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 database 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 Conclusions, we will point out the amount of data used in the study. In the Results and Discussion, we will discuss in more detail the uncertainty in the results due to the limited statistics.

[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] Thank you for encouraging us to look more carefully at the particle number size distributions. After going back and investigating this data in more detail we conclude that the gap between the SMPS and APS data is more likely due to (1) drop off in the efficiency of the APS at size channels below 0.7 \( \mu m \) and (2) uncertainty of the hygroscopic properties at Amphitrite Point used to correct the SMPS data for hygroscopic growth. Regarding (1), drop off in the efficiency of the APS at sizes below 0.7 \( \mu m \) has been often observed (Beddows et al., 2010). To address this issue, in the revised manuscript, the APS data at sizes below 0.7 \( \mu m \) will be omitted, as done previously (Maguhn et al., 2003). Regarding (2), in the revised manuscript, we will use a different method to correct the SMPS data for hygroscopic growth at Amphitrite Point. Specifically, we will use a free parameter to correct for the hygroscopic growth, which results 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 the SMPS and APS data (Beddows et al., 2010).

To address the referee’s comment, we have also compared the SMPS and APS measurements from Amphitrite Point with additional measurements at Amphitrite Point that were carried out during the same field campaign. In the revised manuscript we will include this comparison. In addition, we will include 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).

[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 will be added to 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 will be made clear in the revised manuscript. In addition, to address the referee’s comments, possible inter-annual variability in the model output will be discussed in the revised manuscript.

[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.


Minor and technical comments:

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

[A6] The word “particle” will be added before “size”.

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)

To address the referee’s comment, the following sentence will be added to Section 2.1.2:

“The temperature was decreased to approximately 0 °C, 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 deposit particles.”

equation (1): There seems to be a typo in the formula: N₀ 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.

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) will be separated into two equations. In this case, the consistency between our calculations and the equation in Hader et al. (2014) should be more obvious.

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?

A RH below 2% was reached by using three successive dryers. This low RH was confirmed using a Vaisala hygrometer MI70. This information will be added to the revised manuscript.

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

The units will be changed to SI units (km h⁻¹).

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.

The drying effect due to 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.

page 7, line 6 and page 10, line 12: The “-“ is missing for Vergara-Temprado.
Thanks for pointing out this mistake. It will be corrected.

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.”

Thanks for the advice. The sentence will be rephrased as suggested.

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

Thanks for pointing out this mistake. The date in chapter 2.3.1 will be corrected.

Clearly state how many samples you used for the present study.

To address the referee’s comment, the number of samples will be stated.

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.

To address the referee’s comment, in the revised manuscript we will add the locations of the other studies referenced.

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.

Thanks for pointing this out. The result from Mertes et al. (2007) will be cited in the revised manuscript.

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.

To address the referee’s comment we will clearly state that the \( n_s \) values we report corresponds to the total aerosol particles whereas the \( n_s \) values from Niemand et al. (2012) corresponds to only dust particles.

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, a 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] To address the referee’s comment, stronger motivation will be provided.

[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 will be 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 will use 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 will be corrected in the revised Supplement.

Literature:


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] For typical atmospheric conditions, the equilibration timescale for gas-particle partitioning is on the order of minutes to tens of minutes for secondary organic aerosol (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 species during drying should not have been an issue in our experiments.

[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. In the revised manuscript we will clearly state that the \( n_s \) values we report corresponds to the total aerosol particles, and the \( n_s \) values correspond to a lower limit of the \( n_s \) values of 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 averaging 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] 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 release enough INPs to overwhelm a small INP source from the ocean. To address the referee’s comment, this information will be added to the revised manuscript.

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] This sentence will be re-worded.

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

[A27] A reference will be added.

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

[A28] Stage 2 through stage 8 were analyzed (seven stages in total). This sentence will be reworded to make this point clear in the revised manuscript.

[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 will be added to Section 2.1.2 in the revised manuscript.

[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 °C. Samples were not frozen. To address the referee’s comments, in the revised manuscript, we will point out that additional studies are needed to determine the
effect of storage on 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 will expand the discussion in Section 3.1 on possible terrestrial sources of INP.

[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] A 10-year study of aerosol size distribution at an Arctic station, Zeppelin station, Svalbard, showed that new particle formation events are rather common in the summer Arctic as a result of photochemical production in combination with low condensation sink (Tunved et al., 2013). The seasonal variation of aerosol concentrations reported in this 10-year study showed that the highest concentration was observed in July. In addition, new particle formation has been observed in the Canadian Arctic marine boundary layer during the summer (Burkart et al., 2017). To address the referee’s comments, the standard deviation of the average total number concentration will be added. 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.

[33] P12, l439 (Page 9, line 6 in the ACPD version): Please provide equation for $n_s$.

[A33] An equation for $n_s$ will be added to the revised manuscript.

[34] P14, l409 (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 marker of marine biological activities. This information will be added to 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] Regarding the first point, 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. This information will be added to the revised manuscript.

Regarding the second point, the apparent inconsistency between the results in this manuscript (94 % of the time over the ocean) and the results from Mason et al. (2015a) (88 % of the time over the ocean) is due to the difference in the samples included in the analysis: in the current study, we only used 23 sets of samples, while Mason et al. (2015a) used 34 sets of samples. Nevertheless, the conclusions in our manuscript are consistent with the conclusions in Mason et al. (2015a).

[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 will be changed to a log scale, so the 0-600 m range is easier to differentiate.
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. INP concentrations at the surface might be significantly different and possibly decoupled from INP concentrations above the boundary layer. To address the referee’s comments, this information will be included in the revised manuscript.

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 will be 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.
The limitations of $n_s$ will be discussed in the revised manuscript.

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.

To address the referee’s comments, additional discussion will be added to this location.

Technical correction:

$n_s$ 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.

“Efficiency” will be changed to “ability” in the revised manuscript.

Literature list from referee #3


References for author’s response:


