Prof. Daniel Cziczo
Co-Editor of Atmospheric Chemistry and Physics

Dear Dan,

Listed below are our responses to the comments from the reviewers of our manuscript. For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. ***[1]***). Authors' responses are in red below each referee statement with matching numbers (e.g. ***[A1]***). We thank the reviewers for carefully reading our manuscript and for their helpful suggestions!

Sincerely,

Allan Bertram
Professor of Chemistry
University of British Columbia

Anonymous Referee #1

General Comments

This paper represents a nice contribution to the literature, adding to the limited data on immersion freezing INP spectra in the Arctic region and emphasizing the dominant role that mineral dusts can play when overlain on pristine air masses that are otherwise representative of marine air. I was only curious about the use of a ratio calculation made on the basis of mineral and sea spray aerosol surface areas estimated by CCSEM, rather than actually calculating a surface-active site density on the basis of the aerosol distribution to support what composition is most responsible for the INP activity. While the ratio approach is one to take, it would be good to see actual surface area estimates in order to understand the consistency of the data with previous mineral dust parameterizations, rather than only assuming the validity a priori. For example, only three samples were analyzed, understandable of course given the effort needed to perform the analyses of many particles for composition and size. Given this though, the unknown exact source of the dust, and the fact that what remains active after transport and any scavenging removal could differ from pure minerals tested in the laboratory, it could be interesting to know if the ice activity is truly consistent with dust parameterizations or is reduced and in a more competitive scenario with marine INPs. Otherwise, it is difficult to know if the inference of the dust dominance at all temperatures and loadings is as assumed. To know this, one might need to know mass fractions and surface areas very accurately. Hence, I suggest to add these actual values to the tables, and perhaps some discussion of alignment of data with the published or estimated parameterizations used to quantify assumed mineral influence.

As for the estimates of marine INP contributions, the authors may know that an active site density parameterization is now in the literature (McCluskey et al., 2018), which seems to align quite well with the values they infer from published laboratory data. I was a little curious about two factors in how the data were utilized. First, there is sparse data at -15 to -20 ◦C as emphasized temperatures. And I note on Page 9, lines 13-14, that even 36% of the samples had no INPs at -25 ◦C. Were "zero" or unresolvable data treated in some manner to come up with the average values plotted in Fig. 3? Also, a decision was made to not attempt to correct and discuss INPs for data at lower temperatures where background is present from the pure water. Was the correction simply too large in this region? Finally, I question whether time over marine areas would be expected to positively correlate with INP concentrations in any case. This is not obvious to me given the low strength of the marine source of INPs and the ready ability of terrestrial sources to dominate with any exposure to land emissions, which has been clear in some past studies. Specific questions/comments for addressing before publication are listed below. The paper is otherwise very well written, so these extra comments are limited to the topics overviewed above.

Specific Comments

[1] Page 3, lines 17-20: Could the meaning of the wind directions noted be clarified? Do these imply from open water? Or assumed free from stack contamination? This actually touches on the topic of using remote ocean data and assuming marine only influence of course, about which not much is said.

[A1] The wind directions noted correspond to conditions assumed to be free from stack contamination. To address the referee's comment this information has been added to the revised manuscript. Specifically the following text has been added to the manuscript (Section 2.1):

"The data reported in DeMott et al. (2016) only included sites in Baffin Bay, days where it did not rain and conditions when the samples were assumed to have reduced exposure to ship smokestack contamination i.e. when the apparent wind direction measured on the ship was between 0-90 degrees or 270-360 degrees, where 0/360 corresponds to the bow of ship"

[2] Page 5, lines 18-19: Can you justify not considering the freezing of drops not on a spot as another sort of background freezing spectrum? Is it because you cannot be certain that some small particles did not migrate under this position on the slide?

[A2] Correct - we cannot be certain that some small particles do not migrate under this position on the slide. As stated on Page 5, Lines 19-21, "We assumed these relatively rare occurrences were due to particles < 0.18 μm in diameter that were not focused into spots or due to rebound of a small fraction of the particles off the hydrophobic glass slides". *We do not consider this freezing as another sort of background freezing spectrum since no freezing from the blanks was observed at temperatures ≥-25 °C.*

[3] Page 7, lines 3-5: This statement about the footprint layer confused me because I could not clearly distinguish how this differed from the earlier statement that the terminal point was 0 to 60 m above sea level. Please add, if possible.

[A3] At the start of the simulation the particles are released in a box and are followed backwards in time. For our study the box had the dimensions 100 x 100 m in the horizontal and from 0 to 60 m in the vertical. This initial box is only relevant for the release, after which the particles are free to move in all dimensions. After release of the particles, they are followed backwards in time to determine potential emission sensitivity (PES) plots. Since we are concerned with emissions from the surface we determined PES plots for the footprint layer (0 - 300 m).

We have added text to the manuscript to clarify that 0 - 60 m in the vertical only relates to initial simulation conditions.

[4] Page 7, lines 19-20: One sample with "low" INP was selected, but I see only 4 or 5 samples in Fig. 2 that even have data at -15 ◦C, so if three of these were used, then they are already not low INP samples I would judge. Is there a possible bias in the samples selected? After all, INP concentrations of 0.1 per liter at this temperature are fairly classical continental numbers in the first place. Your qualification about the conclusions being applicable only to the CCSEM cases on page 9 is duly noted. I wonder if you might comment about the influence of selection of samples for microscopy.

[A4] We chose to use two samples that showed high INP concentrations and one sample with a low INP concentration. The 29th July sample did not display freezing activity above -25 °C, which is why it was considered a low INP concentration day. We explored two days with higher INP concentrations, the 25th July and the 21st July. These two days were random choices. We have adjusted the text in section 3.2 to read:

"The two samples corresponding to high [INP(T)] were collected on July 21st and 25th, these days were chosen randomly out of the samples that showed freezing activity at -20 °C and -25 °C. The sample corresponding to a low [INP(T)] was collected on July 29th, this day was chosen randomly out of the samples that did not display any freezing activity above -25 °C. "

[5] Page 7, lines 23-24: Please clarify if fully quantitative EDS analysis is performed to determine atomic percentages. That is the counts are interpreted quantitatively or qualitatively? Figure S4 shows actual concentration values as a means of interpreting mixtures as sea salt or dust, but there are no mixtures. It would be interesting to know how many of the dusts were salty, as processing may potentially alter their assumed behaviors (i.e., compared to parameterized dust).

[A5] A fully quantitative EDX analysis was performed to determine atomic percentages. For the current analysis, we combined particles classified as mineral dust and mixtures of mineral dust and sea salt together, as illustrated in Figure S4. If we used the

classification scheme by Derimian et al. (2017) that explicitly classifies particles as sea salt, mineral dust, and mixed sea salt/mineral dust, for the three samples analysed, no particles are classified as mixed sea salt/mineral dust. The text in the manuscript has been adjusted to the following:

"This classification scheme does not include a mixed mineral dust/sea salt particle category. Rather, any particles that have both sea salt and mineral dust are classified as either mineral dust or sea salt depending on the largest atomic percentage contribution. If we used the classification scheme by Derimian et al. (2017), which explicitly classifies particles as sea salt, mineral dust, and mixed sea salt/mineral dust, no particles would be classified as mixed sea salt/mineral dust particles."

[6] Page 7, lines 27-28: Please also tabulate surface areas attributed to mineral dust and sea salt. It seems that one could also derive ns for each of these. This is critically important to the assumption that these can simply be applied in existing parameterizations that will then be turned into a ratio according to your Eq. 3.

[A6] To address the referee's comment the surface areas attributed to mineral dust and sea salt have been added in Table S2 in the Supplement. Derivation of n_s is discussed below.

[7] Page 8, lines 18-21: Following from what I just said, if you have surface area, can't you compute ns and compare to parameterizations on the assumptions that particles are all SSA or all minerals, or use the mineral ratio to parse it out? Are the results consistent in any way with the parameterization or does the dust act differently than expected? Otherwise, by applying Niemand et al., you are making assumptions about the activity of the dust in these samples that may or may not be true. The same could be said about the marine spectrum, but the dust appears to dominate here, so is the most critical assumption.

[A7] To address the referee's comment we have added calculations of n_s-values of mineral dust to the revised manuscript and we have compared the calculated n_s-values to data in the literature for mineral dust.

[8] Page 9: The correlation analyses are not especially impressive on first glance. Nevertheless, it seems to make sense that residence time over land would have a positive impact on INP concentrations, while time over water would not be expected to matter. This is the natural result when the land surface is a so much more powerful source by the amount that has been noted in past studies. The marine source would only be expected to show up when there is little or no land influence. Both sources would seem to depend more on wind conditions.

[A8] We agree with the referee.

[9] Page 10, lines 11-12: As noted, there is field evidence for marine INP ice active surface site densities in line with what is assumed for the exercise here.

[A9] In the revised manuscript we have referenced the field evidence for marine INP ice active surface site densities.

[10] Page 10, line 14: The comment about biological INPs is kind of a hanging thought. Are you inferring that the results are influenced by these? Your references suggest a range of source types including microbes or macromolecules it seems. While one might agree with the sentiment, it is not clear how it relates to the subject of this paper and why it only appears in the conclusions.

[A10] We have removed the comment about biological INPs.

[11] Page 10, lines 26-27: While the correlation analyses and dispersion modeling support the role of mineral dust especially in case of higher loading (e.g., a few to 10% of surface area represented is a good amount of mineral dust), this does not indicate at what level marine INPs become important for lower INP concentrations does it? In a number of cases, INPs were apparently below detection limits, possibly consistent with typical surface areas and marine INP ns, as well as with limited sample volumes assessed (limited warm temperature assessment). And again, any time spent over land would be expected to influence INPs strongly, while time spent over what might be a more constant and low INP source like the ocean would not be expected to correlate with INP concentrations. Those numbers may be relatively uniform independent of time spent over the marine source, but instead more correlated with marine conditions that influence emission rates. The fractional marine influence does not seem fully evaluated, as it would have required assessing more cases at the lower end of the spectrum of measurements I suspect.

[A11] We agree with the referee.

We have modified this discussion from the following: "This correlation analysis together with the particle dispersion modelling provides further evidence, that sea spray aerosol was likely not the major source of INPs during sampling."

To:

"This correlation analysis together with the particle dispersion modelling suggests that sea spray aerosol was likely not the major source of INPs during sampling, at least not when INP concentrations were high. Sea spray aerosol may still have played a role when the INP concentrations were low during sampling."

References

McCluskey, C. S., Ovadnevaite, J., Rinaldi, M., Atkinson, J., Belosi, F., Ceburnis, D., et al. (2018). Marine and terrestrial organic ice-nucleating particles in pristine marine to continentally influenced Northeast Atlantic air masses. Journal of Geophysical Research: Atmospheres, 123. https://doi.org/10.1029/2017JD028033

Anonymous Referee #2

Summary

This study researched the quantity, chemistry, and potential sources of immersion mode ice nucleating particles in the Canadian Arctic marine boundary layer during the summer of 2014. Aerosol samples were collected at 28 locations on a MOSSI impactor and then analyzed using a droplet freezing technique to quantify the concentration of INPs as a function of temperature. The ratio of mineral dust to sea spray particle surface area was quantified using EDX for three samples. These surface area ratios were then converted to active site density ratios, which revealed that mineral dust was the dominant INP type in the considered samples. Finally, the authors use the FLEXPART particle dispersion model to correlate INP concentration data with air mass back trajectories. This analysis suggests the source of INPs in the arctic marine boundary layer are relatively local mineral or soil dusts.

General Comments

This study makes a substantial contribution to the field by presenting an impressive amount of immersion mode INP concentration data for the Arctic. The locality of the field work is very timely, and the additional analyses on particle composition and back trajectories nicely support the authors' main goal of quantifying immersion mode INP concentrations. The paper is well suited for publication in ACP. Below, I provide a few questions and comments to strengthen the paper and clarify its results.

Note on INP Nomenclature

[12] Please try to be clearer throughout the text that you considered the immersion freezing mode of ice nucleation. This is especially important given your results that mineral dust was an important source of particulate matter in your samples: mineral dust can activate in the deposition freezing mode at the temperatures considered here (e.g. Hoose and Mohler, 2012), but the mode of freezing investigated in this study isn't even included in the abstract.

[A12] We have clarified the mode of ice nucleation to be immersion freezing throughout the manuscript.

Specific Comments

[13] Page 2, Line 9: Please clarify here that sea spray particles actually vary widely in composition, depending on mechanism of formation. For instance, film rupture particles are organic enriched compared to jet droplet particles (e.g. Wang et al. 2017). This will highlight the importance of considering both microlayer and bulk seawater samples, as you suggest in the following sentence.

[A13] We have clarified that sea spray particles vary widely in composition depending on mechanism of formation. This has been done by amending the text to:

"Sea spray aerosol is generated by a bubble bursting mechanism at the ocean surface (Blanchard, 1964) and varies widely in composition, depending on the mechanism of formation. For example, particles from film rupture are enriched in organics compared to particles from jet droplets (Wang et al., 2017)."

[14] Page 2, Line 11: For completeness, the authors may also consider citing Huang et al. 2018 here. This paper follows up on Burrows et al. 2013 by discussing the sensitivity of the relative contribution of sea spray to INP concentrations at high latitudes to a model's choice of active site density parameterizations.

[A14] We have added the citation Huang et al. (2018) to the manuscript.

[15] Page 3, Line 15: Were the metrological data collected with on-ship sensors? If so, can some estimate of the height of these sensors above sea level be given?

[A15] The meteorological data was collected with on-ship sensors. We have added the height of the sensors used to measure RH, wind speed, and temperature to the manuscript.

[16] Page 4, Line 8: Has the transmission efficiency of the louvered total suspended particulate (TSP) inlet been quantified before? For example, it sounds similar to an inlet characterized by Kenny et al. 2005. If the exact loss percentages have not been measured, that's okay; but the authors should note that larger particles (like mineral dust) may be more prone to impaction and thus be undersampled on the MOSSI slides.

[A16] The transmission efficiency of the louvered total suspended particulate inlet has been quantified previously (Kenny et al., 2005). Based on this previous study, the transmission efficiency of 10 μm particles through the inlet is $\geq$ 90 %. To address the referee's comments, this information has been added to the revised manuscript.

[17] Page 6, Line 7: Was the height of the chemical ionization mass spectrometer inlet at a similar height to the inlet of the MOSSI?

[A17] We have added the height of the CIMS to the manuscript.

[18] Page 7, Line 17: The authors state that only particles at the edge of the spots were analyzed with EDX, but how are they sure these particles are representative of the bulk of particles directly below the micro-orifices? For example, the authors mention the particle rebound effect (Page 4, Line 12). Can the authors discuss if perhaps mineral dust is more prone to rebound than the deliquesced salty particles, and therefore may be more likely to end up at the edge of the spot where EDX was performed?

[A18] The particles on the edge of the spots were directly under the nozzles, meaning they were part of the spot. We assume the composition of the edge of the spot is the same as the whole spot; however, we are unable to confirm this assumption. In the revised manuscript we have clearly stated this assumption by adding the following:

"Since the edge was still directly under the nozzle, we assumed the composition of the edge of the spot is the same as the composition of the whole spot, although we are unable to confirm this assumption."

[19] Page 7, Line 22: "First, the atomic percentages of each particle were determined from EDX spectra." Can the authors clarify if only one discrete spot of each particle was examined, or if the EDX data represents an average signal from a raster scan?

[A19] To address the referee's comment we have clarified this point in the manuscript by adding the following sentence:

"The EDX data for an individual particle represents an average signal from a raster scan."

[20] Page 7, Line 23: Do the authors have any way to estimate what fraction of particles were internally mixed; i.e. mineral dust coated with sea salt?

[A20] See [A5] above.

[21] Page 8, Line 4: How were blanks prepared and treated? Were they fresh slides out of the package? Or were they treated the same way as the sampled slides, i.e. placed within the MOSSI only without turning on the pump? The latter would better account for contamination in the sample handling and preparation process; but in either case, more information is needed here.

[A21] We have included more information on how the blanks were prepared and treated in the manuscript.

[22] Page 8, Line 17: "The two samples corresponding to high [INP(T)] were collected on July 21st and 25th." As I read Figure 2, these are among the highest but not the highest INP concentration days. Can the authors provide any more reasoning behind why these particular days were chosen for compositional analysis? How typical was the meteorology on these days? Or perhaps they were chosen to be evenly spaced in time and location? Or was the choice random?

[A22] We analysed two days with higher INP concentrations, the 25th July and the 21st July. These two days were random choices. We have adjusted the text in section 3.2 to read:

"The two samples corresponding to high [INP(T)] were collected on July 21st and 25th, these days were chosen randomly out of the samples that showed freezing activity at -

20 °C and -25 °C . The sample corresponding to a low [INP(T)] was collected on July 29th, this day was chosen randomly out of the samples that did not display any freezing activity above -25 °C."

The 21st and 25th had 5 m/s and 3 m/s average wind speeds, respectively. The average wind speed during sampling was 5 m/s. The samples were not chosen because of the meteorology.

[23] Page 10, Line 14: The jump to biological particles is a bit of a non-sequitur. You might elaborate on how biological activity can influence sea spray particle composition, which in turn effects INP activity.

[A23] We have taken this statement out of the manuscript.

[24] Page 10, Conclusions: Can the authors discuss their findings that mineral dust immersion INPs dominated over sea spray INPs in the context of Irish et al. 2017, which found immersion mode INPs to be abundant in the seawater from this region? E.g. perhaps the INPs were present in the seawater but were never aerosolized because it wasn't windy enough to generated sea spray. This also ties in with Reviewer 1's comment that time spent over open ocean may not correlate with sea spray INP concentration if sea spray particles are not being formed. Then, include a discussion on whether these conditions are typical: how do your wind speeds compare with average (intra- and interannual) wind speeds in the region? Briefly delving into a reanalysis dataset (e.g. ECMWF ERA-Interim) might help you to explore this question in depth, or you could look for historical observational data. This will require a little bit more work on your end, but it would strengthen your analysis immensely by allowing you to hypothesize whether your findings are typical.

[A24] We have addressed the referee's comment by adding the following discussion in the manuscript:

"Previous studies have shown that INPs are ubiquitous in Arctic seawater (Irish et al., 2017; Wilson et al., 2015). Our results show that INPs in the Arctic seawater are not emitted at a high enough rate to compete with mineral dust, at least for the samples with high INP concentrations. The flux of sea spray aerosol to the atmosphere is a function of the wind speed. The average minute wind speed during sampling (5.5 m s^{-1} with 10th and 90th percentiles of 2.1 and 10.0 m s^{-1}) was similar to the average minute wind speed during the whole campaign (5.2 m s^{-1} with 10th and 90th percentiles of 1.5 and 9.3 m s^{-1}). In addition, our average wind speeds were similar to historical monthly average wind speed data from Alert, NU, Canada (3.6 m s^{-1} in July and 3.3 m s^{-1} in August; climate.weather.gc.ca; climate ID: 2400300). The influence of sea spray aerosol may be more important during periods of higher wind speeds. On the other hand, high wind speeds are also likely to increase the flux of mineral dust from local sources."

[25] Page 20, Table 1: Please include in the table caption the length of the back-trajectory (7 days).

[A25] The length of the back-trajectory has been included in the caption of Table 1.

[26] Page 21, Figure 1: Can you also denote in this figure where the samples for chemical/EDX analysis were collected? That will help us visualize the potential sources of these particles in light of the data in Figure 5. Otherwise, this figure looks great!

[A26] We have indicated which sampling locations correspond to samples used for CCSEM-EDX analysis.

[27] Page 23, Figure 3: Please also show the min/max INP concentration at each temperature (in addition to the average and standard deviation you've already shown) to provide more information on the degree of variability. Are the data from DeMott et al. 2015 averages, or discrete measurements?

[A27] We have clarified that the data from DeMott et al. (2016) are discrete measurements. The min/max INP concentrations at each temperature have also been added to the figure.

[28] Page 25, Figure 5: Please increase the font size in this figure to something legible.

[A28] The font size in Figure 5 has been increased.

[29] Supplement, Page 9, Figure S6: Please include units (L^{-1}) for INP concentrations either in the axis label or the figure caption.

[A29] (L^{-1}) has been added to the axis label in Figure S6 (now Figure S7).

References

Hoose, C. and Möhler, O., 2012. Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmospheric Chemistry and Physics, 12, 9817– 9854.
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Kenny, L., Beaumont, G., Gudmundsson, A., Thorpe, A. and Koch, W., 2005. Aspiration and sampling efficiencies of the TSP and louvered particulate matter inlets. Journal of Environmental Monitoring, 7(5), pp.481-487.
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Ice nucleating particles in the marine boundary layer in the Canadian Arctic during summer 2014

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Abstract. Ice nucleating particles (INPs) in the Arctic can influence climate and precipitation in the region; yet our understanding of the concentrations and sources of INPs in this region remain uncertain. In the following we 1) measured concentrations of INPs in the immersion mode in the Canadian Arctic marine boundary layer during summer 2014 on board the CCGS *Amundsen*, 2) determined ratios of surface areas of mineral dust aerosol to sea spray aerosol, and 3) investigated the source region of the INPs using particle dispersion modelling. Average concentrations of INPs at -15, -20 and -25 °C were 0.005, 0.044, and 0.154 L^{-1}, respectively. These concentrations fall within the range of INP concentrations measured in other marine environments. For the samples investigated the ratio of mineral dust surface area to sea spray surface area ranged from 0.03 to 0.09. Based on these ratios and the ice active surface site densities of mineral dust and sea spray aerosol determined in previous laboratory studies, our results suggest that mineral dust is a more important contributor to the INP population than sea spray aerosol for the samples analysed. Based on particle dispersion modelling, the highest concentrations of INPs were often associated with lower latitude source regions such as the Hudson Bay area, eastern Greenland, or northwestern continental Canada. On the other hand, the lowest concentrations were often associated with regions further north of the sampling sites and over Baffin Bay. A weak correlation was observed between INP concentrations and the time the air mass spent over bare land, and a weak negative correlation was observed between INP concentrations and the time the air mass spent over ice and open water. These combined results suggest that mineral dust from local sources is an important contributor to the INP population in the Canadian Arctic marine boundary layer during summer 2014.

Victoria Irish 2018-11-19 1:31 PM
Comment [1]: Addresses referee 2 [12]

1 Introduction

Ice nucleating particles (INPs) initiate the heterogeneous formation of ice in clouds at temperatures warmer than required for homogeneous freezing. INPs are important since they can change the frequency and properties of ice and mixed-phase clouds. The frequency and properties of clouds in the Arctic have been shown to be especially sensitive to concentrations of INPs, yet our understanding of the concentrations and sources of INPs in this region remains uncertain (Coluzza et al., 2017; Creamean et al., 2018; Harrington et al., 1999; Kanji et al., 2017; Korolev et al., 2017).

Examples of atmospherically relevant INPs include, but are not limited to, mineral dust particles and sea spray aerosol (DeMott et al., 2016; Després et al., 2012; Hoose and Möhler, 2012; Murray et al., 2012; Niemand et al., 2012; Szyrmer and Zawadzki, 1997). Sea spray aerosol is generated by a bubble bursting mechanism at the ocean surface (Blanchard, 1964) and varies widely in composition, depending on the mechanism of formation. For example, particles from film rupture are enriched in organics compared to particles from jet droplets (Wang et al., 2017). Recent work has shown that the sea surface microlayer and bulk seawater contain INPs (Irish et al., 2017; Rosinski et al., 1986; Schnell, 1977; Schnell and Vali, 1975, 1976; Wilson et al., 2015). Modelling studies have also suggested that sea spray aerosol can be a significant contributor to the INP population in marine environments when the source of other INPs is small (Burrows et al., 2013; Huang et al., 2018; Vergara-Temprado et al., 2017; Wilson et al., 2015). However our understanding of the flux of INPs from the ocean to the atmosphere is incomplete, and more studies are needed to understand when and where sea spray aerosol are a significant contributor to INP concentrations in the atmosphere.

Victoria Irish 2018-12-17 11:01 AM
Comment [2]: Addresses referee 2 [13]

Victoria Irish 2018-12-6 12:54 PM
Comment [3]: Addresses referee 2 [14]

Mineral dust is transported to the atmosphere by wind erosion, which is sensitive to factors like soil composition, soil moisture, and wind velocity (Ginoux et al., 2001). Mineral dust has been identified as an important contributor to the atmospheric INP population in many field and laboratory studies (Atkinson et al., 2013; Boose et al., 2016; Chen et al., 2018; Conen et al., 2011; Connolly et al., 2009; Creamean et al., 2013; DeMott et al., 2015; Eastwood et al., 2008; Hill et al., 2016; Kaufmann et al., 2016; Klein et al., 2010; Murray et al., 2012; Niedermeier et al., 2010; Niemand et al., 2012; O'Sullivan et al., 2014; Prenni et al., 2009a, 2009b; Rangel-Alvarado et al., 2015; Steinke et al., 2016; Wex et al., 2014; Wheeler et al., 2015). Modelling studies have also confirmed that mineral dust particles are important atmospheric INPs (Alizadeh-Choobari et al., 2015; Atkinson et al., 2013; Burrows et al., 2013; Hendricks et al., 2011; Hoose et al., 2010; Lohmann and Diehl, 2006; Prenni et al., 2009b; Vergara-Temprado et al., 2017).

When glaciers and permafrost in the Arctic melt erodible soil is exposed. The increased areas with erodible soil can be a potential source of mineral dust in the Arctic (Huang et al., 2015). The ice nucleation properties of dry mineral dust from Thule, Greenland were measured by Fenn and Weickmann (1959) and they found it could nucleate ice at temperatures as warm as -5 °C. Groot Zwaaftink et al. (2016) also suggested through a modelling study that during the summer local mineral dust sources in the Arctic (mineral dust from latitudes north of 60 ° N) can dominate the total mineral dust concentrations at the surface. However, previous studies have not yet shown that mineral dust from regional erodible soil could be a major source of INPs in the atmosphere in the Arctic.

In the past 30 years, warming in the Arctic has decreased sea ice and land snow by approximately 20 % and 13 %, respectively (Derksen and Brown, 2012). This may have led to an increase in sea spray particles and mineral dust particles from local sources in the region and, as a result, an increase in INPs. Because of the continuing warming trend in this region, the concentration of INPs from these local sources may continue to increase with important implications for the frequency and properties of ice and mixed-phase clouds as well as climate in the region. To evaluate the scale of this feedback mechanism, studies are needed to determine the concentrations and sources of INPs in the Arctic.

To help address the issues raised above we: 1) determined concentrations of INPs in the immersion mode in the Canadian Arctic marine boundary layer during summer 2014, 2) measured the ratio of surface areas of mineral dust particles to sea spray particles, and 3) investigated the source region of the INPs using a particle dispersion model. The specific goals of this study were to quantify the concentrations of INPs in the Canadian Arctic marine boundary layer and to provide insights into the source of INPs in this region during the summer.

Victoria Irish 2018-11-19 1:31 PM
Comment [4]: Addresses referee 2 [12]

2 Experimental

2.1 Sampling locations

All measurements and sample collections were performed on board the CCGS *Amundsen* as part of the NETwork on Climate and Aerosols; addressing key uncertainties in Remote Canadian Environments (NETCARE). The 28 sampling locations are shown in Fig. 1, and the sampling dates, times, and coordinates are detailed in Table S1. The air temperature, relative humidity, and wind speed during sampling were measured with on-ship sensors at approximately 15 m above sea level and are shown in Fig. S1. INP concentrations from a subset of the locations (indicated with blue rings around the red symbols in Fig. 1) were previously reported in DeMott et al. (2016) but are also included here as they were collected during the same expedition and with the same methodology. The data reported in DeMott et al. (2016) only included sites in Baffin Bay, days where it did not rain and conditions when the samples were assumed to have reduced exposure to ship smokestack contamination, i.e. when the apparent wind direction measured on the ship was between 0-90 degrees or 270-360 degrees, where 0/360 corresponds to the bow of ship. (The apparent wind direction is defined as the wind direction experienced by an observer on the moving ship as opposed to the true wind direction, which is defined as the wind direction experienced by an observer when the ship is stationary.)

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2.2 Quantifications of INPs

To determine the concentration of INPs in the immersion mode, atmospheric particles were collected on hydrophobic glass slides using a micro-orifice single stage impactor (MOSSI; MSP corp., Shoreview, MN, USA). The freezing properties of the collected particles were then determined with the droplet freezing technique (DFT). The combination (MOSSI-DFT) is similar to the micro-orifice uniform deposit impactor droplet freezing technique (MOUDI-DFT) recently used to determine the size resolved concentrations of INPs (Mason et al., 2015b, 2015a, 2016). The main

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difference between the MOSSI-DFT technique and the MOUDI-DFT technique is the use of a single stage impactor compared to a multistage impactor. The use of a single stage impactor simplifies the analysis and reduces collection time but sacrifices size information. The MOSSI-DFT technique is also similar to the technique used by others to measure deposition freezing (Knopf et al., 2010, 2014, Wang et al., 2012b, 2012a).

2.2.1 Micro-orifice single stage impactor

The MOSSI was located on the port side of the bridge on the ship, approximately 10 m in front of the ship's smokestack. During sampling the flow rate through the MOSSI was 10 L min^{-1}, resulting in particles with aerodynamic diameters > 0.18 μm being collected on the hydrophobic glass slides placed within the MOSSI. The collection time of samples with the MOSSI for INP analysis was approximately 20 minutes. The MOSSI sampled air through a louvered total suspended particulate (TSP) inlet, which was approximately 15 m above sea level. The transmission efficiency of 10 μm particles through the TSP inlet is ≥ 90 % (Kenny et al., 2005). The nozzle plate within the MOSSI contained 300 micro-orifices. As a result, particles collected on the hydrophobic glass slides beneath the nozzle plate were concentrated into 300 spots. After collection the hydrophobic glass slides containing the particles were stored at 4 °C for no longer than 3 months prior to analysis.

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Particle rebound can be an issue with an inertial impactor such as the MOSSI. Particle rebound occurs when particles impact the collection substrate but are not retained. Rebound has been shown to be reduced at RH values above 70 %, although this depends on the chemical composition of the particles (Bateman et al., 2013; Chen et al., 2011; Fang et al., 1991; Lawson, 1980; Saukko et al., 2012; Vasiliou et al., 1999; Winkler, 1974). During collection with the MOSSI the RH was always well above 70 % (Figure S1) with an average of 93 %. Furthermore, field measurements of INP concentrations using the MOUDI-DFT (a method similar to the MOSSI-DFT) have shown good agreement with INP concentrations measured with an instrument that is not susceptible to particle rebound (a continuous flow diffusion chamber) when the RH of the sampled aerosol was as low as 40 % (DeMott et al., 2017; Mason et al., 2016). Nevertheless, particle rebound cannot be ruled out, and therefore, the INP concentrations reported here should be considered as lower limits.

2.2.2 Droplet freezing technique

The droplet freezing technique (Koop et al., 1998; Mason et al., 2015b) was used to determine the concentration of INPs in the immersion mode collected on hydrophobic glass slides using the MOSSI. Briefly, the hydrophobic glass slides containing the collected particles were located in a temperature and relative humidity controlled flow cell, coupled to an optical microscope with a charged-coupled device (CCD) camera and a 1.25x objective (Axiolab; Zeiss, Oberkochen, Germany). Typically between 15-25 spots of particles (out of the 300 spots generated by the micro-orifices in the nozzle plate) could be monitored in the CCD field of view, which was approximately 12.25 mm^2 in area. Water was then condensed on the hydrophobic glass slides by decreasing the temperature to 2 °C and flowing a gas (pure Helium) with a dew point of greater than 2 °C over the hydrophobic glass slides. This resulted in water droplets (with diameters between 100 to 500 μm)

condensing on the spots (referred to here as spot droplets) as well as water droplets condensing on other areas of the slides (referred to here as non-spot droplets). After the droplets were condensed, the temperature of the flow cell was decreased at a rate of 10 °C min^{-1}. From videos recorded while the temperature was decreased, the freezing temperature of each droplet was manually determined by observing the change in the droplet's optical properties. The droplets that contained spots of deposited particles were also identified from these videos. For comparison purposes, hydrophobic glass slides that were not exposed to atmospheric particles were also processed in the same way as described above and labelled as blanks.

The number of INPs as a function of temperature, *#INP(T)*, was calculated for each experiment using the following equation:

$$\#INP(T) = \left(-\ln\left(\frac{N_u(T)}{N_s} \right) N_s \right) \quad (1)$$

Where N_u is the number of unfrozen droplets covering the spots, and N_s is the number of spots in the field of view. Equation 1 accounts for the possibility that each droplet covering a spot can contain multiple INPs (Vali, 1971).

Equation 1 assumes that each spot was covered by only one droplet. For cases when more than one droplet formed on a spot, the first droplet that froze was considered in Eq. 1. This was expected to give an equivalent result to the case of only one droplet condensing on the spot. For cases when one droplet contained two spots (this occurred for 2 % of the total number of spots in all experiments), an upper limit to the number of INPs was calculated by assuming two droplets covered the two spots and both droplets froze at the observed freezing temperature. A lower limit was calculated by assuming the two spots were covered by two droplets with one droplet freezing at the observed freezing temperature, and the other droplet freezing at -37 °C (approximately the homogeneous freezing temperature). If one droplet contained 3 or more spots a similar procedure to the above was used to calculate the upper and lower limits to *#INP(T)*.

Freezing of droplets that did not cover spots was a relatively rare occurrence at the temperature range we focused on in this manuscript (≥ -25 °C; see Section 3.1). For example the ratio of frozen non-spot droplets to frozen spot droplets was 0.02 and 0.07 at -25 °C and -20 °C, respectively. We assumed these relatively rare occurrences were due to particles < 0.18 μm in diameter that were not focused into spots or due to rebound of a small fraction of the particles off the hydrophobic glass slides. To take into account the INPs not concentrated into the spots, we added the number of frozen non-spot droplets at each temperature to Eq. 1. Since the freezing of non-spot droplets was a relatively rare occurrence, we did not apply the Vali correction (Vali, 1971) to these freezing events.

Approximately 2 % of the freezing events in our experiments occurred by contact freezing between -16.2 and -34.8 °C. Contact freezing occurred when a frozen droplet grew in size, due to the Wegener-Bergeron-Findeisen process (Findeisen, 1938), and caused the freezing of a neighbouring unfrozen droplet. When calculating concentrations of INPs, contact freezing events were excluded.

The atmospheric concentration of INPs as a function of temperature, *[INP(T)]*, was calculated with the following equation:

$$[INP(T)] = \#INP(T).\frac{300}{N_s}.\frac{1}{V} \quad (2)$$

Where V is the volume of air sampled, and the ratio of $300/N_s$ takes into account that only a fraction of the total number of spots in the sample were observed in a freezing experiment.

2.3 Effect of ship emissions on measured INP

To determine if particles from the ship's smokestack affected the measured INP concentrations, we first investigated the relationship between INP concentrations measured on the ship and the gas-phase HONO concentrations, a product of the reaction between NO_x from the ship smokestack and water (Von Glasow et al., 2003). HONO was measured by a chemical ionisation mass spectrometer located on the bridge of the ship about 5 m in front of the smokestack and approximately 15 m above sea level. No correlation was observed between HONO and INP concentrations at -25 °C ($R = 0.05$, $p = 0.403$).

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Second we separated our INP results into samples that were not exposed to smokestack emissions based on measured wind direction and wind speed, and samples that may have been exposed to smokestack emissions based on measured wind direction and wind speed. When the apparent wind direction measured on the ship was between 0-90° and 270-360° (where 0°/360° = bow of ship) and when the apparent wind speed (minute average) was higher than 2.5 m s^{-1} for the entire collection time, we assumed that the samples were not exposed to smokestack emissions. Within the uncertainty of the measurements, the INP concentrations measured when samples were not exposed to smokestack emissions (based on the apparent wind direction and speed) are the same as INP concentrations measured when samples may have been exposed to smokestack emissions (Fig. S2). Since the criteria discussed above and the results from the HONO analysis do not suggest measured INP concentrations were influenced by the smoke stack emissions, all samples that were collected have been included in this study.

2.4 Particle dispersion modelling

FLEXPART-WRF (Brioude et al., 2013), a version of the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005), was used to investigate the potential emission source regions of the INPs. FLEXPART-WRF is driven by meteorology from the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008), and was run in backward mode. The simulation domain for FLEXPART-WRF is shown in Fig. S3.

WRF 3.5.1 was run for the 2014 Amundsen campaign using initial and boundary conditions from the European Centre for Medium-Range Weather Forecasts (ECMWF) operational analysis (a grid resolution on 0.25°). The ECMWF

wind, temperature, and RH were used to nudge the WRF run every 6 hours above the atmospheric boundary layer. A full list of the parameterizations and options used for the WRF simulations is given in Table 1 of Wentworth et al. (2016).

FLEXPART-WRF was run in backward mode at 20 minute intervals along the ship track. At the start of a simulation 100,000 particles were released from the ship's location in an initial box of volume of 100 x 100 m in the horizontal, and from 0 to 60 m above mean sea level in the vertical. After the particles were released from the initial box, they were followed backward for seven days with output generated hourly.

As mentioned above, each INP sample was collected over a period of approximately 20 minutes. As a result, one FLEXPART-WRF run overlapped with each INP sampling period. The FLEXPART-WRF runs that overlapped in time with the INP sampling periods were used to produce potential emission sensitivity (PES) plots for the INP samples. A PES plot was produced by integrating the FLEXPART-WRF output over the 7 days prior to the release of particles. The value of the PES in a particular grid cell is proportional to the particles residence time in that cell. Since this study is concerned with INP sources from the surface, only particles within the footprint layer (0 to 300 m altitude) are considered when calculating PES values, and we report PES values for the footprint layer.

2.5 Statistical analyses

Pearson correlation analysis was used to compute correlation coefficients (R). P values were also calculated to determine if the correlations were statistically significant at the 95 % confidence level ($p < 0.05$).

2.6 Computer controlled scanning electron microscopy with energy dispersive X-ray spectroscopy (CCSEM-EDX)

Immediately following the collection of each sample for INP analysis, additional particle samples were collected to determine the ratio of mineral dust surface area to sea salt surface area by computer controlled scanning electron microscopy with energy dispersion X-ray spectroscopy (CCSEM-EDX). Particles were collected on transmission electron microscopy (TEM) grids (carbon 200 mesh; Ted Pella) using the same MOSSI used to collect INP samples. Collection time of samples for CCSEM-EDX was approximately 20 minutes. The samples were kept at room temperature for a maximum of 38 months before analysis. Since a long collection time was used (20 minutes), particles in the spots directly below the micro-orifices of the nozzle plate in the MOSSI were too close together to identify individual mineral dust and sea salt particles using CCSEM-EDX. To overcome this issue, we only analysed particles on the edge of the spots directly below the micro-orifices of the nozzle plate with CCSEM-EDX. Since the edge was still directly under the nozzle, we assumed the composition of the edge of the spot is the same as the composition of the whole spot, although we are unable to confirm this assumption.

Due to time constraints, only three samples (two with a high [INP(T)] and one with a low [INP(T)]) were analysed by CCSEM-EDX. The method of using CCSEM-EDX to study atmospheric particles is described by Laskin et al. (2006). Particles with sizes between 0.15 to 5 µm (area equivalent diameters) were analysed. First, the atomic percentages of each particle were determined quantitatively from EDX spectra. The EDX data for an individual particle represents an average signal from a raster scan. Then, based on the atomic percentages of each particle, particles were classified as sea salt, mineral

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dust, or other using the scheme shown in Fig. S4, which is based on the work by Laskin et al. (2012). This classification scheme does not include a mixed mineral dust/sea salt particle category. Rather, any particles that have both sea salt and mineral dust are classified as either mineral dust or sea salt depending on the largest atomic percentage contribution. If we used the classification scheme by Derimian et al. (2017), which explicitly classifies particles as sea salt, mineral dust, and mixed sea salt/mineral dust, no particles would be classified as mixed sea salt/mineral dust particles.

After each particle was classified, the surface areas of mineral dust particles and sea salt particles were determined using 2D projected images recorded by SEM. Note that the actual surface area of mineral dust is underestimated using this method due to surface irregularities and complex topology. The ratio of mineral dust surface area to sea salt surface area was then determined by dividing the surface area of mineral dust for each sample by the surface area of sea salt for the same sample.

3 Results and discussion

3.1 Measured INP concentrations

The measured concentrations of INPs in the immersion mode, *[INP(T)]*, sampled in the Arctic are shown in Figs. 2b and 2c. The measured *[INP(T)]* on new hydrophobic glass slides taken straight from the package, cleaned with ultra pure water, but not exposed to atmospheric particles (referred to as "blanks") are shown in red in Figs. 2a and 2c. Freezing of the blanks occurred over the range of -25.9 °C to -38.4 °C. For the droplet sizes and cooling rates used here homogeneous freezing occurs at approximately -37 °C. Therefore the freezing that occurred in the blanks at temperatures above approximately -37 °C was due to heterogeneous freezing likely caused by the hydrophobic glass slides. In the following we will focus on *[INP(T)]* at temperatures of -25 °C and warmer since no freezing from the blanks was observed in this temperature range. A full time series of *[INP(T)]* at -15 °C, -20 °C, and -25 °C are reported in Fig S5.

In Fig. 3 we compare recent measurements of *[INP(T)]* from several field campaigns in marine environments with the average concentrations measured in the current study. Figure 3 illustrates that the average INP concentrations measured in the current study fall within the range of INP concentrations measured in other marine environments. This observation, however, does not confirm that sea spray aerosol was the major source of INPs during the studies reported here.

3.2 Measured ratios of mineral dust surface area to sea salt surface area

For three samples (two with high *[INP(T)]* and one with low *[INP(T)]*), we calculated the ratios of mineral dust surface area to sea salt surface area using CCSEM-EDX. The two samples corresponding to high *[INP(T)]* were collected on July 21st and 25th, these days were chosen randomly out of the samples that showed freezing activity at -20 °C and -25 °C. The sample corresponding to a low *[INP(T)]* was collected on July 29th, this day was chosen randomly out of the samples that did not display any freezing activity above -25 °C. In Table S2 we report the total number of particles analysed by CCSEM-EDX for each sample, the fraction of particles classified as mineral dust and sea salt particles, and the surface area

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corresponding to mineral dust and sea salt particles. Shown in Fig. 4a are the calculated ratios of mineral dust surface area to sea salt surface area using surface area measurements from CCSEM-EDX. For the three samples, this ratio ranged from 0.03 to 0.09. Using this ratio we estimated the ratio of *[INP(T)]* from mineral dust, $[INP(T)]_{MD}$, to *[INP(T)]* from sea spray, $[INP(T)]_{SS}$, using the following equation:

$$\frac{[INP(T)]_{MD}}{[INP(T)]_{SS}} = \frac{n_s(MD).S_{MD}}{n_s(SS).S_{SS}} \quad (3)$$

Where $n_s(SS)$ is the ice active surface site density for sea spray aerosol, $n_s(MD)$ is the ice active surface site density for mineral dust, and S_{SS} and S_{MD} are the total surface areas measured by CCSEM-EDX for sea salt and mineral dust, respectively. The $n_s(SS)$-values were determined using laboratory data from DeMott et al. (2016). Recent studies show that $n_s(SS)$-values determined from DeMott et al. (2016) are consistent with values determined in pristine marine environments (McCluskey et al., 2018). The $n_s(MD)$-values were calculated using the exponential function reported by Niemand et al. (2012) that was determined from freezing data of Asian dust, Saharan dust, Canary Island dust, and Israel dust. For details see Section S1.

The ratios of INP concentrations based on Eq. 3 for freezing temperatures of -25, -20, and -15 °C, are shown in Figs. 4b, 4c, and 4d respectively. These ratios suggest that for the three samples when CCSEM-EDX measurements were performed, the $[INP(T)]_{MD}$ are higher than the $[INP(T)]_{SS}$ (ratios were between 10 to 10^3, inclusive of errors, at -15, -20 and -25 °C), assuming the n_s-values used are applicable for the field studies reported here. These results also suggest that mineral dust is a more important contributor to the INP population than sea spray aerosol for the times and locations corresponding to the CCSEM-EDX measurements.

Above we assumed that the ice active surface site density for mineral dust, $n_s(MD)$, in our studies can be calculated with the exponential function reported by Niemand et al. (2012) that was determined from freezing data of Asian dust, Saharan dust, Canary Island dust, and Israel dust. To test this assumption, we calculated $n_s(MD)$-values using the CCSEM-EDX measurements on July 21st and 25th. Details on how $n_s(MD)$-values were calculated is given in Section S2, and the results are shown in Fig. S6. In short, within the uncertainty of the measurements, our calculated $n_s(MD)$-values are consistent with the results from Niemand et al. (2012).

3.3 Particle dispersion modelling

Figure 5a shows the averaged PES for the footprint layer for all samples combined and suggests that the source of INPs sampled during the campaign was local (i.e. the Canadian Arctic Archipelago, Baffin Bay, and eastern Greenland).

Figure 5b shows the averaged PES for the footprint layer for samples that had the highest INP concentrations (top 36 % of the samples) at -25 °C. Figure 5c shows the average PES for the footprint layer for samples that had the lowest INP concentrations (bottom 36 % of the samples) at -25 °C. A cut-off of 36 % was selected since no freezing was observed in 36 % of the samples at -25 °C. Figure 5d shows the ratio of the average PES for the highest INP concentrations to the average

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PES for all samples. Figure 5e shows the ratio of the average PES for the lowest INP concentrations to the average PES for all samples. Previous work has shown that ratios of average PES are useful for identifying likely sources (Hirdman et al., 2010). Considering all figures together, the highest INP concentrations are associated with lower latitude regions such as the Hudson Bay area, eastern Greenland, or northwestern continental Canada. On the other hand, the lowest concentrations (below the detection limit at -25 °C) were often associated with regions further north and over Baffin Bay.

Figures 5f and 5g show maps of surface cover type (i.e. bare land, open water, sea ice, and snow cover) from the first and last days of the campaign, respectively, based on data from the National Snow and Ice Data Center (NSIDC, 2008). The maps of surface coverage were combined with the PES values in the footprint layer from FLEXPART to determine the total residence time over each surface type for a given INP sample. Specifically, surface coverage data from the NSIDC was downloaded in GEOtiff format and converted to vector shapefiles. The fraction of each FLEXPART grid cell that was over each surface type category (e.g. bare land, open water) was then calculated using these vector shapefiles. The residence time in a grid cell was then multiplied by the fraction of the cell in each surface type category, and then the results were summed over all grid cells to determine the relative time spent over each surface type.

Correlations between the total residence time over each surface type and the concentration of INPs for each sample at -15, -20, and -25 ºC were then investigated (Table 1 and Fig S7). This correlation analysis showed statistically significant ($p < 0.05$) positive correlations between the total residence time over bare land in the footprint layer, and both *[INP(T)]* at -15 °C ($R = 0.5$) and -25 °C ($R = 0.4$). On the other hand, a statistically significant negative correlation was observed between the total residence time in the footprint layer over sea ice and both *[INP(T)]* at -20 °C ($R = -0.4$) and -25 °C ($R = -0.3$). Furthermore, a statistically significant negative correlation was observed between the total residence time in the footprint layer over open water and *[INP(T)]* at -20 °C ($R = -0.4$). These negative correlations can be explained by a stronger source of INPs from bare land compared to sea ice or open water. Related, Bigg (1996) observed a correlation between INP concentrations measured in the high Arctic and the time since the sampled air mass was last in contact with open ocean ($R = -0.54$, $p < 10^{-4}$). In contrast Bigg and Leck (2001) observed no correlation between INP concentrations and the time since the sampled air mass was last in contact with open ocean.

4 Conclusions

Concentrations of INPs in the marine boundary layer were measured at 28 different locations in the Canadian Arctic. Results showed that the concentrations of INPs are similar to concentrations measured in other marine environments.

For three collected samples the ratio of mineral dust surface area to sea spray surface area ranged from 0.03 to 0.09. Based on these ratios, and the ice active surface site densities of mineral dust and sea spray aerosol determined in previous laboratory studies, mineral dust is a more important contributor to the INP population than sea spray aerosol for the samples analysed (ratios were between 10 to 10^3, inclusive of errors, at -15 °C, -20 °C and -25 °C). This result suggests that INPs

from mineral dust are more important contributors to the INP population than sea spray aerosol for the times and locations during sampling.

Particle dispersion modelling suggested that the INPs sampled in this study were likely not from long-range transport. For the days when *[INP(T)]* was high, a likely source was the Hudson Bay area, eastern Greenland, or north-western continental Canada. For days when *[INP(T)]* was low, the air mass spent more time over regions further north and over Baffin Bay. Correlation analyses showed that there were statistically significant positive correlations between *[INP(T)]* at -15 and -25 °C and the time the air mass spent over land. Statistically significant negative correlations were observed between *[INP(T)]* at -20 and -25 °C and the time the air mass spent over sea ice, and *[INP(T)]* at -20 °C and the time the air mass spent over open water. This correlation analysis together with the particle dispersion modelling provides further evidence that mineral dust are a more important contributor to the INP population than sea spray aerosol, at least when INP concentrations were high. Sea spray aerosol may still have played a role when the INP concentrations were low during sampling.

Previous studies have shown that INPs are ubiquitous in Arctic seawater (Irish et al., 2017; Wilson et al., 2015). Our results show that INPs in the Arctic seawater are not emitted at a high enough rate to compete with mineral dust, at least for the samples with high INP concentrations. The flux of sea spray aerosol to the atmosphere is a function of the wind speed. The minute average wind speed during sampling (5.5 m s^{-1} with 10^{th} and 90^{th} percentiles of 2.1 and 10.0 m s^{-1}) was similar to the minute average wind speed during the whole campaign (5.2 m s^{-1} with 10^{th} and 90^{th} percentiles of 1.5 and 9.3 m s^{-1}). In addition, our average wind speeds were similar to historical monthly average wind speed data from Alert, NU, Canada (3.6 m s^{-1} in July and 3.3 m s^{-1} in August; climate.weather.gc.ca; climate ID: 2400300). The influence of sea spray aerosol may be more important during periods of higher wind speeds. On the other hand, high wind speeds are also likely to increase the flux of mineral dust from local sources. As warming increases in the Arctic, more erodible soil will be exposed for longer periods of time (Huang et al., 2015). These results, together with our freezing results suggest that warming in the Arctic will increase concentrations of INPs from mineral dust in the region, with possible implications for cloud properties and climate. Additional studies, including modelling and field studies are needed to quantify the importance of this feedback process for climate in the region.

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Acknowledgements

We would like to thank the scientists, officers, and crew of the CCGS *Amundsen* for support during the expedition in 2014. We acknowledge the use of data products or imagery from the Land, Atmosphere Near real-time Capability for EOS (LANCE) system operated by the NASA/GSFC/Earth Science Data and Information System (ESDIS) with funding provided by NASA/HQ.

The CCSEM-EDX analysis was performed at Environmental Molecular Sciences Laboratory at PNNL (Ringgold ID 47937 and 48781), a DOE Office of Science User Facility sponsored by the Office of Biological and Environmental Research. PNNL is operated for DOE by Battelle Memorial Institute under Contract DEAC06-76RL0 1830. We would also like to thank the Natural Sciences and Engineering Research Council of Canada and Fisheries and Oceans Canada for funding.

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	-15 °C			-20 °C			-25 °C		
	R	**p**	**n**	**R**	**p**	**n**	**R**	**p**	**n**
Bare land residence time	**0.5**	**0.008**	**27**	0.3	0.058	27	**0.4**	**0.033**	**27**
Open water residence time	-0.1	0.240	27	**-0.4**	**0.023**	**27**	-0.3	0.054	27
Sea ice residence time	-0.2	0.180	27	**-0.4**	**0.028**	**27**	**-0.3**	**0.041**	**27**
Snow cover residence time	-0.2	0.162	27	0.2	0.183	27	0.0	0.449	27

Table 1: Correlation coefficients (R), p and n values for correlation analysis between *[INP(T)]* (L^{-1}) at -15, -20, and -25 °C and the time the air mass spent in the previous 7 days over different surface types within 0-300 m of the surface. Numbers in bold indicate correlations that are statistically significant ($p < 0.05$).

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Comment [23]: Addresses referee 2 [25]

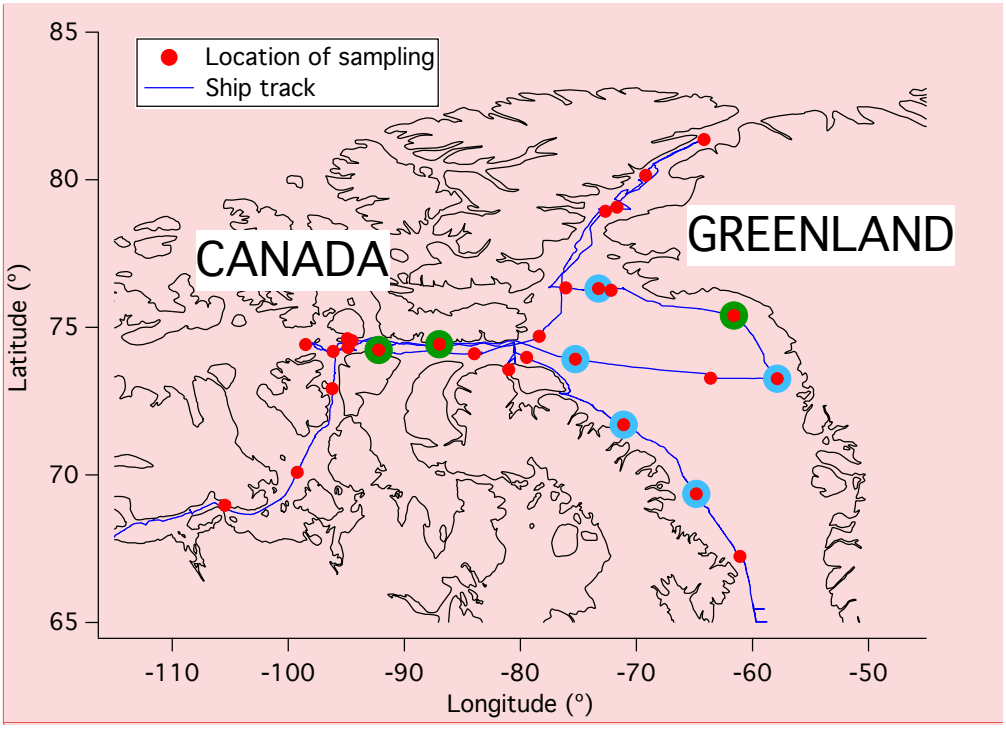

Figure 1: Locations of sampling. Blue circles around the red dots indicate the locations of samples used in DeMott et al. (2016). Green circles around the red dots indicate the locations of samples used for CCSEM-EDX analysis. Information on specific geographical coordinates is given in Supplemental Table S1.

Victoria Irish 2018-11-19 2:42 PM
Comment [24]: Addresses referee 2 [26]

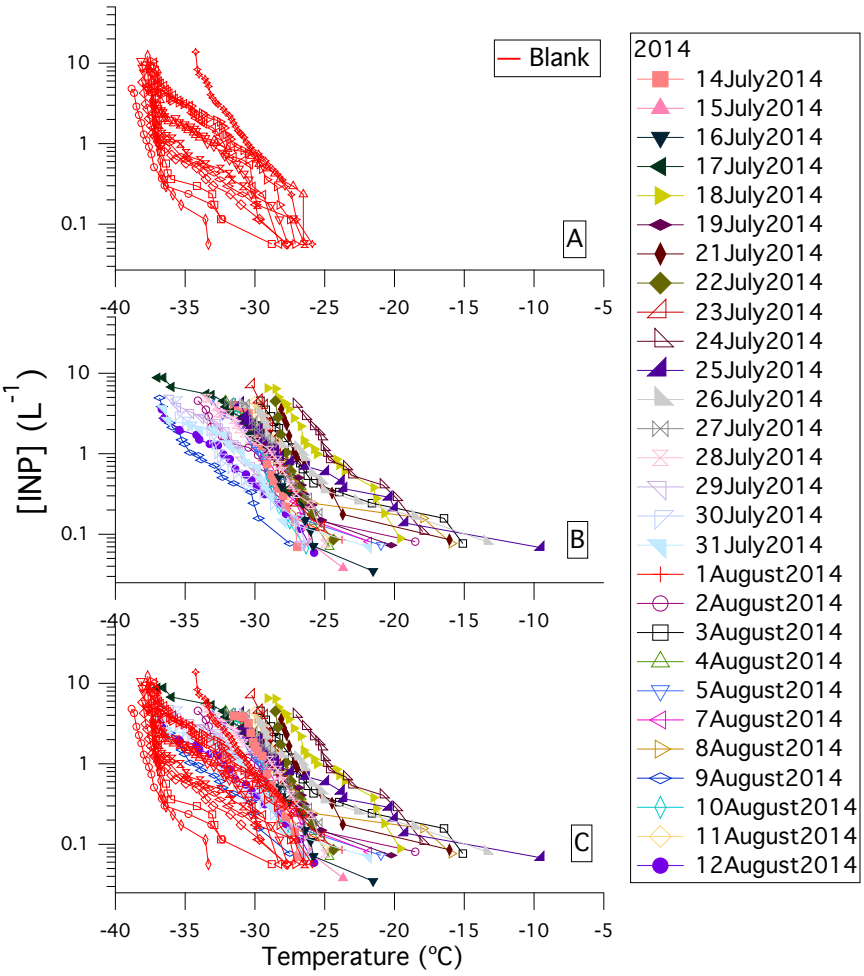

Figure 2: Plot of *[INP]* (L^{-1}) as a function of temperature (°C) for A) the blanks, B) the samples, and C) the blanks and samples. The *[INP]* (L^{-1}) of 11 blanks are shown in Panels A and C. Each blank was performed on a separate hydrophobic glass substrate. Error bars are not shown to improve the visibility of the data in the plot. Error bars in the x direction are ± 0.3 °C for each data point. Error bars in the y direction were calculated using nucleation statistics following Koop et al. (1997); the errors for our measured *[INP]* (L^{-1}) can be seen in Figure S5.

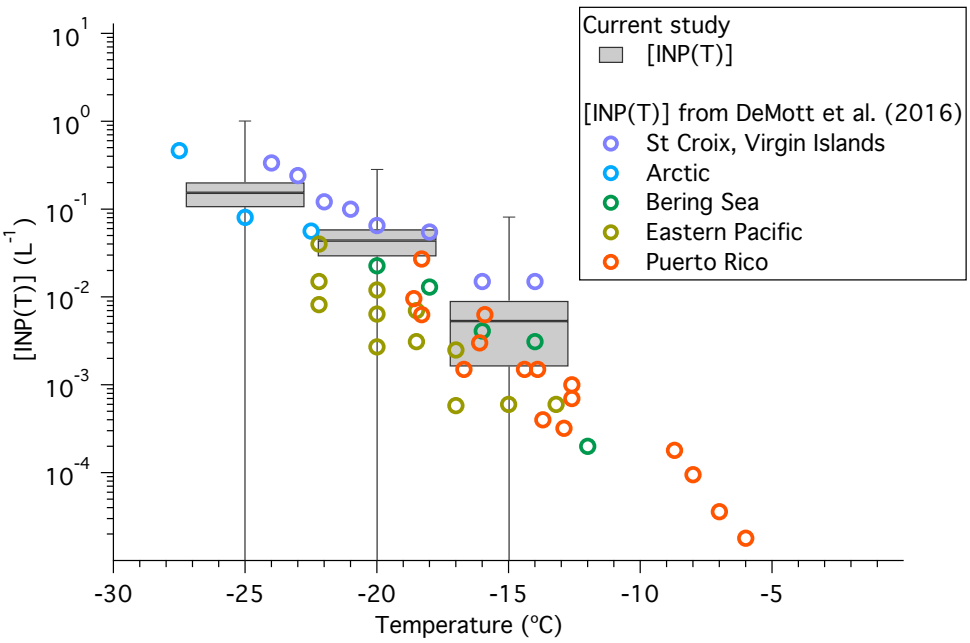

Figure 3: Plot of *[INP(T)]* (L^{-1}) as a function of temperature including a comparison to discrete field measurements from several recent field studies in marine environments reported in DeMott et al. (2016). The grey boxes represent the average *[INP(T)]* reported in this study as well as the standard error of the mean at -15, -20, and -25 °C. The whiskers represent the minima and maxima values of *[INP(T)]* measured in the current study. Note the minimum *[INP(T)]* measured for all temperatures in the current study was 0. Other coloured circles represent INP measurements from field studies in marine environments reported in DeMott et al. 2016, with the locations indicated in the legend.

Victoria Irish 2018-12-18 10:52 AM
Comment [25]: Addresses referee 2 [27]

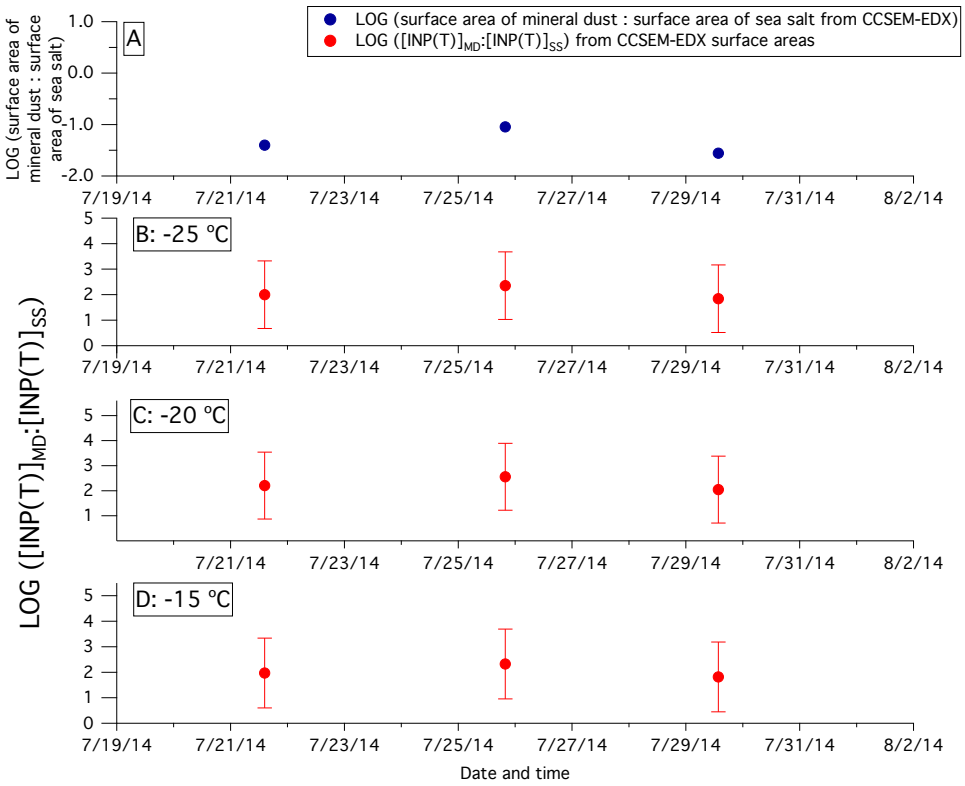

Figure 4: A) Ratios of the surface area of mineral dust particles to the surface area of sea salt particles measured by CCSEM-EDX (blue circles). Ratios of predicted $[INP(T)]_{MD}$ to the predicted $[INP(T)]_{SS}$ calculated using CCSEM-EDX measurements (red circles) at B) -25 °C, C) -20 °C, and D) -15 °C.

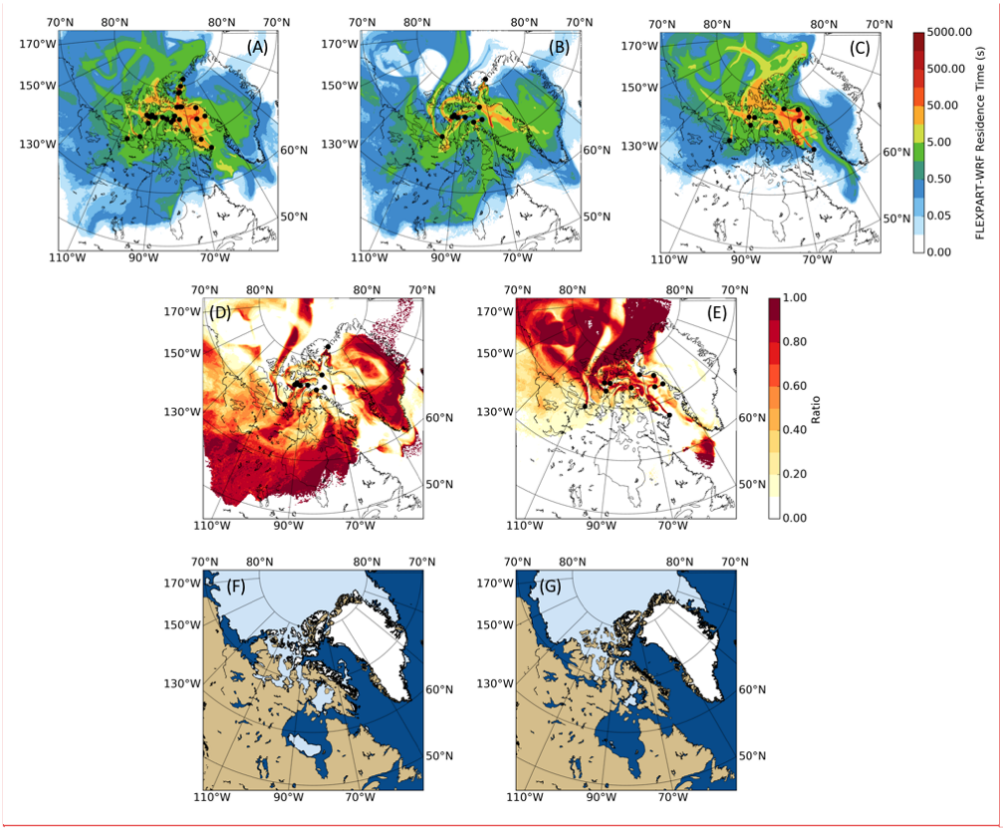

Figure 5: Top row - Average FLEXPART-WRF footprint potential emission sensitivities (PES) plots for A) all sampling days, B) the 36 % of samples with the highest *[INP(T)]* (L^{-1}), and C) the 36 % of samples with the lowest *[INP(T)]* (L^{-1}). Black circles indicate the ship's position at the sampling mid-time. Middle row - Maps showing the ratios of D) plot B to plot A, and E) plot C to plot A. Bottom row - Maps showing the surface cover type on F) the first day of sampling (14th July), and G) the last day of sampling (12th August). Bare land, open water, sea ice, and snow cover are shown as beige, dark blue, light blue, and white, respectively. Note that for this study, lakes are included in the bare land category.

Victoria Irish 2018-11-19 2:55 PM
Comment [26]: Addresses Reviewer 2 [28]