerosol samples collected between end of March until July in Alert.  $N_{\text{INP}}$  derived from these Alert samples were slightly below values reported for other more southerly stations (mostly in Canada) in that study. They also found that at all stations large fractions of INP were contributed by supermicron particles. These fractions generally were largest at the highest  $T$  at which measurements were made, i.e., at  $-15^{\circ}\text{C}$ , where, for the Alert samples,  $> 90\%$  and  $70\%$  of all INP were  $> 1\ \mu\text{m}$  and  $> 2.5\ \mu\text{m}$ , respectively. Similarly, Si et al. (2018) found a size dependent ability of particles nucleating ice for samples collected mostly in coastal areas in southern Canada and one sample collected in Lancaster Sound in the Canadian Arctic, with larger particles being more ice active. They also concluded that sea spray aerosol was not a major contributor to INP for the samples taken in southern Canada. Based on concentrations of K-feldspar taken from a global model,  $N_{\text{INP}}$  measured at  $-25^{\circ}\text{C}$  were modelled well, while INP ice active at  $-15^{\circ}\text{C}$  were missing in this model. Also Creamean et al. (2018) foundreport a strong size dependence of the ice activity in samples collected on land in the northern Alaskan Arctic, where again the largest particles in the supermicron size range were the most efficient INP. During sampling phases from March until mid May, when grounds were covered in snow and ice, number concentrations of the supermicron INP were lower than those in late May, by up to two orders of magnitude. The increase in  $N_{\text{INP}}$  was suggested to originate from open leads in the sea ice and from open tundra. Similarly, for a coastal mountain station in northern Norway (at  $70^{\circ}\text{N}$ ), based on four filters sampled during July, Conen et al. (2016) observed that air masses were enriched in INP ice active at  $-15^{\circ}\text{C}$  when they had passed over land. An origin of these INP from decaying leaves was suggested. Irish et al. (2019) derived  $N_{\text{INP}}$  during a ship cruise in the Canadian Arctic marine boundary layer in summer. They suggest that mineral dust contributed more strongly to the observed INP than sea spray, with mineral dust particles likely originating in the Arctic (Hudson Bay, eastern Greenland, north-west continental Canada).

Different substances are known to contribute to atmospheric INP, as given in a number of review articles (Szyrmer and Zawadzki, 1997; Hoose and Moehler, 2012; Murray et al., 2012; Kanji et al., 2017). In general, it is known that  $N_{\text{INP}}$  increases roughly exponentially with decreasing  $T$ , although at higher  $T$  steep increases may be observed, followed by a weaker increase or even a plateau region down to roughly  $-20^{\circ}\text{C}$  (as seen in e.g., Petters and Wright, 2015; O’Sullivan et al., 2018; Creamean et al., 2019). Ice nucleation at higher  $T$  typically is related to macromolecules from biogenic entities as bacteria, fungal spores, lichen, pollen and marine biota. These ice active macromolecules nucleate ice from just below  $0^{\circ}\text{C}$  down to roughly  $-20^{\circ}\text{C}$  (Murray et al., 2012; Kanji et al., 2017; O’Sullivan et al., 2018). But Biogenic INP typically occur in low concentrations in the atmosphere, but nevertheless, at remote marine locations as the Southern Ocean, where  $N_{\text{INP}}$  is generally low, marine biogenic INP might make up a large fraction or even the entire INP population (Burrows et al., 2013; McCluskey et al., 2018a). At less remote locations, the majority of atmospheric INP consists of mineral dust particles originating from deserts or soils. Pure mineral dust particles of atmospheric relevant sizes typically are ice active below  $-15^{\circ}\text{C}$  (Murray et al., 2012; Kanji et al., 2017) or even below  $-20^{\circ}\text{C}$  (Augustin-Bauditz et al., 2014) and, with the above mentioned exception of remote marine locations, typically occur at much higher concentrations than biogenic INP (Murray et al., 2012; Petters and Wright, 2015). However, mineral dust particles might also occur together with biogenic ice active material (Tobo et al., 2014; O’Sullivan et al., 2014; Hill et al., 2016), and such a mixed particle acts like a biogenic INP (Augustin-Bauditz et al., 2016) and should be attributed to the aforementioned group of biogenic INP.

The existence of particularly high fractions of supercooled water observed in Arctic stratiform clouds, as e.g., observed in Costa et al. (2017) in the temperature range above  $-20^{\circ}\text{C}$ , in comparison to more convective clouds in midlatitudes and the tropics, could be expected to be linked to a lack of biogenic INP in the Arctic, due to sparse biological activity. However, it is known that biogenic INP are contained in sea water (Schnell, 1977) and the oceanic surface microlayer (SML) (Wilson et al., 2015; Irish et al., 2017) and are emitted to the atmosphere by sea spray production (DeMott et al., 2016). An increase in INP concentrations in the SML (Wilson et al., 2015) and for the biosphere in general (Schnell and Vali, 1976) from equatorial regions towards the poles has been observed. Also, present in the Arctic are fungi (Fu et al., 2013), lichen and bacteria (Santl-Temkiv et al., 2018) which could potentially contribute biogenic INP.

In the present study, we aimed at increasing the knowledge on Arctic surface concentrations of INP active in the immersion freezing mode, as described in the following. The immersion freezing mode was examined as it has been described as the most important heterogenous ice nucleation mode in mixed-phase cloud (Ansmann et al., 2009; Wiacek et al., 2010; de Boer et al., 2011). No specific measurement campaign was organized to do the examinations described herein. Instead, use was made of already existing filter samples. Besides for deriving temperature spectra of  $N_{\text{INP}}$  for 104 filter samples (Sec. 3.1), we also determined possible sources for INP that are ice active at high  $T$  for selected samples. For that, correlations between  $N_{\text{INP}}$  and some chemical compounds were made (Sec. 3.2.1), and an analysis was done concerning possible regions of origin of INP that are ice active at high  $T$  (Sec. 3.2.2) on a selection of samples. The results will also be discussed in light of literature data (Sec. 4).



## 2 MethodsMeasurements

Quartz fiber filters were sampled regularly at the four Arctic measurement stations of Alert, Ny Ålesund, Utqiagvik, and Villum Research Station (VRS) during the past years. Fig. 1 shows the locations of these four stations, which are all in close proximity to the ocean (<3 km). A portion of the filters was provided for the herein presented analysis. In the following, some detail will be given on these four different stations, including the filter handling, and on the measurement and evaluation method used to obtain INP number concentrations. We also describe in detail the temperature history of the filters, although it is not known with certainty up to date how the temperature during storage will affect INP concentrations. Generally, the filters were kept frozen whenever possible. Transport from the four institutes where the samples had been kept to TROPOS was done in insulated boxes, together with cooling elements. The shipment was organized such that transport was fast (one to three days) and that upon arrival at TROPOS the temperature in the boxes was still below 0°C. At TROPOS, samples were again stored at -18°C until the measurements were done. These measures during storage and transport are precautions, as for biogenic INP, storage at temperatures above 0°C or even storage under freezing conditions has been described to reduce their ice activity (Wex et al., 2015; Polen et al., 2016, respectively). In this study, until mentioned otherwise, all samples from all stations (including also field blanks) were treated similar during all procedures. In the next sections, peculiarities of the separate four stations are described, followed by details on the measurements and its evaluation.

### 2.1 Alert

A custom-built high-volume aerosol sampler was used at the Dr. Neil Trivett Global Atmosphere Watch Observatory in Alert, Canada (82°30'N 62°22'W, 210 m above sea level (a.s.l.)), to collect 38 samples between April 2015 and April 2016. The sampler is installed at a walk-up deck, about 4 m above the ground. Flow rate is approximately 1.4 m<sup>3</sup>/min at STP condition. Quartz filters (Pall Life Sciences, PallFlex Filters, 8 x 10 in, USA) were pre-fired at 900°C overnight and then shipped to Alert while already being loaded on cartridges. During transport and storage, the filter containing cartridges were wrapped by aluminum foil and they were inside of sealed plastic bags. Sampling time on those filters was either one week or two weeks, the latter being used from August until October (due to operational issues, no filter was sampled in July). A total of 9 field blanks (roughly one every month) were collected. These field blanks were treated similar to the other filters, i.e., inserted into the sampler, however, without an airflow through them. They were also stored similarly to the sampled filters at all times. After sampling, filters were stored (at room temperature ≈ 20°C) in their sampling cartridges (wrapped in aluminum foil inside sealed plastic bags) at the Alert station and shipped in cardboard boxes (containing 5 sampling cartridges each) to the Toronto lab at Environment Climate Change Canada where they were stored frozen at < -30°C. From these filters, a circular piece with 47 mm in diameter was providedshipped to Leipzig for this study.

The total sampling area on the filters was 17.8 cm x 22.8 cm. For the measurements at TROPOS, described in detail in Sec. 2.5 below, circles with 1 mm diameter were punched out from the samples using sterile biopsy punches, and immersed in ultra-pure water separately. The volume of air sampled per 1 mm piece of filter wasdiffered for the different samples and varied from roughly 270 to 540 L. transport to TROPOS was done in a cooler, together with cooling elements, where preparations



were done such that transport took less than 3 days and the box was stored at freezing temperatures during delay times at airports and customs. Upon arrival at TROPOS the temperature was just around 0°C. At TROPOS, samples were again stored at -18°C until the analysis was done.

## 2.2 Ny Ålesund

- 5 Filter sampling in Ny Ålesund on Svalbard (at 78°55'N 11°55'E, 10 m a.s.l.) is done by the University of Florence, Italy. Quartz fiber filters have been sampled regularly since 2010, using a high-volume sampler with quartz microfiber filters (CHMLAB GROUP QF1 grade, Barcelona, Spain). The filters were pre-treated at 400°C prior to sampling. The filters had a diameter of 47 mm, of which one quarter was provided for the present study. Sampling duration was 4 days in 2012, and of these filters, 13 sampled from late March until beginning of September were examined in the present study, together with two field blanks.
- 10 The total air volume collected on each filter was roughly 200 m<sup>3</sup>. Each circular 1 mm filter piece used for the analysis sampled particles from roughly 130 L. Once sampled, filters were stored in a freezer at the Italian base in Ny Ålesund and then shipped to Italy via cargo. At the home university, they were then stored in a cold room at -20°C. Transport to TROPOS was done in a Styrofoam box, together with cooling elements. The transport took little more than 24 hours and temperature inside the box was well below 0°C upon arrival. At TROPOS, samples were again stored at -18°C until the analysis was done.

## 15 2.3 Utqiagvik (formerly known as Barrow)

- Filter sampling in Utqiagvik, Alaska (at 71°18'N, 156°46'W, 11 m a.s.l.) is done by Baylor University, US as described in Barrett and Sheesley (2017). For the present study, quartz fiber filters were used that had been sampled regularly in an annual campaign from June 2012 to June 2013, using a high-volume sampler (Tisch Environmental, Cleves, OH, USA). Filters were stored frozen prior to and immediately following all sampling. Two rectangular filter pieces (with a lateral length of 1.5 cm)
- 20 from each of 41 different filters and two field blanks were provided for the present study. The sampled area of each filter was 399 cm<sup>2</sup>. Sampling on each filter was done for 4 up to 13 days (7 days on average), collecting particles from a total air volume of roughly 6000 to 23000 m<sup>3</sup>. This yields an air volume of roughly 120 to 450 L collected on each circular 1 mm filter piece used for the analysis. Prior to sampling, filters were pre-treated at 500°C. After sampling, the filters were stored in a freezer on site and transported to the home university in coolers with cooling elements, where they were then stored at -18°C. Transport
- 25 to TROPOS was done in a cooler, together with cooling elements, where preparations were done such that transport took less than 2 days and the box was stored at freezing temperatures during delay times at airports and customs, such that filters arrived still well frozen. At TROPOS, samples were again stored at -18°C until the analysis was done.

## 2.4 Villum Research Station

- Villum Research Station (VRS) at Station Nord in northern Greenland (at 81°36'N, 16°40'W, 10 m a.s.l.) is operated by Aarhus
- 30 University, Denmark (in cooperation with the Danish Defense, the Arctic Command). Quartz fiber filters are sampled regularly since 2008, using a high-volume sampler (Digitel, Hegnau, Switzerland), employing weekly sampling Bossi et al. (2016). The

filters had an exposed area of 154 cm<sup>2</sup> and sampled a total air volume of roughly 5000 m<sup>3</sup>. From filters sampled in 2015, a 2 cm diameter piece was cut from each of the filters and provided for this study. Due to the large interest in shares of the filters, only samples from 11 different filters, all from different months in 2015 and one from December 2013, could be used herein. As for all samples used in this study, from the 2 cm pieces punches of 1 mm in diameter were cut [at TROPOS directly prior to doing the measurements](#). The resulting small pieces were then used for INP analysis. The area of these 1 mm pieces corresponds to a sample volume of 255 L of air. Prior to sampling in the field, filters were pre-treated at 450°C. Storage of the filters at VRS was done in freezers. Filters are transported from Greenland to Denmark around 3 times per year by the Danish Royal Air Force and then shipped to Roskilde, where they were then stored at -18°C. [Transport to TROPOS was done in a Styrofoam box, together with cooling elements, where the transport took roughly one day, and the temperature inside the box was well below 0°C upon arrival. At TROPOS, samples were again stored at -18°C until analysis was done.](#)

## 2.5 Freezing device INDA, the Ice Nucleation Droplet Array

For freezing experiments examining immersion freezing, a device comparable to one introduced in Conen et al. (2012) was used, but deploying PCR-trays (Hill et al., 2016) instead of separate tubes. The same device had been used in Chen et al. (2018). From each filter [piece that had been shipped to TROPOS](#), circles with a diameter of 1 mm were punched out directly before measurements were done, and each of the 96 wells of a PCR tray was filled with such a filter piece together with 50 µL of ultra-pure water. (Background measurements of ultra-pure water are given in the supporting information (SI)). After sealing the PCR tray with a transparent foil, it was immersed into a bath thermostat such that the water table in the wells was below the surface of the liquid in the thermostat. The bath of the thermostat was then cooled with a cooling rate of 1 K/min, and the freezing process was monitored by a camera, taking a picture every 6 seconds. A LED light source installed below the PCR tray ensured that wells in which the water was still liquid could be easily distinguished from frozen ones. Frozen fractions ( $f_{ice}$ ) were then determined as the number of frozen tubes divided by the total number of tubes. Typically, fresh water to be used in the experiments was taken once a day and stored in a glass bottle. Whenever fresh water was taken, an experiment was run with this water in the tubes only, to assure that the water was satisfyingly clean. Similarly, experiments were run with field blank filters that had the same history as the samples but without sampling (see SI), and signals from the field blanks were well below those of the sampled filters. [A subtraction of the signals of the field blank from those of the measurements was not done. This is justified in a detailed discussed in the SI. The interpretation of the results from the filters presented in this study is the same for both uncorrected and background-corrected samples.](#)

## 2.6 Deriving $N_{INP}$

Equation 1 was used to derive  $N_{INP}$  from the measured  $f_{ice}$  (Vali, 1971; Conen et al., 2012). This equation accounts for the possibility of the presence of multiple INP in one vial by assuming that the INP are Poisson distributed. Additionally it normalizes the values resulting from the measurement with the air volume sampled on each 1 mm filter piece. This yields

concentrations of ice nucleating particles per volume of sampled air:

$$N_{\text{INP}} = -(\ln(1 - f_{\text{ice}})) / (F * A_p / A_f) \quad (1)$$

$F$  is the total volume of air drawn through the filter,  $A_p$  and  $A_f$  are the surface area of a single 1 mm filter piece and the whole sampled area of the filter, respectively.

- 5 The temperature and concentration regions for which data were obtained for the different samples depend on a number of factors. The measured value  $f_{\text{ice}}$  is a fraction ranging from 0 to 1. Therefore,  $N_{\text{INP}}$ , as derived using equation 1, can only take on a limited range of values. This range is based on the negative natural logarithm of 0.01 and 0.99 (4.6 to 0.01). The absolute values of  $N_{\text{INP}}$  then also depend on the volume of air sampled onto each 1 mm filter piece, i.e., on the volume of air drawn through the filter during the sampling period and on the relation of the surface area of one filter piece to the total sampled
- 10 surface area. The volume collected per 1 mm filter piece was within a factor of 4.5 for all filters (120 to 540 L). Altogether, the range of  $N_{\text{INP}}$  that can be obtained herein is roughly from  $2 \cdot 10^{-5} \text{ L}^{-1}$  to  $0.04 \text{ L}^{-1}$ . From this limitation, it also follows that also the range of  $T$  for which  $N_{\text{INP}}$  could be obtained is limited, as it is tied to the concentrations that can be measured. Times with more ice active INP show up as  $N_{\text{INP}}$  at higher  $T$ . The highest  $T$  at which ice activity was observed was close to  $-5^\circ\text{C}$ , as will be shown in the next section.
- 15 It should also be noted that samples that had less than 60 L of air volume collected on each 1 mm filter piece were also examined, but  $f_{\text{ice}}$  was close to the background and therefore these samples were not considered in this study. Results from background measurements are given in the SI. Measurement uncertainty as shown in this work was derived based on Harrison et al. (2016), i.e., following the assumption that the INP are Poisson distributed between the different examined droplets. The first few droplets that freeze in each experiment therefore show the highest uncertainties.

## 20 2.7 Using back-trajectories and satellite maps

- A more in depth analysis concerning possible INP source regions was done for a selection of filter samples from each measurement station. For that, 5-day back-trajectories were calculated with HYSPLIT (Stein et al., 2015) to determine the origin of sampled air masses. These calculations were based on GDAS (Global Data Assimilation System) meteorological data, using an hourly time resolution. A new trajectory was started every 6 hours during the whole time for which sampling was done on the
- 25 respective filter. Back-trajectories were initiated at an altitude of 100 m above the sampling locations, as this altitude still has a high likelihood of being connected to the ground, and as lower elevations are more prone to uncertainties. In the supplementary information (SI), these back-trajectories are shown separately for the selected examined filter samples.

- Using these back-trajectories, it was examined over which ground the air masses collected on the filters had passed in the 5 days prior to arrival at the measurement station. The aim was to see contributions from open land or open water for the different
- 30 selected filter samples. Therefore, it was distinguished between snow, open land, sea ice and open water. To do so, maps from the Interactive Multisensor Snow and Ice Mapping System (IMS) (Helfrich et al., 2007; National-Ice-Center, 2008) were used. IMS maps are a composite product produced by NOAA/NEDIS (National Oceanic and Atmospheric Administration's National Environmental Satellite Data and Information Service) combining information on both sea ice and snow cover. Information

from 15 different sources of input are included in the production of these maps (Helfrich et al., 2007). Currently these maps have been provided for 20 years. We used the daily northern hemisphere maps with a resolution of 4 km (National-Ice-Center, 2008). For each time step we applied nearest-neighbor interpolation in space and time to find the corresponding satellite coordinate along the back-trajectory. With that, for each back-trajectory, we determined the conditions on ground during the passage of the air mass, i.e., if the ground was covered by snow or ice or if open water or open land was present. The resulting information is shown exemplarily in Fig. 4 for the filter sample collected in Alert starting June 10, 2015. Using these maps, it was counted how often air masses that were collected on one filter were above open land or open water while the air mass was below 100 m. (As will be discussed in detail in Sec. 3.2.2, for Utqiaġvik, results reported here always refer to an upper altitude of 500 m and 10-day back-trajectories.) An altitude restriction was used as we were trying to geographically locate INP sources on the surface. Additionally, the back-trajectories were only considered back in time until an integral amount of 2 mm of precipitation (taken from the information included in the back-trajectories) was reached. This was done as precipitation formation occurs via the ice phase, so that precipitation is assumed to lead to a wash out of INP.

### 3 Results

In the following,  $N_{\text{INP}}$  derived from filter samples will shortly be introduced. A correlation to some available chemical composition data is made. Finally an analysis on air mass origins is introduced to analyze possible source regions for INP that are ice active at high  $T$ . A comparison to literature data and further discussion of the results are then presented in Sec. 4 and 5, respectively.

#### 3.1 Arctic atmospheric INP concentrations

Quartz fiber filters from the four different Arctic stations shown in Fig. 1 were analyzed to derive  $N_{\text{INP}}$ . Fig. 2 shows  $N_{\text{INP}}$  for all different samples, separately for the four sampling locations. Due to the comparably large number of filters analyzed for Alert and Utqiaġvik, separate curves can not be seen easily in Fig. 2. Therefore, Fig. 3 shows time-series of  $N_{\text{INP}}$  for  $T$  at  $-7^{\circ}\text{C}$ ,  $-10^{\circ}\text{C}$ ,  $-13^{\circ}\text{C}$  and  $-9.5^{\circ}\text{C}$  for Alert, Utqiaġvik, Ny Ålesund and VRS, respectively.  $T$  was chosen such that  $N_{\text{INP}}$  could be obtained for the largest possible number of all curves (data for the three samples with the lowest  $N_{\text{INP}}$  are missing for Alert (April 29 and May 27, 2015 and April 4, 2016), and the one with the highest  $N_{\text{INP}}$  for Utqiaġvik (May 3, 2013), indicated by arrows in Fig. 3). The yellow background shows for which samples an more in-depth analysis is presented in Sec. 3.2.2. Error bars in Fig. 3 show the 95% confidence interval.

It is worth noticing that once a sample has a comparably high concentration at one  $T$  this will be generally true is generally observed at all  $T$  at which measurements are available, and vice versa, i.e., curves do not intersect much (see Fig. 2). Therefore, the curves shown in Fig. 3 can be used to discuss observed trends for INP that are ice active at high  $T$ . It should also be noted that for Ny Ålesund, data only exists for March until September, and that for VRS, there is mostly only one data-point per month, if any (no data exists for February, March and May).

In Figs. 2 and 3 it can be seen that in general  $N_{\text{INP}}$  obtained for the summer months is higher than for the winter months. A decrease of  $N_{\text{INP}}$  is observed starting in fall (October or November). For months with the lowest observed ice activity, which generally are winter and early spring months, values for  $N_{\text{INP}}$  were measured down to below  $-20^{\circ}\text{C}$ . From June until September mostly INP that were ice active between  $-5^{\circ}\text{C}$  and  $-15^{\circ}\text{C}$  were detected (see Fig. 2), and for Utqiaġvik and VRS such highly ice active INP were observed as early as in April. These highly ice active INP will be in the focus of this study in the next two sections (Sec. 3.2.1 and 3.2.2). Similarly Such highly ice active INP have been assumed suggested to be biogenic in origin based on tests such as heat treatment (Hill et al., 2016; O’Sullivan et al., 2018), tests which due to the limited available amount of filter material could not be done in the present study. and these will be in the focus of this study in the next two sections (Sec. 3.2.1 and 3.2.2).

## 3.2 Sources of INP

### 3.2.1 Correlation to chemical composition

Typical  $N_{\text{INP}}$  measured in the atmosphere are several orders of magnitude below total particle number concentrations, and therefore mass concentrations of INP are so small that a correlation between bulk chemical composition and  $N_{\text{INP}}$  might not be expected, particularly not for the very rare INP that are ice active at high  $T$ . This is in line with recent findings for a long term study on INP at Cape Verde by Welti et al. (2018). There, no correlation between  $N_{\text{INP}}$  and bulk chemical composition was found for  $T$  down to  $-16^{\circ}\text{C}$  for a number of different compounds, which included  $\text{Ca}^{2+}$ ,  $\text{Na}^{+}$  and elemental carbon as tracers of continental, marine and combustion sources, respectively. At a lower  $T$  of  $-25^{\circ}\text{C}$ , recently Si et al. (2019) reported that mineral dust tracers correlated with INP, which suggests that mineral dust was a major contributor to the INP population at that  $T$ . Si et al. (2018) found that for three coastal sites in Canada a model based on K-feldspar as the only INP calculated  $N_{\text{INP}}$  that fit measurements well at  $-25^{\circ}\text{C}$  while at  $-15^{\circ}\text{C}$  measurements were under-predicted, suggesting a missing source of INP that are active at higher  $T$ . In the following it will be examined if there is a correlation between the INP detected in the present study that are ice active at high  $T$  and chemical composition.

The examined filters pieces were not particularly sampled for this study, and were entirely needed to do the above described INP analysis. No dedicated chemical analysis could be done additionally. But as other parts of most of the filters were also used in other studies, some information on chemical composition was available. This was used to derive correlations with  $N_{\text{INP}}$  shown in Fig. 3, i.e., with those INP that are ice active at high  $T$ . Tab. 1 shows values for  $R$ ,  $R^2$  and  $p$  for linear correlations between  $N_{\text{INP}}$  and different bulk chemical properties.

In general, no correlations were found. The highest values for  $R^2$  ( $> 0.3$ ) together with values for  $p < 0.05$  were found for potassium and sulphate in Ny Ålesund and The only case with a positive value for  $R$  (0.59) and a low value for  $p$  (0.01) was found for POC+CC (pyrolyzed organic carbon and carbonate carbon) in Alert. While it was a positive correlation between  $N_{\text{INP}}$  and POC+CC, there were anti-correlations between  $N_{\text{INP}}$  and both potassium and sulphate. And even these highest values for  $R^2$  are still low. Nevertheless, a short explanation should be given on POC+CC. Both POC and CC contain carbon that pyrolyzes at  $870^{\circ}\text{C}$  in a pure He stream (Huang et al., 2006). CC might indicate the presence of soil dust (Huang et al.,

2006). POC includes some charred carbon formed at 550°C, which is the lower temperature step of the applied analysis (EnCan-total-900 method, Huang et al., 2006; Chan et al., 2010) and highly oxidized organic compounds / high molecular weight refractory carbon. Based on previous studies, the POC mass is proportional to the oxygen mass in organic aerosols (Chan et al., 2010), releasing as carbon monoxide at 870°C. POC was observed to form from sucrose and glucose (Huang et al., 2006), and therefore might be indicative of biogenic material. This might points towards a direction in which more detailed studies should be undertaken in the future. In Sec. 5, we will discuss a range of possible sources for the observed INP.

$N_{\text{INP}}$  are several orders of magnitude below total particle number concentrations, and therefore mass concentrations of INP are so small that a correlation between bulk chemical composition and  $N_{\text{INP}}$  might not be expected, particularly not for the very rare INP that are ice active at high  $T$ . This is in line with recent findings for a long term study on INP at Cape Verde by Welti et al. (2018). There, also no correlation between  $N_{\text{INP}}$  and bulk chemical composition was found for  $T$  down to -16°C for a number of different compounds, which included  $\text{Ca}^{2+}$ ,  $\text{Na}^{+}$  and elemental carbon as tracers of continental, marine and combustion sources, respectively.

### 3.2.2 Determination of possible source regions

A more in depth analysis concerning possible INP source regions was done for a selection of filter samples from each measurement station. For that, samples were chosen that had been collected in spring, directly before and after the transition from typical winter to typical summer conditions (see yellow background in Fig. 3). 17 separate filter samples were included in the following analysis, two for VRS, four for Alert and Ny Ålesund each, and seven for Utqiagvik.

5-day back-trajectories were calculated for all of these samples with HYSPLIT (Stein et al., 2015), based on GDAS (Global Data Assimilation System) meteorological data, using an hourly time resolution and starting a new trajectory every 6 hours during the whole time for which sampling was done on the selected filters. Back-trajectories were initiated at an altitude of 100 m above the sampling locations, as this altitude still has a high likelihood of being connected to the ground, and as lower elevations are more prone to uncertainties. In the supplementary information (SI), these back-trajectories are shown separately for the 17 examined filter samples.

Using these back-trajectories, it was examined over which ground the air masses collected on the filters had passed in the 5 days prior to arrival at the measurement station. The aim was to see if contributions from open land or open water were potentially more pronounced during times when INP active at high  $T$  were observed. Therefore, it was distinguished between snow, open land, sea ice and open water. To do so, maps from the Interactive Multisensor Snow and Ice Mapping System (IMS) (Helfrich et al., 2007; National-Ice-Center, 2008) were used. IMS maps are a composite product produced by NOAA/NEDIS (National Oceanic and Atmospheric Administration's National Environmental Satellite Data and Information Service) combining information on both sea ice and snow cover. Information from 15 different sources of input are included in the production of these maps (Helfrich et al., 2007). Currently these maps have been provided for 20 years. We used the daily northern hemisphere maps with a resolution of 4 km (National-Ice-Center, 2008). For each time step we applied nearest-neighbor interpolation in space and time to find the corresponding satellite coordinate along the back-trajectory. With that, for each back-trajectory, we determined the conditions on ground during the passage of the air mass, i.e., if the ground was

covered by snow or ice or if open water or open land was present. The resulting information is shown exemplarily in Fig. 4 for the filter sample collected in Alert starting June 10, 2015. Using these maps, it was counted how often air masses that were collected on one filter were above open land or open water while the air mass was below 100 m. (As will be discussed in detail below, for Utqiagvik, results reported here always refer to an upper altitude of 500 m and 10-day back-trajectories.) An altitude restriction was used as we were trying to geographically locate INP sources on the surface. Additionally, the back-trajectories were only considered back in time until an integral amount of 2 mm of precipitation (taken from the information included in the back-trajectories) was reached. This was done as precipitation formation occurs via the ice phase, so that precipitation leads to a wash out of INP.

Results from the more in depth analysis concerning possible INP source regions, based on back-trajectories and satellite maps as described in Sec. 2.7, is presented in the following. For this analysis, samples were chosen that had been collected in spring, directly before and after the transition from typical winter to typical summer conditions (see yellow background in Fig. 3). 17 separate filter samples were included, two for VRS, four for Alert and Ny Ålesund each, and seven for Utqiagvik. The aim was to see if contributions from open land or open water were potentially more pronounced during times when INP active at high  $T$  were observed.

Fig. 5 shows the number of time steps when air masses were over open land or open water for the separate filter samples. Additionally, gray bars in the background indicate the percentage of time the air masses collected on one filter were below 100 m for Alert, VRS and Ny Ålesund or below 500 m for Utqiagvik. It can already be seen, that, besides for Utqiagvik, the presence of highly ice active INP on a filter is related to air masses that fulfill the above given criteria, i.e., that traveled over open land or open water at a low altitude. It also can be seen that this was not found for Utqiagvik. Initially, no open land and hardly any open water had been found for this site when 5-day back-trajectories were used, together with an altitude restriction of 100 m, which means that air masses did not travel over open land or open water at altitudes below 100 m. To check if the length of the back-trajectory or the chosen maximum altitude influenced our results for Utqiagvik, an analysis was also done using 10-day back-trajectories and 500 m, although here 10-day instead of 5-day back-trajectories were used, and although 500 m was taken as the altitude limit, presented in Fig. 5. This extension The latter simply only resulted in larger percentages of time for which the air masses were below this altitude limit. But still not a large number of time steps was found for which air masses traveled over open land or open water for Utqiagvik. We will get back to this again below.

Four separate rows in Fig. 6 (from A to D) show the spectra of  $N_{\text{INP}}$  (called INP spectra for simplicity from now on) for the samples included in this analysis (right panels) and the locations where the respective air masses traveled over open land or open water at altitudes below 100 m (or 500 m for Utqiagvik) (left panels). Three different types of INP spectra can be distinguished: First there are those for which we observed the start of ice activation only at around  $-10^{\circ}\text{C}$  and which went down to well below  $-15^{\circ}\text{C}$ . For these, INP spectra and locations are depicted in magenta or orange. Second, there are INP spectra for which we observed ice nucleation from roughly  $-5^{\circ}\text{C}$  to above  $-15^{\circ}\text{C}$ . For these, INP spectra and locations are depicted in greenish colors. The third category was used only for Utqiagvik for INP spectra with medium ice activity depicted in blueish. Error bars shown in Fig. 6 show the 95% confidence interval.



For Alert, VRS and Ny Ålesund, the absence or scarcity of orange and magenta marks on the maps in Fig. 6 (maps on the left at A, B and C) shows that almost no open land or open water contributed to air masses sampled on the respective filters. This is in accordance with the corresponding INP spectra, which showed comparably low ice activity. The magenta locations close to Svalbard for the Alert sample correspond to the sample from May 20, 2015, for which somewhat more ice active INP were found than for the subsequent sample from May 27, 2015. For this latter sample, no contributions from open land or open water were observed, and it is, in fact, the sample with the lowest ice activity observed in this study (see Fig. 2). Locations depicted in greenish colors potentially contributed INP that are ice active at high  $T$ . They can be found on open land as well as on open water. In connection to filters sampled at Alert or at VRS they show up in North Greenland, on Ellesmere Island (on which Alert is located), in the Baffin Bay and along the southern part of the west coast of Greenland. Concerning filters sampled at Ny Ålesund, greenish marks show up on Svalbard and the adjacent sea.

The above analysis shows that coastal regions may be particularly important as source for highly ice active INP, including open waters close to coasts. Indeed, highly ice active biogenic INP were found in Arctic surface waters before (e.g., Wilson et al., 2015; Irish et al., 2017). For the highly ice active samples collected on Ny Ålesund on June 12 and June 28, 2012, the surrounding of the measurement station was completely snow free during the times when these samples were collected, whereas for all other cases there was at least partial or total snow cover around the stations. In other words, local terrestrial sources close to the measurement station may also contribute as sources for highly ice active INP, as already discussed in Creamean et al. (2018). Also Irish et al. (2019) describe Arctic land masses to be the source for observed Arctic INP (ice active at  $-15^{\circ}\text{C}$ ,  $-20^{\circ}\text{C}$  and  $-25^{\circ}\text{C}$ ), and these INP were suggested to be mineral dust. On Svalbard, Tobo et al. (2019) found higher atmospheric  $N_{\text{INP}}$  in July than in March, and they additionally described glacial outwash sediments in Svalbard to be highly ice active. This ice activity was assumed to be connected to small amounts of organic (likely biogenic) material. Based on these findings, Tobo et al. (2019) suggest the higher  $N_{\text{INP}}$  in summer to be connected to organic (biogenic) components in glacially sourced dust. Some coastal regions in the Arctic, e.g., the west coast of Greenland together with the region around Baffin Bay and the Canadian Arctic Archipelago as well as the area around the Bering Strait and also Svalbard are known for their abundance of sea bird colonies (Croft et al., 2016). These regions partially coincide with regions highlighted as possible INP sources in Fig. 6. These regions are known to emit ammonia, which plays a role in new particle formation in the Arctic (Croft et al., 2016). But clearly, newly formed particles are not expected to contribute to atmospheric INP at the temperatures examined in this study, and INP are likely simply also emitted from regions with high biological activity. In Sec. 5 we will discuss possible INP sources in more detail.

Some regions in the Arctic are known for their abundance of sea bird colonies, e.g., the west coast of Greenland together with the region around Baffin Bay and the Canadian Arctic Archipelago as well as the area around the Bering Strait and also Svalbard (Croft et al., 2016). These colonies emit ammonia which plays a role in new particle formation in the Arctic (Croft et al., 2016). Newly formed particles are not expected to contribute to atmospheric INP at the temperatures examined in this study, however, it is striking that these regions are also those that were found here as possible sources for highly ice active INP. The available open land does not only attract sea birds but also seems to enable the emission of highly ice active INP. Similarly, open water close to coasts did show up as possible locations contributing to highly ice active INP, too. Highly ice



active biogenic INP were found in Arctic surface waters before (Wilson et al., 2015; Irish et al., 2017, e.g., ), and below, in Sec. 5 we will discuss possible INP sources in more detail. It should also be mentioned that for the highly ice active samples collected on Ny Ålesund on June 12 and June 28, 2012, the surrounding of the measurement station was completely snow free while these samples were collected, whereas for all other cases there was at least partial or total snow cover around the stations.

5 In other words, local terrestrial sources close to the measurement station may also contribute as sources for highly ice active INP, as already discussed in Creamean et al. (2018).

For Utqiagvik, altogether data from seven filters were included in the analysis. Two of them showed INP spectra at comparably low  $T$ , three at medium  $T$  and two at high  $T$ . For all types of INP spectra, no contribution from open land was observed with the back-trajectory analysis. Only minor contributions from open water were found for the latter two types, although the analysis was extended to include 10-day back-trajectories and the maximum altitude up to which air masses were considered was relaxed to 500 m. Air masses did travel below 100 m, and even more often below 500 m (see Fig. 5 and the back-trajectories for Utqiagvik and their heights profiles in the SI). But the transition to filters on which INP active at comparably high  $T$  were found to happen earlier at Utqiagvik than at the other three measurement locations, already towards the end of March. IMS maps almost exclusively identified the ground as sea ice and snow in the regions that were crossed by the air masses, even until beginning of May 2013. And in general, air masses spent more time over sea ice than over snow (see back-trajectories in the SI). There has to be a source for highly ice active INP that was not revealed in the analysis done here. Polynyas and open leads may contribute to explain this inconsistency. The resolution of the IMS maps used may be too coarse so that open water related to polynyas and open leads could have gone unnoticed.

While the analysis introduced here shows regions that potentially may have contributed highly ice active INP to the sampled air masses, it does not make any statement about other regions. Other regions potentially could be sources, too, but might only have been crossed by air masses at high altitudes, or may not have been crossed at all.

## 4 Comparison with literature

Fig. 7 shows the ranges of  $N_{\text{INP}}$  observed for the four stations as shaded areas. Data from the different stations cover a rather similar range. As explained above (Sec. 2.6),  $N_{\text{INP}}$  could only be measured up to some  $10^{-2} \text{ L}^{-1}$ , depending on the volume of air sampled onto one 1 mm filter piece. Hence the upper concentration limit of our data is determined by the measurement method. The gray background shows literature data of  $N_{\text{INP}}$  determined from precipitation samples collected mostly in North America and Europe (Petters and Wright, 2015). In that data set, those samples showing the highest  $N_{\text{INP}}$  at  $T > -20^\circ\text{C}$  originate from rain and hail samples collected in North Carolina (US) and Alberta, Montreal (CA). Some of the INP spectra we detected at the highest  $T$ , observed particularly in Alert and Utqiagvik, show values for  $N_{\text{INP}}$  that are similar or only about 1 order of magnitude below data reported in Petters and Wright (2015). At the lowest temperatures at which we detected INP spectra,  $N_{\text{INP}}$  are lower than data from Petters and Wright (2015), i.e.,  $N_{\text{INP}}$  in the Arctic may, at times, be lower than at other locations. lowest Arctic  $N_{\text{INP}}$ , as those we observed in the winter months, might be below the lowest values observed on

continents in mid-latitudes. It is worthwhile adding that still lower concentrations were observed in marine remote locations in the Southern Ocean (McCluskey et al., 2018a) and for clean marine air in the North East Atlantic (McCluskey et al., 2018b).

Fig. 7 shows additional data on Arctic  $N_{\text{INP}}$  from literature that was already discussed in the Introduction (Borys, 1983, 1989; Bigg, 1996; Bigg and Leck, 2001; Rogers et al., 2001; Prenni et al., 2007; Mason et al., 2016; Conen et al., 2016; DeMott et al., 2016). Not every single data point from these papers is shown, as Fig. 7 aims at giving an overview of the range of data that exists. Data on Arctic  $N_{\text{INP}}$  in general is still scarce, which is particularly true for data at high  $T$ . Also, data scatters over a wide range. Highest values of  $N_{\text{INP}}$  shown in Fig. 7 originate from one set of aircraft measurements made in May (Rogers et al., 2001), while a second set of aircraft measurements, taken in September and October on-board an aircraft flying out of Alaska, agrees with our data at the highest  $T$  (Prenni et al., 2007). For the data taken from (Prenni et al., 2007), the reported measurement uncertainty is shown, representative for typical uncertainties for the type of instrumentation used in Rogers et al. (2001) and Prenni et al. (2007). Error bars indicate one standard deviation at the higher end. The lower end is indicative of the detection limit, and for a substantial fraction of measurements no INP were detected in Prenni et al. (2007).

Going back to Fig. 7, literature data from ground-based measurements that are in the same range of  $N_{\text{INP}}$  in which we measured are also within the same  $T$  range. But besides for data from Conen et al. (2016), these data are at the lower end of  $T$  that we observed. This also holds for the data from DeMott et al. (2016), which were obtained during summer ship cruises in the Baffin Bay and in the central Bering Sea.

Data shown for Borys (1983) were taken in Ny Ålesund and Utqiagvik. A slight tendency towards higher  $N_{\text{INP}}$  in summer months, compared to winter months, can be seen. Similarly, as said in the Introduction, Bigg and Leck (2001) found a decreasing trend for  $N_{\text{INP}}$  at  $-15^{\circ}\text{C}$  from July to September. The observed decrease was roughly one order of magnitude, however, the scatter from sample to sample was almost as large as that trend. Nevertheless, these data sets are early indications of the annual trend that has been found very clearly in the present study.

Concerning possible sources for INP, Bigg (1996) assumed that mainly oceanic sources contributed to the observed INP, with only a weak contribution from land. Bigg and Leck (2001) discussed a marine origin of at least some of the INP they analyzed. These latter two studies were ship based. The land based study by Conen et al. (2016) showed INP that were ice active at higher  $T$  than those observed in Bigg (1996) and Bigg and Leck (2001). Conen et al. (2016) traced these INP back to terrestrial contributions, possibly decaying leaves. During aircraft measurements, Rogers et al. (2001) identified some INP as mineral dust particles and others as containing low-molecular-weight components. These latter might have been connected to biogenic INP.

These different studies report very diverse  $N_{\text{INP}}$ . In these studies, different instrumentation was used and sometimes different ice nucleation mechanisms were probed. Also instrumental limitations typically determine the ranges of  $T$  and  $N_{\text{INP}}$  that can be probed. All of this might add to the diversity in the data. But, as can be seen in Fig. 3, a difference of up to 2 orders of magnitude in  $N_{\text{INP}}$  was measured between winter and summer for a single temperature in our data set. Therefore, the diversity in  $N_{\text{INP}}$  reported in previous studies will also originate from different times of the year when these studies were made.

All of our samples were collected on land, and regions that showed up as possible sources for highly ice active INP in Fig. 6 were on or close to land. While, as said above (Sec. 3.2.2) other regions can not be excluded as sources, it will be interesting to see in the future if  $N_{\text{INP}}$  detected further away from terrestrial sources will consistently be lower than those obtained on land.

Concerning an influence of INP emitted from ground on higher altitudes, Herenz et al. (2018) recently compared aerosol particle number size distributions (PNSD) measured on ground and during overflights at different heights in May 2014 in Tuktoyaktuk (Northwest Territories of Canada on the Arctic Ocean). PNSD measured on ground and in heights up to 1200 m generally agreed were similar, while PNSD obtained at larger altitudes were clearly different. Therefore, the atmospheric aerosol, including INP, can be similar in heights up to levels where cloud formation is observed. Hence, INP detected by ground-based measurements may well be able to influence ice formation in clouds, at least during times when the cloud layers are coupled to the surface.

Compared to previous literature data introduced in this study, the new long term data introduced in the here presented here study extends the range of Arctic  $N_{\text{INP}}$  towards higher  $T$ . It also clearly shows that Arctic INP concentrations vary throughout the year, with a regular presence of INP that are ice active at  $T$  well above  $-10^{\circ}\text{C}$  throughout the summer months at all four terrestrial measurement stations.

## 5 Discussion

The annual cycle observed for  $N_{\text{INP}}$  in this study is not in tune with what is known for particle number concentrations and size distributions occurring across the Arctic (Tunved et al., 2013; Nguyen et al., 2016; Freud et al., 2017). This is not too surprising: Recent studies, including Arctic locations, found that a large fraction of the number of INP are super-micron in size (Mason et al., 2016; Si et al., 2018; Creamean et al., 2018), while the majority of the number of particles are in the sub-micron size range.

Concerning the annual Arctic aerosol cycle, there is a maximum in particle number concentrations in early spring, before precipitation sets in, caused by accumulating anthropogenic pollution known as Arctic haze (Shaw, 1995).  $N_{\text{INP}}$  are low during that time of the year, which might indicate that anthropogenic pollution does not contribute to atmospheric INP, at least in the  $T$  range examined in this study. This is in line with the observation that Arctic haze particles are not efficient INP (Borys, 1989), and also with a recent study showing that anthropogenic pollution did not contribute to INP in very polluted air in Beijing (Chen et al., 2018). It is also in agreement with observations by Hartmann et al. (2019), based on ice cores from Svalbard and Greenland, who found that  $N_{\text{INP}}$  in the Arctic did not increase over the past 500 years (from roughly 1480 to 1990), while tracers for anthropogenic pollution did increase markedly.

The formation of Arctic haze is related to the fact that during winter months, air masses from mid latitudes can transport aerosol particles into the Arctic (Heidam et al., 1999; Stohl, 2006). In contrast, in the summer months the Arctic lower atmosphere is effectively isolated and transport of atmospheric aerosol particles into the Arctic is low (Heidam et al., 1999; Stohl, 2006). Hence, the here observed increase in  $N_{\text{INP}}$  in late spring and summer has to originate from Arctic sources, including local ones situated close to the measurement site.

Once Arctic haze is removed by precipitation in spring (Browse et al., 2012), Arctic particles are mostly newly formed particles in the size range up to 100 nm (Lange et al., 2018), originating from gaseous precursors (Engvall et al., 2008; Leaitch et al., 2013; Croft et al., 2016; Wentworth et al., 2016; Dall'Osto et al., 2018). Number concentrations of these particles roughly peak in July and August (Freud et al., 2017). New particle formation ceases in late summer or early fall, and until Arctic haze sets in again later in winter, the total particle number concentration is at its lowest, well below  $100 \text{ cm}^{-3}$ . But newly formed particles do not contribute to INP unless subject to particular conditions at cirrus level (Kanji et al., 2017). Nor does their annual cycle follow what we observed for  $N_{\text{INP}}$ , as concentrations for newly formed particles cease much earlier in the year than observed for  $N_{\text{INP}}$ .

Arctic concentrations of mineral dust particles have been measured and modeled (Fan, 2013; Zwaafink et al., 2016) and were described to largely depend on long-range transport, with largest concentrations occurring in spring. However, dust from local sources may also contribute (Zwaafink et al., 2016). Mineral dust particles themselves likely only become ice active at  $T$  at the lower end of or below those examined in the present study, but they may be carriers of biogenic ice active macromolecules (Conen et al., 2011; Tobo et al., 2014; O'Sullivan et al., 2014; Augustin-Bauditz et al., 2016; O'Sullivan et al., 2016; Hill et al., 2016). Continental sources of fungal spores were found to contribute to the organic aerosol observed in the Arctic in summer (Fu et al., 2013), and it is known that spores of some fungal species are ice active at comparably high  $T$  (Pummer et al., 2015). The same also holds for lichen (Moffett et al., 2015) and bacteria (Hartmann et al., 2013). Sources and abundance of bacteria were examined for an area in southwest Greenland (Santl-Temkiv et al., 2018), and roughly half of all airborne bacterial cells were described to originate from local terrestrial environments as e.g., surface soils, while the other half was said to have been long-range transported, originating from marine, glaciated, and terrestrial surfaces. In summary, microorganisms originating from continental sources in general could contribute to the highly ice active INP observed in this study. This is in line with the observation presented in Fig. 6, where particularly for VRS and Ny Ålesund, highly ice active INP were observed for air masses that traveled over open land.

Particle generation at open leads in the Arctic has been observed (Held et al., 2011), related to sea spray from bubble production mechanisms that exist independently of wind (Norris et al., 2011). Leck and Bigg (2005) found particles in the Arctic aerosol which could be attributed to algal exopolymer secretions and suggest they became airborne via bubble bursting. This agrees with observations by Orellana et al. (2011) and Fu et al. (2015), showing that Arctic marine microgels from algae, present both in surface water and SML, contribute to Arctic atmospheric particles. Such microgels were assumed to be related to INP observed in marine SML (Wilson et al., 2015). It was recently described that ice activity in Arctic seawater was negatively correlated with salinity (Irish et al., 2017), possibly indicating that these aquatic INPs were associated with melting sea ice releasing ice active biogenic material into the ocean. Marginal ice zones are known to be of importance for phytoplankton blooms due to melting sea ice releasing iron (Wang et al., 2014). Overall, although the above mentioned particle production mechanism in Arctic open leads was found to be only of minor importance for the overall atmospheric particle number concentration (Held et al., 2011), this, together with wind driven sea spray production, could be a source for at least some of the highly ice active INP observed in our measurements. This likely holds for Alert, but it may also be the case in the example of Utqiagvik, where in April and May very little contributions of open land or open sea, as defined by the IMS maps,

was found, while highly ice active INP were present. These could potentially originate from open leads or polynyas. It should be added that Creamean et al. (2018) found a similarly large increase in highly ice active INP during May at Oliktok Point in northern Alaska, only roughly 300 km east of Utqiagvik. This increase was related to INP from tundra surfaces and open water, particularly the marginal ice zone. Polynyas 700 km away from the measurement site were not found to contribute, likely due to settling of particles during the long traveling times in the slow moving air masses that were observed.

In summary, while anthropogenic pollution and new particle formation do not explain highly ice active INP observed in this study, these INP can originate both from terrestrial or marine sources in the Arctic. These sources are strong in summer and weak or absent in winter, depending on the conditions on the ground. Should terrestrial sources be found to contribute stronger than marine ones,  $N_{\text{INP}}$  further away from land will be lower than those observed here.

Concerning Arctic mixed-phase clouds at temperatures above  $-20^{\circ}\text{C}$ , recently Norgren et al. (2018) observed a depression of cloud ice in these clouds when they are polluted, as observed during Arctic haze. Under these conditions, they have a lower amount of cloud ice mass for a given amount of condensed liquid mass. Based on the results of the here presented study, this can be explained by the lower concentrations of INP during times of Arctic haze - a reasoning that sheds light on the importance of the here reported findings which could not yet have been discussed in Norgren et al. (2018). With respect to recent modeling results, Solomon et al. (2018) did large-eddy simulations (LES) of Arctic mixed-phase stratocumulus clouds and find indications that changes in INP in the Arctic aerosol dominate over changes in cloud condensation nuclei (CCN). This shows the potential importance of determining INP and their changes due to climate change in the Arctic. However, Taylor et al. (2018) examined the Arctic annual cycle in cloud amounts from 24 different models and found significant disagreements. They conclude that the parameterization of the ice microphysics in the models contribute to the observed differences. Overall, the field of Arctic mixed-phase clouds and related ice nucleation currently is one of intense research, and the herein presented results contribute to our understanding and lay ground for improved descriptions of INP in modeling in the future.

## 6 Summary and Conclusions

This study shows a yearly cycle of  $N_{\text{INP}}$  at four different Arctic sites, an observation that has not been seen such clearly in previous studies. Maximum values were observed roughly from late spring until well into fall, minimum values were observed during winter and early spring. INP spectra for the most ice active INP are close to some observed at continental locations outside of the Arctic (Petters and Wright, 2015). Potential source regions for INP that are ice active at the higher examined  $T$  were determined based on combining air mass back-trajectories with IMS satellite maps. Such source regions were found in the Arctic on open land as well as on open water, particularly in the Baffin Bay, along the southern part of the west coast of Greenland and on Svalbard and the adjacent sea. Contributions from these regions could explain the increase in highly ice active INP observed in spring in Alert, Ny Ålesund and VRS. However, a possible source region for the highly ice active INP observed in Utqiagvik could not be identified. Here, highly ice active INP appeared already earlier in the year than at the other stations. Open leads and polynyas present in the Arctic sea ice are not identified as open water in IMS maps and may have contributed to these INP.

Regions that were not identified as source regions for highly ice active INP in this study may still contribute such INP. It should still be mentioned that source regions determined here were mostly on or close to land. We showed that there is a large scatter in literature values observed for  $N_{\text{INP}}$  in the Arctic. As we find a yearly cycle in Arctic  $N_{\text{INP}}$ , this large scatter may partially originate in different times of the year when samples were taken. It may also depend on the proximity to land where  
5 the sampling was done, if terrestrial sources and sources in close proximity to land should show to be dominant sourced for highly ice active INP in the future.

Independent of their origin, these observed INP might be transported certain distances and might have more than only local influences. This can become important, as biogenic emissions in general can be expected to increase in the Arctic in coming years due to Arctic amplification, which amplifies marine primary production (Arrigo et al., 2008; Ardyna et al., 2014), and  
10 changes Arctic microbial communities (Deslippe et al., 2012). Feedback mechanisms involving ice formation and with this radiative properties and lifetimes of Arctic stratiform clouds may exist, related to biogenic INP. Therefore, more thorough studies concerning Arctic INP are needed. Determining the current status of INP in the Arctic and future changes that are to be expected might help to understand aerosol cloud interactions in the Arctic and their significance for the observed strong Arctic warming.

*Data availability.* In case of acceptance, data will be made available on Pangaea.

*Author contributions.* H.W. and F.S. designed research; R.J.S., T.E.B., R.T., S.B., R.B., H.S., L.H., W.Z., and H.H. did field campaign management and/or filter sampling; T.E.B., C.E.M., and W.Z. did chemical analysis; M.H. calibrated and J.L., M.L., O.L. operated the ice nucleation measurement device; P.H. calculated trajectories; A.H. did the work on the satellite data; H.W. analyzed the data and wrote the paper with contributions from L.H., H.H., R.J.S., C.E.M., R.T., F.S., M.H., P.H., M.L., R.B., H.S. and W.Z..

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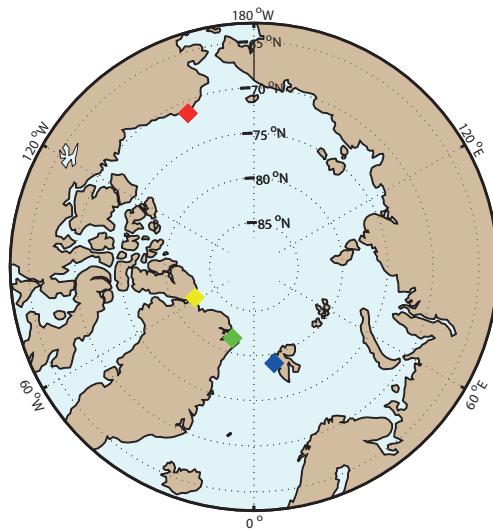
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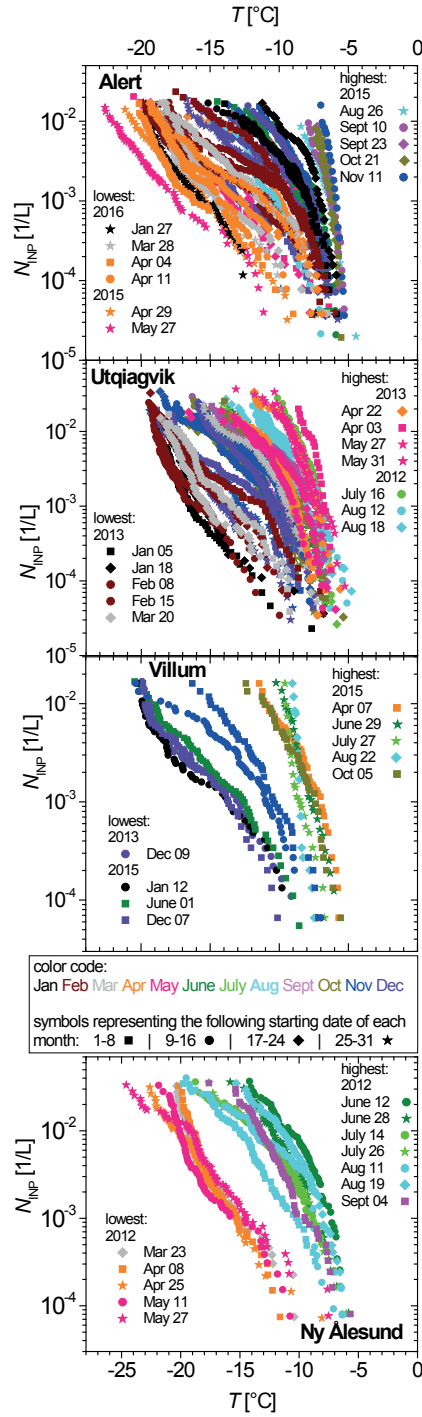
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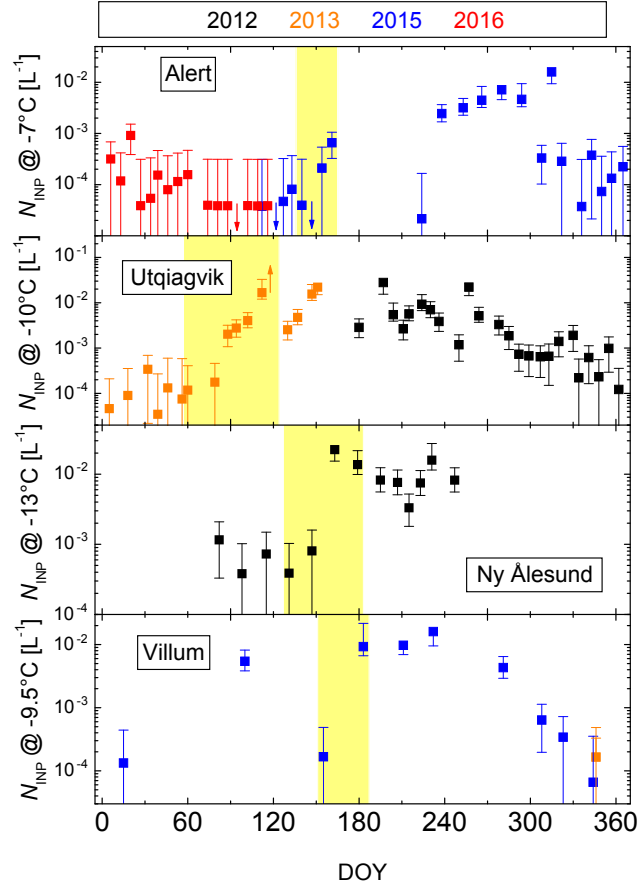


**Figure 1.** The location of the four stations from which filter samples were included herein: Utqiagvik (red diamond), Alert (yellow diamond), VRS (green diamond) and Ny Ålesund (blue diamond).

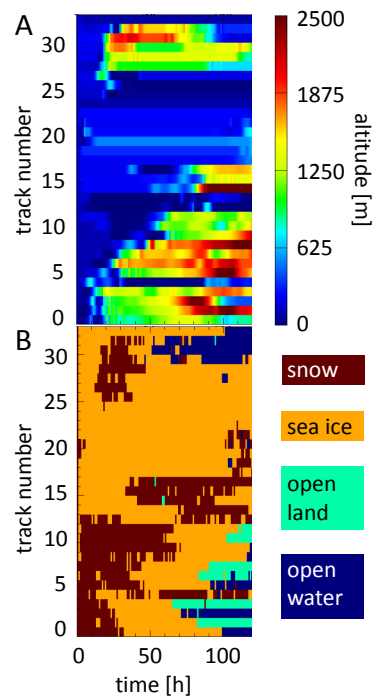




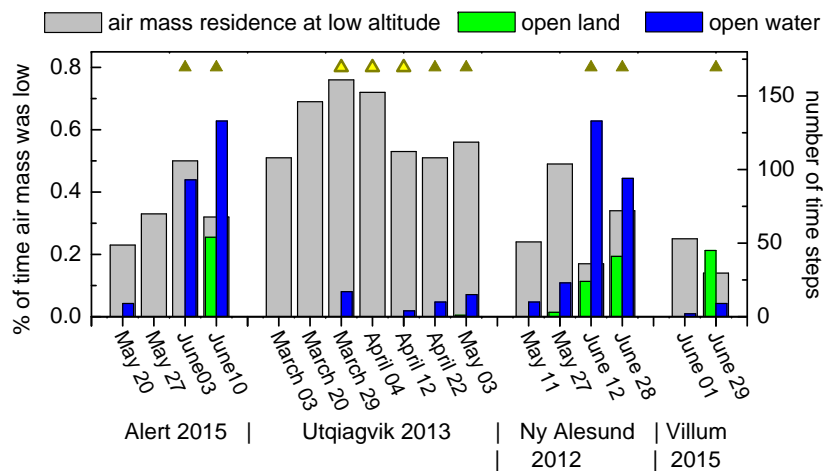
**Figure 2.**  $N_{\text{INP}}$  for all samples. Symbol color distinguishes between data obtained for the different months, while symbol type indicates the day of the month when sampling started. Curves with particularly high and low  $N_{\text{INP}}$  are explicitly listed in the legend. (The  $T$  axis is the same for the upper three panels and different for the lower panel, see values for  $T$  given on top and bottom of the figure, respectively.)



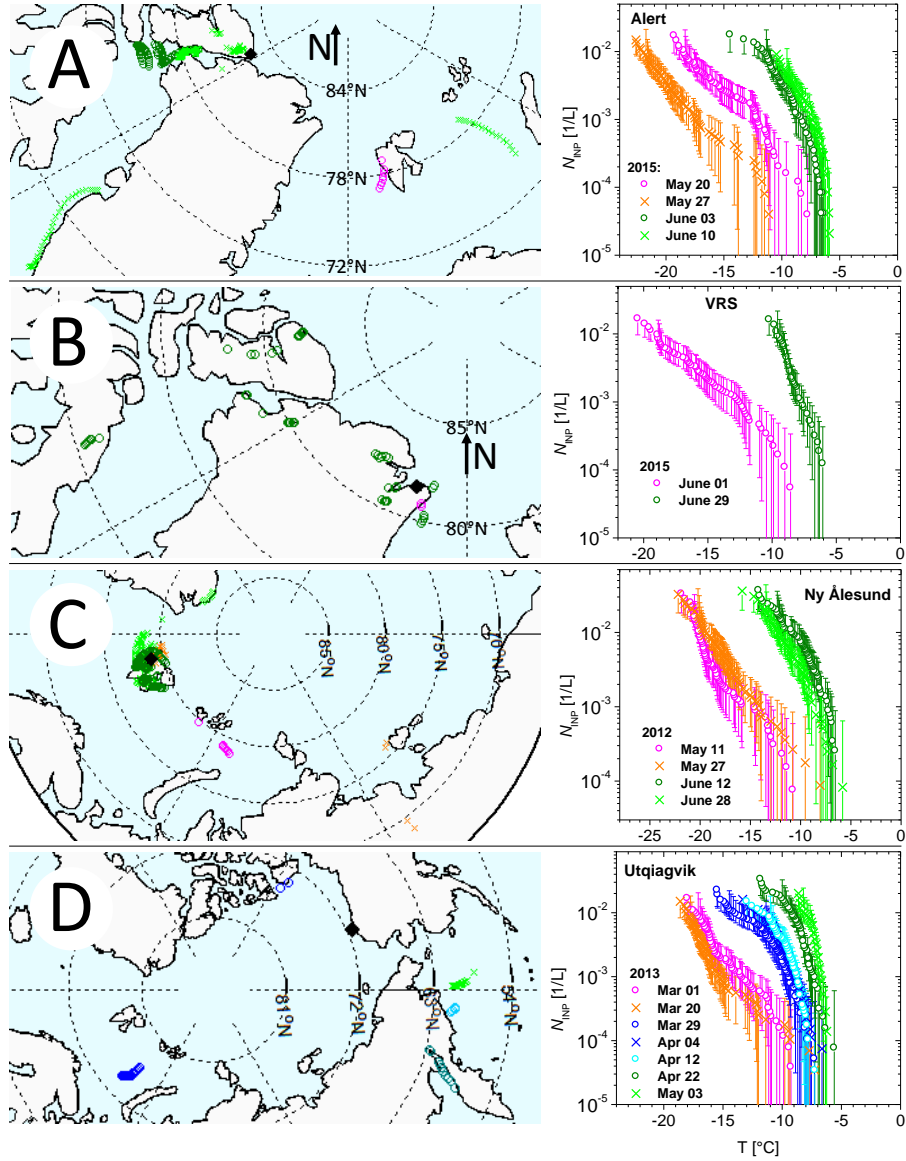
**Figure 3.** Time-series of  $N_{\text{INP}}$  at  $T$  of  $-7^{\circ}\text{C}$ ,  $-10^{\circ}\text{C}$ ,  $-13^{\circ}\text{C}$  and  $-9.5^{\circ}\text{C}$  for Alert, Utqiagvik, Ny Ålesund and VRS, respectively. Small arrows in the panels indicate times when values were either below (for Alert) or above (for Utqiagvik) the detection limit. The yellow background shows for which samples an more in-depth analysis is presented in Sec. 3.2.2. Error bars show the 95% confidence interval.



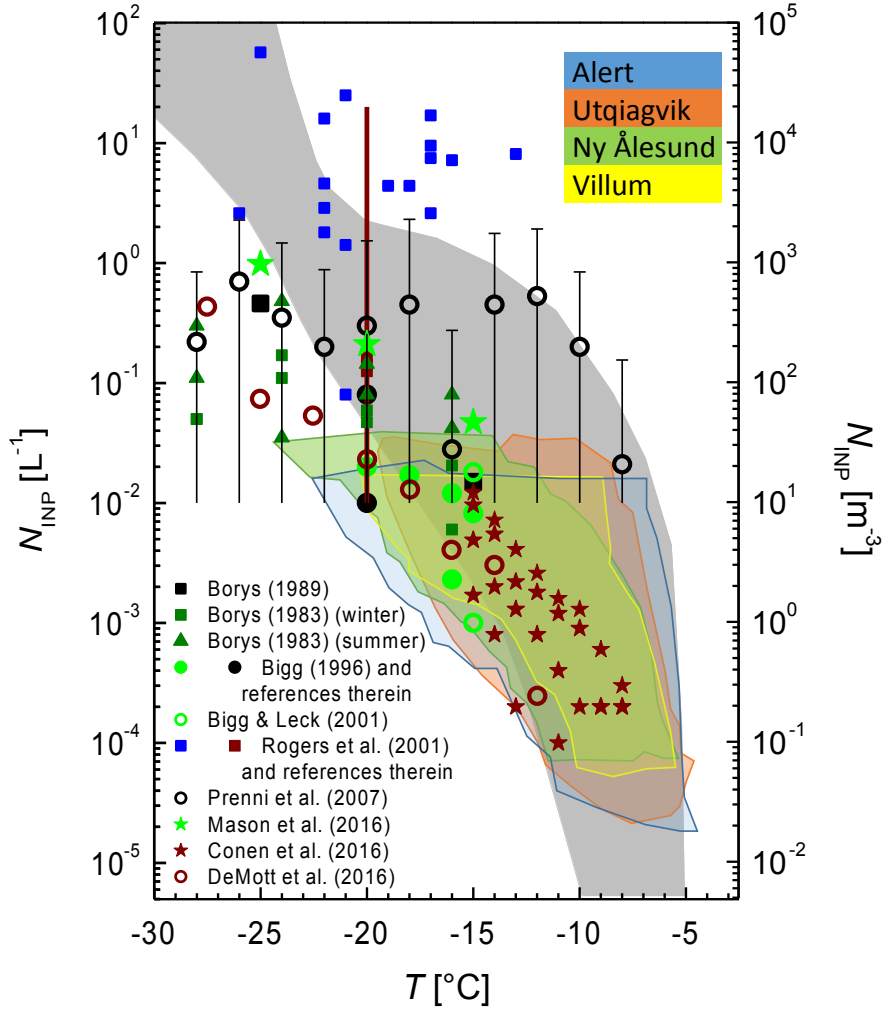
**Figure 4.** Exemplary results from the analysis of the IMS maps for the filter collected in Alert starting June 10, 2015. In both panels, each row (indicated as track number) represents one trajectory, going back in time for 5 days, starting from 0 (when sampling took place) and displaying 120 separate time steps. The colors indicate the altitude of the air mass for the different time steps in panel A and the respective nature of the ground at the location of the air mass in panel B.



**Figure 5.** The number of time steps when air masses were at low altitudes and over open land or open water are shown in green and blue, respectively, for different filter samples. The analysis shown was done for altitudes up to 100 m for Alert, Ny Ålesund and VRS, and up to 500 m for Utqiagvik. Gray bars in the background indicate the percentage of time the air masses collected on each filter were below that altitude. Triangles indicate samples for which highly ice active INP were detected, i.e., for which INP spectra were measured at high  $T$  (single-colored triangles) and at medium  $T$  (triangles with yellow interior). The respective INP spectra are shown in Fig 6.



**Figure 6.** Rows A, B, C and D each show a map on the left side in which locations are indicated where air masses collected on different filters crossed over open land or open water while being at a low altitude (below 100 m for Alert, Ny Ålesund and VRS, below 500 m for Utqiagvik). Black diamonds indicate the location of the measurement site. (Please note: the maps in the two lower rows are rotated by 90° compared to the those in the two upper rows.) The right panels show the INP spectra for the corresponding filters.



**Figure 7.** Comparison of  $N_{\text{INP}}$  determined in this study for the Arctic with literature data by Petters and Wright (2015) (gray background) and Borys (1983, 1989); Bigg (1996); Bigg and Leck (2001); Rogers et al. (2001); Prenni et al. (2007); Mason et al. (2016); Conen et al. (2016); DeMott et al. (2016). Green and brown symbols represent data from [ground surface](#) based measurements, black and blue ones from airborne measurements. For Rogers et al. (2001), brown data indicate data cited from literature, with the vertical bar indicating the extent of the reported values.

**Table 1.** R,  $R^2$  and p values for linear correlations between  $N_{\text{INP}}$  (as shown in Fig. 2) and different bulk chemical properties.

location	species	R	$R^2$	p
Utqiagvik	EC	-0.36	0.13	0.02
	OC	-0.04	<0.01	0.82
	fluoride	0.14	0.02	0.41
	chloride	0.13	0.02	0.43
	nitrite	0.22	0.05	0.19
	bromide	-0.01	<0.01	0.98
	sulfate	-0.12	0.01	0.47
	nitrate	0.03	<0.01	0.86
Ny Ålesund	PM10	-0.36	0.13	0.22
	OC	-0.39	0.15	0.18
	ammonium	-0.44	0.20	0.13
	K	-0.57	0.32	0.04
	Mg	-0.36	0.13	0.22
	nitrite	0.15	0.02	0.62
	nitrate	-0.16	0.03	0.59
	sulphate	-0.60	0.36	0.03
	Na	-0.30	0.09	0.32
	Ca	-0.50	0.25	0.08
	Cl	-0.13	0.02	0.68
	MSA	0.18	0.03	0.55
Alert	POC+CC	0.59	0.35	0.01
	OC	-0.12	0.01	0.62
	EC	0.05	<0.01	0.84

# ***Supplement of*** **Annual variability of ice nucleating particle concentrations at different Arctic locations**

by Wex et al.

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## **1 Background measurements**

Following a recommendation given in Polen et al. (2018), Figs. S1 to S3 show frozen fractions ( $f_{ice}$ ), i.e., the measured parameter, for field blanks together with some spectra of  $f_{ice}$  for pure water and for filter samples that were sampled in the days directly before and after the field blank was taken. Field blanks were treated similar to filters onto which sampling was done, only were they not subjected to sampling air through them. They did, however, spend time in the sampler. The three different panels in Fig. S1 clearly show, that the background level is influenced by the atmospheric INP concentrations, as field blanks taken from April until October show a much larger signal than those collected during the other months. Therefore, and as the availability of field blanks varied between the different measurement stations (there were none for VRS, two for Ny Ålesund and Utqiagvik each, and 9 from Alert), signals from the field blanks were not subtracted from the signals from the samples. However, in all cases and at all stations, the background was low enough to not have influenced the interpretation of the results from the filters presented in this study.

To show the influence of the background on the measurements in more detail, for five filter samples from Alert the background was subtracted. These five samples were selected such that the background had the highest possible impact. From the upper panel of Figs. S1, both samples were included (April 29 and May 07), relating to the same field blank which had been taken on May 07. From the middle panel, the higher one of the two field blanks was taken, together with the lower one of the two samples that it was collected closest to (October 21). Similarly, from the lowest panel, the highest field blank with the lowest of the two related samples (June 10) and the lowest sample (January 27) with the related field blank were included. The five spectra of  $f_{ice}$  for the filter samples are shown together with those for the field blanks in the two left panels of Fig. S4. Subtraction of the background was done by converting  $f_{ice}$  to concentrations of INP per volume of water/suspension (known as  $K(T)$  in the nomenclature by Vali, 2019), following Polen et al. (2018).  $K(T)$  from the field blanks was then subtracted from that of the filter samples, and the result was converted to background corrected atmospheric INP number concentrations. Ultimately this procedure can be summarized as:

$$N_{INP,corr} = (-\ln(1 - f_{ice,s}) + \ln(1 - f_{ice,b}))/V \quad (1)$$

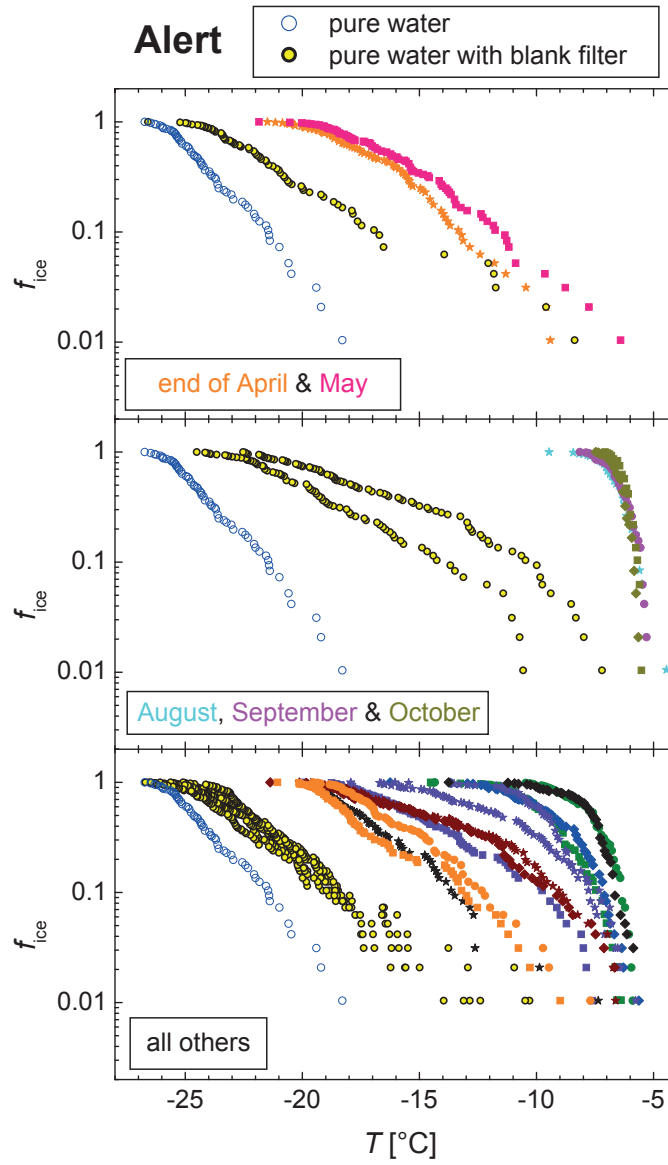


The corrected atmospheric INP number concentration is  $N_{INP,corr}$ , the frozen fractions measured for the filter samples and the field blanks are  $f_{ice,s}$  and  $f_{ice,b}$ , respectively, and  $V$  is the volume of air sampled onto a 1 mm filter piece which was immersed in each examined droplet.

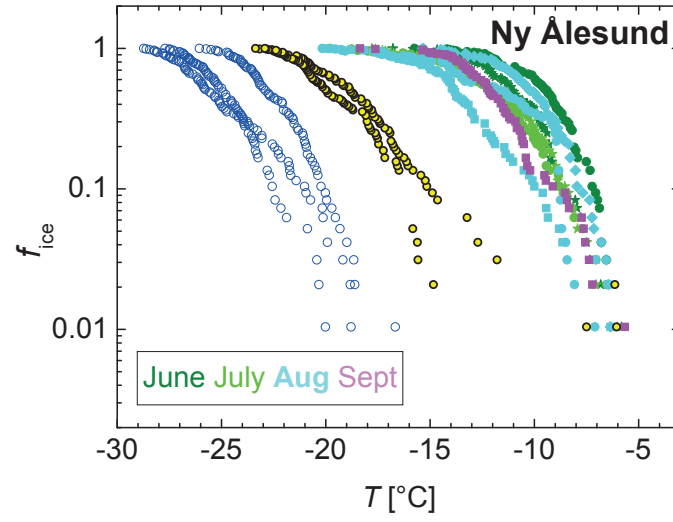
Uncorrected and corrected INP concentrations are shown in the two right panels of Fig. S4.  $N_{INP,corr}$  was lower than the uncorrected value by less than 2% for two samples (June 03 and October 21) and between 5% and 20% for January 27 and for the samples from April 29 and May 07 below -14°C. Above -14°C, this difference was between 20% and 40% for May 07, which, compared to the range the overall signals span, is still small and within the uncertainty given in the main text. For the sample from April 29 above -14°C, there are clear differences between uncorrected and corrected values, going up to 80%, and at the highest temperatures there were two values for which the field blank showed higher  $f_{ice}$  than the sample (see red circle in lower right panel). The respective field blank had been taken on May 07, i.e., at the end of the period when the sample from April 29 was collected. Also, the sample from April 29 was amongst the lowest measured (it could not be included in the time series shown in Fig. 3 in Sec. 3.1 in the main text). Indeed, the sample of April 29 was the only one in this whole study for which a background signal was above the respective measurement. As it was shown above, the signal from the field blanks varied along with the signals from the sampled filters. The sample from April 29 was collected at the end of the season with low INP concentrations, and possibly the field blank taken on May 07 does not represent the background that was present during the time when the sample from April 29 was collected. Therefore, for the case of April 29, rather the attribution of this field blank to the filter, rather than the data obtained from that filter itself, might be considered problematic.

With a background as variable as that observed here, subtracting a background may induce errors. A subtraction of the background was not done in this study, for the following reasons: 1) As discussed above, even for these examples with a comparably high influence from the background, the difference between corrected and uncorrected INP concentrations was generally low. 2) A clear attribution of field blanks to related samples would, in all cases, be needed, due to the variable background, but this seems not even possible for Alert, for which the largest amount of field blanks was available. 3) The interpretation of the results from the filters presented in this study is the same for both uncorrected and background-corrected samples.

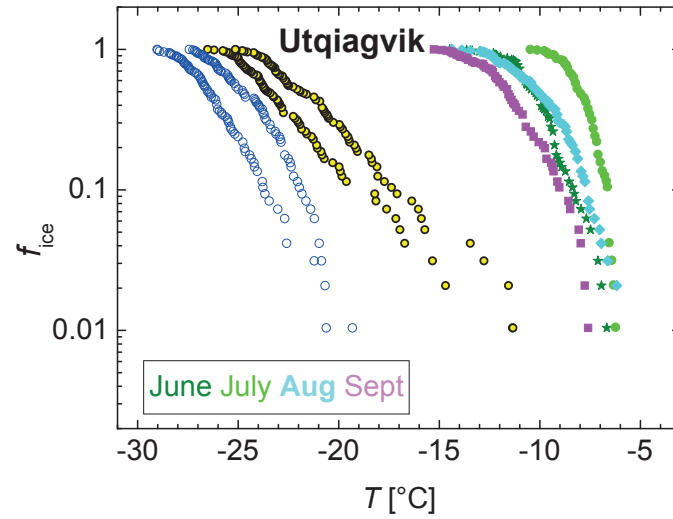
Some filters sampled during spring 2017 in Alert and from March until September 2015 in Ny Ålesund also had been examined. However, these filters had shorter sampling times of only one day and less than 60 L of air had been sampled onto each single circular 1 mm filter piece.  $f_{ice}$  determined for these filters were so close to the background determined from the blank filters, and therefore that these data were not used in this study. It should be added that for cases where a lower background can be achieved or where air with high concentrations of INP is sampled, 60 L of air sampled into each one of the examined droplets may suffice to get values that are well enough separated from the background.



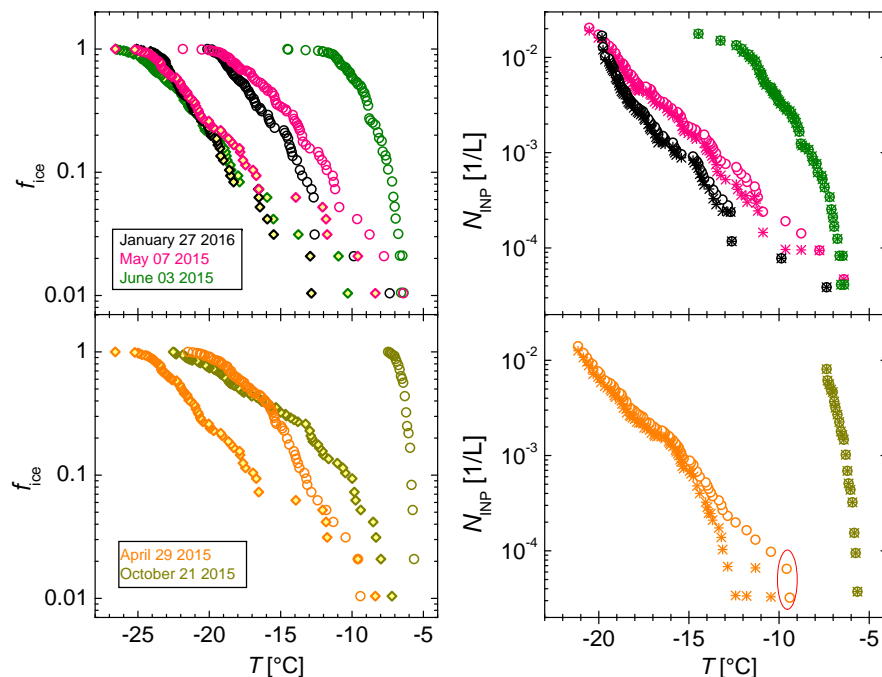
**Figure S1.**  $f_{ice}$  derived for measurements of pure water (blue open circles), blank filters (black circles with yellow filling) and filter samples sampled prior and after the blank filter was taken. The color code for the filter samples is the same as used in Fig. 1.



**Figure S2.** Similar to Fig. S1, but for Ny Ålesund.



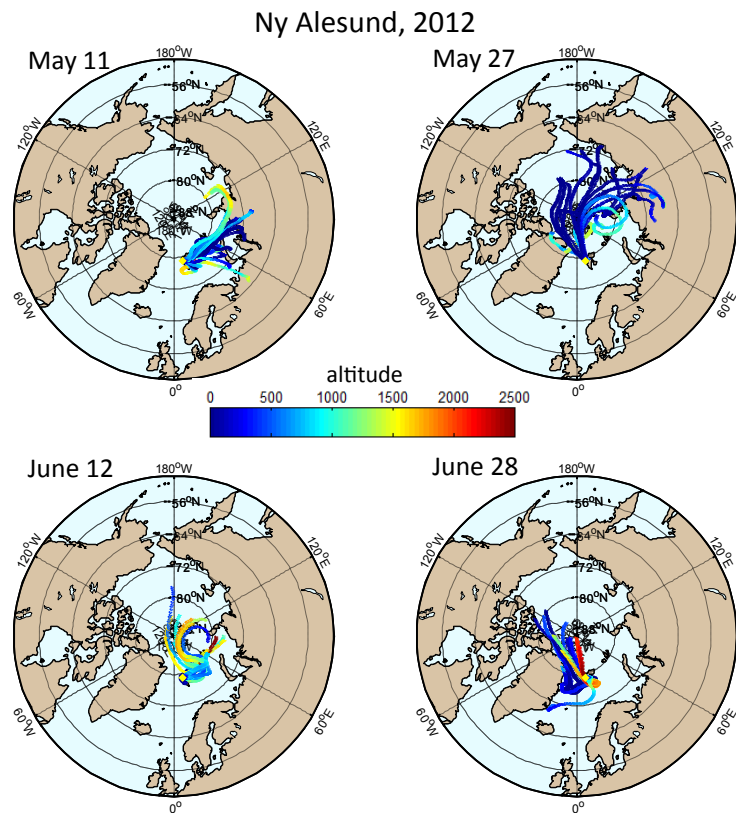
**Figure S3.** Similar to Fig. S1, but for Utqiagvik.



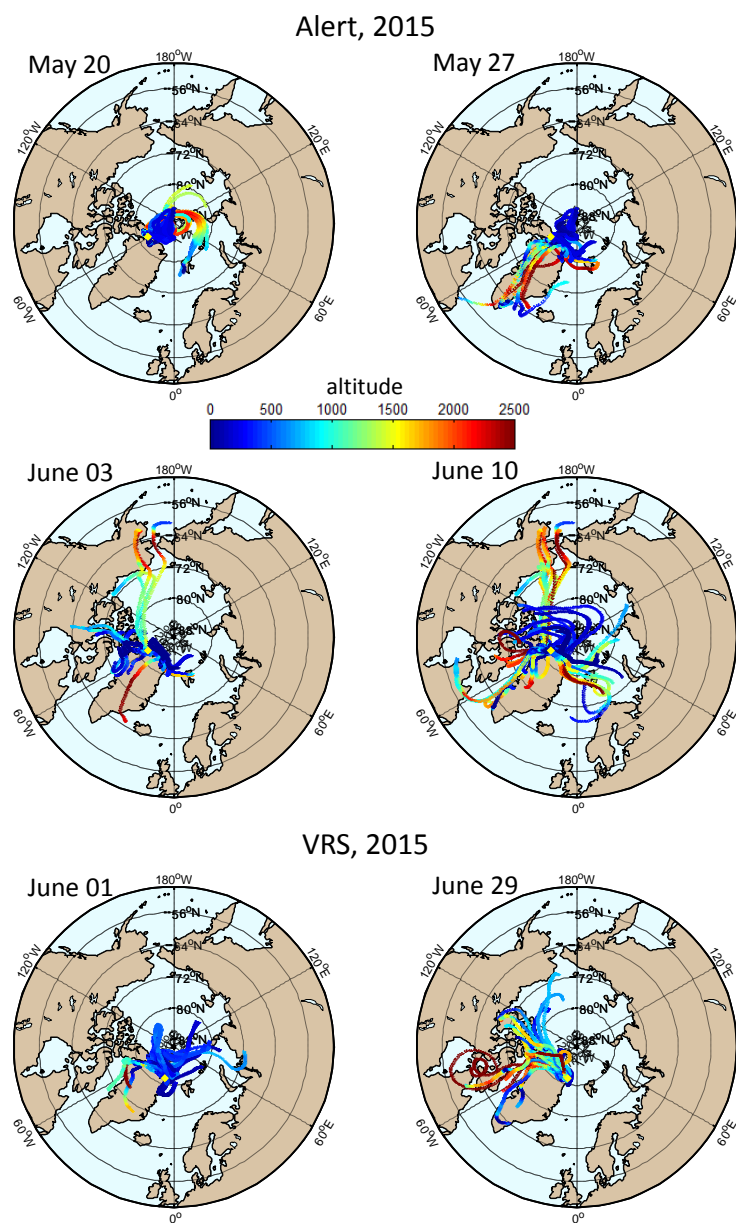
**Figure S4.** The two panels to the left show frozen fractions measured for five different filter samples (open circles), together with the respective field blanks (diamonds, filled in yellow). Symbol colors indicate the day when sampling of the filter sample was started. (Two separate panels were used to increase the visibility of the separate curves.) The two right panels show uncorrected and background-corrected INP concentrations (open circles and stars, respectively). The red circle in the lower right panel shows the only time observed during this study when a background signal was above the measured signal.

## 2 Back-trajectories

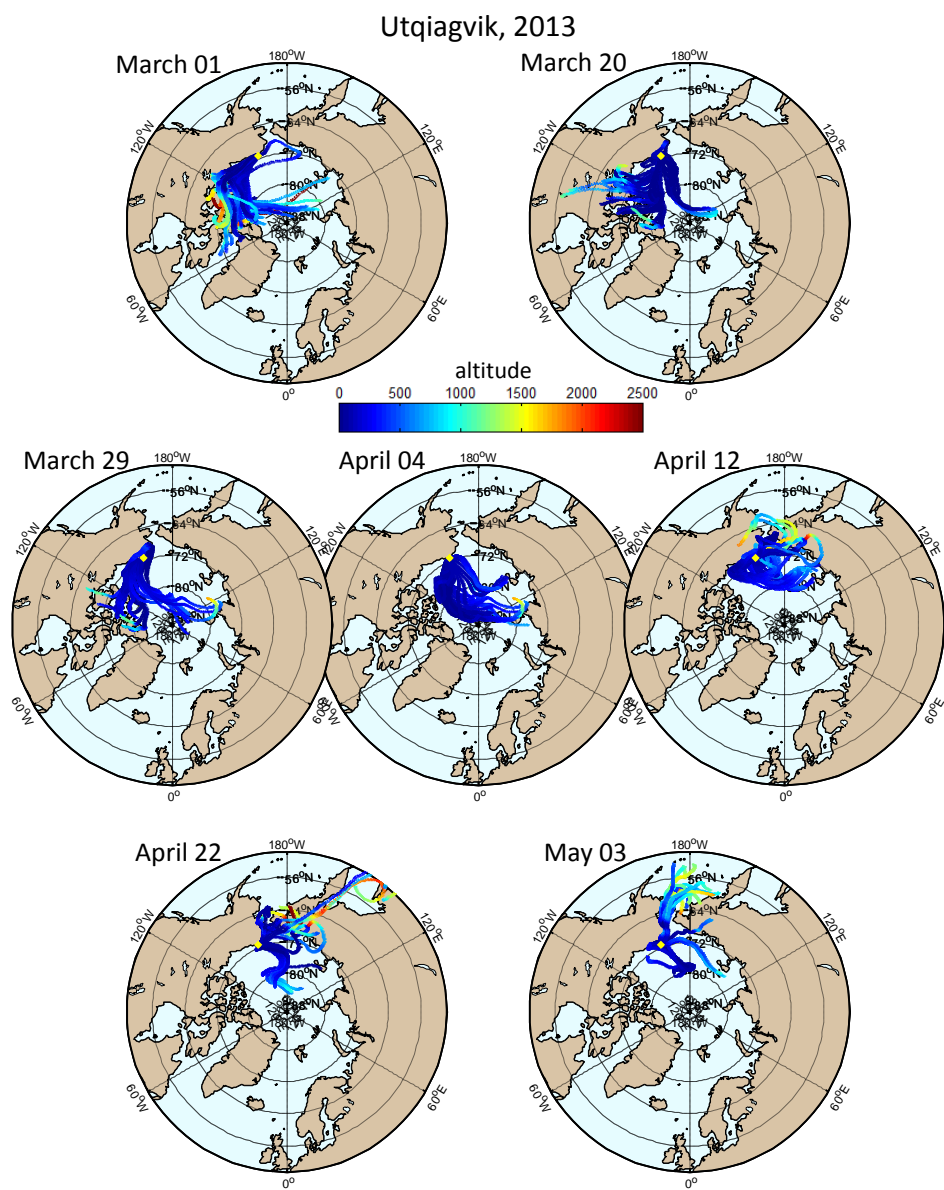
Figs S6 to S8 show the back-trajectories that were derived for the 17 selected filters examined in detail in Sec. 3.3. For Utqiagvik, 5-day and 10-day back-trajectories are shown separately in Figs S7 and S8 and the information on the altitude of these back-trajectories is explicitly shown in Fig. S9.



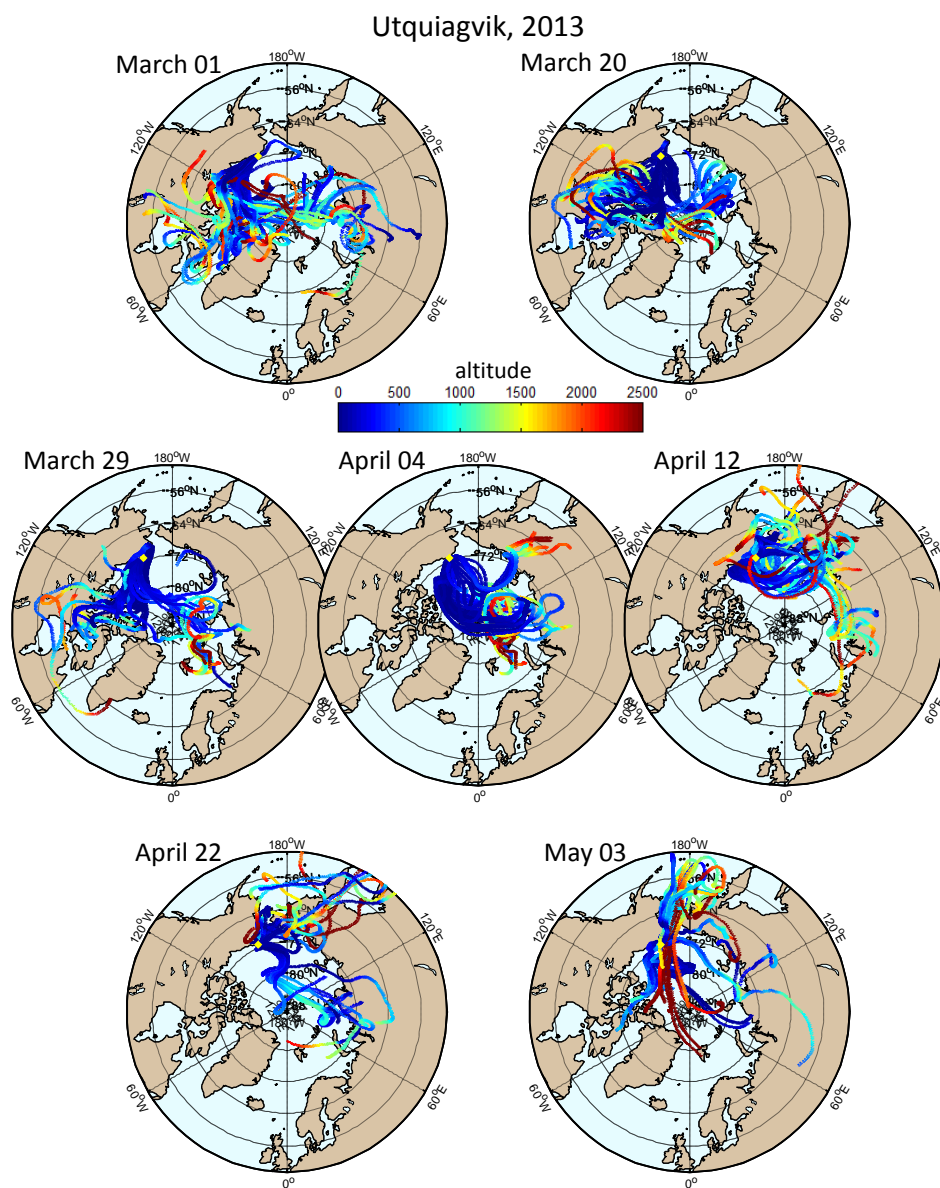
**Figure S5.** 5-day back-trajectories for the filter samples from Ny Ålesund discussed in Sec. 3.3.



**Figure S6.** Similar to Fig. S5, but for Alert and VRS.

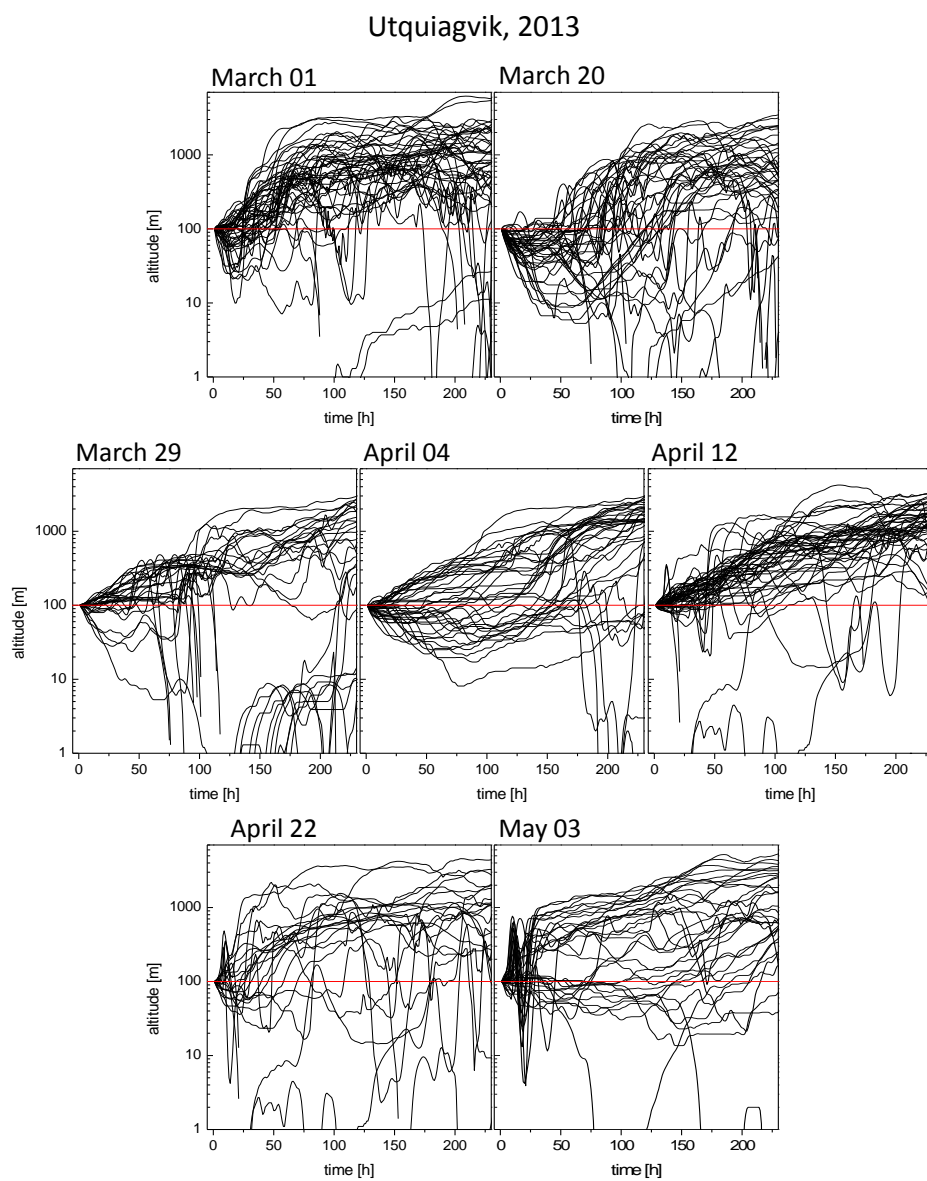


**Figure S7.** Similar to Fig. S6, but for Utqiagvik.



**Figure S8.** Similar to Fig. S7, but showing 10-day back-trajectories.





**Figure S9.** Altitudes of the 10-day back-trajectories displayed in Fig. S8.

### 3 Recommendations

We felt it could help future research if we shared some recommendations, based on lessons we learned. These are the following:

- It could be advantageous to sample on filters that allow for washing off particles, as this enables to do dilution series. With this, obtained data can cover a broader  $T$  range (e.g., polycarbonate membrane filters, Price et al., 2018), compared to that obtained in the present study.

- A higher time resolution used in the filter sampling will facilitate source apportionment. Still, care has to be taken to sample enough material to be above the detection limit, as for the present study samples on which less than 60 L of air had been sampled onto each single circular 1 mm filter piece (i.e., into each examined droplet) could not be used as they were too close to the background (see above, SI 1). Therefore shorter sampling times have to be counterbalanced by higher flow rates during sampling. Suppressing the filter background, if possible, would be of advantage, too. Recently, Polen et al. (2018) gave a number of recommendations related to use of and data from droplet freezing techniques as the one applied in this study, with a focus on working cleanly.

- Sampling with in-situ devices (Rogers et al., 2001; Prenni et al., 2007) can complement off-line filter analysis, measuring down to lower  $T$ . However, they need comparably high  $N_{INP}$  to overcome their detection limits and hence typically do not obtain contribute values at higher lower  $T$  where  $N_{INP}$  is higher lower. Concentration of the aerosol prior to the in-situ sampling, done e.g., in Tobo et al. (2013), can help to increase that  $T$  up to which measurements can be made, increasing the range of  $T$  for which overlap between in-situ and off-line techniques can be obtained.

- Parallel sampling of additional sufficient material to derive chemical composition enables more indepth testing of possible components present in INP. This might help to connect INP to their sources or to at least enable to corroborate the biogenic nature of those INP active at high  $T$ , using e.g., a test of the heat sensitivity of INP for the latter. It should be added that also the choice of filter material is important. Polycarbonate filters can be washed off and different analysis, including heat treatment, can be done on the suspensions. Teflon filters were found to not work well for washing off in Chen et al. (2018) and also could not be punched. When considering heat treatment, parts of sampled quartz fiber filters could be heated prior to punching filter pieces for the analysis, while suspensions can not be made as INP are retained by the fibers (Conen et al., 2012).

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