Prof. Paul Zieger
Co-Editor of Atmospheric Chemistry and Physics

Dear Paul,

Listed below are our responses to the comments from the referees 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]*). Author's responses are offset in blue below each referee statement with matching numbers (e.g. *[A1]*). We thank the referees for carefully reading our manuscript and for their helpful comments!

Sincerely,

Allan Bertram,
Professor, Department of Chemistry
University of British Columbia

Interactive comment on **"Ice-nucleating efficiency of aerosol particles and possible sources at three coastal marine sites"** ***by*** **Meng Si et al.**

Anonymous Referee #1

Received and published: 2 April 2018

Review of "Ice-nucleating efficiency of aerosol particles and possible sources at three coastal marine sites" by Si et al., submitted to ACPD:

The study described in this manuscript is an interesting addition to similar work by the group in which M. Si is working. It interprets measurements of atmospheric aerosol wrt. concentrations of ice nucleation particles (INP) and their possible sources, as well as effects of particle size on INP activity. The work is interesting and timely. However, I have a few major comments (besides for a number of smaller ones) that need to be addressed before the work can be published. The major comments mainly concern the amount of data (which is rather low) and a possible malfunctioning of one of the size spectrometers used and related consequences on the results.

But altogether, the study merits publication once again my comments will have been considered and changes will have been implemented adequately.

Major comments:

[1] The first comment concerns the amount of data used for the study. For two locations, only one measurement was made, while for the third location, the data used has already been used in a different study on INP before. This is properly stated in the text. And the results obtained herein evaluate these data in a new way, yielding more results. But the abstract had raised high expectations, and I was quite disappointed when I realized that the abundance of data included in this study is rather low. It should be made clear already in the abstract and again in the conclusions that the data base is not very strong. This might also influence the results, as one measurement does not deliver good statistics, and this has to be dealt with offensively and should be discussed.

> ***[A1]*** To address the referee's comment, in the Abstract and Summary and conclusions we have added the following sentence:
>
> "For Amphitrite Point, 23 sets of samples were analyzed, and for Labrador Sea and Lancaster Sound, one set of samples was analyzed for each location".
>
> In addition, we have added the following sentence in the Summary and conclusions:
>
> "Since only one sample was analyzed for both Labrador Sea and Lancaster Sound, additional samples should be collected and analyzed at these locations to determine the general applicability of the results presented here for these locations".

[2] I am also concerned about the misfit in the particle number size distributions between those number concentrations measured by the SMPS and by the APS. There is a large gap at ~ 500nm which implies that one of the two instruments might have not worked well. As the majority of the particles is in the SMPS size range, as usual, and as these seem to fit well with literature (at least that is what is said on page 8, line 14), it might have been the APS, measuring roughly one order of magnitude too low concentrations (that's roughly the size of the gap, larger for Lancaster Sound, a little less for Amphitrite Point and only ~*2 in the Labrador Sea). This would translate to the same magnitude of error (i.e., overestimation in this case) in INP concentrations and surface site densities, affecting many statements/results reported in the text. The observed strong increase in n_s for particles < 500nm to particles > 500nm likely is (at least in part) related to this gap between number concentrations as measured by SMPS and APS. Is there a way to find out what the problem might have been? Was there a total particle number counter deployed that could shed light onto this? In any case, this problem has to be critically discussed and related changes in the interpretation of the data has to be included in the revised version of the manuscript.

> ***[A2]*** For short periods of time during the Amphitrite Point campaign, there was an additional SMPS measurement and an additional APS measurement. During these short periods of time, the total number concentrations measured by the two SMPS instruments agreed within 10 % for the size range relevant for this paper and where overlap occurred (0.18-0.3 µm). In addition, the total number concentrations measured by the two APS measurements agreed within 10 % for the size range of 0.7-10 µm. This agreement is within the uncertainty of the instruments.
>
> After going back and investigating the size distribution data used in this study in more detail, we conclude that the gap between the SMPS and APS data is most likely due to (1) a drop off in the efficiency of the APS at size channels below 0.7 µm, and (2) an uncertainty in the hygroscopic properties at Amphitrite Point used to correct

the SMPS data for hygroscopic growth. Regarding (1), drop off in the efficiency of an APS at sizes below 0.7 µm has often been observed previously (Beddows et al., 2010). To address this issue, in the revised manuscript, the APS data at sizes below 0.7 µm has been omitted, as done previously (Maguhn et al., 2003). Regarding (2), in the revised manuscript, we have used a different method to correct the SMPS data for hygroscopic growth at Amphitrite Point. Specifically, we used a free parameter to correct for the hygroscopic growth, which resulted in the optimal overlap between the SMPS and APS data at Amphitrite Point. This type of approach has been used successfully in the past to merge SMPS and APS data (Beddows et al., 2010). After limiting the APS data to sizes of 0.7-10 µm and using a free parameter to correct for the hygroscopic growth at Amphitrite Point, the misfit in the particle number size distribution is much less.

In addition, we have included a comparison between the size distributions measured at all three sites with the size distributions measured previously at a Mid-latitude North-Atlantic marine boundary layer site by O'Dowd et al. (2001). Please see changes in Section 2.4 and Fig. S3 in the revised Supplement.

[3] page 9, line 9: It is interesting that you find n_s depends on size. But why could that be? - The larger particles would have to consist of a material that is more ice active (per surface area) than the smaller ones. How should this come about? (A mechanism would have to be that e.g., clay minerals make smaller particles, and then, the larger the mineral dust particles get, the higher becomes their feldspar content, and isn't this unlikely?) - This observed increase could be a measurement bias (as mentioned above and again in my comment concerning Fig. 4). - This needs to be discussed!

[A3] The size distribution of n_s can be qualitatively explained by considering four different types of aerosol particles each having progressively larger geometric mean diameters and n_s values. As an example, consider a mixture of: a) sulfate aerosols internally mixed with black carbon with a small n_s and small geometric mean diameter, b) sea salt aerosols with a larger n_s and larger geometric mean diameter, c) clay particles with a larger n_s and larger geometric mean diameter, and d) biological particles from terrestrial sources with the largest n_s and largest geometric mean diameter. To address the referee's comments, this information has been added to the Summary and conclusions in the revised manuscript.

[4] page 6, line 30: Was the model really run for 2001? If yes, why not for the respective month of 2013 and 2014, i.e., when the sampling was done? Can it really be assumed that the average monthly INP concentration is the same every year? How much variation could there be expected, and where within this variation are your data? Could this influence your results?

[A4] Model data from the year 2001 was used because this model output was available from previous studies. This has been made clear in the revised manuscript. In addition, to address the referee's comments, the following information has been added to Section 2.6 of the revised manuscript:

> “As mentioned above and as done previously, the model output from the year 2001 was compared with measurements from different years. The inter-annual variability of aerosol concentrations simulated in the model is expected to be up to a factor of 2 due to differences in meteorological conditions (Marmer and Langmann, 2007). Model output for the year 2001 has been found to be able to reproduce the mass concentrations of mineral dust and marine organic aerosols within an order of magnitude with observations made in various years (Vergara-Temprado et al., 2017). Furthermore, the model output for the year 2001 was able to reproduce 62 % of the INP concentrations measured from studies spanning from 1973 to 2016 within an order of magnitude, which is the uncertainty in the predicted INP concentrations reported here (Fig. 8).”

[5] Figure 4: Again, as this is where I first noticed it: There is a VERY pronounced gap between number concentrations measured with the SMPS and the APS. Do you have any explanation? This could potentially influence the derived n_s values and the fraction of INP per particle as well as your comparison with the model, particularly if it was the APS that did not operate well.

> ***[A5]*** Please see ***[A2]*** above.

Minor and technical comments:

[6] page 1, line 17: Add "particle“ before "size“.

> ***[A6]*** The word “particle” has been added before “size”.

[7] chapter 2.1.2: Also mention the temperature and RH at which droplet formation was done (the text has to be complete, i.e., readers should not have to look this up in another paper)

> ***[A7]*** To address the referee’s comment, the following sentence has been added to Section 2.1.2:
>
> “The temperature was decreased to approximately 0 ºC, and the relative humidity was increased to above water saturation using a humidified flow of He (99.999 %, Praxair), resulting in the condensation and growth of water droplets on the collected particles.”

[8] equation (1): There seems to be a typo in the formular: N_0 should not be there as a factor. The value resulting from this equation has a wrong dimension (assuming the corrections factors are dimensionless, which is how they are given in the Supplement). Please check this carefully – see also e.g., Hader et al., 2014.

> ***[A8]*** Thanks for checking the equation in our manuscript. The units used in Equation (1) are correct and consistent with Hader et al. (2014). To address the referee’s comments, in the revised manuscript Equation (1) in Section 2.1.2 has been separated into two equations. In this case, the consistency between our calculations and the equation in Hader et al. (2014) should be more obvious.

[9] page 5, line 24: Reaching an RH below 2% by a silica gel diffusion dryer is quite low (even when the silica gel is changed every 24 hours), unless the dew point of the outside air is quite low, anyway. – Did you estimate this value or check it?

> ***[A9]*** This value was not measured on site, but has been checked in the lab with the same technique. To address the referee's comment, the following has been added to Section 2.3.1 of the revised manuscript:
>
> "Three successive diffusion dryers were used prior to sampling with the SMPS, and the silica was exchanged and dried in an oven every 24 h. Although not measured on site in this campaign, this technique has been found to always reduce the RH to less than 20 %, and usually to less than 2 % (Ladino et al., 2014; Yakobi-Hancock et al., 2014)."

[10] page 6, line 6: Please give wind-speed in SI units – knots is a unit many (including myself) may not be familiar with.

> ***[A10]*** The unit has been changed to SI unit (km h^{-1}).

[11] page 6, line 10: As the MOUDI was inside at least at Amphitrite Point, drying will automatically have occurred, as it will have been warmer in the container than it was outside. This typically leads to a quick drying. The way you formulated this here is correct, however, it may be good to hint at the fact that the RH will also not have been the outside one.

> ***[A11]*** The drying effect due to the warmer temperature inside the container at Amphitrite Point was discussed in Section 2.3.1. Does the referee want us to repeat this information again in Section 2.4? Sorry, this was not clear to us.

[12] page 7, line 6 and page 10, line 12: The "-" is missing for Vergara-Temprado.

> ***[A12]*** Thanks for pointing out this mistake. It has been corrected in the revised manuscript.

[13] page 7, line 17-18: It might be better to formulate it in a different way. Basically you are saying that you don't know where the INP came from (marine or terrestrial and maybe even from further away), so please say something like: "Therefore it is not possible to determine if the INP are of marine or terrestrial origin and they may even have been long-range transported from sources more than three days away."

> ***[A13]*** Thanks for the advice. The sentence has been rephrased as following:
>
> "Therefore, it was not possible to determine if the INPs are of marine or terrestrial origin based on the back trajectories alone. INPs may even have been long-range transported from sources that were not reached by the 3-day back trajectories".

[14] page 7, line 24: Check with chapter 2.3.1 – you give different dates for the sampling period at Amphitrite Point.

[A14] Thanks for pointing out this mistake. The date in chapter 2.3.1 has been corrected.

[15] page 7, line 26: Clearly state how many samples you used for the present study.

[A15] To address the referee's comment, the number of samples has been clearly stated in Section 3.2 of the revised manuscript.

[16] page 8, line 14: Marine sites may differ depending on the sea spray production typical for the area. Were the two studies you cite here done at locations that are similar to yours in this regard? Please mention in the text if they are.

[A16] To address the referee's comment, in the revised manuscript we have added the location of the study referenced. In addition, a figure comparing the size distributions from the current study and the study referenced has been added to the Supplement (Fig. S3). Finally, additional details of the study referenced have been included in the Supplement (figure caption of Fig. S3).

[17] page 8, line 25 ff: This effect was already reported by Mertes et al. (2007), which therefore should be cited here or in the following paragraph.

[A17] Thanks for pointing this out. The result from Mertes et al. (2007) has been cited in Section 3.4 of the revised manuscript.

[18] page 9, line 26-27: Niemand et al. (2012) report n_s for dust samples in which all particles were dust. But your way to determine n_s relates the ice activity to the total particle number concentrations that were measured, hence, this is not the exact same parameter. This needs to be clearly stated here. BTW: In DeMott et al. (2016), INP concentrations for an assumed total particle number concentration of 150 cm-3 were reported (at least that's the value the laboratory data are normalized to - this is said in the caption of Fig. 1 to which you are referring), which is a factor of 2 to 4 below your values – this should also at least be mentioned, although, admittedly, this will not change your results.

[A18] To address the referee's comment, we have added the following statement to Section 3.5:

"Note, the data from Niemand et al. (2012) corresponds to the n_s values of only mineral dust particles, whereas the n_s values reported here correspond to the total aerosol particles, as mentioned above".

Regarding the normalization factor in DeMott et al. (2016), this is certainly relevant when reporting the concentrations of INPs (Fig. 1 in DeMott et al., 2016), but we do not think this is relevant when discussing the n_s values (Fig. 3 in DeMott et al., 2016), which is the focus of our manuscript. In other words, scaling to 150 cm^{-3} was not used when calculating n_s. Please let us know if we misunderstood the referee's comment.

[19] page 9, line 29-30: It should be motivated a bit stronger why you make these statements here. My comment above this one might be one reason, but I am sure you had more in mind when writing these lines.

> ***[A19]*** We hope our response to ***[18]*** provided a stronger motivation.

[20] figure 1: The insets of the pictures of the ship and measurement container might not be visible any more in a printed version, so if you care for them, you might want to make them larger (there is enough “empty” space on the map).

> ***[A20]*** The insets in Fig. 1 have been made larger to be more visible.

[21] chapter S1: Add values for the growth factors used (and / or for values for kappa).

> ***[A21]*** In the revised Supplement, we have used a different method to correct for hygroscopic growth (see ***[A2]*** above).

[22] figure S3: In the caption you say that “Each data point was calculated by adding together the numbers from Fig. 4.” Did you really simply add the data points? Looking at the number, this does not seem to have been the case, and it would have been totally wrong.

> ***[A22]*** Each data point was calculated by averaging the numbers in each size bin from Fig. 4. This has been corrected in the caption of Fig. S4 of the revised Supplement.

Literature:

Hader, J. D., T. P. Wright, and M. D. Petters (2014), Contribution of pollen to atmospheric ice nuclei concentrations, Atmos. Chem. Phys., 14(11), 5433-5449, doi:10.5194/acp-14-5433-2014.

Mertes, S., B. Verheggen, S. Walter, P. Connolly, M. Ebert, J. Schneider, K. N. Bower, J. Cozic, S. Weinbruch, U. Baltensperger, and E. Weingartner (2007), Counterflow virtual impactor based collection of small ice particles in mixed-phase clouds for the physico-chemical characterization of tropospheric ice nuclei: sampler description and first case study, Aerosol Sci. Technol., 41, 848-864.

Anonymous Referee #2

Si et al. present a comprehensive observational and modeling study evaluating size-resolved INPs at multiple coastal locations. They found a relationship between particle diameter and fraction of INPs, indicating the larger particles were more efficient ice nucleators. Size-resolved ice nucleation studies such as this are needed to better characterize INP sources. Although this study provides valuable insight into INPs, I have outlined a few issues below that should be addressed prior to publication.

General comments:

[23] Drying the sample flow to 2% seems quite extreme and is far below the GAW standard of 40% for the SMPS. Can the authors comment on how this dry of a sample flow may affect the ambient aerosol? I would assume these sort of conditions would remove semi-volatile species from the aerosol in addition to water, especially at these sizes. Although the authors do describe the corrections to the different diameter types and hygroscopic growth, the very large discrepancy between the APS and SMPS sampling conditions might not make them directly comparable given the possibility of other semi-volatile species that may have been removed.

> ***[A23]*** To address the referee's comment, the following has been added to Section 2.3.1:
>
> "For typical atmospheric conditions, the equilibration timescale for gas-particle partitioning of semivolatile organic species is on the order of minutes to tens of minutes (Saleh et al., 2013). In contrast, the residence time in the dryers during sampling in the current study was approximately 10s. Therefore, removal of semi-volatile organic species during drying may not have been a large issue but cannot be completely ruled out."

[24] I realize n_S has been commonly used to represent INP data, but how representative is n_S of the actual INP surface sites? The equation takes into account the surface area of *all* aerosols within a given size range, but if only 1 in 10^6 particles are INPs as the authors define for 0.2 um particles, is n_S realistic for the INP fraction? The authors should discuss any potential biases. Also, how was a definite size of INPs determined, given the MOUDI measures size ranges? In this case, shouldn't the aerosol surface area be defined by the same range of sizes from the SMPS and APS?

> ***[A24]*** The n_s values reported here were calculated with the total surface area of the aerosol. In this case, the n_s values correspond to a lower limit of the n_s values of the INPs. To address the referee's comment, in Section 3.5 we have added the following sentence:
>
> "Since this equation considers the surface area of all aerosol particles, rather than the surface area of just the INPs, the calculated n_s values are lower limits to the n_s values for the INPs".

Regarding the second comment, yes, the sizes of the INPs were determined from the MOUDI size ranges. The aerosol surface area is indeed calculated by integrating the surface area measured by the SMPS and APS within the same size range as INPs.

[25] There seems to be disagreement between the air mass sources (especially at Amphitrite Point) and the source apportionment results (i.e., Fig 7). Can the authors comment on why the INPs appear to be of a more terrestrial origin yet air masses were predominantly from over the ocean? What sort of very localized sources could influence the samples?

[A25] INPs of terrestrial origin could come from local vegetation or from long-range transport. Local vegetation could potentially release enough INPs to overwhelm a small INP source from the ocean. Long-range transported desert dust may still dominate the INP population after several days of transport over the ocean (Vergara-Temprado et al., 2017). To address the referee's comment, the following text has been added to Section 3.1 of the revised manuscript:

"At Amphitrite Point, although the air masses were predominantly from the ocean based on the back trajectory analysis, the air masses did pass over local vegetation including western coastal hemlock. This local vegetation could potentially release enough INPs to overwhelm a small INP source from the ocean. Therefore, it was not possible to determine if the INPs are of marine or terrestrial origin based on the back trajectories alone. INPs may even have been long-range transported from sources that were not reached by the 3-day back trajectories (Vergara-Temprado et al., 2017)."

Minor comments:

[26] P2 l 39-43 (Page 1, line 28-30 in the ACPD version): The -35 C statement is redundant from the sentence above. Also, this statement should be reworded since INPs can initiate ice formation below -35 C (e.g., glassy organics, soot, sea salt).

[A26] The sentences in the Introduction have been reworded in the revised manuscript as following:

"Aerosol particles are ubiquitous in the atmosphere, yet only a small fraction of these particles, referred to as ice nucleating particles (INPs), are able to initiate the formation of ice at temperatures warmer than homogeneous freezing temperatures. INPs may impact the frequencies, lifetime, and optical properties of ice and mixed-phase clouds (Andreae and Rosenfeld, 2008; Cziczo and Abbatt, 2001; Lohmann and Feichter, 2005)".

[27] P2 l 44-45 (Page 2, line 3 in the ACPD version): Please provide a reference for this statement.

[A27] A reference has been added to the revised manuscript.

[28] P4, l104 (Page 3, line 12 in the ACPD version): Which 2 stages were analyzed?

> ***[A28]***. Stages 2 through 8 were analyzed (seven stages in total). This sentence has been reworded as following to make this point clear in the revised manuscript:
>
> "Stages 2 through 8 of the MOUDI were analyzed for this study (seven stages in total)".

[29] P4, l125 (Page 3, line 27 in the ACPD version): How many droplets? What was the spacing? Were any neighboring droplet freezing effects apparent? For example, if droplets are too close, they can induce freezing in neighboring droplets.

> ***[A29]*** On average, approximately 40 droplets were analyzed in each experiment. The spacing between droplets varied, but was roughly 100 µm on average. Freezing of neighboring droplets was apparent in some cases, and this was taken into account while calculating the INP concentrations. This information has been added to Section 2.1.2 of the revised manuscript as following:
>
> "On average, approximately 40 droplets were analyzed in each experiment. The final droplet size was approximately 50-150 µm in diameter, and the spacing between droplets was roughly 100 µm, on average. After the formation of droplets, the flow cell was cooled down to -40 ºC at a rate of -10 ºC min^{-1} while images of the droplets were recorded. During this process, most freezing events occurred by immersion freezing, while approximately 10 % occurred by contact freezing, which refers to the freezing of liquid droplets caused by contact with neighbouring frozen droplets. When calculating INP concentrations, the contact freezing was accounted for in two ways: (i) an upper limit to the fraction frozen by immersion freezing was calculated by assuming all the contact freezing droplets froze by immersion freezing; (ii) a lower limit to the fraction frozen by immersion freezing was calculated by assuming all the contact freezing droplets remained liquid until the homogeneous freezing temperature was reached".

[30] P7, l188 and P8 l 222-223 (Page 5, line 14 and page 6, line 7 in the ACPD version): Was there any issues with artifacts from storing the dishes at room temperature as opposed to freezing the samples? Also, could the authors comment on how there could be issues comparing samples from the different locations given the different storage conditions and duration?

> ***[A30]*** Samples were stored under dry conditions and at either room temperature or 4 ºC. Samples were not frozen. To address the referee's comments, in the revised manuscript, we have pointed out that additional studies are needed to determine the effect of storage on INP concentrations. Specifically, the following has been added to Section 2.3.2:
>
> "In contrast, the samples collected at Amphitrite Point were stored at room temperature and relative humidity for less than 24 h prior to INP analysis, as mentioned above. Studies are needed to determine the effect of sample storage conditions on measured INP concentrations".

[31] P9, l264 on: Since the measurements were conducted at coastal locations, there is a likelihood that terrestrial sources of INPs may also influence the air sampled, especially given air mass trajectories show not all air masses originated from over the ocean. Can the authors comment on how this possible interference may have been dealt with, aside from the brief statement on the end of section 3.1?

> ***[A31]*** We fully agree with the referee that terrestrial sources of INPs may also influence the air sampled. Both the comparison with literature n_s values and the comparison with simulated INP concentrations addressed the importance of terrestrial sources to the INP population. To further address the referee's comment, we have expanded the discussion in Section 3.1 on possible terrestrial sources of INP. Specifically, the following has been added:
>
> "At Amphitrite Point, although the air masses were predominantly from the ocean based on the back trajectory analysis, the air masses did pass over local vegetation including coastal western hemlock. This local vegetation could release enough INPs to overwhelm a small INP source from the ocean. Therefore, it was not possible to determine if the INPs are of marine or terrestrial origin based on the back trajectories alone. INPs may even have been long-range transported from sources that were not reached by the 3-day back trajectories".

[32] P11, l 313 (Page 8, line 11 in the ACPD version): These concentrations seem fairly high for an Arctic marine atmosphere. What was the error or standard deviation of these averages? Were they just from when air masses originated over the ocean? Was new particle formation observed?

> ***[A32]*** To address the referee's comments, the standard deviation of the average total number concentration has been added to the revised manuscript. The majority of the air masses at Lancaster Sound were from over the ocean (63 % of the time), though the air masses also passed over the land. The air mass sources were discussed in Section 3.1. New particle formation has been observed in the Canadian Arctic marine boundary layer during the summer (Burkart et al., 2017). This information has been added to Section 3.3 of the revised manuscript.

[33] P12, l 349 (Page 9, line 6 in the ACPD version): Please provide equation for n_S.

> ***[A33]*** An equation for n_s has been added as Eq. (3) to Section 3.5 of the revised manuscript.

[34] P14, l 409 (Page 10, line 19 in the ACPD version): How was "marine biological activities" defined?

> ***[A34]*** In Mason et al. (2015a), methanesulfonic acid (MSA) was used as a tracer of marine biological activities. This information has been added to Section 3.6 of the revised manuscript.

[35] P14, l411 (Page 10, line 20 in the ACPD version): But air masses originated from over the ocean 94% of the time, so how would terrestrial sources be a dominant source of INPs? There seems to be some inconsistency between air mass sources in this manuscript as compared to the results from Mason et al. (2015a).

[A35] See ***[A31]*** above.

[36] Figure 2: Given the MBL can often be quite low, especially in the Arctic, the color scale should be adjusted so that the 0 – 600 m range is easier to differentiate in the figure.

[A36] The color scale in Figure 2 has been changed to a log scale so that the 0-600 m range is easier to differentiate.

Anonymous Referee #3

Review of “Ice-nucleating efficiency of aerosol particles and possible sources at three coastal marine sites”

Si et al. (2018) investigate sources of ice nucleating particles (INPs) from three coastal sites with a combined measurement-modeling approach. Measurements were taken with a suite of well- established instrumentation, that allowed quantification of INPs by an active site density function n_s. The results were compared with the output of a global INP model, and it was found that the two INP model of K-feldpsar and marine organics missed a high temperature INP source. Speculation as to what this source is was carried out reasonably. The paper is well written and the figures are clear. I think this study merits publication in ACP after some minor concerns are addressed.

[37] The first pertains to the global INP model. I would appreciate the inclusion of a more critical account of the limitations of the model when being compared to ground based measurements. There is a big jump in the conclusion that there is a missing source of INP. For example, how can the authors be sure the measurements aren’t artificially inflating the INP activity at higher temperatures by sampling from the ground? The global INP model is supposed to shed light on what, statistically and on long/large enough scales, INPs matter. The measurements on the other hand are happening locally from boundary layer air. Please investigate this point further.

[A37] The measurements were compared with predictions in the lowest level in the model. Since the model includes a parameterization of boundary layer turbulence (Holtslag and Boville, 1993), a comparison between measurements at the surface with predictions in the lowest level of the model should be appropriate. To address the referee’s comments, this information has been included in Section 2.6 of the revised manuscript. In addition, we have also added the following caveat to the Summary and conclusions:

“In addition, since the results presented here correspond to surface measurements, similar studies as a function of altitude are needed to determine if these results are applicable to higher altitudes and to the free troposphere.”

Other comments are specific to the text and are outlined below.

[38] P2 L1-2: The studies cited do not conclude that INPs “significantly impact the frequencies, lifetime, and optical properties of ice and mixed-phase clouds”. Consider changing to something less assertive like “may impact”.

[A38] Thank you for the suggestion. The wording has been changed to “may impact”.

[39] P9 L5: n_s as a function of size is a useful approach here. However, there are issues with surface area corrections that make n_s not without shortcomings. Studies by Beydoun et al. (2016), Emersic et al. (2015), and Hiranuma et al. (2015) discuss these shortcoming and should be included in an additional discussion on what kind of limitations the authors expect when analyzing n_s against surface area.

> ***[A39]*** The limitation of n_s has been discussed in the revised manuscript. Specifically, the following has been added to Section 3.5 of the revised manuscript:
>
> "The surface active site density, n_s, represents the number of ice nucleation sites per surface area (Connolly et al., 2009; Hoose and Möhler, 2012; Vali et al., 2015). This parameterization assumes that freezing is independent of time and can be scaled with surface area. Although these assumptions may not be accurate in all cases (Beydoun et al., 2016; Emersic et al., 2015; Hiranuma et al., 2015), n_s is commonly used to describe freezing data due, in part, to its simplicity".

[40] P11 L5: The authors can do a better job here of synthesizing their results and suggesting a way forward. For example, on the measurement side, samples can be investigated with a chemical composition analysis. On the modelling side, large eddy simulations can discern whether boundary layer INP are different than free atmospheric INPs simulated by the global model. So I think there's a bit more room here for discussing future efforts.

> ***[A40]*** To address the referee's comments, we have added the following text to the end of the Summary and conclusions:
>
> "Since only one sample was analyzed for both Labrador Sea and Lancaster Sound, additional samples should be collected and analyzed at these locations to determine the general applicability of the results presented here for these locations. In addition, since the results presented here correspond to surface measurements, similar studies as a function of altitude are needed to determine if these results are applicable to higher altitudes and to the free troposphere. Comparison with predictions of INPs from a high-resolution model would also be useful to assess the importance of local INP sources. Studies of the chemical composition of the INPs are also needed to test the conclusions reached in the current study".

Technical correction:

[41] n_s is a surface area density, not an efficiency. It has units of m^{-2} and does not range from 0 to 1 (like an efficiency would). You may also want to consider changing that in the title as well. Please refer to Vali et al. (2014) to ensure INP specific terminology is consistent.

> ***[A41]*** "Efficiency" has been changed to "ability" in the revised manuscript.

Literature list from referee #3

Beydoun, H., Polen, M., & Sullivan, R. C. (2016). Effect of particle surface area on ice active site densities retrieved from droplet freezing spectra. *Atmospheric Chemistry and Physics*, *16*(20), 13359–13378. https://doi.org/10.5194/acp-16-13359-2016

Emersic, C., Connolly, P. J., Boult, S., Campana, M., & Li, Z. (2015). Investigating the discrepancy between wet suspension and dry-dispersion derived ice nucleation efficiency of mineral particles. *Atmospheric Chemistry and Physics*, *15*(19), 11311–11326. https://doi.org/10.5194/acp-15-11311-2015

Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., et al. (2015). A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques. *Atmospheric Chemistry and Physics*, *15*(5), 2489–2518. https://doi.org/10.5194/acp-15-2489-2015

Vali, G., DeMott, P., Möhler, O., & Whale, T. F. (2014). Ice nucleation terminology. *Atmospheric Chemistry and Physics Discussions*, *14*, 22155–22162. https://doi.org/10.5194/acpd-14-22155-2014

References for author’s response:

Andreae, M. O. and Rosenfeld, D.: Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active aerosols, Earth-Science Rev., 89(1–2), 13–41, doi:10.1016/j.earscirev.2008.03.001, 2008.

Beddows, D. C. S., Dall’osto, M. and Harrison, R. M.: An Enhanced Procedure for the Merging of Atmospheric Particle Size Distribution Data Measured Using Electrical Mobility and Time-of-Flight Analysers, Aerosol Sci. Technol., 44(11), 930–938, doi:10.1080/02786826.2010.502159, 2010.

Beydoun, H., Polen, M. and Sullivan, R. C.: Effect of particle surface area on ice active site densities retrieved from droplet freezing spectra, Atmos. Chem. Phys., 16(20), 13359–13378, doi:10.5194/acp-16-13359-2016, 2016.

Burkart, J., Willis, M. D., Bozem, H., Thomas, J. L., Law, K., Hoor, P., Aliabadi, A. A., Köllner, F., Schneider, J., Herber, A., Abbatt, J. P. D. and Leaitch, W. R.: Summertime observations of elevated levels of ultrafine particles in the high Arctic marine boundary layer, Atmos. Chem. Phys., 17(8), 5515–5535, doi:10.5194/acp-17-5515-2017, 2017.

Connolly, P. J., Möhler, O., Field, P. R., Saathoff, H., Burgess, R., Choularton, T. and Gallagher, M.: Studies of heterogeneous freezing by three different desert dust samples, Atmos. Chem. Phys., 9(8), 2805–2824, doi:10.5194/acp-9-2805-2009, 2009.

Cziczo, D. J. and Abbatt, J. P. D.: Ice nucleation in NH4HSO4, NH4NO3, and H2SO4 aqueous particles: Implications for circus cloud formation, Geophys. Res. Lett., 28(6), 963–966, doi:10.1029/2000GL012568, 2001.

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F. and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, Proc. Natl. Acad. Sci., 113(21), 5797–5803, doi:10.1073/pnas.1514034112, 2016.

Emersic, C., Connolly, P. J., Boult, S., Campana, M. and Li, Z.: Investigating the discrepancy between wet-suspension- and dry-dispersion-derived ice nucleation efficiency of mineral particles, Atmos. Chem. Phys., 15(19), 11311–11326, doi:10.5194/acp-15-11311-2015, 2015.

Hader, J. D., Wright, T. P. and Petters, M. D.: Contribution of pollen to atmospheric ice nuclei concentrations, Atmos. Chem. Phys., 14(11), 5433–5449, doi:10.5194/acp-14-5433-2014, 2014.

Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., Diehl, K., Dreischmeier, K., Ebert, M., Frank, F., Hoffmann, N., Kandler, K., Kiselev, A., Koop, T., Leisner, T., Möhler, O., Nillius, B., Peckhaus, A., Rose, D., Weinbruch, S., Wex, H., Boose, Y., DeMott, P. J., Hader, J. D., Hill, T. C. J., Kanji, Z. A., Kulkarni, G., Levin, E. J. T., McCluskey, C. S., Murakami, M., Murray, B. J., Niedermeier, D., Petters, M. D., O'Sullivan, D., Saito, A., Schill, G. P., Tajiri, T., Tolbert, M. A., Welti, A., Whale, T. F., Wright, T. P. and Yamashita, K.: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques, Atmos. Chem. Phys., 15(5), 2489–2518, doi:10.5194/acp-15-2489-2015, 2015.

Holtslag, A. A. M. and Boville, B. A.: Local Versus Nonlocal Boundary-Layer Diffusion in a Global Climate Model, J. Clim., 6(10), 1825–1842, doi:10.1175/1520-0442(1993)006<1825:LVNBLD>2.0.CO;2, 1993.

Hoose, C. and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmos. Chem. Phys., 12(20), 9817–9854, doi:10.5194/acp-12-9817-2012, 2012.

Ladino, L. A., Zhou, S., Yakobi-Hancock, J. D., Aljawhary, D. and Abbatt, J. P. D.: Factors controlling the ice nucleating abilities of α -pinene SOA particles, J. Geophys. Res. Atmos., 119(14), 9041–9051, doi:10.1002/2014JD021578, 2014.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5(3), 715–737, doi:10.5194/acp-5-715-2005, 2005.

Maguhn, J., Karg, E., Kettrup, A. and Zimmermann, R.: On-line Analysis of the Size Distribution of Fine and Ultrafine Aerosol Particles in Flue and Stack Gas of a Municipal Waste Incineration Plant: Effects of Dynamic Process Control Measures and Emission Reduction Devices, Environ. Sci. Technol., 37(20), 4761–4770, doi:10.1021/es020227p, 2003.

Marmer, E. and Langmann, B.: Aerosol modeling over Europe: 1. Interannual variability of aerosol distribution, J. Geophys. Res., 112(D23), D23S15, doi:10.1029/2006JD008113, 2007.

Mason, R. H., Si, M., Li, J., Chou, C., Dickie, R., Toom-Sauntry, D., Pöhlker, C., Yakobi-Hancock, J. D., Ladino, L. A., Jones, K., Leaitch, W. R., Schiller, C. L., Abbatt, J. P. D., Huffman, J. A. and Bertram, A. K.: Ice nucleating particles at a coastal marine boundary layer site: correlations with aerosol type and meteorological conditions, Atmos. Chem. Phys., 15(21), 12547–12566, doi:10.5194/acp-15-12547-2015, 2015.

Mertes, S., Verheggen, B., Walter, S., Connolly, P., Ebert, M., Schneider, J., Bower, K. N., Cozic, J., Weinbruch, S., Baltensperger, U. and Weingartner, E.: Counterflow Virtual Impactor Based Collection of Small Ice Particles in Mixed-Phase Clouds for the Physico-Chemical Characterization of Tropospheric Ice Nuclei: Sampler Description and First Case Study, Aerosol Sci. Technol., 41(9), 848–864, doi:10.1080/02786820701501881, 2007.

O'Dowd, C. D., Becker, E. and Kulmala, M.: Mid-latitude North-Atlantic aerosol characteristics in clean and polluted air, Atmos. Res., 58(3), 167–185, doi:10.1016/S0169-8095(01)00098-9, 2001.

Saleh, R., Donahue, N. M. and Robinson, A. L.: Time Scales for Gas-Particle Partitioning Equilibration of Secondary Organic Aerosol Formed from Alpha-Pinene Ozonolysis, Environ. Sci. Technol., 47(11), 5588–5594, doi:10.1021/es400078d, 2013.

Vali, G., DeMott, P. J., Möhler, O. and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem. Phys., 15(18), 10263–10270, doi:10.5194/acp-15-10263-2015, 2015.

Vergara-Temprado, J., Murray, B. J., Wilson, T. W., O&apos;Sullivan, D., Browse, J., Pringle, K. J., Ardon-Dryer, K., Bertram, A. K., Burrows, S. M., Ceburnis, D., DeMott, P. J., Mason, R. H., O&apos;Dowd, C. D., Rinaldi, M. and Carslaw, K. S.: Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations, Atmos. Chem. Phys., 17(5), 3637–3658, doi:10.5194/acp-17-3637-2017, 2017.

Yakobi-Hancock, J. D., Ladino, L. A., Bertram, A. K., Huffman, J. A., Jones, K., Leaitch, W. R., Mason, R. H., Schiller, C. L., Toom-Sauntry, D., Wong, J. P. S. and Abbatt, J. P. D.: CCN activity of size-selected aerosol at a Pacific coastal location, Atmos. Chem. Phys., 14(22), 12307–12317, doi:10.5194/acp-14-12307-2014, 2014.

Ice-nucleating ability of aerosol particles and possible sources at three coastal marine sites

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Abstract. Despite the importance of ice-nucleating particles (INPs) for climate and precipitation, our understanding of these particles is far from complete. Here, we investigated INPs at three coastal marine sites in Canada, two at mid-latitude (Amphitrite Point and Labrador Sea), and one in the Arctic (Lancaster Sound). For Amphitrite Point, 23 sets of samples were analyzed, and for Labrador Sea and Lancaster Sound, one set of samples was analyzed for each location. At all three sites, the ice-nucleating ability on a per number basis (expressed as the fraction of aerosol particles acting as an INP) was strongly dependent on the particle size. For example, at diameters of around 0.2 µm, approximately 1 in 10^6 particles acted as an INP at -25 ºC, while at diameters of around 8 µm, approximately 1 in 10 particles acted as an INP at -25 ºC. The ice-nucleating ability on a per surface area basis (expressed as the surface active site density, n_s) was also dependent on the particle size, with larger particles being more efficient at nucleating ice. The n_s values of supermicron particles at Amphitrite Point and Labrador Sea were larger than previously measured n_s values of sea spray aerosol, suggesting that sea spray aerosol was not a major contributor to the supermicron INP population at these two sites. Consistent with this observation, a global model of INP concentrations under-predicted the INP concentrations when assuming only marine organics as INPs. On the other hand, assuming only K-feldspar as INPs, the same model was able to reproduce the measurements at a freezing temperature of -25 ºC, but under-predicted INP concentrations at -15 ºC, suggesting that the model is missing a source of INPs active at a freezing temperature of -15 ºC.

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1 Introduction

Aerosol particles are ubiquitous in the atmosphere, yet only a small fraction of these particles, referred to as ice nucleating particles (INPs), are able to initiate the formation of ice at temperatures warmer than homogeneous freezing temperatures.

INPs may impact the frequencies, lifetime, and optical properties of ice and mixed-phase clouds (Andreae and Rosenfeld, 2008; Cziczo and Abbatt, 2001; Lohmann and Feichter, 2005).

It is now well established that mineral dust particles represent a large fraction of INPs in the atmosphere (Hoose et al., 2010). For example, laboratory studies have shown that mineral dust particles are efficient at nucleating ice (Atkinson et al., 2013; Boose et al., 2016a; Broadley et al., 2012; Eastwood et al., 2008; Field et al., 2006; Hartmann et al., 2016; Hiranuma et al., 2015; Kanji and Abbatt, 2010; Knopf and Koop, 2006; Murray et al., 2011; Wex et al., 2014). Field measurements have shown that mineral dust is a main component of INPs at different locations (Boose et al., 2016b; DeMott et al., 2003; Klein et al., 2010; Prenni et al., 2009; Worringen et al., 2015). Modeling studies have also suggested that mineral dust particles are a major contributor to INP concentrations in many locations around the globe (Hoose et al., 2010; Vergara-Temprado et al., 2017).

Recent studies also suggest that sea spray aerosol may be an important source of INPs in some remote marine regions (Wilson et al., 2015). For example, field and laboratory measurements have shown that seawater contain particles that can nucleate ice (Alpert et al., 2011a, 2011b; Irish et al., 2017; Knopf et al., 2011; Schnell, 1977; Schnell and Vali, 1976, 1975; Wilson et al., 2015), and these INPs in seawater are thought to be emitted into the atmosphere by the bubble bursting mechanism (DeMott et al., 2016; Wang et al., 2015). Field measurements suggest that ambient INPs collected in marine environment can come from marine origin (DeMott et al., 2016; Rosinski et al., 1986, 1988; Schnell, 1982), and modeling studies have shown that sea spray aerosol is a major source of INPs in some remote marine environments (Burrows et al., 2013; Vergara-Temprado et al., 2017; Wilson et al., 2015). Modeling studies have also suggested that INPs from the ocean can significantly modify the properties of mixed-phase clouds in the atmosphere, with implications for radiative forcing predictions (Yun and Penner, 2013). Despite the growing evidence indicating that sea spray aerosol is an important type of INPs, our understanding of when and where sea spray aerosol is an important component of the total INP population is far from complete. Additional field measurements of INPs in marine environments would help improve our understanding of this topic.

Here we report INP measurements in the immersion mode from three coastal marine sites. Immersion freezing refers to freezing initiated by INPs immersed in liquid droplets (Vali et al., 2015), and this freezing mode is considered to be the most relevant for mixed-phase clouds (Ansmann et al., 2009; de Boer et al., 2011; Westbrook and Illingworth, 2011). The three coastal marine sites investigated were Amphitrite Point, Labrador Sea, and Lancaster Sound (Fig. 1). For two of these sites (Amphitrite Point and Labrador Sea), the size distributions of INPs in the immersion mode have been reported previously (Mason et al., 2015a, 2016). In the following, we build on these previous measurements by reporting the following for all three coastal marine sites: 1) the size distribution of INPs, 2) the fraction of aerosol particles acting as an INP as a function of size, and 3) the surface active site density, n_s, as a function of size. In addition, we compare the INP measurements to predictions from a recently developed global model of INP concentrations (Vergara-Temprado et al., 2017). We use this combined information to help determine if sea spray aerosol or mineral dust are the major sources of INPs at these three sites. This type of information is needed to help constrain future modeling studies of INPs and mixed-phase clouds.

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2 Methods

2.1 Measurements of INP concentrations as a function of size

Concentrations of INPs as a function of size were measured with the micro-orifice uniform deposit impactor-droplet freezing technique (MOUDI-DFT; Mason et al., 2015b). This technique involves collecting size-fractionated aerosol particles on hydrophobic glass slides with a micro-orifice uniform deposit impactor (MOUDI; Marple et al., 1991), and determining the freezing properties of collected aerosol particles with the droplet freezing technique (DFT). Details are given below.

2.1.1 Aerosol particle sampling with a MOUDI

A MOUDI (model 110R or 120R; MSP Corp., Shoreview, MN, USA) was used to collect size-fractionated aerosol particles. Aerosol particles were sampled at a flow rate of 30 L min^{-1}. The MOUDI has eleven stages, and each stage consists of a nozzle plate and an impaction plate. Aerosol particles were collected by inertial impaction onto hydrophobic glass slides (HR3-215; Hampton Research, USA) positioned on top of each impaction plate. Custom substrate holders were used to position the glass slides within the MOUDI. See Mason et al. (2015b) for details on the substrate holders. Stages 2 through 8 of the MOUDI were analyzed for this study (seven stages in total), corresponding to aerodynamic diameters of 5.6-10 µm, 3.2-5.6 µm, 1.8-3.2 µm, 1.0-1.8 µm, 0.56-1.0 µm, 0.32-0.56 µm and 0.18-0.32 µm, respectively, where the bounds are 50 % cut-off efficiencies (Marple et al., 1991).

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Particle rebound from the substrate is an issue when sampling particles with an inertial impactor. Rebound occurs when the kinetic energy of the particles striking the impactor substrate exceeds the adhesion and dissipation energies at impact (Bateman et al., 2014). Rebound can alter the number concentration and size distribution of the INPs determined with the MOUDI-DFT. Previous work has shown that particle rebound can be reduced when relative humidity (RH) is above 70 % (Bateman et al., 2014; Chen et al., 2011; Fang et al., 1991). In addition, good agreement between INP concentrations measured by the MOUDI-DFT and INP concentrations measured by a continuous flow diffusion chamber (a technique that is not susceptible to rebound) has been observed in previous field campaigns when the RH of the sampled aerosol stream was as low as 40-45 % (DeMott et al., 2017; Mason et al., 2015b).

2.1.2 Droplet freezing experiments

The freezing properties of the collected aerosol particles were determined using the DFT (Iannone et al., 2011; Mason et al., 2015b; Wheeler et al., 2015). Briefly, the hydrophobic glass slides with the collected particles were placed in a temperature- and humidity-controlled flow cell coupled to an optical microscope (Axiolab; Zeiss, Oberkochen, Germany). The temperature was decreased to approximately 0 ºC, and the relative humidity was increased to above water saturation using a humidified flow of He (99.999 %, Praxair), resulting in the condensation and growth of water droplets on the collected particles. On average, approximately 40 droplets were analyzed in each experiment. The final droplet size was approximately 50-150 µm in diameter, and the spacing between droplets was roughly 100 µm, on average. After the formation of droplets,

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$$\#INPs(T) = \left(-ln\left(\frac{N_u(T)}{N_0}\right)N_0\right) f_{nu,0.25-1\ mm}, \qquad (1)$$

where $N_u(T)$ is the number of unfrozen droplets at temperature T; N_0 is the total number of droplets analyzed within an experiment; $f_{nu,0.25-0.1\ mm}$ is a correction factor for the non-uniformity of particle concentrations across the sample deposit at a scale of 0.25-0.1 mm (see Mason et al. (2015b) for details). Equation (1) accounts for the possibility of multiple INPs in one droplet (Vali, 1971).

The number concentration of INPs in the atmosphere, [INPs(T)], was then determined using the following equation:

$$[INPs(T)] = \#INPs(T)\left(\frac{A_{deposit}}{A_{DFT}V}\right) f_{nu,1\ mm}, \qquad (2)$$

where $A_{deposit}$ is the total area of the sample deposit on each MOUDI impaction plate; A_{DFT} is the area analyzed in the droplet freezing experiment; V is the total volume of air sampled by the MOUDI; $f_{nu,1\ mm}$ is a correction factor for the non-uniformity of particle concentrations across the sample deposit at a scale of 1 mm (see Mason et al. (2015b) for details). The values of $f_{nu,1\ mm}$ and $f_{nu,0.25-0.1\ mm}$ are given in Table S1 in the Supplement.

2.2 Measurements of aerosol particle number and surface area size distributions

The combination of an aerodynamic particle sizer (APS) and a scanning mobility particle sizer (SMPS) was used to measure the aerosol number and surface area as a function of size. The APS (model 3321, TSI, Shoreview, MN, USA) measures diameters using the time-of-flight technique (Baron, 1986). At all three sites, the APS was operated with a sample flow of 1 L min^{-1} and a sheath flow of 4 L min^{-1}. The aerodynamic diameter range measured by the APS was 0.54-20 µm. Due to possible drop off in the sampling efficiency of the APS at sizes below 0.7 µm (Beddows et al., 2010), only APS data at sizes above 0.7 µm is used here, as done previously (Maguhn et al., 2003). The SMPS measures diameters based on the mobility of a particle in an electric field (Asbach et al., 2009; Hoppel, 1978). At Amphitrite Point, the SMPS (model 3936, TSI) was operated at 0.57 L min^{-1} sample flow with 2 L min^{-1} sheath flow, and was used to measure particles with mobility diameters from 18.4 to 930.6 nm. At Labrador Sea and Lancaster Sound, the SMPS (model 3034, TSI) was operated at 1 L min^{-1} sample flow rate with 4 L min^{-1} sheath flow and was used to measure particles with mobility diameters from 10 to 487 nm. The sampling condition and strategy is discussed below for each site.

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2.3 Locations of sampling

Sampling occurred at three coastal marine sites: Amphitrite Point (48.92º N, 125.54º W) on Vancouver Island in British Columbia, Canada, Labrador Sea (54.59º N, 55.61º W) off the coast of Newfoundland and Labrador, Canada, and Lancaster Sound (74.26º N, 91.46º W) between Devon Island and Somerset Island in Nunavut, Canada (Fig. 1 and Table 1). All measurements were conducted as part of the NETwork on Climate and Aerosols: addressing key uncertainties in Remote Canadian Environments (NETCARE). Average sampling times, ambient RH values, ambient temperatures, and wind speeds during sampling are summarized in Table 1. Additional details about the three coastal marine sites are given below.

2.3.1 Amphitrite Point

Measurements at Amphitrite Point were carried out at a marine boundary layer site operated by Environment and Climate Change Canada, BC Ministry of the Environment, and Metro Vancouver. This site, which is frequently influenced by marine background air (McKendry et al., 2014), is located on the west coast of Vancouver Island, British Columbia, Canada, and is approximately 2.3 km south of the town of Ucluelet (population 1627), with the Pacific Ocean to its west and south, and Barkley Sound to its southeast and east.

MOUDI samples were collected from 6 to 27 August 2013 (18 day samples, 16 night samples) as part of a larger campaign that focused on cloud condensation nuclei and INPs at a marine coastal environment (Ladino et al., 2016; Mason et al., 2015b, 2015a; Yakobi-Hancock et al., 2014). The average INP concentrations as a function of size for the entire campaign have been reported previously as well as the INP concentrations for each sample (Mason et al., 2015a). In the following, we focus on a subset of these measurements (12 day samples, 11 night samples) corresponding to the time period when MOUDI-DFT, APS, and SMPS data are all available.

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The MOUDI, APS, and SMPS were located within a mobile trailer (herein referred to as the NETCARE trailer) that was approximately 100 m from the rocky shoreline of the Pacific Ocean, separated by a narrow row of trees and shrubs approximately 2-10 m in height (Mason et al., 2015a). Aerosol particles were sampled through louvered total suspended particulate (TSP) inlets (Mesa Labs Inc., Butler, NJ, USA) that were approximately 25 m above sea level. The MOUDI and APS sampled directly from ambient air without drying, whereas the SMPS sampled ambient air through diffusion dryers. After MOUDI samples were collected, they were stored in petri dishes at room temperature and analyzed for INP concentrations within 24 h of collection.

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Meteorological parameters were measured at a lighthouse that was approximately halfway between the NETCARE trailer and the Pacific Ocean. The ambient temperature and RH were measured with an HMP45C probe (Campbell Scientific, Logan, UT, USA). Wind speed was determined by a model 05305L Wind Monitor (R. M. Young, Traverse City, Michigan, USA). The temperature and RH within the NETCARE trailer were monitored using a temperature/RH sensor probe (Acurite 00891W3). The average temperature inside the NETCARE trailer during INP sampling period was 25 ºC, compared to an average ambient temperature of 14 ºC. As a result, the average RH of the air sampled by the MOUDI and APS inside the trailer was lower than ambient RH. Based on the average ambient temperature and RH and average temperature within the trailer, the average RH in the sampling line for the MOUDI and APS was approximately 50 %. Three successive diffusion dryers were used prior to sampling with the SMPS, and the silica was exchanged and dried in an oven every 24 h. Although not measured on site in this campaign, this technique has been found to always reduce the RH to less than 20 %, and usually to less than 2 % (Ladino et al., 2014; Yakobi-Hancock et al., 2014). For typical atmospheric conditions, the equilibration timescale for gas-particle partitioning of semivolatile organic species is on the order of minutes to tens of minutes (Saleh et al., 2013). In contrast, the residence time in the dryers during sampling in the current study was approximately 10s. Therefore, removal of semivolatile organic species during drying may not have been a large issue but cannot be completely ruled out.

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2.3.2 Labrador Sea and Lancaster Sound

Measurements at Labrador Sea and Lancaster Sound were carried out onboard the Canadian Coast Guard Service (CCGS) vessel Amundsen. Amundsen serves as both an icebreaker for shipping lanes and a research vessel. The APS and MOUDI were located next to each other on top of the bridge of this vessel. Sampling occurred through louvered TSP inlets that were approximately 15 m above sea level. The SMPS was positioned behind the bridge, approximately 20 m away from the APS and MOUDI, and sampled aerosol particles through 3/8” outside diameter stainless steel tube with an inverted U-shaped inlet that was approximately 15 m above sea level. Meteorological parameters were measured with sensors on a tower deployed on the foredeck of the Amundsen. Wind speed and direction were monitored at a height of 16 m above sea surface using a conventional propeller anemometer (RM Young Co. model 15106MA). Temperature and RH were measured using an RH/Temperature probe (Vaisala model HMP45C212) housed in a vented sunshield.

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One MOUDI sample was collected on 11 July 2014 while in the Labrador Sea off the coast of Newfoundland and Labrador. Results of this sample have been reported in Mason et al. (2016). A second MOUDI sample was collected on 20 July 2014 while in the Lancaster Sound between Devon Island and Somerset Island. When the two MOUDI samples were collected, the apparent wind direction was ±90º of the bow and the wind speed was > 9.3 km h^{-1}, suggesting that ship emissions did not influence the samples (Johnson et al., 2008). After collection, the samples were vacuum-sealed and stored in a 4 ºC fridge for 45-46 days prior to analysis. In contrast, the samples collected at Amphitrite Point were stored at room temperature and relative humidity for less than 24 h prior to INP analysis, as mentioned above. Studies are needed to determine the effect of sample storage conditions on measured INP concentrations.

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2.4 Conversion of mobility diameter to aerodynamic diameter and corrections for hygroscopic growth

At Labrador Sea and Lancaster Sound, a dryer was not used prior to sampling with the MOUDI, APS, and SMPS. Hence, for these two sites, all data correspond to the RH and temperatures during the sampling. The sizes measured by the MOUDI and the APS were in aerodynamic diameter, while the SMPS measured mobility diameter. To allow comparison between the INP

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At Amphitrite Point, a dryer was also not used when sampling with the MOUDI and APS. On the other hand, dryers were used prior to sampling with the SMPS. To allow comparison between the INP data, APS data and SMPS data at this site, a free parameter was used to convert the SMPS data under dry conditions to aerodynamic diameters at the RH and temperature during the sampling. The free parameter was determined from the optimal overlap between the SMPS and APS data. This type of approach has been used successfully in the past to merge SMPS and APS data (Beddows et al., 2010; Khlystov et al., 2004) (see Sect. S2 for details).

2.5 Back trajectory analysis

For each MOUDI sample collected for INP analysis, a 3-day back trajectory was calculated using the HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) model of the NOAA Air Resources Laboratory (Stein et al., 2015). The GDAS (Global Data Assimilation System) 1 degree meteorological data was used as input. Back trajectories were initiated at the beginning of each MOUDI sampling period and at every hour until the end of the sampling period. The initiating height was the same as the height of the MOUDI sampling inlets as mentioned in Sect. 2.3. Back trajectories were also initiated at heights of 50 m and 150 m a.g.l. for each location to determine if the trajectories were sensitive to the height of initiation.

2.6 Global model of INP concentrations

A global model of INP concentrations relevant for mixed-phase clouds was used to predict concentrations of INPs at the three sampling sites (Vergara-Temprado et al., 2017). The model considers ice nucleation by K-feldspar, associated with desert dust, and marine organics, associated with sea spray aerosol, as INPs. In this model (GLOMAP-mode), aerosol number and mass concentration of several aerosol species are simulated in seven lognormal modes (3 insoluble and 4 soluble). The model has a horizontal resolution of 2.8 x 2.8 degrees with 31 vertical levels, and it is run for the year 2001 with meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF). Model output for the year 2001 was used since this model output was available from previous studies. The model includes a parameterization of boundary layer turbulence (Holtslag and Boville, 1993). The aerosol components are emitted internally mixed with the species of their mode, and several aerosol microphysical processes including new particle formation, particle growth, dry deposition and wet scavenging are represented (Mann et al., 2014). The INP concentrations are determined using a laboratory-based temperature-dependent density of active sites (active sites per unit surface area) for K-feldspar (Atkinson et al., 2013) and a parameterization for marine organics based on the INP content of microlayer samples (expressed as active sites per unit mass of organic carbon) (Wilson et al., 2015) following the method shown in Vergara-Temprado et al. (2017) Appendix 2.

To predict INP concentrations at the three coastal marine sites, we used the output of the model for the grid cells that overlapped with the measurement locations. Since the measurements were carried out at the surface, output from the lowest

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level of the model was used. We calculated the mean concentrations of INPs from K-feldspar and marine organics for the months when measurements were made. For the simulations at Amphitrite Point, Labrador Sea, and Lancaster Sound, the months of August, July, and July were used, respectively.

As mentioned above and as done previously, the model output from the year 2001 was compared with measurements from different years. The inter-annual variability of aerosol concentrations simulated in the model is expected to be up to a factor of 2 due to differences in meteorological conditions (Marmer and Langmann, 2007). Model output for the year 2001 has been found to be able to reproduce the mass concentrations of mineral dust and marine organic aerosols within an order of magnitude with observations made in various years (Vergara-Temprado et al., 2017). Furthermore, the model output for the year 2001 was able to reproduce 62 % of the INP concentrations measured from studies spanning from 1973 to 2016 within an order of magnitude, which is the uncertainty in the predicted INP concentrations reported here (Fig. 8).

3 Results and discussion

3.1 Air mass sources from back trajectories

Figure 2 shows the 3-day back trajectories initiated for every hour during the MOUDI sampling at the three sites. The initiation heights were the same as the MOUDI sampling inlet heights. Similar results were obtained using initiation heights of 50 m and 150 m a.g.l. (see Fig. S1-S2 in the Supplement). When considering all the back trajectories, at Amphitrite Point, 94 % of the time was spent over the ocean, at Labrador Sea, 40 % of the time was spent over the ocean, and at Lancaster Sound, 63 % of the time was spent over the ocean. The rest of the time was spent over the land. At Amphitrite Point, although the air masses were predominantly from the ocean based on the back trajectory analysis, the air masses did pass over local vegetation including coastal western hemlock. This local vegetation could potentially release enough INPs to overwhelm a small INP source from the ocean. Therefore, it was not possible to determine if the INPs are of marine or terrestrial origin based on the back trajectories alone. INPs may even have been long-range transported from sources that were not reached by the 3-day back trajectories (Vergara-Temprado et al., 2017).

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3.2 INP concentrations as a function of size

In Fig. 3, the average INP number concentration is plotted as a function of size for the freezing temperatures of -15 ºC, -20 ºC, and -25 ºC. These three temperatures were chosen because freezing events were rare at temperatures warmer than -15 ºC, and for some MOUDI stages, all the droplets were frozen at temperatures lower than -25 ºC, making calculations of INP concentrations using Eq. (1)-(2) not possible at temperatures lower than -25 ºC. Mason et al. (2015a) previously reported the average INP number concentrations as a function of size at Amphitrite Point for the time period of 6-27 August 2013. Here we report the average INP number concentrations as a function of size at the same site for a subset of the measurements (23 out of 34 samples) from Mason et al. (2015a) when both APS and SMPS data were available. Not surprisingly, the results shown here are very similar to the results shown by Mason et al. (2015a). The result for Labrador Sea shown in Fig. 3 has also been reported previously in Mason et al. (2016), while the result for Lancaster Sound is new and represents the first report of INP concentrations as a function of size in the Arctic marine boundary layer. Lancaster Sound had the lowest INP concentrations among the three sites with average concentrations of INPs of 0 L^{-1}, 0.16 L^{-1}, and 0.67 L^{-1} for the freezing temperatures of -15 ºC, -20 ºC, and -25 ºC, respectively. These numbers are consistent with several previous measurements reported in the Arctic. For example, Mason et al. (2016) reported the following mean concentrations at a surface site in Alert, Nunavut: 0.05 L^{-1}, 0.22 L^{-1} and 0.99 L^{-1} for freezing temperatures of -15 ºC, -20 ºC, and -25 ºC, respectively. Bigg (1996) reported mean INP concentration of 0.01 L^{-1} at -15 ºC on an icebreaker in the Arctic. Fountain and Ohtake (1985) measured mean INP concentrations of 0.17 L^{-1} at -20 ºC at a surface site in Barrow, Alaska.

At Amphitrite Point and Labrador Sea, the majority of INPs measured were > 1 µm in diameter at all the temperatures studied. At Lancaster Sound, the majority of INPs were also > 1 µm at -25 ºC. At -15 ºC, the concentrations of INPs were not above detection limit at any of the sizes, while at -20 ºC, freezing was only observed for sizes between 0.56 and 1 µm.

3.3 Size distributions of ambient aerosols

As mentioned above, the concentrations of aerosol number and surface area as a function of size were determined from measurements with a SMPS and an APS. The results are shown in Fig. 4. The size distributions were consistent with the size distributions measured at a mid-latitude North-Atlantic marine boundary layer site by O'Dowd et al., (2001) (see Fig. S3 in the Supplement). The average total number concentrations were 1487 ± 512 cm^{-3}, 3020 ± 128 cm^{-3}, and 946 ± 254 cm^{-3} for Amphitrite Point, Labrador Sea, and Lancaster Sound, respectively. The number concentration at the Arctic site Lancaster Sound may have been influenced by the new particle formation in the summer Arctic marine boundary layer (Burkart et al., 2017; Tunved et al., 2013). For the size range of measured INPs (0.18-10 µm), on average, < 3 % of the number concentration was supermicron in diameter, and < 47 % of the surface area concentration was supermicron in diameter.

3.4 Ice-nucleating ability on a per number basis

The ice-nucleating ability on a per number basis is represented as the fraction of aerosol particles acting as an INP. Shown in Fig. 5 is the fraction of aerosol particles acting as an INP as a function of size. To generate Fig. 5, first the aerosol number concentrations (Fig. 4a) was binned using the same bin widths as the MOUDI, resulting in the total aerosol number concentration in each size bin (Fig. S4a). Then the INP concentration (Fig. 3) was divided by the aerosol number concentration (Fig. S4a). Figure 5 shows that the fraction of particles acting as an INP is strongly dependent on the size. For Amphitrite Point and Labrador Sea, and for diameters of around 0.2 µm, approximately 1 in 10^6 particles acted as an INP at -25 ºC. On the other hand, at the same sites and for diameters of around 8 µm, approximately 1 in 10 particles acted as an INP at -25 ºC. A similar trend may be present at Lancaster Sound, but at the smaller sizes investigated, the concentrations of INPs were below detection limit. The results in Fig. 5 show that the large particles at the three sites studied are extremely efficient at nucleating ice, and as a result, even though the number concentration of large particles might be small in the atmosphere, they can make an important contribution to the total INP number concentrations.

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The strong dependence on the size shown in Fig. 5 is consistent with the small number of previous studies that investigated the fraction of aerosol particles acting as an INP as a function of size. Berezinski et al. (1988) studied INPs collected at 100-500 m above ground level in the southern part of the European territory of the former USSR. At a freezing temperature of -20 ºC and for a diameter of 0.1 µm, approximately 1 in 10^5 particles acted as an INP, while for a diameter of 10 µm approximately 1 in 100 particles acted as an INP. A study of residuals of mixed-phase clouds by Mertes et al. (2007) found that 1 in 10 supermicron particles acted as an INP, while only 1 in 10^3 submicron particles acted as an INP. Huffman et al. (2013) studied INPs collected at a semi-arid pine forest in Colorado, United States. At a freezing temperature of -15 ºC and for a diameter of 2 µm approximately 1 in 10^3 particles acted as an INP, while at the same freezing temperature but for a diameter of 10 µm, more than 1 in 100 particles acted as an INP.

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3.5 Surface active site density, n_s, as a function of size

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The surface active site density, n_s, represents the number of ice nucleation sites per surface area (Connolly et al., 2009; Hoose and Möhler, 2012; Vali et al., 2015). This parameterization assumes that freezing is independent of time and can be scaled with surface area. Although these assumptions may not be accurate in all cases (Beydoun et al., 2016; Emersic et al., 2015; Hiranuma et al., 2015), n_s is commonly used to describe freezing data due, in part, to its simplicity. Here we use the following equation to calculate n_s as a function of size from our experimental data:

$$n_s(T) = \frac{[INPs(T)]}{S_{tot}}, \qquad (3)$$

where $[INPs(T)]$ is the INP concentration at temperature T determined from Eq. (2) in a given size range, and S_{tot} is the total surface area of all aerosol particles in the same size range. Since this equation considers the surface area of all aerosol particles, rather than the surface area of just the INPs, the calculated n_s values are lower limits to the n_s values for the INPs.

Shown in Fig. 6 are the measured n_s values as a function of size determined with Eq. (3). To generate Fig. 6, first the aerosol surface area concentration (Fig. 4b) was binned using the same bin widths as the MOUDI, resulting in the total aerosol surface area concentration in each size bin (Fig. S4b). Following Eq. (3), the INP concentration (Fig. 3) was then divided by the total aerosol surface area concentration (Fig. S4b), resulting in n_s values as a function of size. Figure 6 shows that n_s is dependent on the size, with the larger particles being more efficient at nucleating ice. For Amphitrite Point and Labrador Sea, at a freezing temperature of -25 ºC, n_s was approximately two orders of magnitude higher for 8 µm particles compared to 0.2 µm particles. The dependence of n_s on size can be qualitatively explained by considering four different types of aerosol particles each having progressively larger geometric mean diameters and larger n_s values.

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To determine whether sea spray aerosol or mineral dust are the major sources of INPs at the three sites, the measured n_s values were compared to n_s values of sea spray aerosol and mineral dust at -15 ºC, -20 ºC, and -25 ºC, respectively (Fig. 7). The n_s values of sea spray aerosol in Fig. 7 are from field studies in the marine boundary layer and laboratory studies of sea spray aerosol as reported in DeMott et al. (2016). Specifically, the data in Fig. 1A in DeMott et al. (2016) were re-plotted and fitted using linear regression (see Fig. S5 in the Supplement). Since the reported n_s values in DeMott et al. (2016)

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correspond to dry conditions, these values should be considered as upper limits to the n_s values for sea spray aerosol exposed to high RH values. Figure 7 shows that the n_s values of sea spray aerosol are smaller than the measured n_s values in the supermicron range at all freezing temperatures at Amphitrite Point. This is also the case for Labrador Sea at freezing temperatures of -20 and -25 ºC. For Lancaster Sound, the n_s values of sea spray aerosol are smaller than the measured n_s values for sizes of 5.6-10 µm and a freezing temperature of -25 ºC. These combined results suggest that sea spray aerosol was not the major contributor to the supermicron INP population at Amphitrite Point and Labrador Sea, and not a contributor to the largest INPs (5.6-10 µm in size) observed at Lancaster Sound.

The n_s values of mineral dust particles shown in Fig. 7 are based on laboratory measurements with five different dust samples: Asian dust, Saharan dust, Canary Island dust, Israel dust, and Arizona test dust (Niemand et al., 2012). Specifically, the data in Fig. 6 in Niemand et al. (2012) were re-plotted and fitted using linear regression (see Fig. S6 in the Supplement). Figure 7 shows that the n_s values for mineral dust are greater than or equal to the measured n_s values at all three sites. These results suggest that mineral dust could be a possible source of the supermicron INPs at the three sites studied. However, these results do not confirm mineral dust as a major contributor of supermicron INPs nor do they rule out other types of particles as a major contributor of supermicron INPs. Note, the data from Niemand et al. (2012) corresponds to the n_s values of only mineral dust particles, whereas the n_s values reported here correspond to the total aerosol particles, as mentioned above.

3.6 Comparison between measured and simulated INP concentrations

Shown in Fig. 8 is a comparison between the measured INP concentrations and the simulated INP concentrations at the surface at the three sites using a global model of INP concentrations based on the ice nucleation of K-feldspar and marine organics. When considering only marine organics as INPs in the model, predicted INP concentrations are less than measured INP concentrations in all cases except for Amphitrite Point at a freezing temperature of -25 ºC. This suggests that sea spray aerosol is not the dominant source of INPs at the three coastal marine sites studied for all three temperatures, which is consistent with conclusions reached in Sect. 3.5. When considering only K-feldspar, associated with desert dust, as INPs in the model, the predicted INP concentrations at -25 ºC are consistent with the measurements at all three sites, but at -15 ºC and -20 ºC the predicted INP concentrations are less than measured. When considering both marine organics and K-feldspar as INPs in the model the predicted INP concentrations at -25 ºC are consistent with measurements, but at warmer temperatures, the predicted INPs are still less than measured. The underestimation of INP concentrations at warmer temperatures of the model could be explained by a missing source of INPs that are active at temperatures warmer than -25 ºC, as hypothesized in Vergara-Temprado et al. (2017) based on the comparison with measurements at other sites. Possible sources missing in the model that could explain the high-temperature INPs include bacteria, fungal material, agricultural dust or biological nanoscale fragments attached to mineral dust particles (Fröhlich-Nowoisky et al., 2015; Garcia et al., 2012; Haga et al., 2013; Mason et al., 2015a; Möhler et al., 2008; Morris et al., 2004, 2013, O'Sullivan et al., 2014, 2015, 2016; Spracklen and Heald, 2014; Tobo et al., 2013, 2014).

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Recently Mason et al. (2015a) investigated the source of INPs at Amphitrite Point using correlations between INP number concentrations, atmospheric particles, and meteorological conditions. Correlations between INP number concentrations and marine aerosol (sodium as a tracer) and marine biological activities (methanesulfonic acid as a tracer) were not statistically significant. On the other hand, a strong correlation was observed between INP concentrations and fluorescent bioparticles, suggesting biological particles from terrestrial sources were likely a dominant source of INPs at this site. These results are consistent and complementary to the studies presented above.

4 Summary and conclusions

The INP number concentrations in the immersion freezing mode as a function of size were determined at three coastal marine sites in Canada: Amphitrite Point (48.92º N, 125.54º W), Labrador Sea (54.59º N, 55.61º W), and Lancaster Sound (74.26º N, 91.46º W). For Amphitrite Point, 23 sets of samples were analyzed, and for Labrador Sea and Lancaster Sound, one set of samples was analyzed for each location. The result for Lancaster Sound is the first report of INP number concentrations as a function of size in the Arctic marine boundary layer. The freezing ability of aerosol particles as a function of size was investigated by combining the size-resolved concentrations of INPs and the size distributions of aerosol number and surface area. We found that the fraction of aerosol particles acting as an INP is strongly dependent on the particle size. At -25 ºC and for Amphitrite Point and Labrador Sea, approximately 1 in 10^6 particles acted as an INP at diameters around 0.2 µm, while approximately 1 in 10 particles acted as an INP at diameters around 8 µm. We also found that the surface active site density, n_s, is dependent on the particle size. At -25 ºC and for Amphitrite Point and Labrador Sea, n_s was approximately two orders of magnitude higher for 8 µm particles compared to 0.2 µm particles. The size distribution of n_S can be qualitatively explained by considering four different types of aerosol particles each having progressively larger geometric mean diameters and n_S values. As an example, consider a mixture of a) sulfate aerosols internally mixed with black carbon with a small n_S and small geometric mean diameter, b) sea salt aerosols with larger n_S and larger geometric mean diameter, c) clay particles with a larger n_S and larger geometric mean diameter, and d) biological particles from terrestrial sources with the largest n_S and largest geometric mean diameter.

Sea spray aerosol and mineral dust were investigated as the possible sources of INPs. Sea spray aerosol was not the major source of INPs based on the comparison of the measurements with the n_s values of sea spray aerosol, and the INP concentrations predicted by a global model. On the other hand, the mineral dust may be a main source of INPs at the three sites and at a freezing temperature of -25 ºC based on the comparison of the measured INP concentrations with the predictions of a global model. However, the under-prediction of the INP concentrations at -15 ºC and -20 ºC suggests the existence of other possible sources of INPs such as biological particles from terrestrial sources or agricultural dust. Since only one sample was analyzed for both Labrador Sea and Lancaster Sound, additional samples should be collected and analyzed at these locations to determine the general applicability of the results presented here for these locations. In addition, since the results presented here correspond to surface measurements, similar studies as a function of altitude are needed to

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determine if these results are applicable to higher altitudes and to the free troposphere. Comparison with predictions of INPs from a high-resolution model would also be useful to assess the importance of local INP sources. Studies of the chemical composition of the INPs are also needed to test the conclusions reached in the current study.

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References:

Alpert, P. A., Aller, J. Y. and Knopf, D. A.: Ice nucleation from aqueous NaCl droplets with and without marine diatoms, Atmos. Chem. Phys., 11(12), 5539–5555, doi:10.5194/acp-11-5539-2011, 2011a.

Alpert, P. A., Aller, J. Y. and Knopf, D. A.: Initiation of the ice phase by marine biogenic surfaces in supersaturated gas and supercooled aqueous phases, Phys. Chem. Chem. Phys., 13(44), 19882, doi:10.1039/c1cp21844a, 2011b.

Andreae, M. O. and Rosenfeld, D.: Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active aerosols, Earth-Science Rev., 89(1–2), 13–41, doi:10.1016/j.earscirev.2008.03.001, 2008.

Ansmann, A., Tesche, M., Seifert, P., Althausen, D., Engelmann, R., Fruntke, J., Wandinger, U., Mattis, I. and Müller, D.: Evolution of the ice phase in tropical altocumulus: SAMUM lidar observations over Cape Verde, J. Geophys. Res., 114(D17), D17208, doi:10.1029/2008JD011659, 2009.

Asbach, C., Kaminski, H., Fissan, H., Monz, C., Dahmann, D., Mülhopt, S., Paur, H. R., Kiesling, H. J., Herrmann, F., Voetz, M. and Kuhlbusch, T. A. J.: Comparison of four mobility particle sizers with different time resolution for stationary exposure measurements, J. Nanoparticle Res., 11(7), 1593–1609, doi:10.1007/s11051-009-9679-x, 2009.

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D. and Malkin, T. L.: The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds, Nature, 498(7454), 355–358, doi:10.1038/nature12278, 2013.

Baron, P. A.: Calibration and use of the aerodynamic particle sizer (APS 3300), Aerosol Sci. Technol., 5(1), 55–67, doi:10.1080/02786828608959076, 1986.

Bateman, A. P., Belassein, H. and Martin, S. T.: Impactor apparatus for the study of particle rebound: Relative humidity and capillary forces, Aerosol Sci. Technol., 48(1), 42–52, doi:10.1080/02786826.2013.853866, 2014.

Beddows, D. C. S., Dall'osto, M. and Harrison, R. M.: An Enhanced Procedure for the Merging of Atmospheric Particle Size Distribution Data Measured Using Electrical Mobility and Time-of-Flight Analysers, Aerosol Sci. Technol., 44(11), 930–938, doi:10.1080/02786826.2010.502159, 2010.

Berezinski, N. A., Stepanov, G. V. and Khorguani, V. G.: Ice-forming activity of atmospheric aerosol particles of different sizes, Atmos. Aerosols Nucleation, 309, 709–712, 1988.

Beydoun, H., Polen, M. and Sullivan, R. C.: Effect of particle surface area on ice active site densities retrieved from droplet freezing spectra, Atmos. Chem. Phys., 16(20), 13359–13378, doi:10.5194/acp-16-13359-2016, 2016.

Bigg, E. K.: Ice forming nuclei in the high Arctic, Tellus B, 48(2), 223–233, doi:10.1034/j.1600-0889.1996.t01-1-00007.x, 1996.

de Boer, G., Morrison, H., Shupe, M. D. and Hildner, R.: Evidence of liquid dependent ice nucleation in high-latitude stratiform clouds from surface remote sensors, Geophys. Res. Lett., 38(1), n/a-n/a, doi:10.1029/2010GL046016, 2011.

Boose, Y., Welti, A., Atkinson, J., Ramelli, F., Danielczok, A., Bingemer, H. G., Plötze, M., Sierau, B., Kanji, Z. A. and Lohmann, U.: Heterogeneous ice nucleation on dust particles sourced from nine deserts worldwide - Part 1: Immersion freezing, Atmos. Chem. Phys., 16(23), 15075–15095, doi:10.5194/acp-16-15075-2016, 2016a.

Boose, Y., Sierau, B., García, M. I., Rodríguez, S., Alastuey, A., Linke, C., Schnaiter, M., Kupiszewski, P., Kanji, Z. A. and Lohmann, U.: Ice nucleating particles in the Saharan Air Layer, Atmos. Chem. Phys., 16(14), 9067–9087, doi:10.5194/acp-16-9067-2016, 2016b.

Broadley, S. L., Murray, B. J., Herbert, R. J., Atkinson, J. D., Dobbie, S., Malkin, T. L., Condliffe, E. and Neve, L.: Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust, Atmos. Chem. Phys., 12(1), 287–307, doi:10.5194/acp-12-287-2012, 2012.

Burkart, J., Willis, M. D., Bozem, H., Thomas, J. L., Law, K., Hoor, P., Aliabadi, A. A., Köllner, F., Schneider, J., Herber, A., Abbatt, J. P. D. and Leaitch, W. R.: Summertime observations of elevated levels of ultrafine particles in the high Arctic marine boundary layer, Atmos. Chem. Phys., 17(8), 5515–5535, doi:10.5194/acp-17-5515-2017, 2017.

Burrows, S. M., Hoose, C., Pöschl, U. and Lawrence, M. G.: Ice nuclei in marine air: Biogenic particles or dust?, Atmos. Chem. Phys., 13(1), 245–267, doi:10.5194/acp-13-245-2013, 2013.

Chen, S. C., Tsai, C. J., Chen, H. D., Huang, C. Y. and Roam, G. D.: The Influence of Relative Humidity on Nanoparticle Concentration and Particle Mass Distribution Measurements by the MOUDI, Aerosol Sci. Technol., 45(5), 596–603, doi:10.1080/02786826.2010.551557, 2011.

Connolly, P. J., Möhler, O., Field, P. R., Saathoff, H., Burgess, R., Choularton, T. and Gallagher, M.: Studies of heterogeneous freezing by three different desert dust samples, Atmos. Chem. Phys., 9(8), 2805–2824, doi:10.5194/acp-9-2805-2009, 2009.

Cziczo, D. J. and Abbatt, J. P. D.: Ice nucleation in NH4HSO4, NH4NO3, and H2SO4 aqueous particles: Implications for circus cloud formation, Geophys. Res. Lett., 28(6), 963–966, doi:10.1029/2000GL012568, 2001.

DeMott, P. J., Sassen, K., Poellot, M. R., Baumgardner, D., Rogers, D. C., Brooks, S. D., Prenni, A. J. and Kreidenweis, S. M.: African dust aerosols as atmospheric ice nuclei, Geophys. Res. Lett., 30(14), 26–29, doi:10.1029/2003GL017410, 2003.

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F. and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, Proc. Natl. Acad. Sci., 113(21), 5797–5803, doi:10.1073/pnas.1514034112, 2016.

DeMott, P. J., Hill, T. C. J., Petters, M. D., Bertram, A. K., Tobo, Y., Mason, R. H., Suski, K. J., McCluskey, C. S., Levin, E. J. T., Schill, G. P., Boose, Y., Rauker, A. M., Miller, A. J., Zaragoza, J., Rocci, K., Rothfuss, N. E., Taylor, H. P., Hader, J.

D., Chou, C., Huffman, J. A., Pöschl, U., Prenni, A. J. and Kreidenweis, S. M.: Comparative measurements of ambient atmospheric concentrations of ice nucleating particles using multiple immersion freezing methods and a continuous flow diffusion chamber, Atmos. Chem. Phys., 17(18), 11227–11245, doi:10.5194/acp-17-11227-2017, 2017.

Eastwood, M. L., Cremel, S., Gehrke, C., Girard, E. and Bertram, A. K.: Ice nucleation on mineral dust particles: Onset conditions, nucleation rates and contact angles, J. Geophys. Res., 113(D22), D22203, doi:10.1029/2008JD010639, 2008.

Emersic, C., Connolly, P. J., Boult, S., Campana, M. and Li, Z.: Investigating the discrepancy between wet-suspension- and dry-dispersion-derived ice nucleation efficiency of mineral particles, Atmos. Chem. Phys., 15(19), 11311–11326, doi:10.5194/acp-15-11311-2015, 2015.

Fang, C. P., McMurry, P. H., Marple, V. A. and Rubow, K. L.: Effect of Flow-induced Relative Humidity Changes on Size Cuts for Sulfuric Acid Droplets in the Microorifice Uniform Deposit Impactor (MOUDI), Aerosol Sci. Technol., 14(2), 266–277, doi:10.1080/02786829108959489, 1991.

Field, P. R., Möhler, O., Connolly, P., Krämer, M., Cotton, R., Heymsfield, A. J., Saathoff, H. and Schnaiter, M.: Some ice nucleation characteristics of Asian and Saharan desert dust, Atmos. Chem. Phys., 6(10), 2991–3006, doi:10.5194/acp-6-2991-2006, 2006.

Fountain, A. G. and Ohtake, T.: Concentrations and Source Areas of Ice Nuclei in the Alaskan Atmosphere, J. Clim. Appl. Meteorol., 24(4), 377–382, doi:10.1175/1520-0450(1985)024<0377:CASAOI>2.0.CO;2, 1985.

Fröhlich-Nowoisky, J., Hill, T. C. J., Pummer, B. G., Yordanova, P., Franc, G. D. and Pöschl, U.: Ice nucleation activity in the widespread soil fungus *Mortierella alpina*, Biogeosciences, 12(4), 1057–1071, doi:10.5194/bg-12-1057-2015, 2015.

Garcia, E., Hill, T. C. J., Prenni, A. J., DeMott, P. J., Franc, G. D. and Kreidenweis, S. M.: Biogenic ice nuclei in boundary layer air over two U.S. High Plains agricultural regions, J. Geophys. Res. Atmos., 117(D18), D18209, doi:10.1029/2012JD018343, 2012.

Haga, D. I., Iannone, R., Wheeler, M. J., Mason, R., Polishchuk, E. A., Fetch, T., Van Der Kamp, B. J., McKendry, I. G. and Bertram, A. K.: Ice nucleation properties of rust and bunt fungal spores and their transport to high altitudes, where they can cause heterogeneous freezing, J. Geophys. Res. Atmos., 118(13), 7260–7272, doi:10.1002/jgrd.50556, 2013.

Harrison, A. D., Whale, T. F., Carpenter, M. A., Holden, M. A., Neve, L., O'Sullivan, D., Vergara-Temprado, J. and Murray, B. J.: Not all feldspars are equal: a survey of ice nucleating properties across the feldspar group of minerals, Atmos. Chem. Phys., 16(17), 10927–10940, doi:10.5194/acp-16-10927-2016, 2016.

Hartmann, S., Wex, H., Clauss, T., Augustin-Bauditz, S., Niedermeier, D., Rösch, M. and Stratmann, F.: Immersion Freezing of Kaolinite: Scaling with Particle Surface Area, J. Atmos. Sci., 73(1), 263–278, doi:10.1175/JAS-D-15-0057.1, 2016.

Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., Diehl, K., Dreischmeier, K., Ebert, M., Frank, F., Hoffmann, N., Kandler, K., Kiselev, A., Koop, T., Leisner, T., Möhler, O., Nillius, B., Peckhaus, A., Rose, D., Weinbruch, S., Wex, H., Boose, Y., DeMott, P. J., Hader, J. D., Hill, T. C. J., Kanji, Z. A., Kulkarni, G., Levin, E. J. T., McCluskey, C. S., Murakami, M., Murray, B. J., Niedermeier, D., Petters, M. D., O'Sullivan, D., Saito, A., Schill, G.

P., Tajiri, T., Tolbert, M. A., Welti, A., Whale, T. F., Wright, T. P. and Yamashita, K.: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques, Atmos. Chem. Phys., 15(5), 2489–2518, doi:10.5194/acp-15-2489-2015, 2015.

Holtslag, A. A. M. and Boville, B. A.: Local Versus Nonlocal Boundary-Layer Diffusion in a Global Climate Model, J. Clim., 6(10), 1825–1842, doi:10.1175/1520-0442(1993)006<1825:LVNBLD>2.0.CO;2, 1993.

Hoose, C. and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmos. Chem. Phys., 12(20), 9817–9854, doi:10.5194/acp-12-9817-2012, 2012.

Hoose, C., Kristjánsson, J. E., Chen, J.-P. and Hazra, A.: A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model, J. Atmos. Sci., 67(8), 2483–2503, doi:10.1175/2010JAS3425.1, 2010.

Hoppel, W. A.: Determination of the aerosol size distribution from the mobility distribution of the charged fraction of aerosols, J. Aerosol Sci., 9(1), 41–54, doi:10.1016/0021-8502(78)90062-9, 1978.

Huffman, J. A., Prenni, A. J., DeMott, P. J., Pöhlker, C., Mason, R. H., Robinson, N. H., Fröhlich-Nowoisky, J., Tobo, Y., Després, V. R., Garcia, E., Gochis, D. J., Harris, E., Müller-Germann, I., Ruzene, C., Schmer, B., Sinha, B., Day, D. A., Andreae, M. O., Jimenez, J. L., Gallagher, M., Kreidenweis, S. M., Bertram, A. K. and Pöschl, U.: High concentrations of biological aerosol particles and ice nuclei during and after rain, Atmos. Chem. Phys., 13(13), 6151–6164, doi:10.5194/acp-13-6151-2013, 2013.

Iannone, R., Chernoff, D. I., Pringle, A., Martin, S. T. and Bertram, A. K.: The ice nucleation ability of one of the most abundant types of fungal spores found in the atmosphere, Atmos. Chem. Phys., 11(3), 1191–1201, doi:10.5194/acp-11-1191-2011, 2011.

Irish, V. E., Elizondo, P., Chen, J., Chou, C., Charette, J., Lizotte, M., Ladino, L. A., Wilson, T. W., Gosselin, M., Murray, B. J., Polishchuk, E., Abbatt, J. P. D., Miller, L. A. and Bertram, A. K.: Ice-nucleating particles in Canadian Arctic sea-surface microlayer and bulk seawater, Atmos. Chem. Phys., 17(17), 10583–10595, doi:10.5194/acp-17-10583-2017, 2017.

Johnson, M. T., Liss, P. S., Bell, T. G., Lesworth, T. J., Baker, A. R., Hind, A. J., Jickells, T. D., Biswas, K. F., Woodward, E. M. S. and Gibb, S. W.: Field observations of the ocean-atmosphere exchange of ammonia: Fundamental importance of temperature as revealed by a comparison of high and low latitudes, Global Biogeochem. Cycles, 22(1), GB1019, doi:10.1029/2007GB003039, 2008.

Kanji, Z. A. and Abbatt, J. P. D.: Ice Nucleation onto Arizona Test Dust at Cirrus Temperatures: Effect of Temperature and Aerosol Size on Onset Relative Humidity, J. Phys. Chem. A, 114(2), 935–941, doi:10.1021/jp908661m, 2010.

Khlystov, A., Stanier, C. and Pandis, S. N.: An Algorithm for Combining Electrical Mobility and Aerodynamic Size Distributions Data when Measuring Ambient Aerosol Special Issue of Aerosol Science and Technology on Findings from the Fine Particulate Matter Supersites Program, Aerosol Sci. Technol., 38(sup1), 229–238, doi:10.1080/02786820390229543, 2004.

Klein, H., Nickovic, S., Haunold, W., Bundke, U., Nillius, B., Ebert, M., Weinbruch, S., Schuetz, L., Levin, Z., Barrie, L. A.

and Bingemer, H.: Saharan dust and ice nuclei over Central Europe, Atmos. Chem. Phys., 10(21), 10211–10221, doi:10.5194/acp-10-10211-2010, 2010.

Knopf, D. A. and Koop, T.: Heterogeneous nucleation of ice on surrogates of mineral dust, J. Geophys. Res., 111(D12), D12201, doi:10.1029/2005JD006894, 2006.

Knopf, D. A., Alpert, P. A., Wang, B. and Aller, J. Y.: Stimulation of ice nucleation by marine diatoms, Nat. Geosci., 4(2), 88–90, doi:10.1038/ngeo1037, 2011.

Ladino, L. A., Zhou, S., Yakobi-Hancock, J. D., Aljawhary, D. and Abbatt, J. P. D.: Factors controlling the ice nucleating abilities of α -pinene SOA particles, J. Geophys. Res. Atmos., 119(14), 9041–9051, doi:10.1002/2014JD021578, 2014.

Ladino, L. A., Yakobi-Hancock, J. D., Kilthau, W. P., Mason, R. H., Si, M., Li, J., Miller, L. A., Schiller, C. L., Huffman, J. A., Aller, J. Y., Knopf, D. A., Bertram, A. K. and Abbatt, J. P. D.: Addressing the ice nucleating abilities of marine aerosol: A combination of deposition mode laboratory and field measurements, Atmos. Environ., 132, 1–10, doi:10.1016/j.atmosenv.2016.02.028, 2016.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5(3), 715–737, doi:10.5194/acp-5-715-2005, 2005.

Maguhn, J., Karg, E., Kettrup, A. and Zimmermann, R.: On-line Analysis of the Size Distribution of Fine and Ultrafine Aerosol Particles in Flue and Stack Gas of a Municipal Waste Incineration Plant: Effects of Dynamic Process Control Measures and Emission Reduction Devices, Environ. Sci. Technol., 37(20), 4761–4770, doi:10.1021/es020227p, 2003.

Mann, G. W., Carslaw, K. S., Reddington, C. L., Pringle, K. J., Schulz, M., Asmi, A., Spracklen, D. V., Ridley, D. A., Woodhouse, M. T., Lee, L. A., Zhang, K., Ghan, S. J., Easter, R. C., Liu, X., Stier, P., Lee, Y. H., Adams, P. J., Tost, H., Lelieveld, J., Bauer, S. E., Tsigaridis, K., van Noije, T. P. C., Strunk, A., Vignati, E., Bellouin, N., Dalvi, M., Johnson, C. E., Bergman, T., Kokkola, H., von Salzen, K., Yu, F., Luo, G., Petzold, A., Heintzenberg, J., Clarke, A., Ogren, J. A., Gras, J., Baltensperger, U., Kaminski, U., Jennings, S. G., O'Dowd, C. D., Harrison, R. M., Beddows, D. C. S., Kulmala, M., Viisanen, Y., Ulevicius, V., Mihalopoulos, N., Zdimal, V., Fiebig, M., Hansson, H.-C., Swietlicki, E. and Henzing, J. S.: Intercomparison and evaluation of global aerosol microphysical properties among AeroCom models of a range of complexity, Atmos. Chem. Phys., 14(9), 4679–4713, doi:10.5194/acp-14-4679-2014, 2014.

Marmer, E. and Langmann, B.: Aerosol modeling over Europe: 1. Interannual variability of aerosol distribution, J. Geophys. Res., 112(D23), D23S15, doi:10.1029/2006JD008113, 2007.

Marple, V. A., Rubow, K. L. and Behm, S. M.: A Microorifice Uniform Deposit Impactor (MOUDI): Description, Calibration, and Use, Aerosol Sci. Technol., 14(4), 434–446, doi:10.1080/02786829108959504, 1991.

Mason, R. H., Si, M., Li, J., Chou, C., Dickie, R., Toom-Sauntry, D., Pöhlker, C., Yakobi-Hancock, J. D., Ladino, L. A., Jones, K., Leaitch, W. R., Schiller, C. L., Abbatt, J. P. D., Huffman, J. A. and Bertram, A. K.: Ice nucleating particles at a coastal marine boundary layer site: correlations with aerosol type and meteorological conditions, Atmos. Chem. Phys., 15(21), 12547–12566, doi:10.5194/acp-15-12547-2015, 2015a.

Mason, R. H., Chou, C., McCluskey, C. S., Levin, E. J. T., Schiller, C. L., Hill, T. C. J., Huffman, J. A., DeMott, P. J. and

Bertram, A. K.: The micro-orifice uniform deposit impactor–droplet freezing technique (MOUDI-DFT) for measuring concentrations of ice nucleating particles as a function of size: improvements and initial validation, Atmos. Meas. Tech., 8(6), 2449–2462, doi:10.5194/amt-8-2449-2015, 2015b.

Mason, R. H., Si, M., Chou, C., Irish, V. E., Dickie, R., Elizondo, P., Wong, R., Brintnell, M., Elsasser, M., Lassar, W. M., Pierce, K. M., Leaitch, W. R., MacDonald, A. M., Platt, A., Toom-Sauntry, D., Sarda-Estève, R., Schiller, C. L., Suski, K. J., Hill, T. C. J., Abbatt, J. P. D., Huffman, J. A., DeMott, P. J. and Bertram, A. K.: Size-resolved measurements of ice-nucleating particles at six locations in North America and one in Europe, Atmos. Chem. Phys., 16(3), 1637–1651, doi:10.5194/acp-16-1637-2016, 2016.

McKendry, I., Christensen, E., Schiller, C., Vingarzan, R., Macdonald, A. M. and Li, Y.: Low Ozone Episodes at Amphitrite Point Marine Boundary Layer Observatory, British Columbia, Canada, Atmosphere-Ocean, 52(3), 271–280, doi:10.1080/07055900.2014.910164, 2014.

Mertes, S., Verheggen, B., Walter, S., Connolly, P., Ebert, M., Schneider, J., Bower, K. N., Cozic, J., Weinbruch, S., Baltensperger, U. and Weingartner, E.: Counterflow Virtual Impactor Based Collection of Small Ice Particles in Mixed-Phase Clouds for the Physico-Chemical Characterization of Tropospheric Ice Nuclei: Sampler Description and First Case Study, Aerosol Sci. Technol., 41(9), 848–864, doi:10.1080/02786820701501881, 2007.

Möhler, O., Georgakopoulos, D. G., Morris, C. E., Benz, S., Ebert, V., Hunsmann, S., Saathoff, H., Schnaiter, M. and Wagner, R.: Heterogeneous ice nucleation activity of bacteria: new laboratory experiments at simulated cloud conditions, Biogeosciences, 5(5), 1425–1435, doi:10.5194/bg-5-1425-2008, 2008.

Morris, C. E., Georgakopoulos, D. G. and Sands, D. C.: Ice nucleation active bacteria and their potential role in precipitation, J. Phys. IV, 121, 87–103, doi:10.1051/jp4:2004121004, 2004.

Morris, C. E., Sands, D. C., Glaux, C., Samsatly, J., Asaad, S., Moukahel, A. R., Gonçalves, F. L. T. and Bigg, E. K.: Urediospores of rust fungi are ice nucleation active at > −10 °C and harbor ice nucleation active bacteria, Atmos. Chem. Phys., 13(8), 4223–4233, doi:10.5194/acp-13-4223-2013, 2013.

Murray, B. J., Broadley, S. L., Wilson, T. W., Atkinson, J. D. and Wills, R. H.: Heterogeneous freezing of water droplets containing kaolinite particles, Atmos. Chem. Phys., 11(9), 4191–4207, doi:10.5194/acp-11-4191-2011, 2011.

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J. and Leisner, T.: A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles, J. Atmos. Sci., 69(10), 3077–3092, doi:10.1175/JAS-D-11-0249.1, 2012.

O'Dowd, C. D., Becker, E. and Kulmala, M.: Mid-latitude North-Atlantic aerosol characteristics in clean and polluted air, Atmos. Res., 58(3), 167–185, doi:10.1016/S0169-8095(01)00098-9, 2001.

O'Sullivan, D., Murray, B. J., Malkin, T. L., Whale, T. F., Umo, N. S., Atkinson, J. D., Price, H. C., Baustian, K. J., Browse, J. and Webb, M. E.: Ice nucleation by fertile soil dusts: relative importance of mineral and biogenic components, Atmos. Chem. Phys., 14(4), 1853–1867, doi:10.5194/acp-14-1853-2014, 2014.

O'Sullivan, D., Murray, B. J., Ross, J. F., Whale, T. F., Price, H. C., Atkinson, J. D., Umo, N. S. and Webb, M. E.: The relevance of nanoscale biological fragments for ice nucleation in clouds, Sci. Rep., 5(1), 8082, doi:10.1038/srep08082, 2015.

O'Sullivan, D., Murray, B. J., Ross, J. F. and Webb, M. E.: The adsorption of fungal ice-nucleating proteins on mineral dusts: a terrestrial reservoir of atmospheric ice-nucleating particles, Atmos. Chem. Phys., 16(12), 7879–7887, doi:10.5194/acp-16-7879-2016, 2016.

Prenni, A. J., Petters, M. D., Kreidenweis, S. M., Heald, C. L., Martin, S. T., Artaxo, P., Garland, R. M., Wollny, A. G. and Pöschl, U.: Relative roles of biogenic emissions and Saharan dust as ice nuclei in the Amazon basin, Nat. Geosci., 2(6), 402–405, doi:10.1038/ngeo517, 2009.

Rosinski, J., Haagenson, P. L., Nagamoto, C. T. and Parungo, F.: Ice-forming nuclei of maritime origin, J. Aerosol Sci., 17(1), 23–46, doi:10.1016/0021-8502(86)90004-2, 1986.

Rosinski, J., Haagenson, P. L., Nagamoto, C. T., Quintana, B., Parungo, F. and Hoyt, S. D.: Ice-forming nuclei in air masses over the Gulf of Mexico, J. Aerosol Sci., 19(5), 539–551, doi:10.1016/0021-8502(88)90206-6, 1988.

Saleh, R., Donahue, N. M. and Robinson, A. L.: Time Scales for Gas-Particle Partitioning Equilibration of Secondary Organic Aerosol Formed from Alpha-Pinene Ozonolysis, Environ. Sci. Technol., 47(11), 5588–5594, doi:10.1021/es400078d, 2013.

Schnell, R. C.: Ice Nuclei in Seawater, Fog Water and Marine Air off the Coast of Nova Scotia: Summer 1975, J. Atmos. Sci., 34(8), 1299–1305, doi:10.1175/1520-0469(1977)034<1299:INISFW>2.0.CO;2, 1977.

Schnell, R. C.: Airborne ice nucleus measurements around the Hawaiian Islands, J. Geophys. Res., 87(C11), 8886, doi:10.1029/JC087iC11p08886, 1982.

Schnell, R. C. and Vali, G.: Freezing nuclei in marine waters, Tellus, 27(3), 321–323, doi:10.1111/j.2153-3490.1975.tb01682.x, 1975.

Schnell, R. C. and Vali, G.: Biogenic Ice Nuclei: Part I. Terrestrial and Marine Sources, J. Atmos. Sci., 33(8), 1554–1564, doi:10.1175/1520-0469(1976)033<1554:BINPIT>2.0.CO;2, 1976.

Spracklen, D. V. and Heald, C. L.: The contribution of fungal spores and bacteria to regional and global aerosol number and ice nucleation immersion freezing rates, Atmos. Chem. Phys., 14(17), 9051–9059, doi:10.5194/acp-14-9051-2014, 2014.

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D. and Ngan, F.: NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, Bull. Am. Meteorol. Soc., 96(12), 2059–2077, doi:10.1175/BAMS-D-14-00110.1, 2015.

Tobo, Y., Prenni, A. J., DeMott, P. J., Huffman, J. A., McCluskey, C. S., Tian, G., Pöhlker, C., Pöschl, U. and Kreidenweis, S. M.: Biological aerosol particles as a key determinant of ice nuclei populations in a forest ecosystem, J. Geophys. Res. Atmos., 118(17), 10,100-10,110, doi:10.1002/jgrd.50801, 2013.

Tobo, Y., DeMott, P. J., Hill, T. C. J., Prenni, A. J., Swoboda-Colberg, N. G., Franc, G. D. and Kreidenweis, S. M.: Organic matter matters for ice nuclei of agricultural soil origin, Atmos. Chem. Phys., 14(16), 8521–8531, doi:10.5194/acp-14-8521-2014, 2014.

Tunved, P., Ström, J. and Krejci, R.: Arctic aerosol life cycle: linking aerosol size distributions observed between 2000 and

2010 with air mass transport and precipitation at Zeppelin station, Ny-Ålesund, Svalbard, Atmos. Chem. Phys., 13(7), 3643–3660, doi:10.5194/acp-13-3643-2013, 2013.

Vali, G.: Quantitative Evaluation of Experimental Results an the Heterogeneous Freezing Nucleation of Supercooled Liquids, J. Atmos. Sci., 28(3), 402–409, doi:10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2, 1971.

Vali, G., DeMott, P. J., Möhler, O. and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem. Phys., 15(18), 10263–10270, doi:10.5194/acp-15-10263-2015, 2015.

Vergara-Temprado, J., Murray, B. J., Wilson, T. W., O&apos;Sullivan, D., Browse, J., Pringle, K. J., Ardon-Dryer, K., Bertram, A. K., Burrows, S. M., Ceburnis, D., DeMott, P. J., Mason, R. H., O&apos;Dowd, C. D., Rinaldi, M. and Carslaw, K. S.: Contribution of feldspar and marine organic aerosols to global ice nucleating particle concentrations, Atmos. Chem. Phys., 17(5), 3637–3658, doi:10.5194/acp-17-3637-2017, 2017.

Wang, X., Sultana, C. M., Trueblood, J., Hill, T. C. J., Malfatti, F., Lee, C., Laskina, O., Moore, K. A., Beall, C. M., McCluskey, C. S., Cornwell, G. C., Zhou, Y., Cox, J. L., Pendergraft, M. A., Santander, M. V., Bertram, T. H., Cappa, C. D., Azam, F., DeMott, P. J., Grassian, V. H. and Prather, K. A.: Microbial Control of Sea Spray Aerosol Composition: A Tale of Two Blooms, ACS Cent. Sci., 1(3), 124–131, doi:10.1021/acscentsci.5b00148, 2015.

Westbrook, C. D. and Illingworth, A. J.: Evidence that ice forms primarily in supercooled liquid clouds at temperatures > −27°C, Geophys. Res. Lett., 38(14), L14808, doi:10.1029/2011GL048021, 2011.

Wex, H., Demott, P. J., Tobo, Y., Hartmann, S., Rösch, M., Clauss, T., Tomsche, L., Niedermeier, D. and Stratmann, F.: Kaolinite particles as ice nuclei: Learning from the use of different kaolinite samples and different coatings, Atmos. Chem. Phys., 14(11), 5529–5546, doi:10.5194/acp-14-5529-2014, 2014.

Wheeler, M. J., Mason, R. H., Steunenberg, K., Wagstaff, M., Chou, C. and Bertram, A. K.: Immersion Freezing of Supermicron Mineral Dust Particles: Freezing Results, Testing Different Schemes for Describing Ice Nucleation, and Ice Nucleation Active Site Densities, J. Phys. Chem. A, 119(19), 4358–4372, doi:10.1021/jp507875q, 2015.

Wilson, T. W., Ladino, L. A., Alpert, P. A., Breckels, M. N., Brooks, I. M., Browse, J., Burrows, S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G., Miller, L. A., Nájera, J. J., Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Temprado, J. V., Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y., Bertram, A. K., Knopf, D. A. and Murray, B. J.: A marine biogenic source of atmospheric ice-nucleating particles, Nature, 525(7568), 234–238, doi:10.1038/nature14986, 2015.

Worringen, A., Kandler, K., Benker, N., Dirsch, T., Mertes, S., Schenk, L., Kästner, U., Frank, F., Nillius, B., Bundke, U., Rose, D., Curtius, J., Kupiszewski, P., Weingartner, E., Vochezer, P., Schneider, J., Schmidt, S., Weinbruch, S. and Ebert, M.: Single-particle characterization of ice-nucleating particles and ice particle residuals sampled by three different techniques, Atmos. Chem. Phys., 15(8), 4161–4178, doi:10.5194/acp-15-4161-2015, 2015.

Yakobi-Hancock, J. D., Ladino, L. A., Bertram, A. K., Huffman, J. A., Jones, K., Leaitch, W. R., Mason, R. H., Schiller, C. L., Toom-Sauntry, D., Wong, J. P. S. and Abbatt, J. P. D.: CCN activity of size-selected aerosol at a Pacific coastal location, Atmos. Chem. Phys., 14(22), 12307–12317, doi:10.5194/acp-14-12307-2014, 2014.

Yun, Y. and Penner, J. E.: An evaluation of the potential radiative forcing and climatic impact of marine organic aerosols as heterogeneous ice nuclei, Geophys. Res. Lett., 40(15), 4121–4126, doi:10.1002/grl.50794, 2013.

Table 1. The three sampling locations used in this study and conditions during sampling including average ambient temperature (T) and relative humidity (RH).

Location	Coordinates	Sampling period	Average sampling time (h)	Average ambient T (ºC)	Average ambient RH (%)	Average wind speed (m/s)
Amphitrite Point, BC, Canada	48.92º N, 125.54º W	6-27 August 2013	7.8	14	97	4.0
Labrador Sea, NL, Canada	54.59º N, 55.61º W	11 July 2014	6.2	10.9	70	5.4
Lancaster Sound, NU, Canada	74.26º N, 91.46º W	20 July 2014	5.3	2.8	95	4.6

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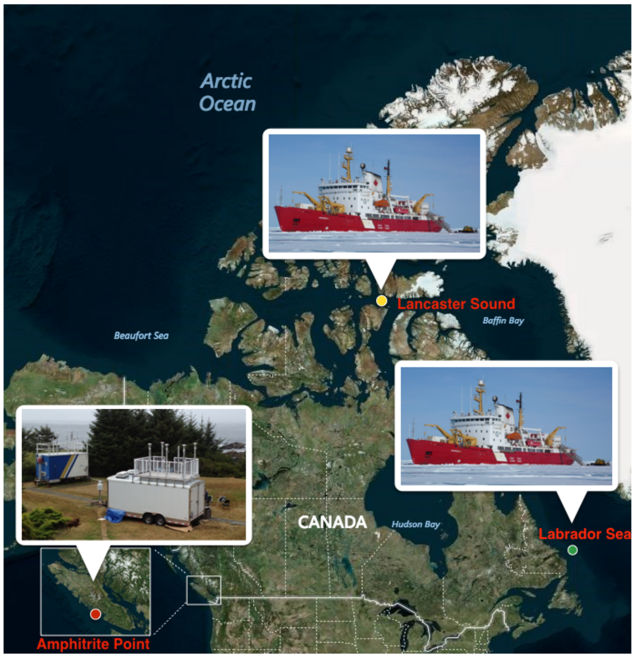

Figure 1. Map showing the three sampling locations: Amphitrite Point (red dot), Labrador Sea (green dot), and Lancaster Sound (yellow dot). Inserts show the images of the sampling platform used at each location.

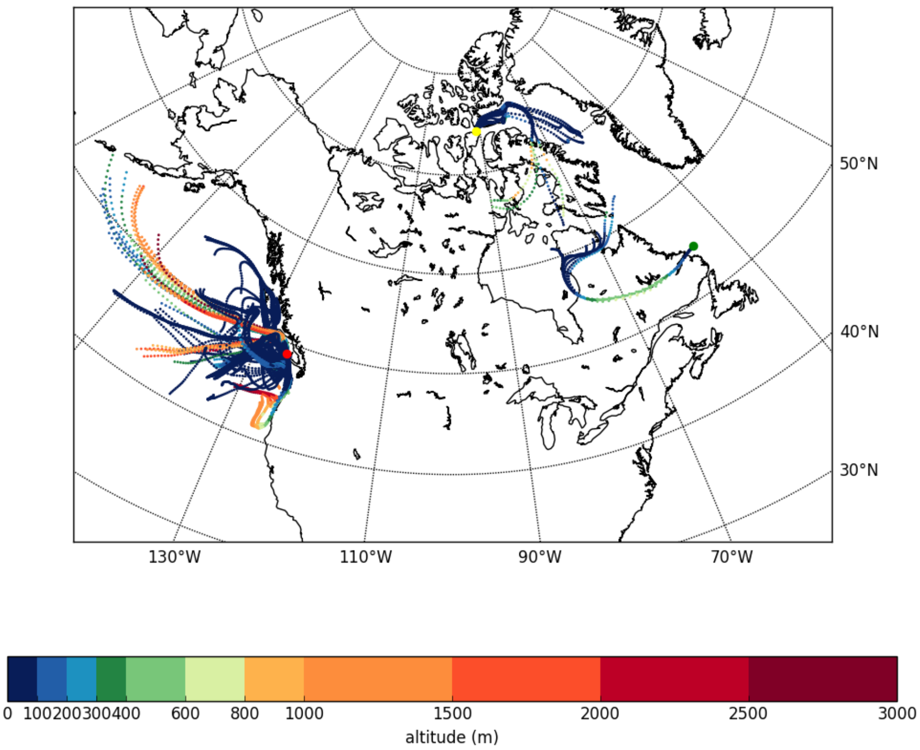

Figure 2. The 3-day HYSPLIT back trajectories for Amphitrite Point (red dot), Labrador Sea (green dot) and Lancaster Sound (yellow dot). The back trajectories were calculated for every hour during the MOUDI sampling period. The altitude is indicated with the colour scale. Global Data Assimilation System (GDAS) meteorological data at 1º x 1º spatial resolution was used as input to calculate the back trajectories using HYSPLIT.

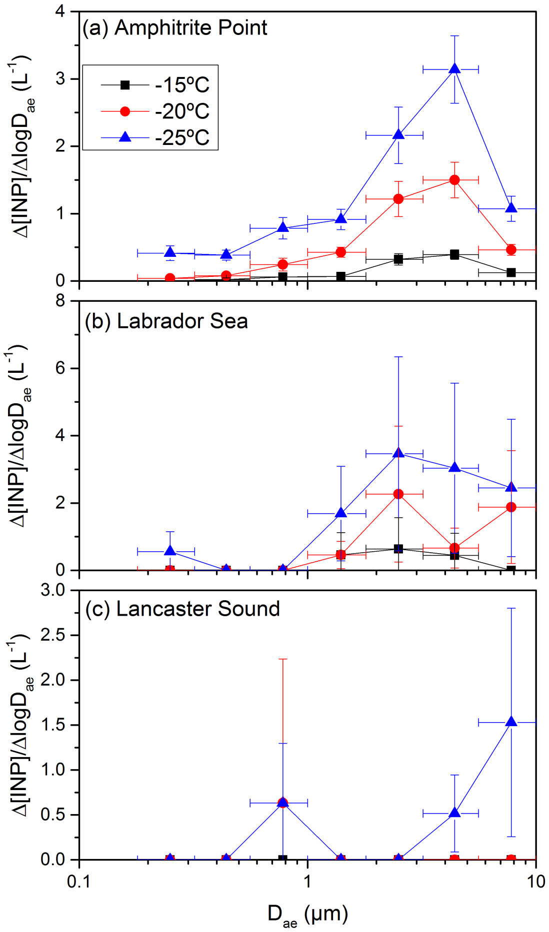

Figure 3. Average INP number concentrations at freezing temperatures of -15 ºC, -20 ºC, and -25 ºC as a function of aerodynamic diameter (*D_{ae}*) for the three sites studied. The x-error bars represent the widths of the size bins from the MOUDI. For the Amphitrite Point samples, standard error of the mean was used to represent the uncertainty of INP concentrations during the month. At both Labrador Sea and Lancaster Sound, only one MOUDI sample was collected, and we assume the monthly INP concentrations have the same normal distribution as the Amphitrite Point samples. Hence for the y error bars at these locations, we assume the relative standard deviation for supermicron and submicron particles were the same as the relative standard deviation for supermicron and submicron particles observed in the Amphitrite Point data.

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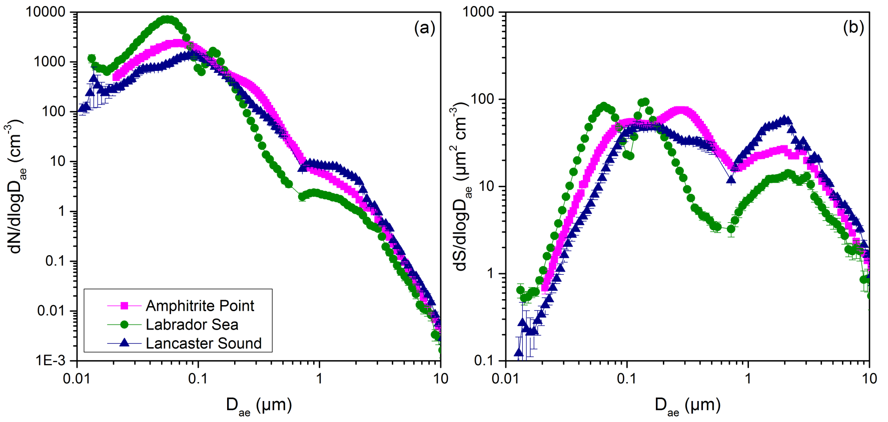

Figure 4. Concentrations of (a) aerosol number, *N*, and (b) surface area, *S*, as a function of aerodynamic diameter, D_{ae}. The y-error bars represent the standard error of the mean for each size bin. In many cases, the error bars are smaller than the size of the symbols. For cases where a gap existed between the SMPS data and the APS data, a straight line was used to extrapolate the data.

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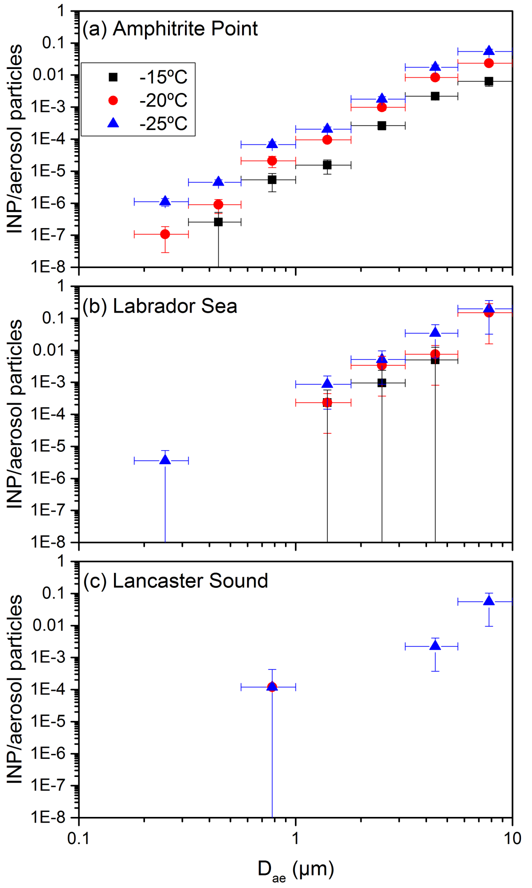

Figure 5. The fraction of aerosol particles acting as an INP as a function of aerodynamic diameter (D_{ae}) at -15 ºC, -20 ºC, and -25 ºC, respectively. The x-error bars represent the widths of the size bins, and the y-error bars are the propagated uncertainties from INP concentrations as a function of size (Fig. 3) and aerosol number concentrations as a function of size (Fig. S4a). In some cases, the y-error bars are smaller than the size of the symbols.

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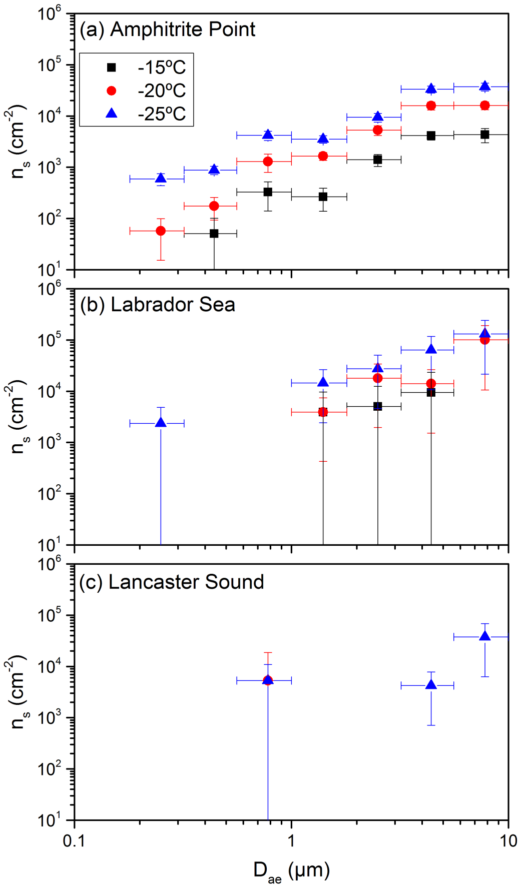

Figure 6. Surface active site density, n_s, as a function of aerodynamic diameter (D_{ae}) at -15 ºC, -20 ºC, and -25 ºC, respectively. The x-errors represent the widths of the size bins, and the y-errors are the propagated uncertainties from INP concentrations as a function of size (Fig. 3) and aerosol surface area concentrations as a function of size (Fig. S4b). In some cases, the y-error bars are smaller than the size of the symbols.

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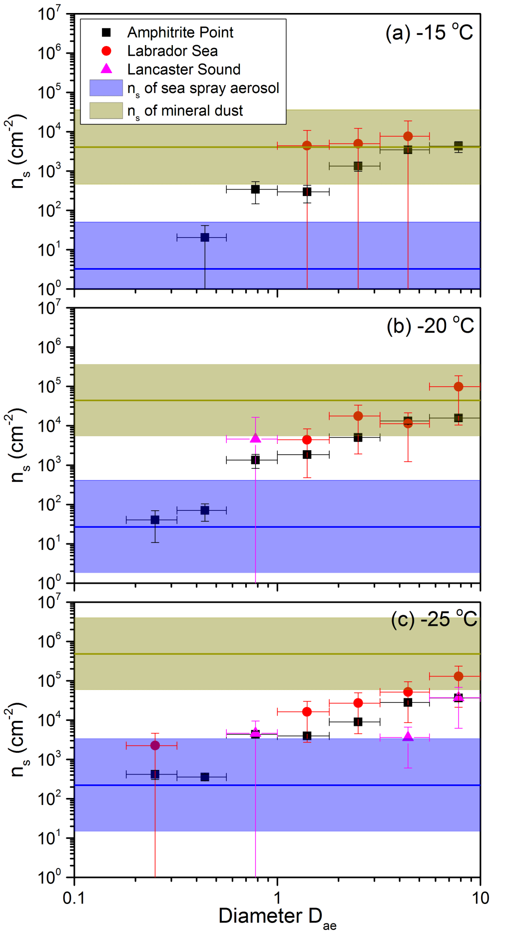

Figure 7. Comparison of measured *n_s* values with previously reported *n_s* values of sea spray aerosol and mineral dust at -15 ºC, -20 ºC, and -25 ºC, respectively. The *n_s* values of sea spray aerosol were taken from DeMott et al. (2016), and the *n_s* values of mineral dust were taken from Niemand et al. (2012). The horizontal lines represent the calculated *n_s* values from linear regression, and the coloured bands represent the 95 % prediction bands (see Fig. S5-S6 in the Supplement). Blue represents sea spray aerosol, and light green represents mineral dust.

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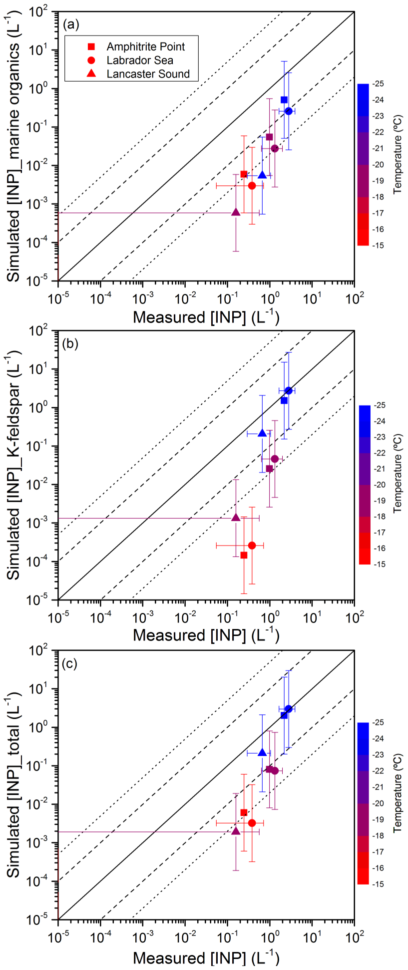

Figure 8. Comparison of measured INP concentrations and (a) simulated INP concentrations from marine organics, (b) simulated INP concentrations from K-feldspar, and (c) simulated INP concentrations from both. The solid lines represent 1:1 ratio, the

dashed and dotted lines represent one order and 1.5 orders of magnitude difference, respectively. The temperature is shown using a color scale. The simulated INP concentrations for Amphitrite Point, Labrador Sea, and Lancaster Sound correspond to mean concentrations for the months of August, July, and July, respectively. The uncertainties in the simulated concentrations are estimated to be around one order of magnitude based on the parameterization and model uncertainty (Harrison et al., 2016; Wilson et al., 2015).

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INP number concentrations were calculated using the following equation:

$$[INPs(T)] = -ln\left(\frac{N_u(T)}{N_0}\right)N_0\left(\frac{A_{deposit}}{A_{DFT}V}\right)f_{nu,1\ mm}f_{nu,0.25-1\ mm}$$,

(1)

Supplemental material

S1 Conversion of mobility diameter measured at Labrador Sea and Lancaster Sound to aerodynamic diameter

The SMPS measured mobility diameter rather than aerodynamic diameter, while both the APS and the MOUDI measured aerodynamic diameter. To allow comparison between the SMPS data, the APS data and the INP data, the mobility diameter, D_m, measured by the SMPS was converted to aerodynamic diameter, D_{ae}, using the following equation (Khlystov et al., 2004):

$$D_{ae} = \sqrt{\frac{\rho_{p,RH}}{\chi \rho_o}} D_m, \qquad \text{(S1)}$$

where χ is the dynamic shape factor that accounts for the non-spherical particle shape; ρ_o is the unit density of 1 g cm^{-3}; and $\rho_{p,RH}$ is the particle density at the sampling RH. In all cases, we assumed a dynamic shape factor of 1. The particle density at the sampling RH, $\rho_{p,RH}$, was calculated using the following equation:

$$\rho_{p,RH} = \rho_w + (\rho_{p,dry} - \rho_w)\frac{1}{gf^3}, \qquad \text{(S2)}$$

where ρ_w is the density of water; $\rho_{p,dry}$ is the density of the dry particles; *gf* is the hygroscopic growth factor. The hygroscopic growth factor was based on the numerical model developed by Ming and Russell (2001) assuming the sampled aerosol consisted of sea spray aerosol with a 30 % organic mass content, following the assumption made in DeMott et al. (2016). This assumption results in growth factors consistent with measurements in the marine boundary layer (Zhou et al., 2001). For the density of the dry particles, we also assumed a sea spray aerosol with a 30 % organic mass content, resulting in a dry density of 1.87 g cm^{-3}. To determine the sensitivity of the size distribution to the assumed composition of the aerosol, calculations were also carried out assuming a sea spray aerosol with a 10 % organic mass content and a 50 % organic mass content. The difference in the resulted size distributions assuming 10 %, 30 %, and 50 % organic mass content is small (see Fig. S7); hence, data shown in the main text only correspond to an assumed composition of a sea spray aerosol with a 30 % organic mass content.

S2 Conversion of mobility diameter to aerodynamic diameter and correction for hygroscopic growth at Amphitrite Point

At Amphitrite Point, dryers were used prior to sampling with the SMPS. As a result, SMPS data needs to be corrected for hygroscopic growth, and the mobility diameter needs to be converted to aerodynamic diameter. The equation to correct for hygroscopic growth is the following:

$$D_{m,RH} = gf \times D_{m,dry}, \qquad \text{(S3)}$$

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where, $D_{m,RH}$ is the mobility diameter at the sampling RH; $D_{m,dry}$ is the mobility diameter under dry condition. The relationship between mobility diameter and aerodynamic diameter is given in Eq. (S1). Combining Eq. (S1) and Eq. (S3) results in the following:

$$D_{ae,RH} = gf\sqrt{\frac{\rho_{p,RH}}{\chi\rho_o}}D_{m,dry} = xD_{m,dry}, \quad \text{(S4)}$$

where $D_{ae,RH}$ is the aerodynamic diameter at the sampling RH, and $x = gf\sqrt{\frac{\rho_{p,RH}}{\chi\rho_o}}$.

Equation (S4) illustrates that the relationship between the dry mobility diameter and the wet aerodynamic diameter is a simple factor x. To determine x, we varied this factor until the optimum fit was obtained between the SMPS and the APS data where overlap occurred (0.7 to 0.93 µm). This type of approach has been used successfully in the past to merge the SMPS and APS data (Beddows et al., 2010; Khlystov et al., 2004). Note, we did not use this approach in Sect. S1 since there was no overlapping size range between the SMPS and APS data measured at Labrador Sea and Lancaster Sound to allow an optimization of the fit.

References:

Beddows, D. C. S., Dall'osto, M. and Harrison, R. M.: An Enhanced Procedure for the Merging of Atmospheric Particle Size Distribution Data Measured Using Electrical Mobility and Time-of-Flight Analysers, Aerosol Sci. Technol., 44(11), 930–938, doi:10.1080/02786826.2010.502159, 2010.

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F. and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, Proc. Natl. Acad. Sci., 113(21), 5797–5803, doi:10.1073/pnas.1514034112, 2016.

Khlystov, A., Stanier, C. and Pandis, S. N.: An Algorithm for Combining Electrical Mobility and Aerodynamic Size Distributions Data when Measuring Ambient Aerosol Special Issue of Aerosol Science and Technology on Findings from the Fine Particulate Matter Supersites Program, Aerosol Sci. Technol., 38(sup1), 229–238, doi:10.1080/02786820390229543, 2004.

Ming, Y. and Russell, L. M.: Predicted hygroscopic growth of sea salt aerosol, J. Geophys. Res. Atmos., 106(D22), 28259–28274, doi:10.1029/2001JD000454, 2001.

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J. and Leisner, T.: A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles, J. Atmos. Sci., 69(10), 3077–3092, doi:10.1175/JAS-D-11-0249.1, 2012.

O'Dowd, C. D., Becker, E. and Kulmala, M.: Mid-latitude North-Atlantic aerosol characteristics in clean and polluted air, Atmos. Res., 58(3), 167–185, doi:10.1016/S0169-8095(01)00098-9, 2001.

Zhou, J., Swietlicki, E., Berg, O. H., Aalto, P. P., Hämeri, K., Nilsson, E. D. and Leck, C.: Hygroscopic properties of aerosol particles over the central Arctic Ocean during summer, J. Geophys. Res. Atmos., 106(D23), 32111–32123, doi:10.1029/2000JD900426, 2001.

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Table S1. The correction factors $f_{nu,1\ mm}$ and $f_{nu,0.25-0.1\ mm}$ for MOUDI stages 2-8 when using substrate holders. The uncertainty in $f_{nu,1\ mm}$ is given as the standard deviation.

MOUDI Stages	$f_{nu,1\ mm}$	$f_{nu,0.25-0.1\ mm}$
2	0.74, +0.18, −0.12	0.1225exp(−11.29μ)+1.065exp(−0.06412μ)
3	0.72, +0.08, −0.08	0.04718exp(−14.15μ)+1.023exp(−0.02347μ)
4	1.18, +0.09, −0.14	0.04252exp(−13.06μ)+1.024exp(−0.02386μ)
5	0.97, +0.03, −0.10	0.03023exp(−14.97μ)+1.015exp(−0.01515μ)
6	0.75, +0.19, −0.02	0.5799exp(−10.57μ)+1.148exp(−0.1408μ)
7	0.84, +0.07, −0.11	0.1151exp(−10.66μ)+1.072exp(−0.07029μ)
8	1.01, +0.03, −0.12	1.03exp(−12.79μ)+1.268exp(−0.2422μ)

$\mu = \frac{N_u(T)}{N_0}$, where $N_u(T)$ is the number of unfrozen droplets at temperature T, and N_0 is the total number of droplets in one freezing experiment.

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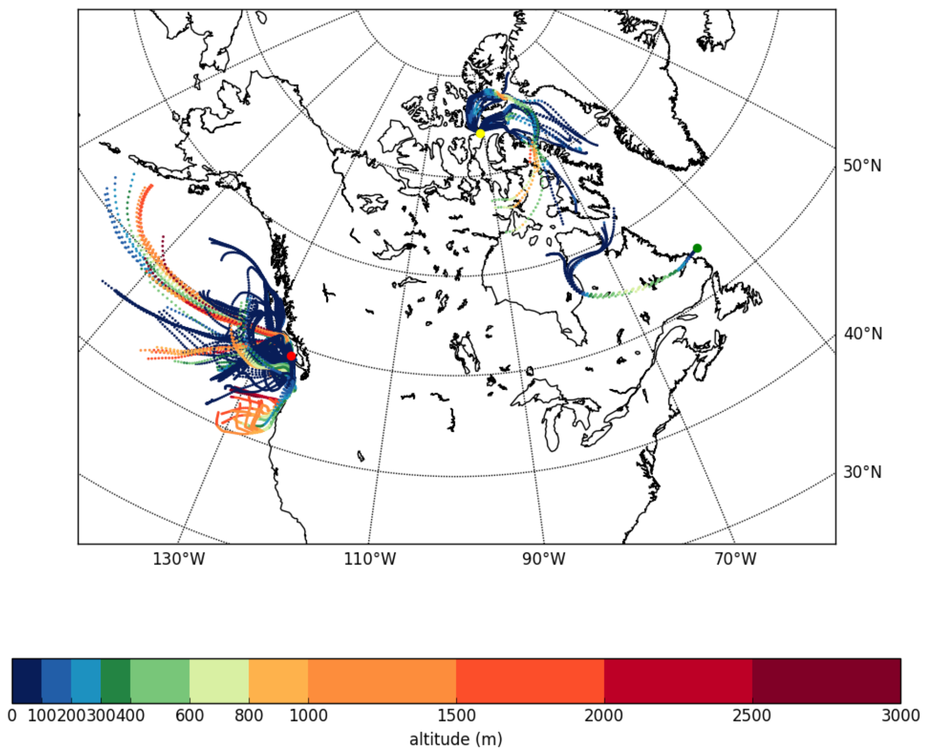

Figure S1. The 3-day HYSPLIT back trajectories initiating at 50 m above ground level for Amphitrite Point (red dot), Labrador Sea (green dot) and Lancaster Sound (yellow dot). The back trajectories were calculated for every hour during the MOUDI sampling period. The starting points are labeled as coloured dots, and the altitude is shown using a colour scale.

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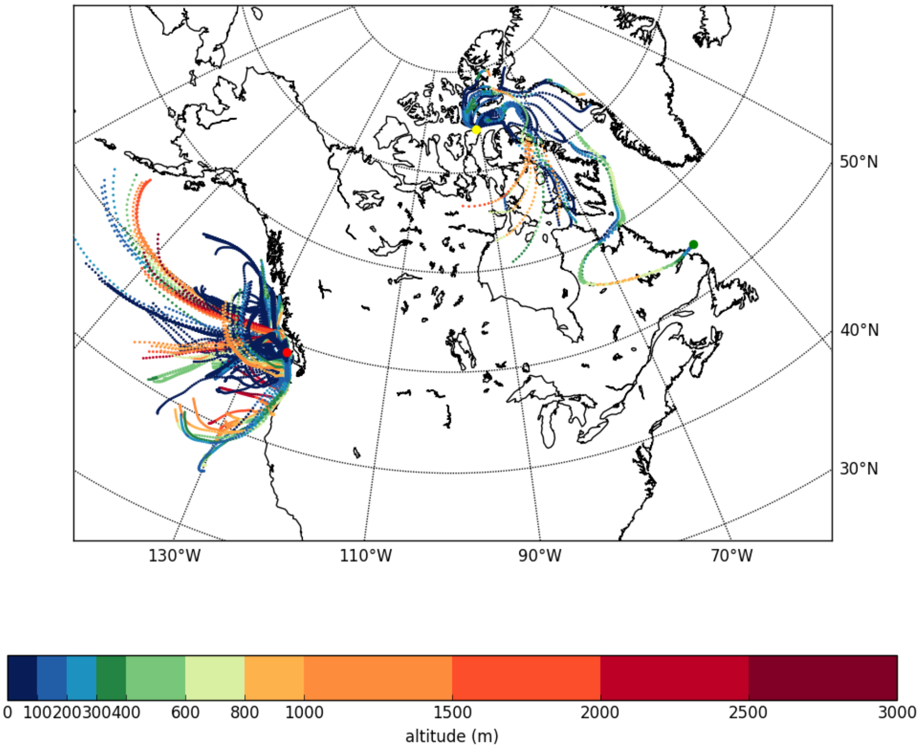

Figure S2. The 3-day HYSPLIT back trajectories initiating at 150 m above ground level for Amphitrite Point (red dot), Labrador Sea (green dot) and Lancaster Sound (yellow dot). The back trajectories were calculated for every hour during the MOUDI sampling period. The starting points are labeled as coloured dots, and the altitude is shown using a colour scale.

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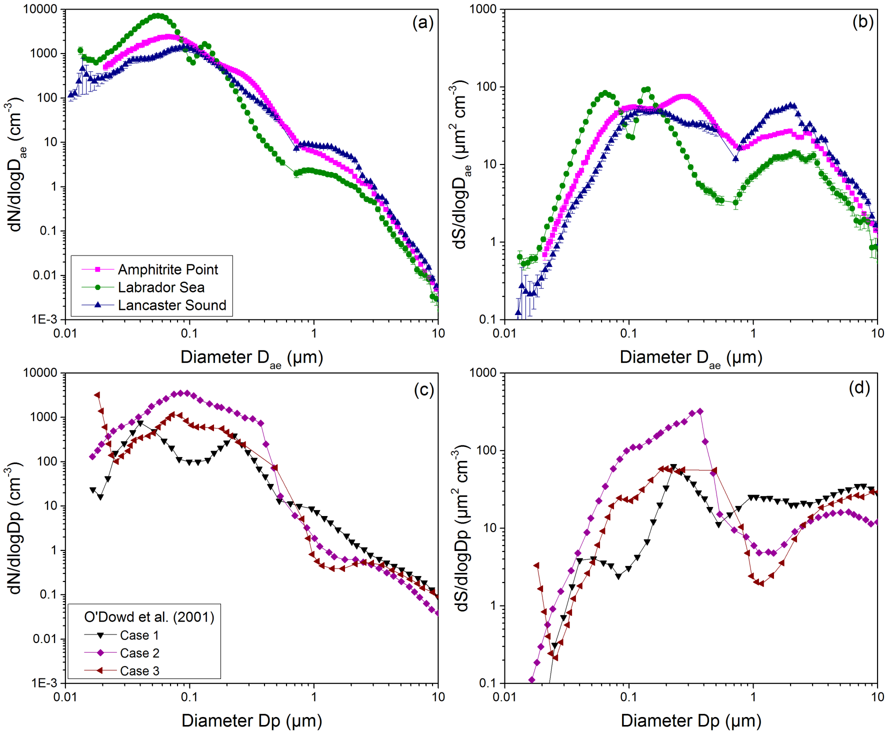

Figure S3. Comparison of aerosol particle number and surface area size distributions measured in this study (a, b) with aerosol particle number and surface area size distributions measured at a mid-latitude North-Atlantic marine boundary layer site (c, d) (O'Dowd et al., 2001)**. Case 1 from** O'Dowd et al. (2001) **corresponds to clean marine air measured under moderate humidity (80 %) and wind speeds (6 m s^{-1}) conditions, Case 2 corresponds to anthropogenically influenced maritime air measured at wind speeds in the order of 2-4 m s^{-1}, and Case 3 corresponds to anthropogenically influenced maritime air during a nucleation burst measured at wind speeds of 4 m s^{-1}**

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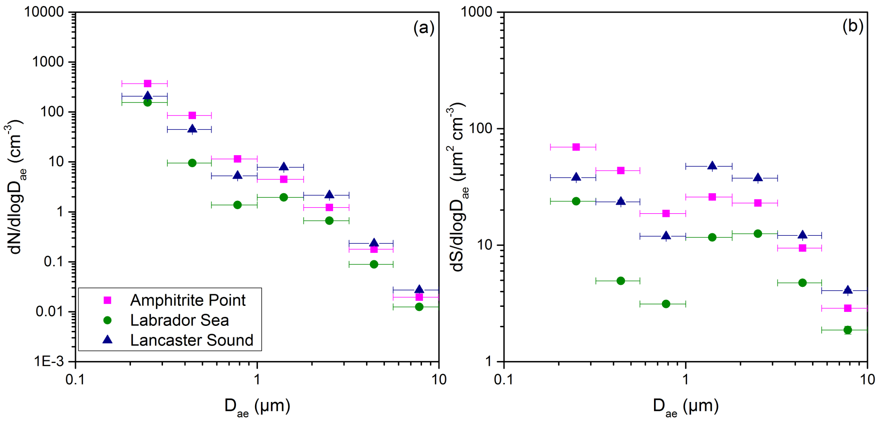

Figure S4. Concentrations of (a) aerosol number, *N*, and (b) surface area, *S*, as a function of aerodynamic diameter, D_{ae}, using the same bin widths as the MOUDI. Each data point was calculated by averaging the numbers from Fig. 4 that were within the corresponding size bin. The x-error bars represent the widths of the size bins, and the y-error bars are propagated uncertainties from the error bars in Fig. 4. In most cases, the y-error bars are smaller than the size of the symbols.

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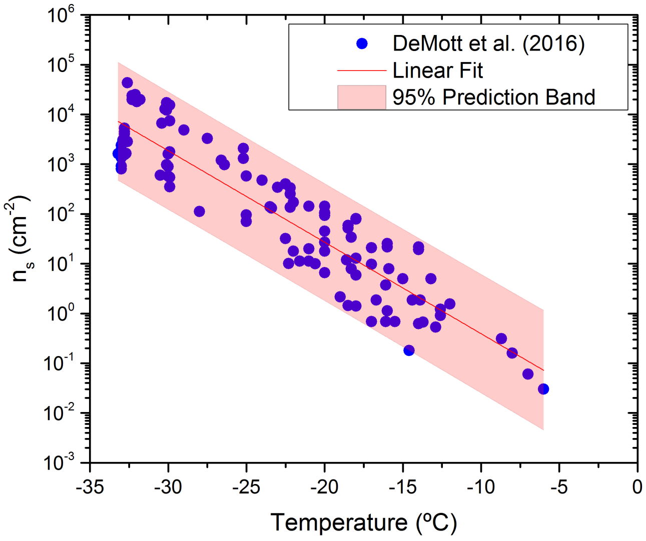

Figure S5. n_s values of sea spray aerosol as a function of temperature taken from DeMott et al. (2016). Shown is a linear fit to the data and 95% prediction bands.

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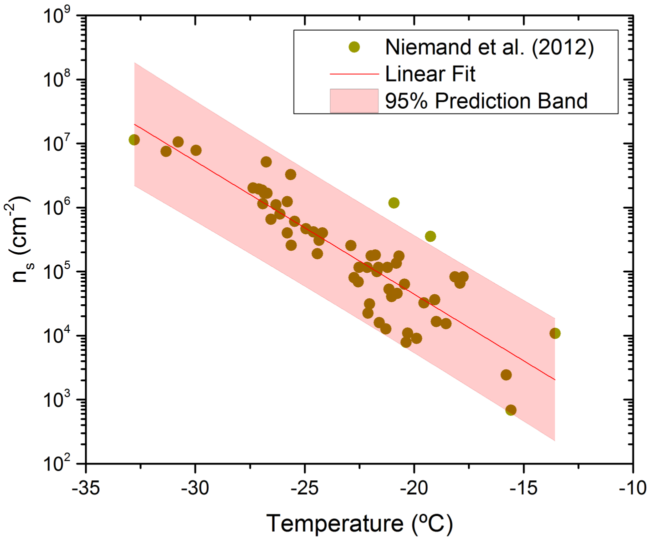

Figure S6. n_s values of mineral dust as a function of temperature taken from Niemand et al. (2012). Shown is a linear fit to the data and 95% prediction bands.

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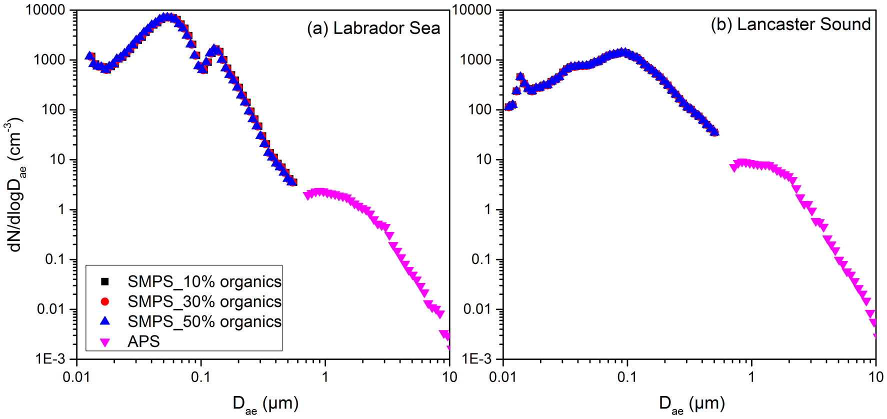

Figure S7. Number size distribution measured at Labrador Sea (a) and Lancaster Sound (b) with the mobility diameter measured by the SMPS converted to aerodynamic diameter assuming a sea spray aerosol with 10 %, 30 %, and 50 % organic mass content.

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S1 Corrections for hygroscopic growth

A dryer was used prior to sampling with the SMPS at the Amphitrite Point site. To allow comparison with other measurements, the SMPS data at Amphitrite Point were corrected for hygroscopic growth using the following equation (Hämeri et al., 2000):

$$gf(RH) = \frac{D_{p,RH}}{D_{p,dry}}, \qquad \text{(S1)}$$

where *gf (RH)* is the hygroscopic growth factor at measured relative humidity (RH); $D_{p,RH}$ **is the particle diameter at measured RH;** $D_{p,dry}$ **is the dry particle diameter. The hygroscopic growth factor was calculated with the numerical model developed by Ming and Russell (2001) assuming the sampled aerosol consisted of sea spray aerosol with a 30% organic mass content, following the assumption made in DeMott et al. (2016). This assumption results in growth factors consistent with measurements in the marine boundary layer (Zhou et al., 2001) and a hygroscopicity parameter, κ, consistent with measurements at Amphitrite Point during the same campaign (Yakobi-Hancock et al., 2014).**

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