We would like to thank both of the reviewers for their insightful feedback regarding our manuscript. We have revised based on their commentary and believe the manuscript is much stronger as a result.

Reviewer 1

Review of "Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location" by Creamean et al. The authors report measurements of ice nucleating particles (INPs) over the period of 3 months in the Arctic during spring. Data from 17 days are presented. Since there are few measurements of INPs in this region and INPs are important for predicting climate, the measurements are certainly valuable. However, most of the conclusions reached by the authors are not well supported by the current analysis. The authors have a nice data set, but a more rigorous analysis is needed to support the conclusions in this manuscript. Specific comments are included below.

Comments:

Abstract. The authors state that the concentrations of coarse mode INP at -15 C were low during the first part of the campaign but then increased by nearly 2 orders during late May. To better illustrate these results, please use a log scale in Fig. 4 to represent the INP concentrations. Currently, the changes in INP concentrations at -15 C are not clear in Fig. 4 due to the use of a linear scale to represent all freezing temperatures.

Thank you for the suggestion. We have changed Figure 4 so that log scale is shown.

Abstract. The authors state that the higher concentrations were attributed to air masses originating from over sea ice leads and tundra surfaces. This conclusion was mainly based on back trajectories and information on snow and ice coverage in the Arctic. Although reasonable, the analysis was not very rigorous. The authors did not rule out anthropogenic pollution from local sources. This should be done in a rigorous and quantitative manner if the authors want to claim the INPs are from natural sources.

The air mass trajectory analysis was used for context for the chemical analysis conducted, both of single particle and bulk compositional information, in addition to other supporting information. First and foremost, we know that the aerosol composition in general in the size ranges relevant to the INP measurements were predominantly sea spray aerosol and dust based on the chemical analyses. Very little influences from soot or fly ash (i.e., local industrial pollution) were observed (4% and 16% of the particles that were > 1.15 µm on 23 May and 28 May, respectively; Figure 7). Second, based on size alone, we would not expect pollution sourced from Prudhoe Bay (which a majority by number are sub-100 nm; Creamean et al. (2018a); Maahn et al (2017)) to overlap with the sizes of the INPs observed (i.e., > 2.96 µm) at Oliktok Point. Third, INPs measured at the warmer end of the temperatures we focus on during our case study are likely biological or dust in origin (e.g., Kanji et al. (2017), Murray et al. (2012)). Fly ash and soot generally form ice at much colder temperatures. We have revised Figure 7 to show wind direction and speed separately, and now discuss this in more detail at the end of section 3.3. We note that although winds were easterly on 28 and 29 May, wind prior to those days were variable and can help explain the aerosol sources during our higher marine and terrestrial INP concentration periods. Thus, based on the combination of freezing temperatures, size, single-particle composition, bulk composition, local meteorology, and air mass transport, we demonstrate that there was indeed little influence from local anthropogenic pollution. We added several sentences discussing these points at the end of section 3.3—that it is possible but unlikely that local pollution largely influenced the INP concentrations during late May.

Section 1.2 (Sample collection). For INP analysis, particles were deposited on Vaseline-coated Mylar. Could the Vaseline influence the INPs and shut off freezing of some of the INPs? Were all INPs extracted

from the Vaseline coating? Since this is a new approach and Vaseline could cause artifacts, the influence of Vaseline on INP concentrations should be discussed.

The fact that Vaseline is used in preparation of the copper plates and contributes very little artifacts to the INPs during testing indicates it likely does not contribute significant artifacts to the samples containing aerosols impacted on Vaseline. The use of the Vaseline coating has already been tested and discussed in Creamean et al. (2018b) as we note in the methods section. Additionally, Tobo et al. (2016) presented drop freezing results using Vaseline with little to no contaminant influences from the Vaseline itself, and observed blank water samples freezing starting at –30 ℃. However, we cannot determine if all the INPs were extracted with certainty. For clarity, we have added, "It is possible not all particles were removed during the extraction process; however, previous control testing indicates sufficient aerosol loading is resuspended (Creamean et al., 2018b)." to section 1.2.

Page 8, lines 19-21. The authors refer to previous studies to suggest that they had relatively clean conditions around late May. But the references are from different years. Hence, these references may not precisely apply to the current study. The authors need to prove that late May was associated with relatively clean conditions. This is not obvious from Figure 2. This is especially important if the authors want to claim the INPs are from natural sources.

The purpose of providing the average values of particle number concentrations in the text, specifically from the CPCs which are indicative of Prudhoe Bay pollution (Creamean et al. (2018a); Maahn et al. (2017)) was to demonstrate how May was relatively clean compared to March and April. To make this clearer, we have revised Figure 2 to show hourly averages and log scale for the particles concentrations to demonstrate the differences between Mar and Apr versus May. We also changed the average values provided in the text to those calculated from hourly concentrations to reflect the values in the figure. One interesting feature that is more prominent is the high variability with very low to concentrations comparable to the previous two months in May. We now discuss this in the text, in addition to highlight such variability as part of the "interesting aerosol events". Please note that we have also removed the wind parameters panel as that was redundant to Figure 7 and we do not discuss the local winds until later when describing the particle compositions.

Page 9, line 11. The larger spread in INP concentrations could also be due to just a larger variation in source strength.

Thank you for pointing this out. We have added this possibility.

Page 9, line 28-29. "particles that are theoretically thought to be too small to serve as efficient INPs." I do not think these references presented a theory that suggested that small particles are not efficient INPs. There are also several laboratory studies that show INPs can have sizes similar to the ranges isolated by stages C and D.

We have revised the wording to instead state that particles of these sizes are relatively less efficient INPs as compared to their larger counterparts.

Page 10, line 6-8. The authors suggest that the back trajectories (Figure 6) illustrate that the transport was slow and remained near the surface. However, there is no information on trajectory height or time in Figure 6. I suggest that Figure 6 be modified to include height information and time information to support the author's claims. Also, the trajectories go outside the plotting area in Figure 6. I suggest that the plotting area be increased so the full trajectories can be observed.

We have revised Figure 6 to include height and time information. We also increased plot size for the full trajectories and included zoomed in panels to show more local scale transport.

Page 10, line 13-15. "Air reaching Oliktok point originated from over a large area of open leads within 30 km off the coast of Utqiagvik (Figure 6d)." This point is not clear from Figure 6d. Where are the open leads in Figure 6d?

We have revised Figure 6 (see response to previous comment) to include an inset of the areas of open water within 30 km and describe this feature more in section 3.3. Open leads are evidenced by the regions that are < 100% sea ice percentage, indicating the presence of open water in the 4 km grid cells. However, we realize that the term "leads" may not accurately represent the area of < 100% sea ice concentrations (i.e. light blue colors directly north of Oliktok Point). The width of leads varies from a couple of meters to over a kilometer, thus, they are difficult to resolve in the 4-km sea ice data we use. It is possible the open water is simply small or larger ice floes that have broken off the pack ice near the ice edge. Thus, we have changed this to "marginal ice zone" (MIZ) as that is a more accurate description of the transition between open water and sea ice during the melt season, but that leads may be a feature within this zone.

Page 10, line 15-16. "sources were generally regional in nature. . ." This statement is not well supported. As the authors point out, mineral dust can be transported to the Arctic from long distances [Stone et al., 2007]. Perhaps the INPs measured by the authors were mineral dust containing biological material transported from long distances? I do not think this was ruled out. If the authors want to rule out long-range transport than more information and discussion is required.

It is possible that the INPs we observed could have originated from mineral dust from distant sources. However, we provide several reasons as to why the INPs were likely from more local or regional sources. Recent work by Kylling et al. (2018) demonstrates surface dust impacts from several midlatitude sources to the entire Arctic is predominant during May. Yet, they also show that the major source of dust at the surface is from North American north of 60 °N, indicating regional source influences. Specific to our region, supermicron mineral dust has been shown to be transported during Arctic Haze as demonstrated by Quinn et al. (2002) at Barrow, but at very low mass concentrations relative to other aerosol species (i.e., < 0.005 μg m^{-3} based on nss-Ca^{2+}). Additionally, given the size in which we observed the INPs (> 2.96 μm), it is unlikely such large particles were transported from very distant sources, especially considering the air mass transport pathways were near the surface in the boundary layer for several days prior to arrival at the site (evidenced by the revised Figure 6). We have added a few sentences ruling this possible source out in section 3.3.

Kylling, A., Zwaaftink, C. D. G., and Stohl, A.: Mineral Dust Instantaneous Radiative Forcing in the Arctic, Geophys Res Lett, 45, 4290-4298, 10.1029/2018gl077346, 2018.

Quinn, P. K., Miller, T. L., Bates, T. S., Ogren, J. A., Andrews, E., and Shaw, G. E.: A 3-year record of simultaneously measured aerosol chemical and optical properties at Barrow, Alaska, J Geophys Res-Atmos, 107, Artn 4130 10.1029/2001jd001248, 2002.

Page 12, lines 1-5. "We present the first INP measurements in an Arctic oilfield location and demonstrate how local and regional transport from marine and terrestrial sources to an industrial region can introduce high concentrations of coarse, warm temperature INPs that are possibly of biological origin." The authors have not ruled out adequately local industrial emissions as the source of INPs. If they want to claim that the sources were local marine and terrestrial, more analysis is required to rule out local industrial emissions. For example, does the INP concentrations correlate with tracers of industrial emissions?

Please see response to the second comment.

Figure 4. As mentioned above, I suggest using a log scale to illustrate the INP concentrations. Currently it is very difficult to follow the trend at -10 C and -15 C due to the linear scale. Also, what is the onset 99th percentile, how was this calculated, and why is it different from the measured onset temperature? Is the measured onset temperature or the onset 99th percentile more relevant? Also, Figure 4 doesn't include error bars. Error bars should be included, otherwise it is hard to tell if the trends are statistically significant.

Please see response to the first comment. The 99th percentile was removed from the figure to avoid confusion, especially since we do not discuss this value in the results and discussion. We have also added error bars to the INP concentrations and onset temperatures.

Figure 27, a, b, c. Why only include SEM-EDX results from the end of May? Results from the first part of May would be very useful to interpret the INP data.

We agree that results from early May would be useful; however, given limited resources and samples, we were not able to analyze any more samples other than those presented. The bulk composition was measured during this time period, and we do compare to the case study time period.

Figure 7, panel e. I suggest a separate panel for the wind speed and direction. Currently the panel is congested.

Done.

Figure caption 8. "Additionally, the bars and whiskers represent INP concentrations at temperatures in Table 2 and at all temperatures, respectively." What temperatures from Table 2 are plotted? What was plotted in the case of "none" in Table 2? Please include this information in the figure caption for clarity.

We have clarified that the bars show the ranges of INP concentrations (i.e., minimum and maximum at the four temperatures shown in Table 2) in the caption.

Reviewer 2

General Comments:

"Marine and Terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location" by Creamean et al. describes results from a 3-month field campaign in Oliktok Point, Alaska in 2017. The field campaign included detailed measurements of in situ aerosol size distribution and number and offline measurements of aerosol composition and ice nucleating particles (INPs). Utilizing size-resolved aerosol impactors, the authors determined the ice nucleation ability of a range of particles sizes. Further, back trajectory modeling and sea ice and snow cover data were used to investigate the influence of various sources on the measured INP concentrations. The authors provide some evidence that changes in sea ice and snow cover may influence INP number concentrations at the measurements site, but lack an explanation of the mechanism that triggers emissions of INPs due to changes in sea ice and snow cover or suggestions on how to explore this in future work. While it is stated that the data demonstrate how "efficient, natural INPs are likely important in such a relatively polluted Arctic location", the supporting evidence for this is not clearly presented and it is not obvious how this study differs from other coastal studies that were found to be influenced by non-marine aerosol sources. Additional analysis and/or details would be beneficial for supporting the conclusions of the paper. Nevertheless, these data are a certainly a substantial contribution to the field given the extreme lack of INP observations in the Arctic and a recent surge of interest in advancing scientific understanding of aerosol cloud interactions in polar regions.

Specific Comments:

Abstract:

P1 – L20 – “radiative properties” were not included in the analysis and discussion in the paper.

True. We have removed “radiative properties” in addition to “chemistry”, since we do not present routine chemistry (i.e., the chemistry presented in the manuscript was measured during the intensive period only).

P1 - L21: What is meant by “efficient” INPs? Do you mean the most efficient based on the nucleation temperature (i.e., coarse mode aerosol froze at a warmer temperature than submicron aerosol samples)? Or do you mean most efficient described by ice nucleation site density (INPs normalized by surface area) or ice nucleation efficiency (INPs normalized by total number of particles)? Or simply that the highest number concentrations of INPs were observed in the coarse mode?

We meant based on freezing temperature and have reworded this line to reflect that.

P1 - L26: Please specify that these data are representative of springtime INP number concentrations at this location (rather than year-round values for the Arctic region). Also, the INP analysis was only performed on 16 days of the 3-month period. This should also be clear.

We clarified that this was springtime. We chose not to add the number of analysis days to the abstract, as we wanted to focus on the results themselves and particularly on the May case study period. The trends we observed are justified by our statements. Details on how many samples were analyzed are clarified throughout the manuscript.

Introduction:

P2 - L9: “Immersion freezing is the most relevant. . . “ - Please provide a reference for this.

We have added Hande and Hoose (2017).

Hande, L. B., and Hoose, C.: Partitioning the primary ice formation modes in large eddy simulations of mixed-phase clouds, Atmos Chem Phys, 17, 14105-14118, 10.5194/acp-17-14105-2017, 2017.

P3 – L11 – Can you elaborate on the terrestrial sources that impacted these other coastal studies? How will this study and approach uniquely address this difficult task of elucidating local terrestrial sources (natural and pollution) from pristine marine sources?

We have added more background regarding the sources impacting the previous coastal studies mentioned in the fourth paragraph of the introduction. Our unique angle is that we use a comprehensive combination of size-resolved INP measurements, single-particle chemistry, bulk chemistry, local meteorology, regional scale transport, and sea ice and land cover conditions to assess INP sources. We now state this at the end of the introduction.

P3 - L27 – What is mean by “natural”?

We clarified that we meant “naturally-sourced”.

P3 – L28 – I think the introduction is very well written. The overview of the different types of INPs is good, but I think some background information on the aerosol composition and sources of the Arctic Region is

also needed. In particular, what are the potential sources of aerosol (i.e., pollution, transported dust, marine organic aerosol, etc.) and what seasonal and conditions are those aerosol sources present? The authors primarily focus on biological particles, but this is not the only aerosol type in the Arctic.

Thank you for the comment. Although general Arctic aerosol composition and sources are certainly important to discuss, a comprehensive review of Arctic aerosol is outside the scope of this manuscript. Our study location is unique in that it is an Arctic oilfield. Additionally, oilfield locations are subject to very different aerosol sources as compared to typical Arctic background locations. Unfortunately, very few aerosol studies have been conducted in oilfield locations, which would be parallel to our measurements. We have elaborated on the local sources of aerosol observed during the summer from our previous studies (Creamean et al. (2018); Maahn et al. (2017)), in addition to a couple other recent studies by Gunsch et al. (2017) and Kirpes et al. (2018) at the end of the introduction to provide more context for what has been observed in the Alaskan Arctic for oilfield aerosol influences.

Methods:

P4 - L13 – Were these collections made at ambient relative humidity? If so, please discuss how this may affect the cut size diameter of each stage.

These were collected at ambient RH, and as a result we have added a sentence stating how RH may affect particle size by making them larger. However, the purpose of collecting at ambient RH is to mimic how the aerosols themselves would nucleate ice in the environmental conditions they existed in.

P5 – L21 – How was the focus period selected? What is mean by “interesting aerosol events”?

This period was chosen based on the shift in air mass sources and large variability particle concentrations and discussed in the results and discussion. We have clarified this in the text as well and revised Figure 2 to show hourly averaged particle concentrations in which the variability of the aerosol is more evident.

P6 - L10 – How were blanks collected? Were multiple blanks collected throughout the study (i.e., at the beginning, during and end?). Only one shown in Figure 3.

Only one blank was collected and tested during the analysis phase. However, previous control study testing has demonstrated the reliability of the pre-treated and prepared PFA (Creamean et al. (2018b)).

Throughout – the section numbers are inconsistent with the rest of the Methods section.

Fixed.

Results and discussion:

P8-L10 – can you say anything about the size distributions of particles during these different atmospheric conditions? E.g., the CPCf/CPCu ratio, UHSAS size distribution, etc?

We have added the hourly-averaged mean particle diameters from the UHSAS to Figure 2b and discuss in section 3.1. Unfortunately, the size ranges of the CPCs and UHSAS are well below the stage A sizes, so we cannot disseminate the mean size results much beyond describing initial conditions (i.e., we cannot use these results to support the 2.96 – > 12 μm INPs). Sizing measurements at sizes relevant to stage A INPs were not available.

P8 – L11 – “general relatively high” – relative to what?

To other locations on the North Slope, which we now have clarified here.

P8 – L20 – "resulted in relatively 'cleaner' conditions" – What is implied by the quotations? Should this simply state that the changes in transport and increased precipitation resulted in lower particle concentrations?

We removed "relatively 'cleaner' conditions" and revised to the suggested change.

P8 – L23 – The predominate wind direction during April and May looks more easterly (mostly red). Perhaps a wind rose plot would be helpful for this discussion?

We removed this sentence since the wind data are more relevant in the following sections when discussing the chemistry (we removed the wind panel from the figure as well). We have also revised Figure 7 to include a panel of just wind direction and speed. We now discuss the winds more closely at the end of section 3.3.

P8 – L25 – If the goal of this study is the examine the role of pollution versus natural aerosol on the INP populations at this site (I think this is correct, though it is not entirely clear), a section is needed that describes the potential influence of natural vs. pollution particles and how you differentiated the difference particle classes. This of course also requires the aerosol composition during the campaign to be summarized earlier. I suggest that the Results and Discussion section be reorganized to first talk about the aerosol composition and influences of natural and anthropogenic aerosol, followed by a discussion on the INP populations measured at the site with a specific section describing the results that support the statement that was in the abstract: ". . .demonstrate strong influences from natural sources despite the relatively high pollution levels in this Arctic environment".

We disagree that our ordering does not support the natural versus pollution sources of INPs and that restructuring the results and discussion would support this. The idea behind the ordering was to first discuss the conditions observed generally during the study, then show the large shift in INPs, followed by the evidence to support why this shift happened and what the likely sources of the INPs were.

We also believe we have enough supporting evidence for demonstrating the INPs were likely from local and regional natural sources during our late May case study. The air mass trajectory analysis was used for context for the chemical analysis conducted, both of single particle and bulk compositional information, in addition to other supporting information. First and foremost, we know that the aerosol composition in general in the size ranges relevant to the INP measurements were predominantly sea spray aerosol and dust based on the chemical analyses. Very little influences from soot or fly ash (i.e., local industrial pollution) were observed (4% and 16% of the particles that were > 1.15 μm on 23 May and 28 May, respectively; Figure 7). Second, based on size alone, we would not expect pollution sourced from Prudhoe Bay (which a majority by number are sub-100 nm; Creamean et al. (2018a); Maahn et al (2017)) to overlap with the sizes of the INPs observed (i.e., > 2.96 μm) at Oliktok Point. Third, INPs measured at the warmer end of the temperatures we focus on during our case study are likely biological or dust in origin (e.g., Kanji et al. (2017), Murray et al. (2012)). Fly ash and soot generally form ice at much colder temperatures. We have revised Figure 7 to show wind direction and speed separately, and now discuss this in more detail at the end of section 3.3. We note that although winds were easterly on 28 and 29 May, wind prior to those days were variable and can help explain the aerosol sources during our higher marine and terrestrial INP concentration periods. Thus, based on the combination of freezing temperatures, size, single-particle composition, bulk composition, local meteorology, and air mass transport, we demonstrate that there was indeed little influence from local anthropogenic pollution. We added several sentences discussing these points at the end of section 3.3—that it is possible but unlikely that local pollution largely influenced the INP concentrations during late May.

Fig 2 – Adding some indicator for days of this campaign that were analyzed with the DFCP will help the reader follow along.

Done.

Fig 3 – Are these blank-corrected spectra? If so, it might be better to show the blank in a supplemental figure. If not, the blank spectra should be shown on all four panels.

They are not blank corrected. We have added the blanks to each of the four panels.

P9 – L30 – The delta T parameter is presented oddly. What is the physical meaning of this parameter? The delta T here is limited by the temperature in which the DFCP saturates (i.e., all droplets freeze), not the "range of freezing temperatures". Is the goal to define a parameter that describe the presence of the "hump" of INPs that are active at warmer temperatures? While the delta T parameter will be lower for spectra that have a "hump" of INPs at warmer temperatures and higher for spectra those do not, the delta T parameter could also be lower for an INP spectra with a steep slope compared to a spectra with lower slope. If the authors want to describe the presence of significant differences in the number of INPs active at warmer temperatures, a better variable to use may be the temperature in which 50% of the wells were frozen. Or, perhaps the authors can clarify the meaning of this parameter. Fig 4. Are there uncertainty bars for the INP number concentrations?

After consideration, we decided to remove ΔT from the manuscript and Figure 5 since we did not discuss it in detail and it did not add additional useful information to the main conclusions. The information from the INP concentrations and onset freezing temperatures sufficiently supports our main conclusions.

P10 – L7 – Please provide trajectory heights in Fig 6, as you refer to the trajectory height in the text and this is one of the main pieces of evidence provided for a connection between the sea ice leads and the observed aerosol.

Done. We have substantially revised Figure 6 to include such information and to support our conclusions in the text.

P10 – L9 – Are these observations of leads and polynyas from satellite, an aircraft, or published? Since the importance of the observed leads are a critical point to your conclusions, these should be provided in some capacity?

As we state in the methods, these data are satellite derived. We realize that the term "leads" may not accurately represent the area of < 100% sea ice concentrations (i.e. light blue colors directly north of Oliktok Point). The width of leads varies from a couple of meters to over a kilometer, thus, they are difficult to resolve in the 4-km sea ice data we use. It is possible the open water is simply small or larger ice floes that have broken off the pack ice near the ice edge. Thus, we have changed this to "marginal ice zone" (MIZ) as that is a more accurate description of the transition between open water and sea ice during the melt season, but that leads may be a feature within this zone. We also have defined that the persistent open water regions west of the Canadian Arctic Archipelago and western Alaska are polynyas.

P10 – L12 – Can you provide more information about the gravitational settling? I think particles in the largest stage could survive such a transit, but this could be calculated.

The basic terminal settling velocities can be calculated, but this information does not take into account external vertical updrafts or downdrafts and how those features may affect particle lifetime. We did do such

calculations and have added discussion on results from such calculations for 16 May and 29 May in the second paragraph in section 3.3, which indicate sources local to Oliktok Point. Additionally, Jaenicke (1980) concludes that particles of these sizes originating within the boundary layer typically reside in the atmosphere for on the order of hours to days (but less than a week). However, for 22 May, air masses did not travel over any substantial open water polynyas rather only over the MIZ north of land, thus, the only possible explanation for the sources on this day is from the open water in this region. Given this information, it is possible these coarse mode INPs could originate from the open water 700 km away, but the more probably scenario is transport from 30 km away. We have also added a couple sentences to elaborate on the gravitational settling based on Jaenicke (1980) and how the distant open water could be a source, but likely not the major source.

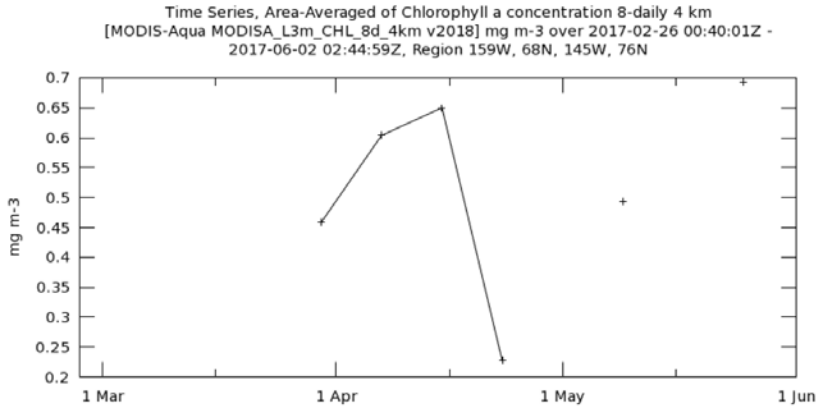
P10 – L19 – on May 29, it looks like there is a portion of the back trajectory (72-73 N and 137-133 W) with lower sea ice percentage compared to any portion of the May 22 source region. How are these two regions distinct? What is considered a significant amount of time to spend over an open lead?

We have substantially revised Figure 6 to show more detail of the transport pathways and have adjusted the text in this transport paragraph accordingly. Regarding the transport over the lead on 29 May, transport from this region occurred 3 days prior and the air mass traveled very close to the surface, indicating possible gravitational settling during transport. However, INP concentrations were still high relative to days in Mar or Apr, which could be a result of transport over the MIZ. We have now added discussion on this topic to the transport paragraph.

P9-L20 – What is the hypothesized mechanism/source of INPs from open Arctic leads? Is organic marine aerosol the suspect? Are there previous studies to suggest that unique aerosol types may be emitted from Arctic leads? How does wind speed play a role? Were Chl a concentrations available for this region?

Recent work by May et al. (2016) has demonstrated that production of sea salt aerosol in the Arctic can occur year-round from leads under elevated wind speeds (i.e., winds speeds > 4 m s^{-1}). The main mechanisms behind aerosol production from open ocean surfaces is bubble bursting from wind-induced wave breaking, although this process is far less studied over leads. A recent study by Gabric et al. (2018) nicely describes generation of marine biogenic aerosol (MBA) from sea ice leads and the MIZ. Thus, other primary aerosols may be generated by the same mechanisms that produce sea salt aerosol and MBA. Recent studies by Wilson et al. (2015) and Irish et al. (2017; and references therein) have shown that the Arctic Ocean surface microlayer and bulk seawater can harbor large concentrations of INPs, indicating physical mechanisms that generate aerosol from the surface waters may eject these INPs into the atmosphere. Additionally, several previous high Arctic ice nucleation studies have demonstrated that leads and other open water sources as vital to influencing atmospheric INP concentrations (Bigg, 1994; Bigg and Leck, 2001). Based on a combination of conclusions from this body of previous work, we conclude that INPs from leads or other open water features (i.e., small ice floe regions) are likely produced via waves and/or bubble bursting and particularly under relatively windy conditions and are likely composed of bacteria or fragments of marine organisms.

We have added discussion on these previous studies to section 3.3 to support our conclusions. However, because we do not have wind speed measurement over the MIZ region, we cannot quantitatively comment on the role of wind speed over the open water we observed. In situ chlorophyll measurements were not available for this region. We did check chl-a concentrations from MODIS (available at an 8-day time resolution), and although chl-a looks to be elevated in the polynya west of Alaska, cloud cover makes it difficult to discern any sort of temporal trend on the scale of the MIZ north of Oliktok Point:



Irish et al. (2017) also evaluated their INP measurements in the context of satellite-based chl-a and were not able to use chl-a concentrations to explain their INP observations.

Gabric, A., Matrai, P., Jones, G., and Middleton, J.: The Nexus between Sea Ice and Polar Emissions of Marine Biogenic Aerosols, Bulletin of the American Meteorological Society, 99, 61-82, 10.1175/Bams-D-16-0254.1, 2018.

May, N. W., Quinn, P. K., McNamara, S. M., and Pratt, K. A.: Multiyear study of the dependence of sea salt aerosol on wind speed and sea ice conditions in the coastal Arctic, J Geophys Res-Atmos, 121, 9208-9219, 10.1002/2016jd025273, 2016.

Summary:

P12 – L9 – "These higher concentrations are attributed to air masses originating from over sea ice leads and tundra surfaces" – Can the authors elaborate on what these particles are exactly? Or provide a hypothesis of what these may be? The single particle and bulk composition measurements suggests significant influence from mineral dusts, but what would be the mechanism for these particles entering the atmosphere via sea ice leads? Particularly for those that were measured in air masses originating from these open Arctic leads? Can the authors elaborate on future needs for understanding more about these significant increases in INPs? How can the scientific understanding of Arctic INP population variability advance? More measurements? Different measurements?

See response to above. Based on previous work and our chemical measurements, we conclude that the INPs from over the open water and land were primary marine aerosol and dust, respectively. Though each analyzed sample's source influences were characterized based on air mass trajectories, the diversity in chemical composition of the aerosol particles in each sample indicate a variety of sources. The observed dust could be due to local road dust in addition to terrestrial dust sources along the air mass back trajectories. We have now noted this in section 3.3.

We already state that additional measurements in seasons other than the spring are needed but have elaborated that such measurements include comprehensive INP concentrations and characterization. A detailed discussion on the future needs is indeed important but outside the scope of this manuscript, and would be better-suited for some sort of Arctic INP review paper.

Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location

Jessie M. Creamean[1,2], Rachel M. Kirpes[3], Kerri A. Pratt[3], Nicholas J. Spada[4], Maximilian Maahn[1,2], Gijs de Boer[1,2], Russell C. Schnell[5], Swarup China[6]

[1]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, 80309, USA
[2]Physical Sciences Division, National Oceanic and Atmospheric Administration, Boulder, CO, 80305, USA
[3]Department of Chemistry, University of Michigan, Ann Arbor, MI, 48109, USA
[4]Air Quality Research Center, University of California, Davis, CA, 95616, USA
[5]Global Monitoring Division, National Oceanic and Atmospheric Administration, Boulder, CO, 80305, USA
[6]Environmental and Molecular Sciences Laboratory, Pacific Northwest National Laboratory, Richland, WA, 99352, USA

Correspondence to: Jessie M. Creamean (jessie.creamean@noaa.gov)

Abstract. Aerosols that serve as ice nucleating particles (INPs) have the potential to modulate cloud microphysical properties and can therefore impact cloud radiative forcing and precipitation formation processes. In remote regions such as the Arctic, aerosol-cloud interactions are severely understudied yet may have significant implications for the surface energy budget and its impact on sea ice and snow surfaces. Further, uncertainties in model representations of heterogeneous ice nucleation are a significant hindrance to simulating Arctic mixed-phase cloud processes. We present results from a campaign called INPOP (Ice Nucleating Particles at Oliktok Point), which took place at a U.S. Department of Energy Atmospheric Radiation Measurement (DOE ARM) facility in the northern Alaskan Arctic. Three time- and size-resolved aerosol impactors were deployed from 1 Mar to 31 May 2017 for offline ice nucleation and chemical analyses and were co-located with routine measurements of aerosol number and ~~,~~ size~~, chemistry, and radiative properties~~. The largest particles (i.e., ≥ 3 μm or "coarse mode") were the most efficient INPs by inducing freezing at the warmest temperatures. During periods with snow- and ice-covered surfaces, coarse mode INP concentrations were very low (maximum of 6×10^{-4} L^{-1} at –15 ˚C), but higher concentrations of warm temperature INPs were observed during late May (maximum of 2×10^{-2} L^{-1} at –15 ˚C). These higher concentrations were attributed to air masses originating from over ~~sea ice leads~~open Arctic Ocean water and tundra surfaces. To our knowledge, these results represent the first INP characterization measurements in an Arctic oilfield ~~location, and~~location and demonstrate strong influences from natural sources despite the relatively high springtime pollution levels ~~in this Arctic environment~~. Ultimately, these results can be used to evaluate the anthropogenic and natural influences on aerosol composition and Arctic cloud properties.

1 Introduction

Aerosols are an important component of the atmospheric system through their various impacts on climate. Depending on the type (Satheesh and Moorthy, 2005), aerosols directly scatter and/or absorb radiation, thereby affecting the atmospheric energy

budget (Boucher et al., 2013). Notably, the largest uncertainty of the energy budget (i.e., radiative forcing estimate) is the aerosol indirect effect. As cloud condensation nuclei (CCN) and ice nucleating particles (INPs), aerosols influence atmospheric radiation through modulation of the microphysics of cloud droplets and ice crystals. Aerosol-induced microphysical modifications influence cloud lifetime and albedo, as well as the production of more or less precipitation (Albrecht, 1989; Twomey, 1977). However, constraining aerosol-cloud impacts in climate models, specifically when parameterizing INPs in mixed phase cloud (MPC) systems, remains a significant challenge due to limited observations (Cziczo et al., 2017; Coluzza et al., 2017; DeMott et al., 2010; Kanji et al., 2017; Korolev et al., 2017).

The efficacy of an aerosol to serve as an INP largely depends on its composition (i.e., chemical, mineral, or biological makeup), morphology, and size, and thus, its source. INPs nucleate ice through pathways dependent upon temperature, saturation with respect to ice, and the INP size and composition (Hoose and Möhler, 2012). Immersion freezing is the most relevant to MPC formation and requires that INPs initially serve as CCN (Hande and Hoose, 2017). Aerosols such as mineral dust, soil dust, sea salt, volcanic ash, black carbon from wildfires, and primary biological aerosol particles (PBAPs) have been shown to serve as INPs (Murray et al., 2012; Hoose and Möhler, 2012; Coluzza et al., 2017; Petters et al., 2009; DeMott et al., 1999; McCluskey et al., 2014; Conen et al., 2011). Among these, dust and PBAPs are the most adroit INPs found in the atmosphere (Murray et al., 2012; Coluzza et al., 2017). PBAPs originating from certain bacteria and vegetative detritus are the most efficient INPs known, capable of initiating freezing near −1 °C, while most PBAPs (e.g., pollen, fungal spores, algae, and diatoms) tend to nucleate ice at temperatures similar to those of mineral dust but warmer than sea salt or volcanic ash (Despres et al., 2012; Murray et al., 2012; Hoose and Möhler, 2012; Tobo et al., 2014; Hader et al., 2014a; O'Sullivan et al., 2014; Creamean et al., 2014; Creamean et al., 2013; Hill et al., 2016; Tesson et al., 2016; Alpert et al., 2011; Knopf et al., 2010; Umo et al., 2015; Durant et al., 2008; Fröhlich-Nowoisky et al., 2015; McCluskey et al., 2014). However, dusts can serve as atmospheric shuttles for small microbes, enabling these particle mixtures to behave more efficiently as INPs compared to the dust alone (Conen et al., 2011; Creamean et al., 2013; O'Sullivan et al., 2014). Consequently, PBAPs have the potential to play a crucial role in cloud ice formation (Creamean et al., 2014; Creamean et al., 2013; Pratt et al., 2009) and precipitation enhancement (Morris et al., 2004; Bergeron, 1935; Christner et al., 2008; Morris et al., 2014; Morris et al., 2017; Stopelli et al., 2014), particularly in the presence of supercooled water or large cloud droplets. PBAPs have been a key focus of recent ice nucleation studies, yet estimating their global impact remains a challenge due to: 1) a dearth of observations in time and space and 2) a poor understanding of sources, flux, and abundance of PBAPs in the Earth-atmosphere system (Hoose et al., 2010a).

Little is known about marine emissions of biological INPs—most studies of such aerosols to date have examined terrestrial sources (Burrows et al., 2013; Murray et al., 2012). Recent laboratory, field, and modelling studies have evaluated the mechanical emission processes, ice nucleation efficiency, and concentrations of INPs generated from marine environments (Knopf et al., 2010; DeMott et al., 2016; McCluskey et al., 2017). Results from these studies imply that PBAPs are most

relevant to ice formation at MPC temperatures (i.e., > −10 °C), whereas sea salt aerosols nucleate ice in both deposition and immersion modes at temperatures relevant for cirrus clouds (i.e., < −38 °C) (Schill and Tolbert, 2014). Conclusions on the role of PBAPs in ice nucleation and precipitation are equivocally based on results from climate modelling, with some studies implying they are insignificant on a global scale (Hoose et al., 2010a; Sesartic et al., 2012; Hoose et al., 2010b), while others have found them to be potentially important (Phillips et al., 2009; Phillips et al., 2008; Vergara-Temprado et al., 2017). An inherent limitation of these pioneering studies is that results were validated by a handful of terrestrial-based observations of biological INPs. This, coupled with the fact that the Earth's surface is 70% covered by ocean, implies that studies using global climate simulations have ignored a potentially significant global source of INPs.

Previous work by Leck and Bigg in the High Arctic were among the first to elucidate the potentially paramount role of the ocean as a ~~microbial~~ source of efficient INPs (Leck and Bigg, 2005; Bigg and Leck, 2001, 2008; Bigg, 1996), suggesting bacteria and fragments of marine organisms were responsible for the ice nucleating contribution to their samples. Schnell (1975) concluded that zones of profuse marine phytoplankton growth may release large numbers of INPs into the atmosphere based on measurements conducted on laboratory-cultured marine phytoplankton. However, new studies that provide information on key sources of INPs *and* their impacts on Arctic MPCs are necessary to assess the direct contribution from the marine environment. Irish et al. (2017) and Wilson et al. (2015) report enhanced ice nucleation activity of particulate matter in the surface microlayer and bulk seawater from the Arctic Ocean. Both studies concluded that the enhanced INP concentrations measured were attributed to heat-labile biological material and organic material associated with phytoplankton cell exudates, respectively. In addition, sources of INPs at Arctic coastal locations have been measured during the summer by Fountain and Ohtake (1985), Mason et al. (2016), and Conen et al. (2016) in Alaska, Canada, and Norway, respectively. ~~, but were likely impacted, if not mostly dominated by, terrestrial sources.~~ Fountain and Ohtake (1985) did not comment on exact sources of INPs they measured at Utqiaġvik (formally Barrow), but determined INPs during episodic increases in concentration to be either from the Alaskan interior or long-range transported from Eurasia. Mason et al. (2016) did not comment on the sources of their size-resolved INPs at Alert between Mar and Jul but determined fewer local sources of aerosols as compared to their midlatitude coastal locations. Conen et al. (2016) attributed a fraction of their observed warm-temperature INPs (i.e., INP concentrations at –8 °C) from May to Sep were fungal spores and that most INPs were aerosolised locally by the impact of raindrops on plant, litter, and soil surfaces. Their measurement site was 219 meters above mean sea level (m a.m.s.l.) and 20 km from the actual coast, thus, could be the reason terrestrial sources were more influential at their site. To date, only a handful of studies have evaluated Arctic INPs in coastal or marine environments.

The Arctic is a remarkable region with regard to many atmospheric processes, but especially with regard to ice nucleation because: 1) the Arctic is mostly ocean, which may be the most prolific regional source of biological INPs, 2) interruption of ocean-atmospheric exchange by sea ice cover likely affects seasonally-dependent patterns in the emission of PBAPs/INPs, and 3) aerosol-cloud interactions could have implications important to understanding declining trends in sea ice and snow coverage.

Overall, limited information on aerosol-cloud processes (Lubin and Vogelmann, 2006; Garrett and Zhao, 2006), contradictory modelling results with regard to the importance of biological INPs (Szyrmer and Zawadzki, 1997), and a critical need to understand the role of aerosol particles in determining cloud phase (Bergeron, 1935; Wegener, 1911; Findeisen, 1938) motivate the need for additional observations to constrain the abundance and ice-nucleating properties of aerosols in the Arctic (Murray et al., 2012). Here, we present a 3-month record of continuous time- and size-resolved INP concentrations at Oliktok Point, a coastal site in ~~Northern~~ Alaska situated within the North Slope of Alaska (Prudhoe Bay) oilfields, which have been shown to be strong and localized sources of pollutants ~~in this region~~. Recent studies have indicated that sources of aerosol in this region include carbonaceous combustion and aged sea spray aerosol measured during the winter and summer (Gunsch et al., 2017; Kirpes et al., 2018) and newly-formed particles from Prudhoe Bay gaseous emissions (Creamean et al., 2018a; Maahn et al., 2017; Kolesar et al., 2017), in which the anthropogenic pollutant aerosol has been linked to increased local cloud droplet concentrations over the Beaufort Sea (Hobbs and Rangno, 1998). Although anthropogenic aerosol may be the dominant type in this region, Gunsch et al. (2017) and Kirpes et al. (2018) also observed a significant fraction of supermicron aerosol to be fresh sea spray aerosol. ~~,~~ Additional sources of aerosol observed at higher altitudes over Prudhoe Bay include regional wildfires and long-range transported pollution, although, these were measured during the summer (Creamean et al., 2018a). ~~(2017)~~However, INPs and their sources have yet to be evaluated in this region. Here, we employ a comprehensive combination of size-resolved INP measurements, single-particle chemistry, bulk aerosol chemistry, local meteorology, regional scale transport, and sea ice and land cover conditions to assess INP sources in Prudhoe Bay. Unique observations of an increase in what were likely marine- and terrestrial-sourced INPs are discussed. This is the first time INP measurements have been conducted in an Arctic oilfield, and we demonstrate how efficient, naturally-sourced INPs are likely important in such a relatively polluted Arctic location.

2 Methods

2.1 Study location and dates

INPOP was conducted at the Third Atmospheric Radiation Measurement (ARM) Mobile Facility (AMF-3) operated by the United States Department of Energy (DOE) in Oliktok Point, Alaska (70.51°N, 149.86°W, 2 ~~meters above mean sea level (m a.m.s.l.)~~ m a.m.s.l.) (Creamean, 2017a). Oliktok Point is located in Prudhoe Bay (Figure 1), the third largest oilfield in North America (U.S. Energy Information Administration, 2015). The study time period was 1 Mar – 31 May 2017. Data from INPOP and the rest of the AMF-3 data record (2013 – present) are available on the ARM data archive (https://www.archive.arm.gov/discovery/) (Creamean, 2017b).

~~1.2~~2.2 Sample collection

For offline ice nucleation analyses, daily aerosol samples were collected 11 Mar – 31 May 2017 (Table 1) using a 4-stage time- and size-resolved Davis Rotating-drum Universal-size-cut Monitoring (DRUM) single-jet impactor (DA400, DRUMAir,

LLC.) (Cahill et al., 1987) housed in a 47 x 35.7 x 17.6 cm Pelican™ case (inset in Figure 1). The DRUM collected aerosol particles at four size ranges (0.15 – 0.34, 0.34 – 1.20, 1.20 – 2.96, and 2.96 – >12 µm in Stokes' equivalent diameter) and sampled at 26.7 L min^{-1}, equalling 38428 total L of air per day (i.e., per sample). Such size ranges cover a wide array of aerosols, particularly those that serve as INPs (DeMott et al., 2010; Fridlind et al., 2012; DeMott et al., 2016). Simultaneously, the large volume of air collected promotes collection of rarer warm temperature biological INPs, which may represent a lower fraction of overall INP concentrations (Mossop and Thorndike, 1966). Samples were deposited onto 20 x 190 mm strips of petrolatum-coated (100%, Vaseline®) Mylar™ (0.02 mm thick; DuPont®) substrate secured onto the rotating drums (20 mm thick, 60 mm in diameter) in each of the four stages at the rate of 7 mm per day (5 mm of sample streaked onto the Mylar followed by 2 mm of blank).

The DRUM was secured inside the AMF-3 container, with an approximately 7-m long sampling line (6.4 mm inner diameter static-dissipative polyurethane tubing; McMaster-Carr®) leading to outside of the container and connected to a plastic funnel inlet covered with loose mosquito netting to prevent rimed ice build-up or blowing snow from clogging the inlet. Aerosol collection was conducted at ambient relative humidity, which may affect particle size. The purpose of not drying the aerosol was to collect at sizes relevant to the particles in true environmental conditions. Daily rotation checks were conducted by disconnecting the DRUM from its pump and removing the stage caps to ensure the drums were rotating at the correct rate and sample substrates remained secured to each drum. Weekly checks of the inlet flow and pressure measured at each stage were conducted to ensure the orifices to each stage were not clogged and the DRUM pump was operating correctly.

The drums rotated for 24 to 26 days before sample substrates were changed (i.e., one full rotation of the drums), equalling three drum changes during INPOP. Sampled substrates were kept on the drums and stored frozen in a standard freezer until transport to Boulder, Colorado and stored frozen in a chest freezer for 5 to 7 months before offline INP analyses (see following section). This is the first time a DRUM has been used for offline INP analyses, but as we demonstrate, it is a useful sample collection method for long-term studies.

For offline single-particle analysis, daily samples were collected at 16.1 L min^{-1} using a 3-stage DRUM impactor with particle size ranges 0.10 – 0.34 µm, 0.34 – 1.15 µm, and >1.15 µm on transmission electron microscopy (TEM) grids (Formvar carbon Type-B copper grids, Ted Pella, Inc.) attached to each of the three drums. The rotation was set at 3 mm per day, such that particles were deposited over the width of one TEM grid per 24-hour sampling period. Drums were changed once over the course of the INPOP study. Sampled substrates were kept on the drums and stored in the dark at ambient temperature until analysis (~3 months) (Laskina et al., 2015). Select daily samples corresponding to interesting case studies were analysed for discussion in the current paper. A third DRUM sampler collecting particles in 8 size bins (0.09 – 0.26 µm, 0.26 – 0.34 µm, 0.34 – 0.56 µm, 0.56 – 0.75 µm, 0.75 – 1.15 µm, 1.15 – 2.5 µm, 2.5 – 5.0 µm, and 5.0 – >12 µm) on Mylar™ for 12 hours per sample was co-located for bulk inorganic analysis (~2 months after collection). The flow rate was maintained at 9.6 L min^{-1} with a drum rotation rate of 0.5 mm per day. Only one set of drums for this sampler was needed for the duration of INPOP.

Samples were stored in the DRUM chambers during shipment and storage until analysis. The same sampling line set ups and routine checks as for the 4-stage DRUM were applied to the 3-stage and 8-stage DRUMs.

~~1.2~~2.3 Drop freezing assays (DFA) for immersion mode INPs

In total, 216 daily samples were collected between the four different stages (herein, stages A, B, C, and D correspond to 2.96 – 12, 1.20 – 2.96, 0.34 – 1.20, and 0.15 – 0.34 µm particles, respectively. Due to contamination issues, stages B and D were not analysed from the first set of daily samples (see Table 1). Other missing data (i.e., samples not analysed) are due to occasional power outages at the AMF-3. Due to the large volume of samples, initially, one daily sample per week was analysed to assess a broader picture of INPs over the course of the spring months. Daily samples were analysed 22 – 29 May 2017 due to interesting aerosol events and source influences (as evidenced in the following sections) observed during this time period as described herein. Immediately prior to analysis, sample strips were cut into their daily segments and stored frozen in 29-mL sterile Whirlpak® bags for up to one week.

Immersion mode freezing was tested using a DFA cold plate apparatus. This cold plate technique was based on previous but slightly modified apparatuses (Hill et al., 2016; Tobo, 2016; Stopelli et al., 2014; Wright and Petters, 2013) and is discussed in detail by Creamean et al. (2018b). For brevity, we call this system the Drop Freezing Cold Plate (DFCP). Before analysing with the DFCP, 2 mL of ultrapure water (UPW; Barnstead™ Smart2Pure™ 6 UV/UF) were added to the sterile Whirlpak® bags containing sample segments to resuspend deposited particles. The bags were sealed and shaken at 500 rpm for two hours (Bowers et al., 2009). It is possible not all particles were removed during the extraction process, however, previous control testing indicates sufficient aerosol loading is resuspended (Creamean et al., 2018b). Copper discs (76 mm in diameter, 3.2 mm thick) were prepared by cleaning with isopropanol (99.5% ACS Grade, LabChem. Inc.), then coating with a thin layer of petrolatum (Tobo, 2016; Bowers et al., 2009). Following sample preparation, a sterile, single-use plastic syringe was used to draw 0.25 mL of the suspension and 100 drops were pipetted onto the petrolatum-coated copper disc, creating an array of ~2.5-µL aliquots. Drops were visually inspected for size; however, it is possible not all drops were the same exact volume, which could lead to a small level of indeterminable uncertainty (Alpert and Knopf, 2016). However, we have previously demonstrated that drop size variability does not impact freezing results (Creamean et al., 2018b). Previous studies have elucidated that drops need to be orders of magnitude different in volume to significantly perturb the freezing temperature from drop size, alone (Hader et al., 2014b; Bigg, 1953; Langham and Mason, 1958). The copper disc was then placed on a thermoelectric cold plate (Aldrich®) and covered with a transparent plastic dome. Small holes in the side of the dome and copper disc permitted placement of up to four temperature probes using an Omega™ thermometer/data logger (RDXL4SD; 0.1 °C resolution and accuracy of ± (0.4% + 1 ºC) for the K sensor types used). During the test, the cold plate was cooled variably within a 1 – 10 °C min^{-1} range from room temperature until around −30 °C. Control experiments with UPW at various cooling rates within this range show no discernible dependency of drop freezing on cooling rate (Creamean et al., 2018b), akin to previous works (Wright and Petters, 2013; Vali and Stansbury, 1966).

A +0.33 °C correction factor was added to any temperature herein and an uncertainty of 0.15 °C was added to the probe accuracy uncertainty based on DFCP characterization testing presented in Creamean et al. (2018b), to account for the temperature difference between the measurement (i.e., in the plate centre) and actual drop temperature. Frozen drops were detected visually, but the freezing temperature and cooling rate of each drop frozen was recorded through custom software. The test continued until all 100 drops were frozen or when the system reached approximately −30 °C. Each sample was tested three times with 100 new drops for each test. From each test, the fraction frozen and percentage of detected frozen drops were calculated. For INPOP, 71 – 100% of the 100 drops were visually detected and recorded as frozen for each test. The results from the triplicate tests were then binned every 0.5 °C to produce one spectrum per sample. Cumulative INP spectra were calculated using the equation posed by Vali (1971) and adjusted to account for the total volume of air per sample:

$$[INPs(T)](L^{-1}) = \frac{\ln N_o - \ln N_u(T)}{V_{drop}} \times \frac{V_{suspension}}{V_{air}}$$

where N_o is the total number of drops, $N_u(T)$ is the number of unfrozen drops at each temperature, V_{drop} is the average volume of each drop, $V_{suspension}$ is the volume of the suspension (i.e., 0.75 mL total for the three tests), and V_{air} is the volume of air per sample (38428 L) (Stopelli et al., 2014; DeMott et al., 2016; Chen et al., 2018; Boose et al., 2016a).

~~1.3~~2.4 Single-particle and bulk aerosol composition

Particles collected on TEM grids using the 3-stage DRUM were analysed for single-particle chemical composition and morphology using computer-controlled scanning electron microscopy with energy dispersive X-ray spectroscopy (CCSEM-EDX). Samples were analysed using a FEI Quanta scanning electron microscope with a field emission gun operating at 20 keV and equipped with a high angle annular dark field detector for particle size and morphology, and an energy dispersive X-ray X-ray spectrometer (EDAX, Inc.) and a Si(Li) detector (10 mm^2) for particle elemental composition of elements including C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Fe, Ni, and Zn. An average of 1000 particles ~~were~~ was analysed per substrate. Data was analysed using k-means cluster analysis of the single-particle EDX spectra (Ault et al., 2012). Fifty clusters were combined into six classes of particle types, including fresh sea spray aerosol, aged sea spray aerosol, dust, organic aerosol, fly ash, and soot, based on similarity of elemental composition (Kirpes et al., 2018; Gunsch et al., 2017; Weinbruch et al., 2012).

Samples collected on the 8-stage DRUM were analysed for elemental concentrations using synchrotron-induced X-ray fluorescence spectrometry (S-XRF) at the Lawrence Berkeley National Laboratory Advanced Light Source facility (VanCuren et al., 2012; Perry et al., 2004). The 1.5 GeV polarized X-ray beam provides very low background bremsstrahlung radiation and high sensitivity of elements including Na, Mg, Al, Si, P, S, Cl, Ar, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Nb, Mo, and Pb. Spectra were acquired with a Vortex-60EX silicon drift detector (Hitachi High Technologies, USA) and were post-processed using both WinAXIL (Canberra, Belgium) and PyMca (Sole et al., 2007) independently, for quality assurance. Further data reduction into timestamped concentrations was performed using custom

software (R Foundation for Statistical Computing, 2017). For specific comparisons, non-soil K was calculated as *non-soil-K* = $K - 0.45 \times Fe$ (Lewis et al., 1988) while soil K was calculated as *soil-K* = *K* – *non-soil-K*. These estimations ignore any contribution from biomass burning, which was a minor component of the observed single-particle designations.

~~1.4~~2.5 Supporting observations and modelling

The AMF-3 at Oliktok Point includes the ARM Aerosol Observing System (AOS), which provides a wide range of aerosol and meteorological measurements (Jefferson, 2011). Data used here are publicly available in the ARM data archive and include aerosol number and size and wind direction and speed. Aerosol number concentrations were measured with a condensation particle counter (CPCf; 10 nm – 3 µm) and ultrafine CPC (CPCu; 3 nm – 3 µm) (TSI, Inc. models 3010 and 3025, respectively). Aerosol size distributions were measured with an ultrahigh-sensitivity aerosol sizer (UHSAS, Droplet Measurement Technologies, Inc.) for particles in the 60 nm – 1 µm size range. The AOS Surface Meteorology (AOSMET) data for atmospheric temperature, humidity, and wind speed and direction were measured via a Vaisala, Inc. WXT520 Weather Transmitter (Kyrouac, 2016). AOS data have a time resolution of 1 second.

Daily snow and ice cover data were obtained from the National Snow & Ice Data Center. Snow cover data (http://nsidc.org/data/g02156#table3, Version 1) were derived from the Ice Mapping System (IMS) daily northern hemisphere snow analysis at a 1 km x 1 km resolution and are derived from a variety of data products including satellite imagery and in situ data (National Ice Center, 2008). Sea ice data (https://nsidc.org/data/g10005, Version 1) were derived from the Multisensor Analyzed Sea Ice Extent Advanced Microwave Scanning Radiometer 2 MASIE-AMSR2 (MASAM2) daily 4 km sea ice concentration product that is a blend of two other daily sea ice data products: ice coverage from the product at a 4 km grid cell size and ice concentration from the AMSR2 at a 10 km grid cell size (Fetterer et al., 2015). MASAM2 was used to meet a need for greater accuracy and higher resolution in ice concentration fields. Air mass backward trajectories (5-day) were calculated using the HYbrid Single Particle Lagrangian Integrated Trajectory model with the SplitR package for RStudio (https://github.com/rich-iannone/SplitR) (Draxler, 1999; Draxler and Rolph, 2011). Data from the Global Data Assimilation System (GDAS; 1˚ latitude-longitude; 3-hourly) from National Centers for Environmental Prediction (NCEP) were used as the meteorological fields in HYSPLIT simulations. Trajectories were initiated at 5 m above ground level (a.g.l.) (i.e., approximate height of the inlet) every 3 hours daily. Only hours within the sample day (i.e., starting at 16:00 UTC) were evaluated per each sample such that 18:00 and 21:00 on the same day and 00:00, 03:00, 06:00 09:00, 12:00, and 15:00 on the following day were considered per sample. Trajectories were only simulated for each day of May during sea ice melt off the Alaskan coast and during the 22 – 29 May case study.

3 Results and discussion

3.1 Atmospheric conditions during INPOP

Shifts in the aerosol population and atmospheric conditions throughout the course of INPOP were evident from the AOS measurements (Figure 2). Particle concentrations were high relative to what has been previously observed in the surrounding areas on the North Slope (e.g., Creamean et al., 2018a; Quinn et al., 2002)~~in general relatively high~~, with the CPCu, CPCf and UHSAS measuring average concentrations of 2065, ~~1589~~1603, and ~~1479~~ 1483 cm^{-3}, respectively, over the entire study. These elevated concentrations are likely the result of the local industrial activities, which are prominent at Oliktok Point (Creamean et al., 2018a; Maahn et al., 2017; Gunsch et al., 2017). Akin to previous work on the North Slope during the Arctic haze, high particle concentrations were observed in April (e.g., Quinn et al., 2007), with averages of ~~2056~~2091, ~~3329~~3286, and ~~1532~~ 1526 cm^{-3} for CPCu and ~~1677~~1702, ~~2769~~2736, and ~~896~~ 907 cm^{-3} for CPCf measured from March, April, and May, respectively. The monthly averaged UHSAS concentration was highest in March, at concentrations much higher than measured in May (~~3050~~3063, ~~1652~~1658, and 975 cm^{-3} for March, April, and May, respectively). Mean particle size from the UHSAS was fairly consistent between the months (156.6±7.6, 149.9±9.9, and 143.6±7.3 nm, respectively). However, data from March may not be representative of the entire month due to a power outage in the first part of that month (i.e., missing data in Figure 2). As documented in previous studies (Law and Stohl, 2007; Stohl, 2006; Garrett et al., 2010; Di Pierro et al., 2013), changes in transport and precipitation patterns resulted ~~in relatively 'cleaner' conditions, including~~in lower particle concentrations around late May. However, one interesting feature is the large variability in particle concentrations, particularly during late May, when hourly averaged concentrations ranged from 13 – 1486, 12 – 1111, and 35 – 1182 cm^{-3} for the CPCu, CPCf, and UHSAS, respectively. Ambient temperature and relative humidity increased steadily over the course of the study and reached values indicative of coastal marine conditions towards the end of May (Cox et al., 2012). ~~March winds frequently originated from the south/southwest, while winds were predominantly northerly/north easterly during April and May. As demonstrated in the current paper, an observed shift in atmospheric conditions at Oliktok Point influenced the aerosol and additionally the INP population.~~

3.2 Observation of a shift in springtime INP glaciation temperatures and concentrations in Prudhoe Bay

INP concentrations from the Arctic are typically very low compared to other regions (Kanji et al., 2017), and industrial regions are not thought to serve as prolific INP sources (e.g., Chen et al., 2018). Figure 3 shows the size-resolved INP concentrations from INPOP. A few key features are evident: (1) larger particles were more efficient INPs, especially at warmer temperatures, (2) larger particles exhibited a larger spread in INP spectra, and (3) late May samples contained particularly high concentrations of warm temperature (i.e., > -10 °C) INPs relative to March and April. INPOP INP concentrations were generally low, falling into the lower end of ranges reported by Kanji et al. (2017) and Petters and Wright (2015). Comparison of our results to previous ice nucleation studies in the Arctic are discussed in section 4.

The first feature discussed above has been reported by other size-resolved Arctic INP studies. Mason et al. (2016) report similar observations in Alert, Canada, where 95% and 70% of INPs were found at sizes > 1 μm and > 2.5 μm in diameter, respectively, and a median diameter of approximately 3 μm for all INPs at –15 ˚C. In general, observational studies in locations outside of the Arctic also report a relationship between temperature and particle size, where supermicron or coarse mode aerosols are the most proficient INPs at warmer temperatures (Vali, 1966; references cited in DeMott et al., 2016; Huffman et al., 2013; Conen et al., 2017). However, modelling studies suggest INPs can be as small as 500 nm (DeMott et al., 2010; DeMott et al., 2015; Fridlind et al., 2012), while observational work suggests that nanometre-sized INPs are typically found attached to larger particles in the atmosphere (O'Sullivan et al., 2015). The second feature (larger spread in INP concentrations between samples with increasing size) indicates a larger variation in different aerosol sources or source strength (Seinfeld and Pandis, 2016; DeMott et al., 2016) versus the smallest INPs observed which have similar spectra, indicating similar sources. Additionally, stage A of the DRUM has a wider size range (~9 μm) versus the smaller stages (~0.2 to 2 μm). The third reported feature was unexpected, given the characteristically polluted atmosphere in the Prudhoe Bay area. The 22 – 29 May period featured INP concentrations up to 4×10^{-3} L^{-1} at −10 °C and 2×10^{-2} L^{-1} at −15 °C, with warm temperature INPs apparent especially for the largest sizes (i.e., 2.96 – > 12 μm (stage A) and 1.21 – 2.96 μm (stage B)). The onset temperatures (up to −5 °C) were also much higher during this period for the two largest size cuts of the DRUM (Figure 4), indicating a presence of biological INPs (Murray et al., 2012; Kanji et al., 2017). The presence of biological INPs in Prudhoe Bay in the spring are somewhat unexpected given the predominantly frozen surfaces. Despite this, the sizes of the observed INPs indicates a more local or regional origin due to reduced atmospheric lifetime (Jaenicke, 1980). The smallest sizes (0.34 – 1.21 μm (stage C) and 0.15 – 0.34 μm (stage D)) did not portray the same trend during late May. There, a higher fraction of particles was active at much colder temperatures (i.e., INP_{-20} and INP_{-25}) as compared to INP_{-10} or INP_{-15}, where temperatures are generally relevant to less efficient biological INPs (i.e., spores) and mineral dust (Murray et al., 2012; Haga et al., 2014; Boose et al., 2016b). A previous study by Stone et al. (2007) demonstrated transport of Asian dust to Utqiaġvik during the spring, thus, introducing sources of colder temperature INPs even though the surface is predominantly frozen.

3.3 Open Arctic ~~leads~~ waters and snow melt over tundra as sources of observed transition in INP properties

Focusing on the month of May, it is clear that there are higher concentrations of warm temperature INPs, particularly for DRUM stages A and B (Figure 4). Stages C and D include, or are completely composed of, particles that are ~~theoretically thought to be too small to serve as efficient INPs~~thought to be less efficient INPs relative to particles with larger diameters (DeMott et al., 2010; DeMott et al., 2015; Fridlind et al., 2012), which is consistent with the low INP activity from such particles during INPOP. Stage A was especially unique, with the range of freezing temperatures becoming smaller over time. ~~To investigate this range in more detail, we define a parameter, ΔT, which is calculated by subtracting the first onset freezing temperature from the lowest freezing temperature detected (Figure 5). There were no notable trends with ΔT from the other stages. Although ΔT was larger on 16 May for stage A, the highest onset freezing temperature from the entire study was observed during this day as indicated in the previous section.~~ Snow coverage decreased sharply within 700 km of Oliktok Point

on 8 May, while sea ice within 700 km dropped in fraction after 13 May (Figure 5e), which roughly aligns with the increase in onset freezing temperature ~~and lower ΔT values~~ for the largest particles in stage A and generally higher onset freezing temperatures for stages B and C.

The observed increase of warm temperature INPs at Oliktok Point can be attributed to air mass transport pathways and timescales over specific sources. Transport of regional air was quite slow and remained predominantly near the surface, as evidenced by the 5-day air mass back trajectories shown in Figure 6. At the beginning of May, frozen surfaces were prominent across the entire region, including the areas over which air transported to Oliktok Point travelled. On 9 May, the marginal ice zone (MIZ) started to develop off the coast of Oliktok Point as evidenced by the < 100% sea ice coverage values in the 4-km data (i.e., areas of exposed open water indicated by the light purple shading in Figure 6b inset). The MIZ may be a combination of ice floes and/or open leads, but we cannot differentiate these features using the 4-km data. Onset temperatures for stages A and B started to increase on 9 May, coincident with the MIZ starting to expose open water. Starting 16 May, ~~several leads and~~ polynyas were observed ~~in the Beaufort Sea~~west of the Canadian Archipelago and northeast of Oliktok Point, about 700 km away. Air mass trajectories show low-altitude transport over this region, corresponding to a slight increase in warm temperature INPs on that day and the warmest onset temperatures (Figure 5a). D~~ However, d~~ue to the distance between Oliktok and the open water, particles within the size range of stage A likely gravitationally settled during the 5-day ~~transit~~transit, yet such sources may still have provided small contributions of aerosol to the observed enhanced INPs as evidenced by the warmest onset temperatures and only slightly elevated INP concentrations as compared to samples collected before 16 May. Jaenicke (1980) showed that particles within the size range of stage A have typical atmospheric lifetimes on the order of minutes to days, particularly when in the boundary layer and near the surface. Based on the minimum and maximum ambient air temperatures observed at Oliktok Point on 16 May (–3 and 2 ˚C, respectively) and altitudes of the trajectories over the polynya northeast of Oliktok Point (40 – 90 hours back in time), basic terminal settling velocity calculations using on the Navier-Stokes equation indicate particles within the stage A range would have gravitationally settled within 11 seconds to 6 hours, indicating the polynyas were not the dominant source and the MIZ closer to Oliktok Point was more likely the source of the larger INPs. However, this simple calculation assumes starting temperature and does not consider vertical updrafts and downdrafts that may affect particle lifetime. On 22 May, there was a significant shift in air mass origin: air reaching Oliktok Point travelled only ~~originated from~~ over the MIZ ~~a large area of open leads within 30 km off the coast of Utqiaġvik~~and not the polynyas to the northeast ~~-~~(Figure 6d inset). This shift corresponded to the highest INP concentrations ~~and lowest ΔT~~ observed from any sample analysed. Sources were generally regional in nature, with locations directly north of Oliktok Point until 26 May, after which they transitioned to originating predominantly south of Oliktok Point, from exposed tundra over the terrestrial North Slope and interior Alaska. ~~ΔT decreased slightly from 26 May onward, but INP concentrations remained high relative to earlier on during INPOP.~~ Ambient air temperatures at Oliktok Point starting 26 May were relatively warm and fluctuated until 29 May (Figure 2), indicating air originating from over warmer surfaces (i.e., darker land versus brighter ice). Onset temperatures decreased following 26 May and INP concentrations remained high, but for colder freezing temperatures (i.e., –25 ˚C; Figure

4a). On 29 May, ~~AT and~~ INP concentrations dropped but were still higher than Mar – early May samples, and air masses transitioned to originating from the northeast. Like 16 May, terminal settling velocity calculations indicate particles on this day were likely from closer to Oliktok Point, ranging from 13 seconds to 7 hours.

Recent work by May et al. (2016) has demonstrated that production of sea salt aerosol in the Arctic can occur under non-stagnant conditions from open leads (i.e., winds speeds > 4 m s^{-1}). The main mechanisms behind aerosol production from open ocean surfaces is bubble bursting from wind-induced wave breaking, although this process is far less studied over leads. A recent study by Gabric et al. (2018) describes generation of marine biogenic aerosol (MBA) from sea ice leads and the MIZ. Thus, other primary aerosols—such as bacteria or marine organisms that have been shown to serve as efficient INPs—may be generated by the same mechanisms that produce sea salt aerosol and MBA. Recent studies by Wilson et al. (2015) and Irish et al. (2017) have shown that the Arctic Ocean surface microlayer and bulk seawater can harbor large concentrations of INPs, indicating physical mechanisms that generate aerosol from the surface waters may eject these INPs into the atmosphere. Additionally, several previous high Arctic ice nucleation studies have demonstrated that leads and other open water sources as vital to influencing atmospheric INP concentrations (Bigg, 1996; Bigg and Leck, 2001, 2008; Leck and Bigg, 2005). Based on our source analysis and a combination of conclusions from these previous studies, we conclude that INPs from open water features are likely produced via bubble bursting and are likely composed of bacteria or fragments of marine organisms. ~~from predominantly over frozen surfaces.~~

~~Colocated~~Collocated single-particle and bulk chemistry measurements support the sources of the air masses during the transition period in late May (Figure 7). Here, we briefly discuss results from the compositional analyses in support of the ice nucleation measurements. Analysis of May periods by CCSEM-EDX identified marine, terrestrial, and combustion influences based on particle composition and air mass trajectories (Figure 7a – d). Particle types from marine (fresh and aged sea spray aerosol (SSA)) and terrestrial (dust) sources comprised the greatest number fractions of supermicron (> 1.15 µm) aerosol particles for all samples (54 % and 37 %, respectively). Each May sample demonstrated a large fraction of marine influence, with nearly 50% of supermicron particles, by number, comprised of fresh and aged SSA. The largest marine influence was observed on 23 May when air masses originated from the north over open water, local wind speeds were low and northerly, and INP concentrations, particularly in the warm temperature regime, were elevated compared to samples prior to 22 May. However, periods with more terrestrial influence were observed, based on increased number fractions of supermicron dust and southerly winds on 24 May (Figure 7e). Additionally, some periods experienced more combustion influence, from local industrial emissions, characterized by relatively greater fractions of organic aerosol, fly ash, and soot (Gunsch et al., 2017; Kirpes et al., 2018) observed by CCSEM-EDX on 28 May. The S-XRF data (0.75 – 5.0 µm) also demonstrated influences from marine and terrestrial sources, but had the highest marine signature (i.e., from fresh sea salt indicated by the Cl mass concentrations) 16 – 20 May when local winds were elevated (~ 10 m s^{-1}) and originated from the north over the Beaufort Sea. The S-XRF indicated the highest dust influence was observed from 25 – 30 May when episodic southerly winds from over the

tundra influenced Oliktok Point (Figure 7e). The observed dust could be due to local road dust in addition to terrestrial dust sources along the air mass back trajectories.

Combined with air mass trajectory analysis, the single-particle and bulk chemistry support the conclusion that regional natural sources largely contributed to the enhanced warm temperature INP concentrations during late May. Very little contributes from soot and fly ash were observed (4% and 16% on 23-May and 28-May, respectively), indicating local pollution was not a large source of the particles in general. Additionally, soot and fly ash can serve as INPs, but at temperatures much lower than –15˚C (Kanji et al., 2017; Murray et al., 2012). Aside from the composition, we would not expect pollution sourced from Prudhoe Bay (which tends to be sub-100 nm) to overlap with the sizes of the INPs observed (i.e., > 2.96 µm) (Creamean et al., 2018a; Maahn et al., 2017). Another possible source that has been shown to influence this region in the spring is long-range transported aerosol. Recent work by Kylling et al. (2018) demonstrates surface dust impacts from several midlatitude sources to the entire Arctic is predominant during May. Yet, they also show that the major source of dust at the surface is from North American north of 60 ˚N, indicating regional source influences. Specific to our region, supermicron mineral dust has been shown to be transported during Arctic Haze as demonstrated by Quinn et al. (2002) at Utqiaġvik, but at very low mass concentrations relative to other aerosol species. Additionally, given the size in which we observed the INPs (> 2.96 µm), it is unlikely such large particles were transported from very distant sources, especially considering the air mass transport pathways were near the surface in the boundary layer for several days prior to arrival at the site. Thus, based on the combination of previous parallel studies, freezing temperatures, size, single-particle composition, bulk composition, local meteorology, and air mass transport, we demonstrate that it is unlikely local pollution largely influenced the INP concentrations during late May.

4 Comparison to other Arctic INP measurements

Immersion mode INP measurements in the Arctic are rare, but here we provide a synopsis of previous measurements in comparison to ours. Table 2 shows ground-based, shipborne, and airborne studies dating back to 1976 in locations throughout the Arctic during various seasons. We excluded a review of diffusional chamber INP measurements as these instruments predominantly measure deposition mode INPs and are thus not directly comparable to our results (Kanji et al., 2017; Cziczo et al., 2017). While the objective of this comparison is to determine whether our measurements align with previous immersion mode INP observations, a detailed review of Arctic INPs is outside the scope of this manuscript.

Overall, our concentrations at selected temperatures were within range of with those previously reported (Bigg, 1996; Borys, 1989; Conen et al., 2016; DeMott et al., 2016; Fountain and Ohtake, 1985; Radke et al., 1976), even though there are: (1) likely dependencies on time of year and location and (2) a spread by several orders of magnitude is apparent. Figure 8 shows the range of our results in comparison with those in Table 2. In general, our INP concentrations agree with or are on the lower end of those previously reported in the spring. One explanation could be that due to the proximity to the Prudhoe Bay oilfield, we

are sampling in a location that is more polluted at the ground than those previously used to document Arctic haze (Borys, 1989; DeMott et al., 2016; Radke et al., 1976). Additionally, Borys (1989) and Radke et al. (1976) measured via airborne platforms, in which they predominantly measured long-range transported Arctic Haze as compared to Arctic boundary layer aerosol. Other sources of variation in the results could be the result of discrepancies in the sample volumes and drop sizes used under different DFA techniques (Vali, 1971; Mossop and Thorndike, 1966). Also, unlike some of the studies presented in Table 2 and Figure 8, we collected substantial volumes of air per sample to enable measurement of the rare warm temperature INPs (38428 L in the present study versus 250 – 32400 L). Despite these differences, our concentrations are consistent with Bigg (1996), Conen et al. (2016), and DeMott et al. (2016), with those studies conducted during summer or fall. Those previous studies were apportioned to either marine or terrestrial sources, resulting in an analogous set of studies to our current effort. Overall, our measured INP concentrations are comparable to those previously conducted in a similar fashion in the Arctic and predominantly fall within the overall global picture of INPs presented by Kanji et al. and (2017) and Petters and Wright (2015).

5 Summary and broader implications

We present the first INP measurements in an Arctic oilfield location and demonstrate how local and regional transport from marine and terrestrial sources to an industrial region can introduce high concentrations of coarse, warm temperature INPs that are possibly of biological origin. Three time- and size-resolved aerosol impactors were deployed from 1 Mar to 31 May 2017 for offline ice nucleation and chemical analyses and were co-located with routine measurements of aerosol number, size, chemistry, and radiative properties. The largest particles (i.e., ≥ 3 μm or "coarse mode") were the most efficient INPs. During periods with snow- and ice-covered surfaces, coarse mode INP concentrations were very low (maximum of 6×10^{-4} L^{-1} at –15 ˚C), but higher concentrations of warm temperature INPs were observed during late May (maximum of 2×10^{-2} L^{-1} at –15 ˚C). These higher concentrations are attributed to air masses originating from over ~~sea ice leads~~open Arctic Ocean water and tundra surfaces, and were likely primary marine aerosol and dust, respectively. In general, our concentrations agree with similar immersion mode ice nucleation evaluations at other Arctic locations, even though such studies vary in terms of location, volume of air per sample, drop freezing technique, and time of year.

Although our measurements were conducted at the ground level, such particles could influence Arctic MPC (AMPC) formation under conditions where the cloud is coupled to the surface by a well-mixed boundary layer. Previous long-term studies have evaluated the annual cycle of AMPC properties and, relevant for our measurements, have concluded that climatologically a large number of AMPC occur over the North Slope of Alaska in May (up to 84 – 90% cloud fraction), with cloud bases over the North Slope reaching down below 500 – 700 m (Shupe, 2011; Shupe et al., 2010; Dong et al., 2010). Specifically, AMPC fraction is at a maximum in the fall, followed by the spring, with the lowest cloud bases occurring over the Arctic Ocean (Shupe et al., 2006; Shupe et al., 2005; Shupe, 2011). Combined, these studies indicate that INPs from surface sources may be important for such low, persistent clouds. Further, average cloud temperatures in the Arctic have been shown to be relatively

warm in the spring and into the summer (up to –10 ˚C on average) (de Boer et al., 2009; Shupe, 2011), indicating that biological INPs from Arctic sources are relevant to AMPC conditions typical for this time of year.

We have demonstrated that the spring to summer transition on the North Slope and beyond can have implications for a shift in INP properties, potentially impacting cloud phase, precipitation amounts, and cloud lifetime. Given significant shifts in broadband radiation regimes starting March manifesting in a sign change in cloud radiative forcing (CRF) from winter and spring months to summer (Dong et al., 2010), understanding aerosol-cloud interactions is crucial during this time of year. Additionally, with increasing likelihood of earlier melting of frozen surfaces in the Arctic in coming decades (Markus et al., 2009; Stroeve et al., 2014; Screen and Simmonds, 2010), the influence of INP on CRF may be shifting in season along with the sign of CRF, with the latter resulting from a complex mix of shifting atmospheric temperature and surface albedos. Understanding the role of INPs in modulating CRF is important as this forcing can act as a positive or negative feedback on the acceleration of surface melt, and the timing of such melt can potentially lead to impacts on a variety of features, midlatitude weather and climate (Cohen et al., 2018). Together, these items demonstrate a strong need to continue to improve understanding of high latitude INPs, with additional INP concentration and characterization measurements required to confirm the findings presented ~~here, and~~here and evaluate this cycle beyond spring months.

Acknowledgements

We acknowledge Fred Helsel, Bruce Edwardson, and Mark Ivey for assistance in study planning, logistics, and setup. Additionally, we thank AMF-3 operators David Oaks, Ben Bishop, Joshua Remitz, and Wessley King for ensuring successful operation of the DRUM during INPOP. We also thank the members of the Oliktok Point Site Science team for their valuable feedback during data ~~analysis, and~~analysis and acknowledge the NOAA Air Resources Laboratory (www.arl.noaa.gov/ready.php) and Rich Iaonnone for the provision of the SplitR HYSPLIT transport model (https://github.com/rich-iannone/SplitR). Andrew Ault (University of Michigan) is acknowledged for discussions of aerosol sampling for INPOP. Funding was provided by US DOE ARM (2016-6841, 2016-6884, and 2016-6875) and Atmospheric Systems Research (ASR) (DE-SC0013306) programs for sample collection/analysis and data interpretation, respectively. Additionally, funding was provided by the NOAA Physical Sciences Division (PSD) to support data and sample analyses. K.A. Pratt and R. Kirpes were supported, in part, by an Early-Career Research Fellowship from the Gulf Research Program of the National Academies of Sciences, Engineering, and Medicine. The content is solely the responsibility of the authors and does not necessarily represent the official views of the Gulf Research Program of the National Academies of Sciences, Engineering, and Medicine. CCSEM-EDX analyses were performed at the Environmental Molecular Sciences Laboratory (EMSL), a national scientific user facility located at the Pacific Northwest National Laboratory (PNNL) and sponsored by the Office of Biological and Environmental Research of the US Department of Energy (DOE). PNNL is operated for DOE by

Battelle Memorial Institute under contract no. DE-AC06-76RL0 1830. Travel funds to PNNL were provided by the Michigan Space Grant Consortium

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Table 1: Dates and start times of sample collection during INPOP (2017) for samples analysed for INPs. Samples correspond to 1 full day (24 hours from the start time). The "INPs" column corresponds to the stages that were analysed via the DFCP per daily sample. A = 2.96 – >12 µm, B = 1.21 – 2.96 µm, C = 0.34 – 1.21 µm, and D = 0.15 – 0.34 µm.

Date	Start (UTC)	INPs
11-Mar	21:00	A, C
18-Mar	20:00	A, C
25-Mar	20:00	A, C
01-Apr	20:00	A, C
17-Apr	16:00	A, B, C, D
24-Apr	16:00	A, B, C, D
02-May	16:00	A, B, C, D
09-May	16:00	A, B, C, D
16-May	16:30	A, B, C, D
22-May	16:30	A, B, C, D
23-May	16:30	A, B, C, D
24-May	16:30	A, B, C, D
25-May	16:30	A, B, C, D
26-May	16:30	A, B, C, D
27-May	16:30	A, B, C, D
28-May	16:30	A, B, C, D
29-May	16:30	A, B, C, D

Table 2: Comparison of atmospheric immersion mode INPs measurements presented here with previous studies in the Arctic. Study reference, location, dates, and average INP concentrations converted to L^{-1} reported for up to the four different temperatures shown. The superscripts g, s, and a represent ground-based, shipborne, and airborne measurements, respectively. "None" refers to no reported concentrations (or observed, in the case of the current study) measured at that temperature.

Study	Location	Dates	Max volume of air (L)	INPs$_{-10}$ (L^{-1})	INPs$_{-15}$ (L^{-1})	INPs$_{-20}$ (L^{-1})	INPs$_{-25}$ (L^{-1})
Bigg (1996)	High Arctic^{g}	early Aug 1991	3000	none	1×10^{-2}	none	none
		Aug – Oct 1991		none	3×10^{-3}	none	none
Borys (1989)	Alaska, Canada, Greenland^{a}	Apr 1986	1400	none	2×10^{-2}	none	5×10^{-1}
Conen et al. (2016)	Norway^{g}	Jul 2015	24000	6×10^{-4}	7×10^{-3}	none	none
DeMott et al. (2016)	Bering Sea^{s}	Summer 2012	13500	none	3×10^{-3}	3×10^{-2}	none
Fountain and Ohtake (1985)	Alaska^{g}	Aug 1978 – Apr 1979	250	none	none	1×10^{-1}	none
Mason et al. (2016)	Canada^{g}	Mar – Jul 2014	32400*	none	5×10^{-2}	2×10^{-1}	1
Radke et al. (1976)	Alaska^{a}	Mar 1970	3000	none	none	2×10^{-2}	none
This study (2018)	Alaska^{g}	Mar – May 2017	38428	8×10^{-4}	5×10^{-3}	2×10^{-2}	4×10^{-2}
		Mar 2017		none	7×10^{-4}	6×10^{-3}	3×10^{-2}
		Apr 2017		7×10^{-5}	2×10^{-3}	1×10^{-2}	7×10^{-2}
		May 2017		1×10^{-3}	8×10^{-3}	2×10^{-2}	4×10^{-2}
		late May 2017		2×10^{-3}	1×10^{-2}	3×10^{-2}	4×10^{-2}

*Not directly provided by citation. Estimated from average total volume of air per sample and standard flow rate specification for the sampler.

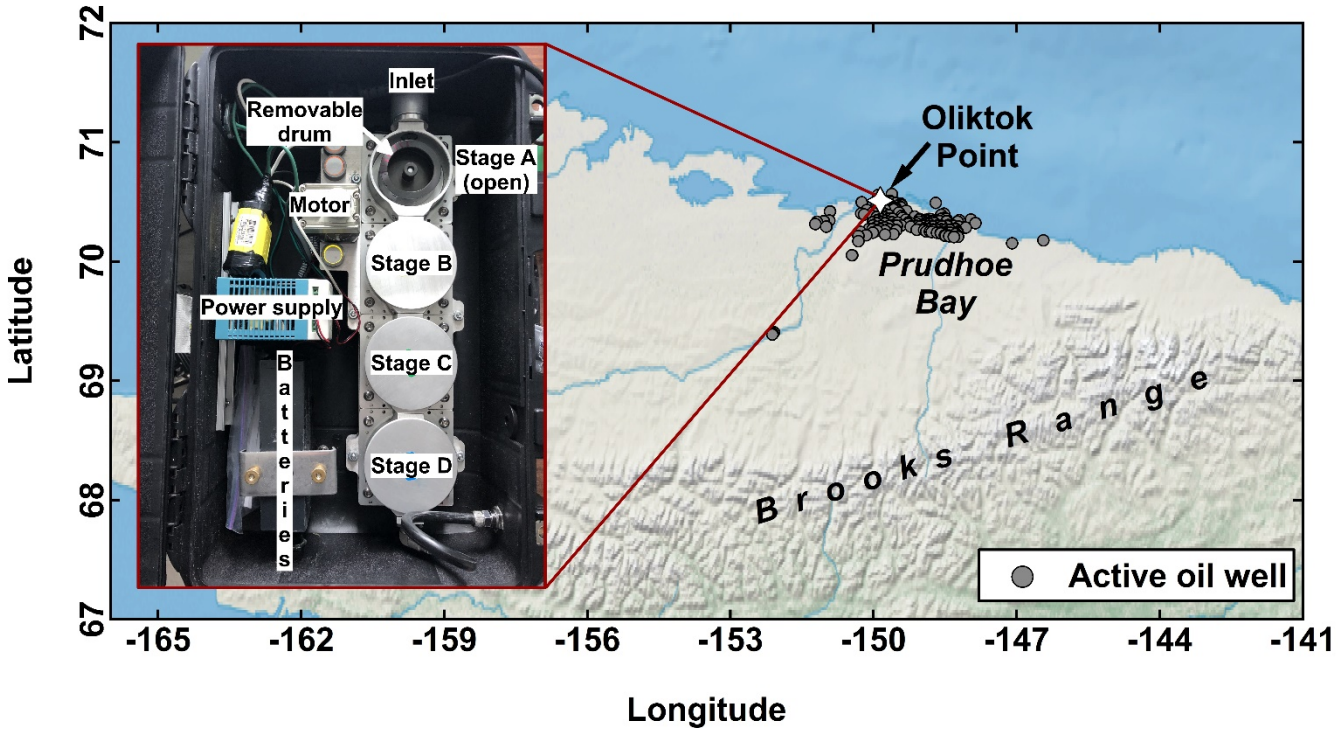
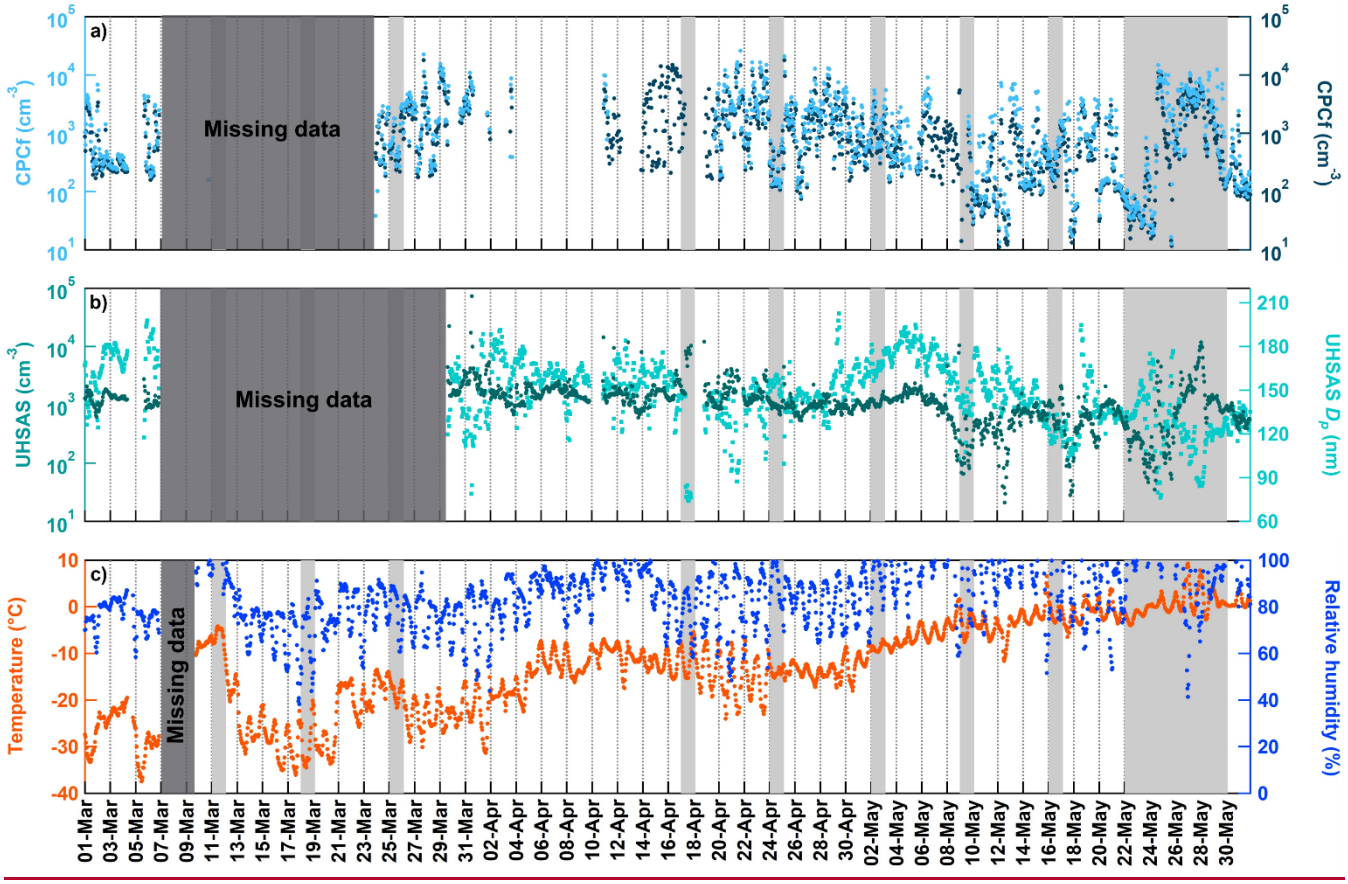


Figure 1: Map of the North Slope of Alaska highlighting Oliktok Point and oil wells that are active in Prudhoe Bay (data obtained from http://doa.alaska.gov/ogc/publicdb.html in Mar 2017). The approximate areas of Prudhoe Bay and the Brooks Mountain range are shown. Inset shows the inside of the DRUM case, with stage A exposed (i.e., cover removed) and major components labelled.

5


a)
Missing data
CPCf (cm^{-3})
b)
Missing data
UHSAS (cm^{-3})
UHSAS D_p (nm)
c)
Missing data
Temperature (°C)
Relative humidity (%)
01-Mar
03-Mar
05-Mar
07-Mar
09-Mar
11-Mar
13-Mar
15-Mar
17-Mar
19-Mar
21-Mar
23-Mar
25-Mar
27-Mar
29-Mar
31-Mar
02-Apr
04-Apr
06-Apr
08-Apr
10-Apr
12-Apr
14-Apr
16-Apr
18-Apr
20-Apr
22-Apr
24-Apr
26-Apr
28-Apr
30-Apr
02-May
04-May
06-May
08-May
10-May
12-May
14-May
16-May
18-May
20-May
22-May
24-May
26-May
28-May
30-May

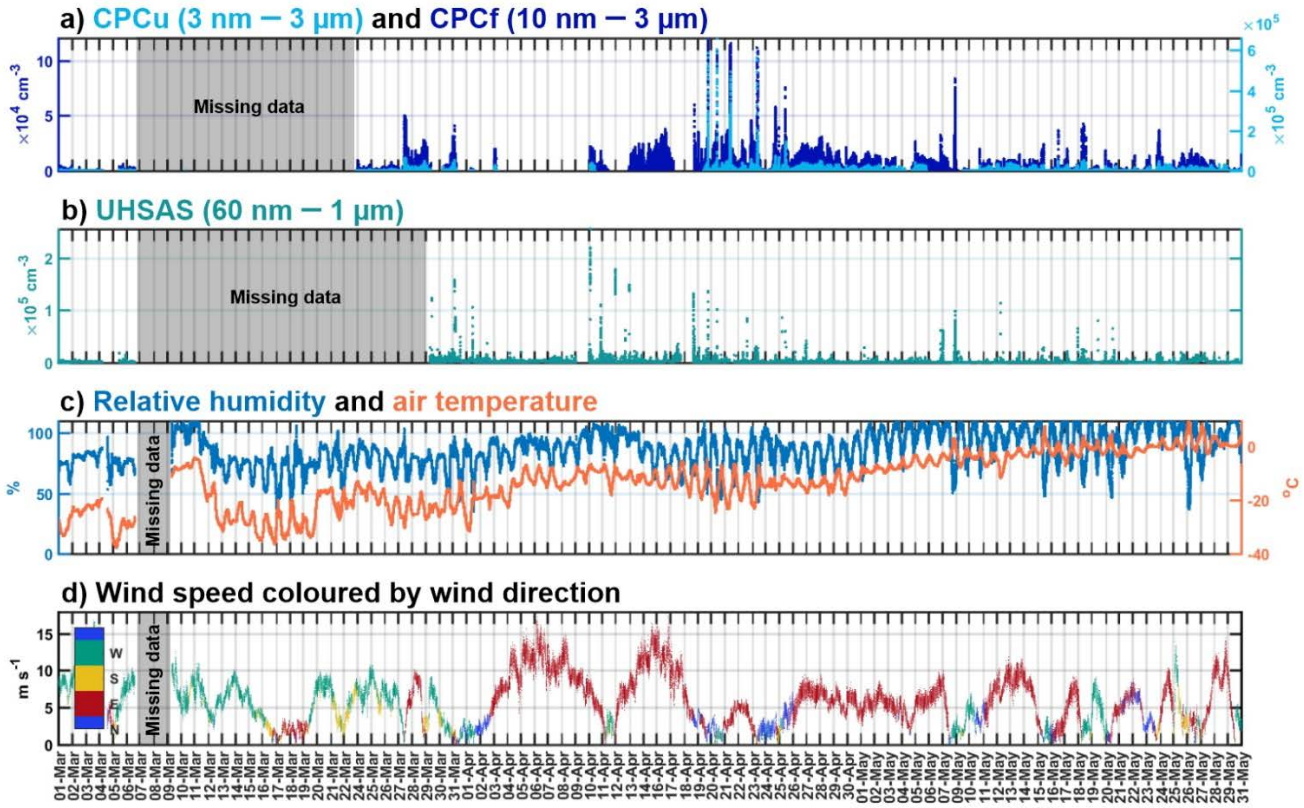
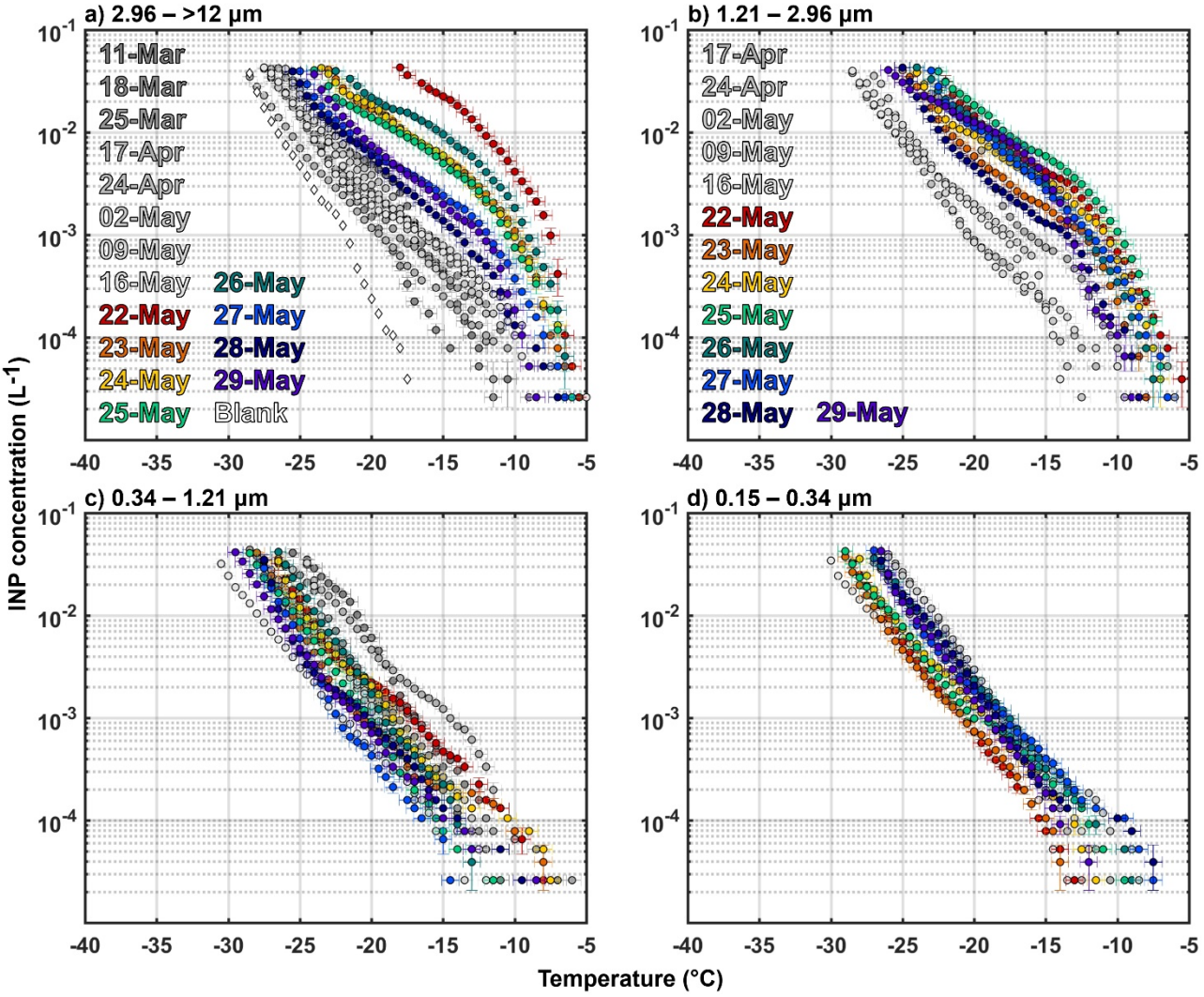


Figure 2: Time series of ~~1 second~~hourly-averaged aerosol number and meteorological variables from AOSMET during 1 Mar – 31 May 2017, including aerosol number concentrations measured by a) the condensation particle counter (CPCf) and ultrafine CPC (CPCu) and b) the ultrahigh-sensitivity aerosol sizer (UHSAS). Average hourly mean particle diameter (D_p) is also shown from the UHSAS in b). Meteorological data include c) relative humidity and air temperature ~~and d) wind speed coloured by wind direction~~. Light grey shading represents days where samples were analysed for INP concentrations. Dark g~~G~~rey shading represents times with missing data.


a) 2.96 – >12 µm
b) 1.21 – 2.96 µm
c) 0.34 – 1.21 µm
d) 0.15 – 0.34 µm
INP concentration (L-1)
Temperature (°C)
11-Mar
18-Mar
25-Mar
17-Apr
24-Apr
02-May
09-May
16-May
22-May
23-May
24-May
25-May
26-May
27-May
28-May
29-May
Blank
17-Apr
24-Apr
02-May
09-May
16-May
22-May
23-May
24-May
25-May
26-May
27-May
28-May
29-May

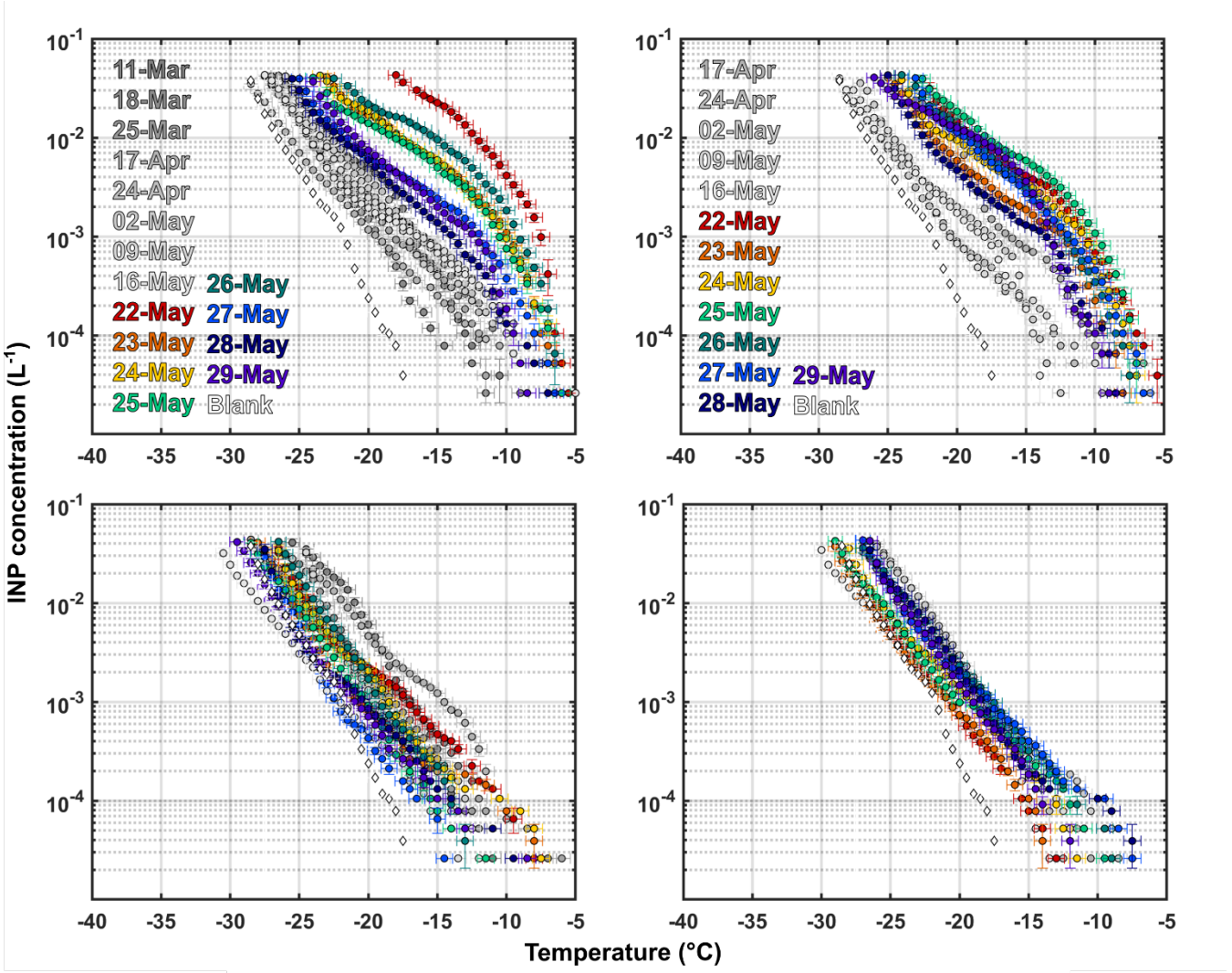
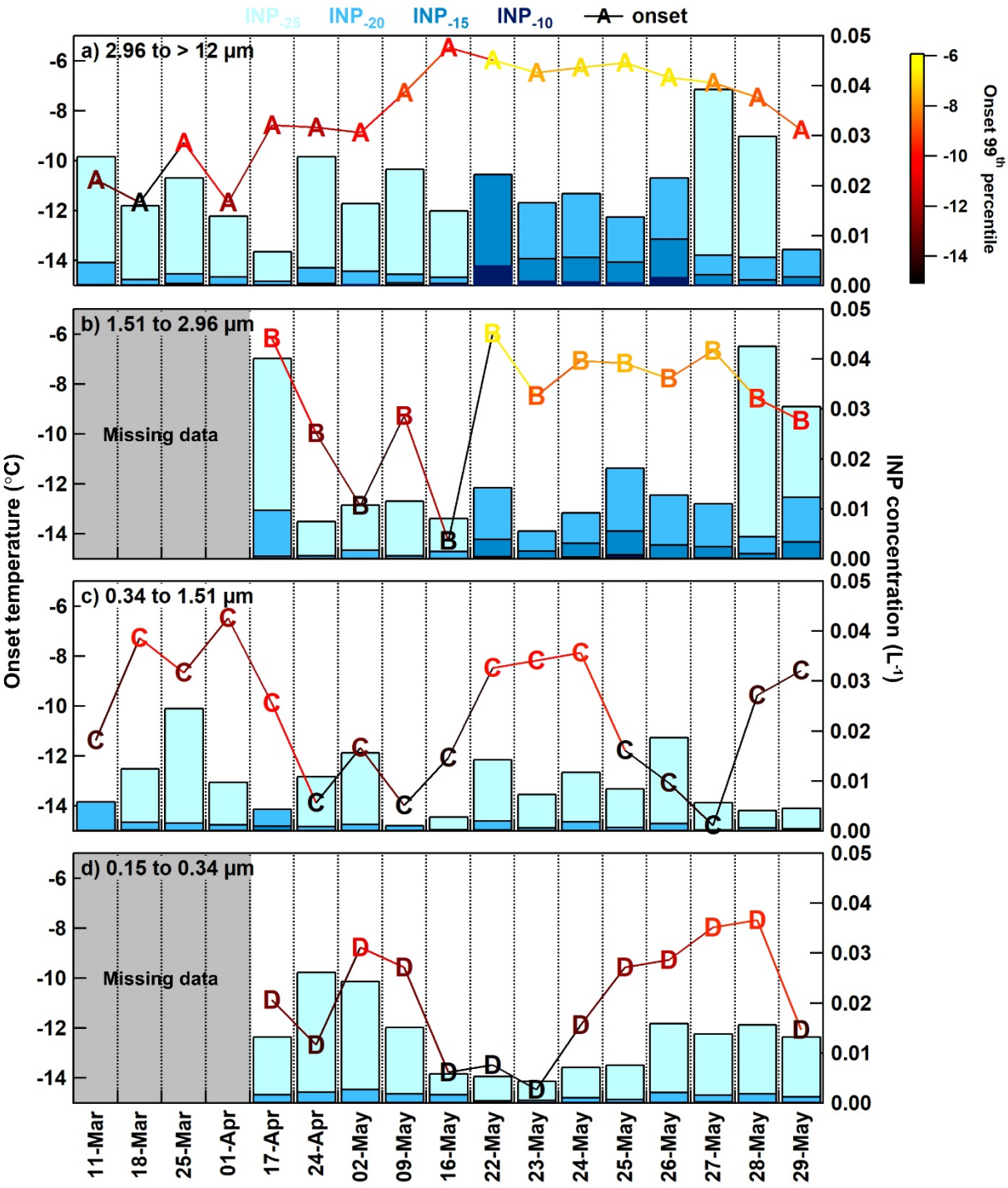


Figure 3: Cumulative INP spectra for the four size-ranged samples of the DRUM from INPOP. One daily sample per week is shown from 11 Mar to 16 May and 17 Apr to 16 May for stages A/C and B/D, respectively. Daily samples are shown from 22 May – 29 May for all stages. White diamond markers ~~in a) only~~ denote the blank Mylar sample in UPW prepared in the same manner as the samples. This blank spectrum applies to all size ranges.


INP-25
INP-20
INP-15
INP-10
onset
a) 2.96 to > 12 µm
b) 1.51 to 2.96 µm
c) 0.34 to 1.51 µm
d) 0.15 to 0.34 µm
Missing data
Missing data
Onset temperature (°C)
INP concentration (L-1)
Onset 99th percentile
11-Mar
18-Mar
25-Mar
01-Apr
17-Apr
24-Apr
02-May
09-May
16-May
22-May
23-May
24-May
25-May
26-May
27-May
28-May
29-May

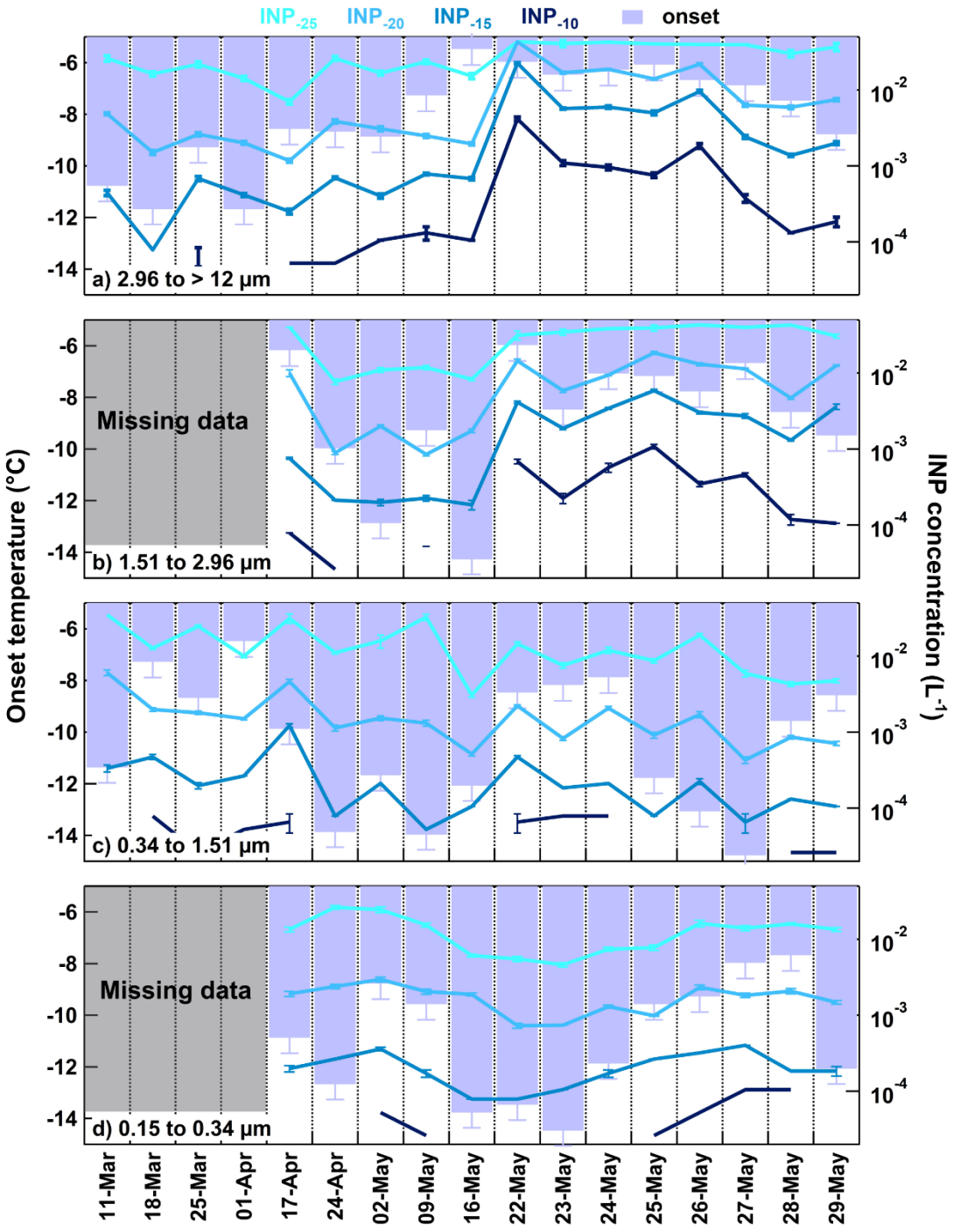
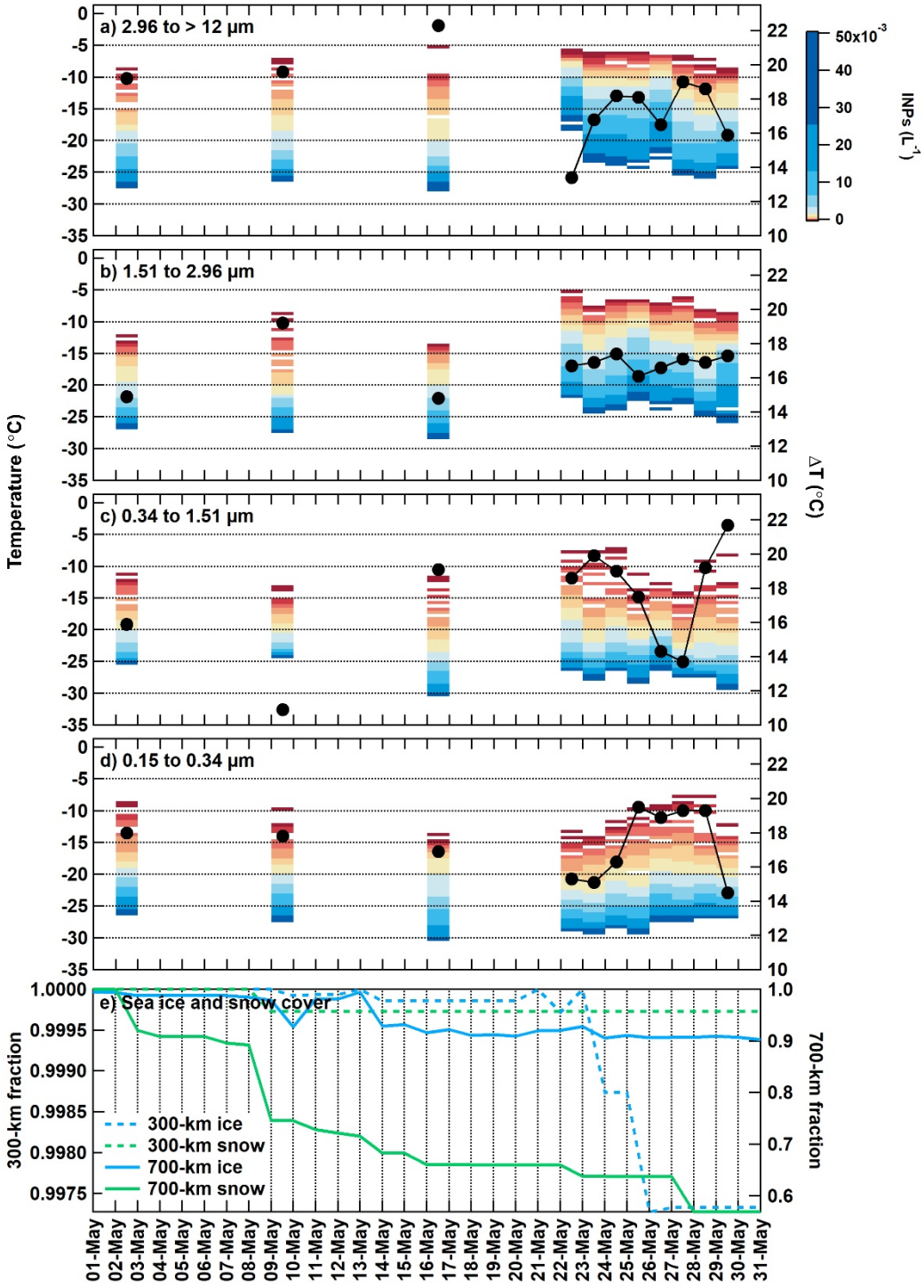


Figure 4: Onset freezing temperatures and INP concentrations at −10 °C, −15 °C, −20 °C, and −25 °C for DRUM stages a) A, b) B, c) C, and d) D for each sample collected during INPOP. Dark ~~. The onset freezing temperatures are coloured by the onset temperature at the 99th percentile. INP -25 concentrations for 22 – 26 May for stages A and B were zero because those all samples~~

~~froze before −25 °C. G~~grey shading represents missing data. Error bars for the onset temperature represent the probe uncertainty and plate correction and standard deviation for the INP concentrations.


a) 2.96 to > 12 µm
b) 1.51 to 2.96 µm
c) 0.34 to 1.51 µm
d) 0.15 to 0.34 µm
e) Sea ice and snow cover
Temperature (°C)
ΔT (°C)
INPs (L-1)
50x10-3
40
30
20
10
0
300-km fraction
700-km fraction
300-km ice
300-km snow
700-km ice
700-km snow
01-May
02-May
03-May
04-May
05-May
06-May
07-May
08-May
09-May
10-May
11-May
12-May
13-May
14-May
15-May
16-May
17-May
18-May
19-May
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26-May
27-May
28-May
29-May
30-May
31-May

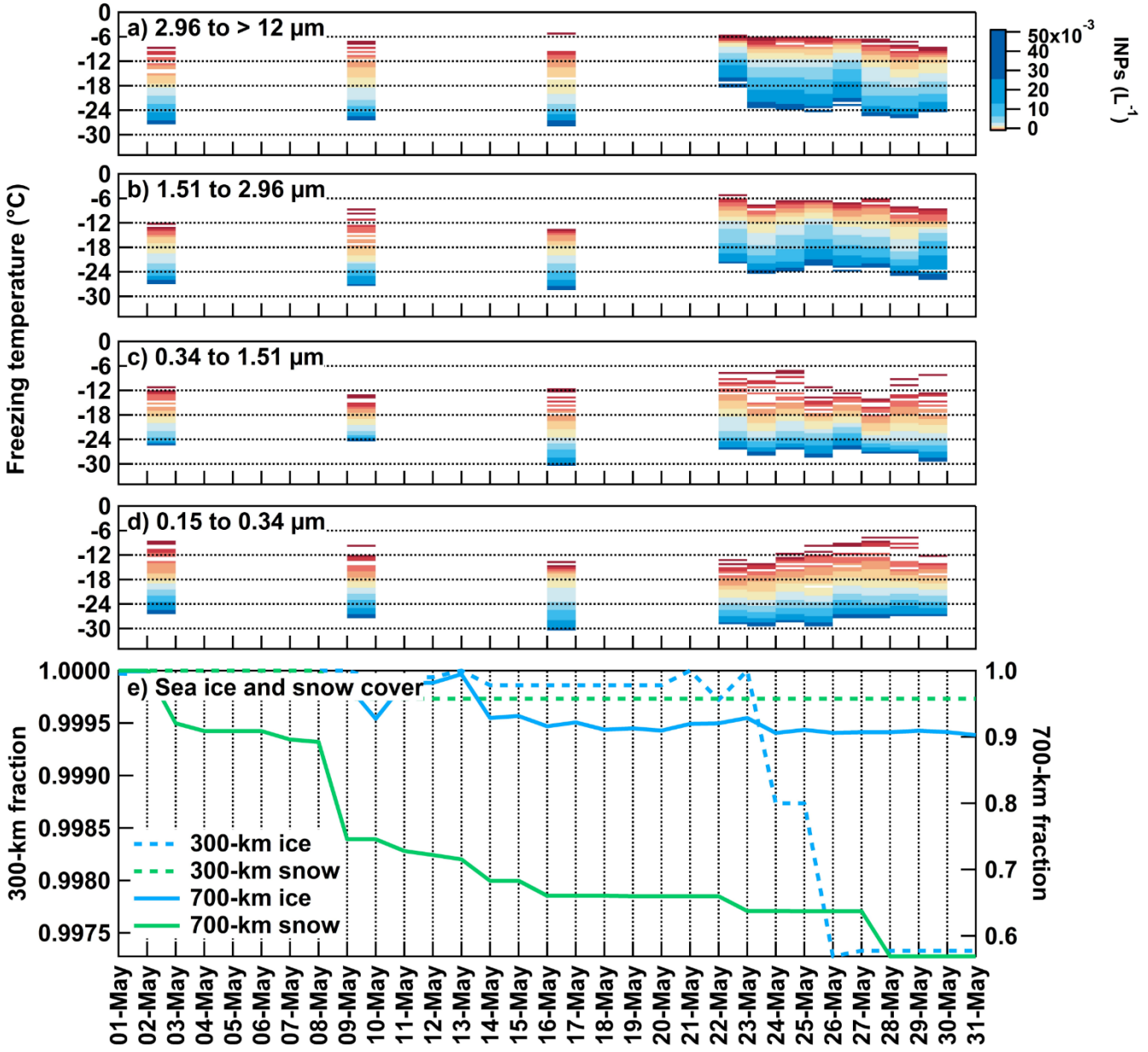
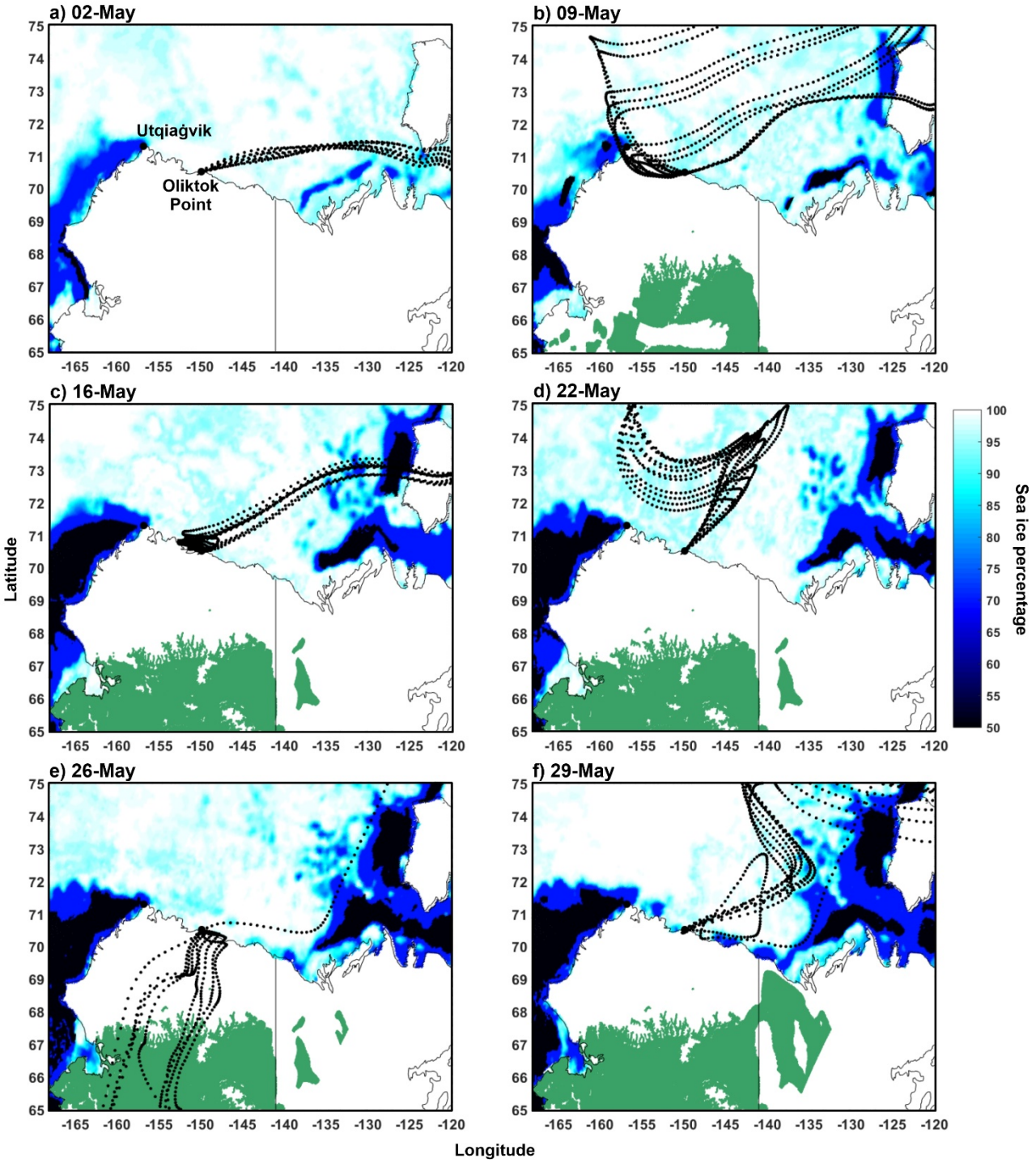


Figure 5: Time series of INP freezing temperatures coloured by INP concentrations (coloured bands, left axis) ~~and ΔT values (black markers, right axis)~~ for DRUM stages a) A, b) B, c) C, and d) D. Also shown are the e) sea ice and snow coverage within a 300-km and 700-km radius from Oliktok Point. Data for the month of May are shown (http://nsidc.org/data/g02156#table3).


a) 02-May
b) 09-May
c) 16-May
d) 22-May
e) 26-May
f) 29-May
Utqiaġvik
Oliktok Point
Latitude
Longitude
Sea ice percentage
75
74
73
72
71
70
69
68
67
66
65
-165 -160 -155 -150 -145 -140 -135 -130 -125 -120
100
95
90
85
80
75
70
65
60
55
50

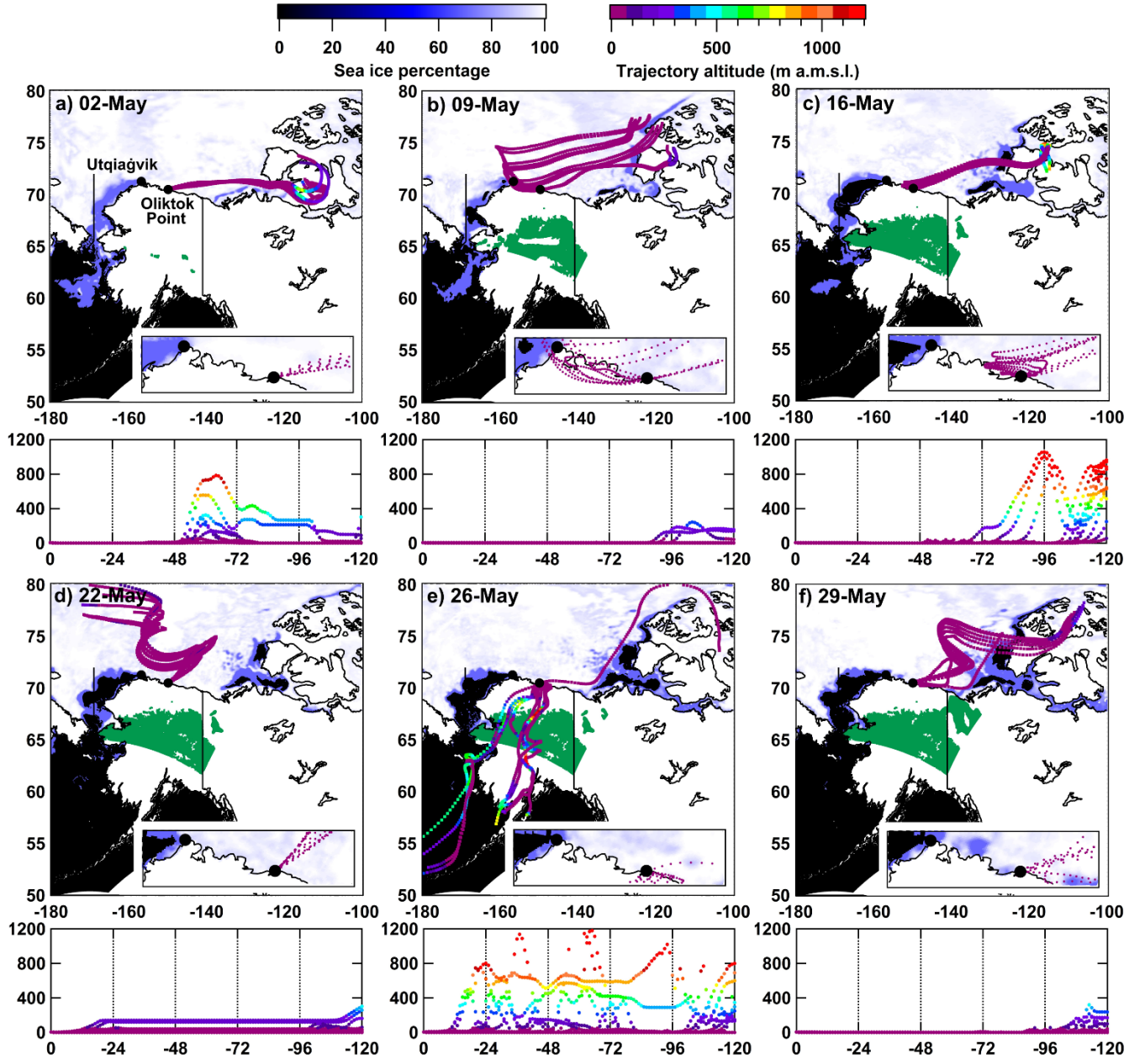
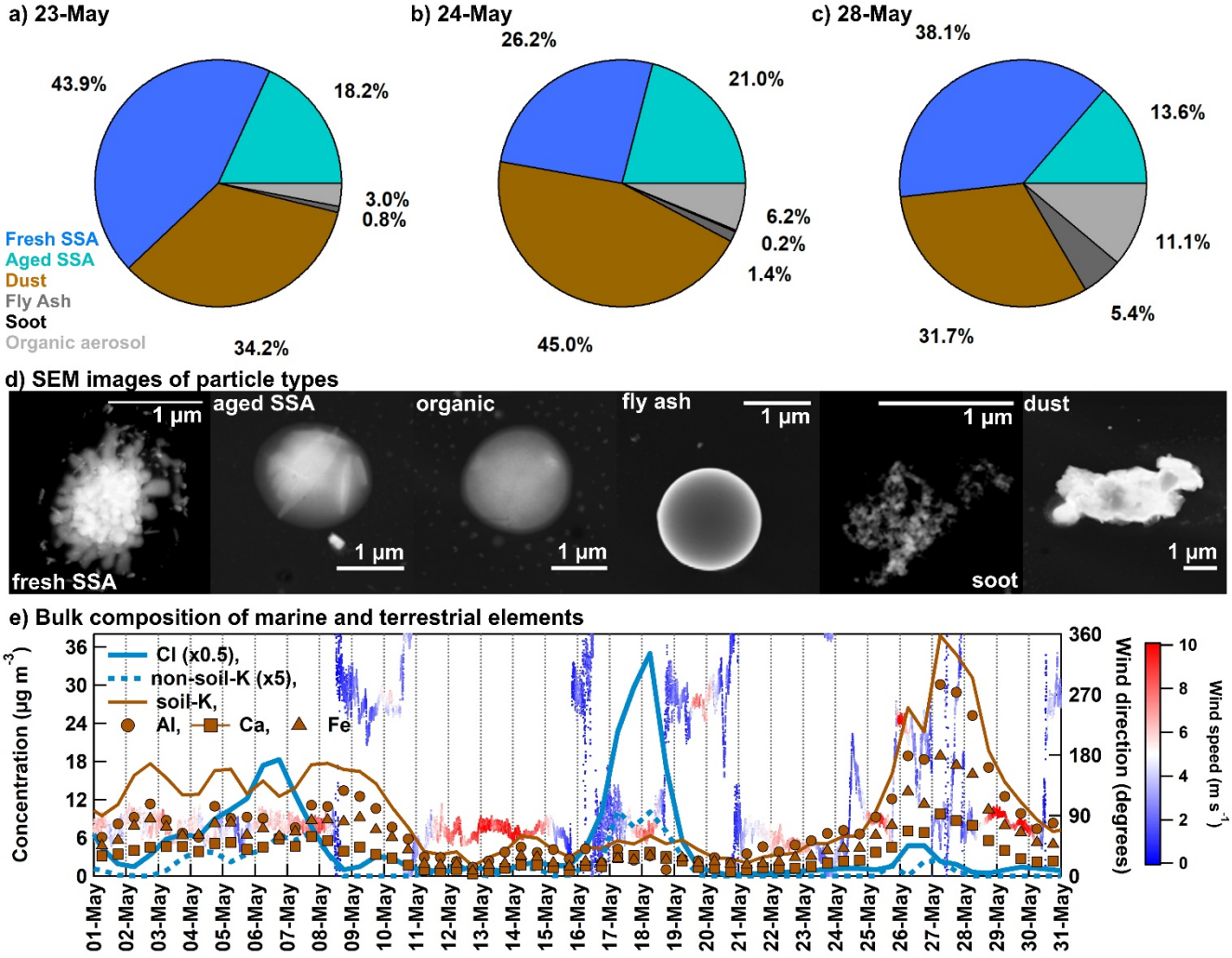


Figure 6: 5-day air mass backward trajectories initiated every 3 hours and land cover (e.g., sea ice, snow, ocean, and open land) for some of the case days in May with INP data. Each date contains the map and corresponding time-height cross section of the 5-day trajectories (shown below each map). The x- and y- axes for the maps represent degrees longitude and latitude, respectively, while those axes for the time-height cross sections represent hours back in time and altitude (m a.m.s.l.), respectively. The locations of Utqiaġvik and Oliktok Point are ~~also~~ labelled in panel a) but indicated in all panels by the black circles. On land, white indicates snow covered surfaces while green indicates open land (e.g., tundra). Land data are missing in southern Alaska and most of Canada.


a) 23-May
43.9%
18.2%
3.0%
0.8%
34.2%
Fresh SSA
Aged SSA
Dust
Fly Ash
Soot
Organic aerosol
b) 24-May
26.2%
21.0%
6.2%
0.2%
1.4%
45.0%
c) 28-May
38.1%
13.6%
11.1%
5.4%
31.7%
d) SEM images of particle types
1 µm
fresh SSA
aged SSA
1 µm
organic
1 µm
fly ash
1 µm
soot
dust
1 µm
e) Bulk composition of marine and terrestrial elements
Concentration (µg m-3)
Cl (x0.5),
non-soil-K (x5),
soil-K,
Al, Ca, Fe
Wind direction (degrees)
Wind speed (m s-1)
01-May
02-May
03-May
04-May
05-May
06-May
07-May
08-May
09-May
10-May
11-May
12-May
13-May
14-May
15-May
16-May
17-May
18-May
19-May
20-May
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26-May
27-May
28-May
29-May
30-May
31-May

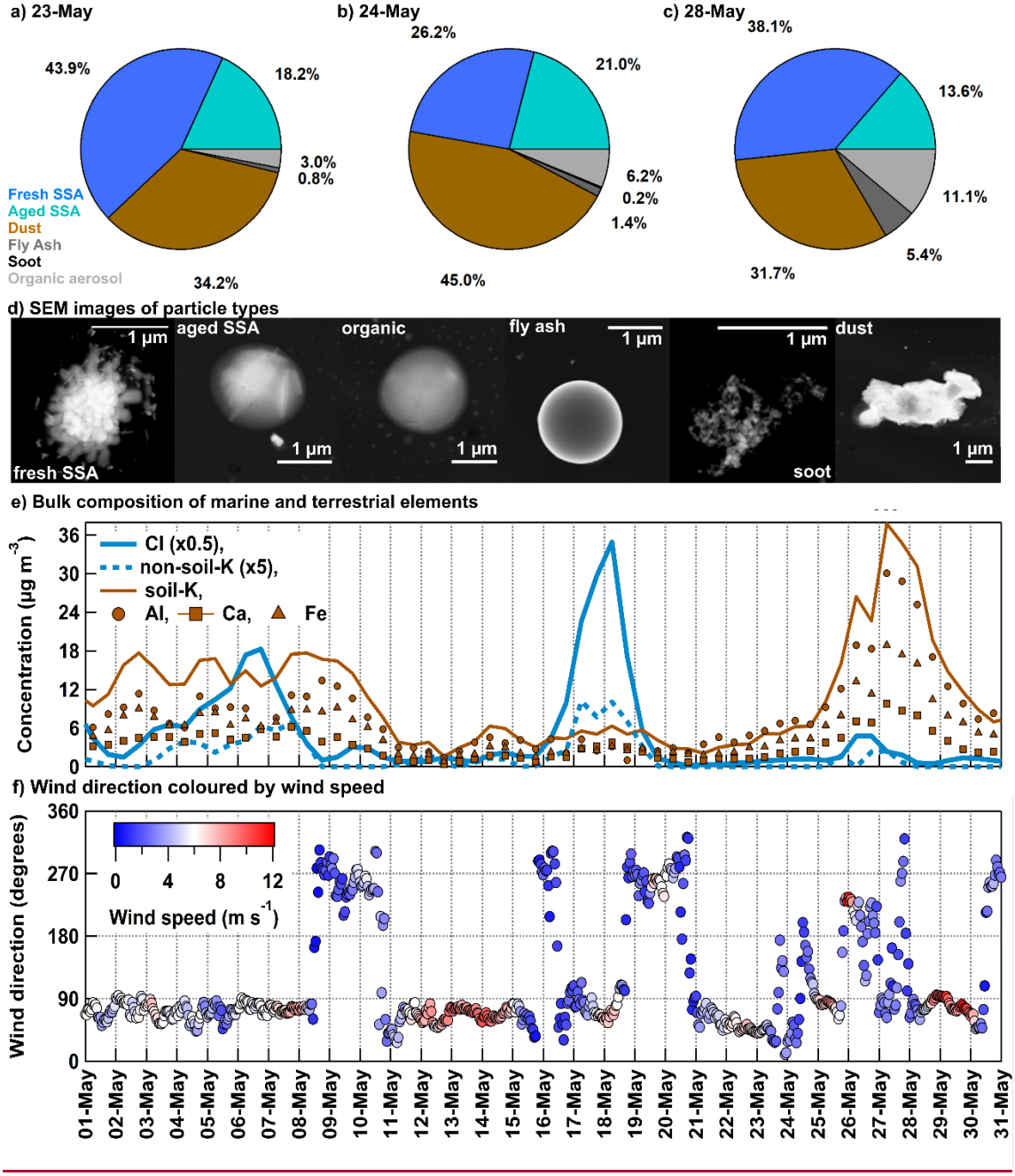

Figure 7: Summary of results from single-particle and bulk compositional measurements during the late May time period. Relative number fractions of CCSEM-EDX single-particle types for samples (> 1.15 µm) for a) 23 May (1156 particles), a) 24 May (918 particles), and c) 28 May (723 particles). SSA represents sea spray aerosol. Examples of SEM images of the particle types classified are shown in d), with the length of the bar scaled to 1 µm. S-XRF results for May for elements in the 0.75 to 5.0 µm size range associated with either marine (Cl and non-soil-K) or terrestrial (soil-K, Al, Ca, and Fe) sources are shown in e). Cl and non-soil-K concentrations are provided as ×0.5 and ×5 their concentrations, respectively, to show on the same axis. f) Wind direction is also shown, coloured by wind speed. For both CCSEM-EDX and S-XRF classifications, shades of blue and brown represent marine and terrestrial sources, respectively.

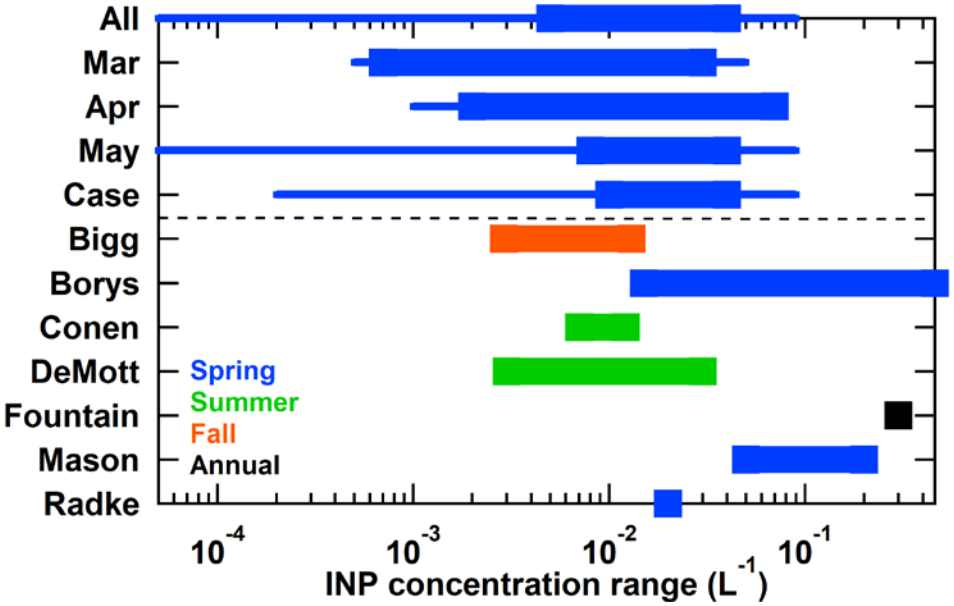

Figure 8: Summary of ranges of atmospheric immersion mode INP concentrations from the current study and those previously reported in the Arctic. Study details can be found in Table 2. For the current study, "all" and "case" correspond to samples from the entire INPOP study and to the 22 – 29 May time period, respectively. Additionally, the bars and whiskers represent the range between the minimum and maximum INP concentrations at the four temperatures in Table 2 per study ~~and at all temperatures, respectively~~. Measurements are coloured to the season in which they are predominantly collected during. The dashed line separates results from the current study from those presented in previous works.