

We would like to thank the reviewer for his/her insightful feedback regarding our manuscript. We have revised based on his/her commentary and believe the manuscript is much stronger as a result.

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 meant 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 meant 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 to 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 different 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 $> 1.15 \mu\text{m}$ 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., $> 2.96 \mu\text{m}$) 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.

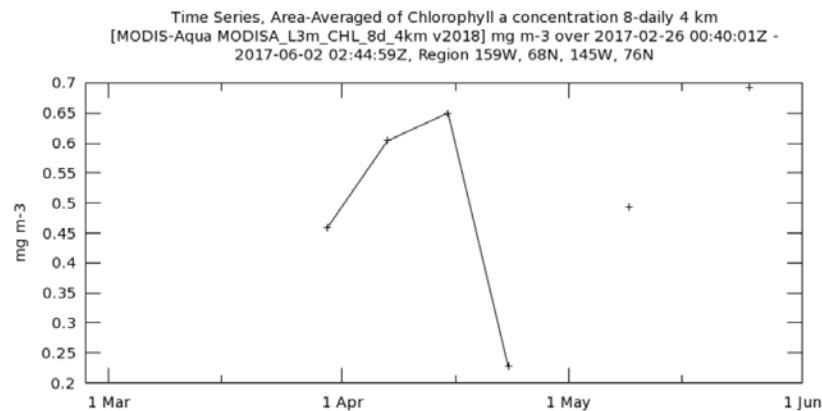
P10 – L19 – on May 29, it looks like there is a portion of the back trajectory (72-73 N and 137-133 W) with lower sea ice percentage compared to any portion of the May 22 source region. How are these two regions distinct? What is considered a significant amount of time to spend over an open lead?

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 $> 4 \text{ m s}^{-1}$). 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.