Title: Size-resolved measurements of ice nucleating particles at six locations in North America and one in Europe

Authors: R. H. Mason[1], M. Si[1], C. Chou[1], V. E. Irish[1], R. Dickie[1], P. Elizondo[1], R. Wong[1], M. Brintnell[2], M. Elsasser[2], W. M. Lassar[3], K. M. Pierce[3], W. R. Leaitch[2], A. M. MacDonald[4], A. Platt[2], D. Toom-Sauntry[2], R. Sarda-Estève[5], C. L. Schiller[6], K. J. Suski[7], T. C. J. Hill[7], J. P. D. Abbatt[8], J. A. Huffman[3], P. J. DeMott[7], A. K. Bertram[1*]

Affiliations:

[1]Department of Chemistry, University of British Columbia, Vancouver, BC, V6T1Z1, Canada

[2]Climate Research Division, Environment Canada, Toronto, ON, M3H5T4, Canada

[3]Department of Chemistry and Biochemistry, University of Denver, Denver, CO, 80208, USA

[4]Air Quality and Processes Research Section, Environment Canada, Toronto, ON, M3H5T4, Canada

[5]Laboratoire des Sciences du Climat et de l'Environnement, CEA/CNRS-UVSQ, 91191, Gif/Yvette, France

[6]Air Quality Science Unit, Environment Canada, Vancouver, BC, V6C3S5, Canada

[7]Department of Atmospheric Sciences, Colorado State University, Fort Collins, CO, 80523, USA

[8]Department of Chemistry, University of Toronto, Toronto, ON, M5S3H6, Canada

[*]Correspondence to: bertram@chem.ubc.ca (A. K. Bertram)

Author response to referees

Anonymous Referee #1
Received and published: 30 August 2015

> ***[A0]*** For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. ***[1]***). Authors' responses are offset in blue below each referee statement with matching numbers (e.g. ***[A1]***). Page and line numbers refer to online ACPD version.

The manuscript by Mason et al. (2015) presents ground-based, size resolved ice nucleating particle (INP) number concentration measurements at seven different locations in North America and Europe. Measurements were conducted with a micro-orifice uniform impactor - droplet freezing technique (MOUDI-DFT) at temperatures of -15, -20 and -25°C. The authors observed a great fraction of INPs at sizes larger than 1 µm (91 % on average at -15°C). The results are compared to earlier observations of INP size distributions as well as to reported INP – aerosol correlations. The results of the study contribute to our understanding of ice nucleation in the atmosphere and the manuscript is thus of interest to the readers of Atmospheric Chemistry and Physics. I recommend this manuscript for publication after the following adjustments are addressed.

> We thank the referee for his/her helpful comments!

General remarks:

[1] The manuscript is very well written and the quantification of INPs larger than 1 µm and 2.5 µm, respectively, is very interesting. Generally, I think, the topic of uncertainty is addressed too little in the manuscript. Measurement uncertainties of the MOUDI-DFT and of equation (1), used to calculate atmospheric INP number concentrations should be given and discussed more. For instance, what are the average and standard deviation of the blank freezing temperature? As the observed INP concentrations are rather small as well as the sample size (especially the marine and agricultural sample), uncertainties should be treated even more carefully.

> ***[A1]*** To address the referee's comments in the revised manuscript we have done the following:
> a) Reported the average freezing temperatures and standard deviation for the blanks as well as the warmest observed freezing temperatures of the blanks.
> b) Discussed other uncertainties in the MOUDI-DFT technique (e.g. discuss uncertainties from non-immersion freezing events, which refers to growth of a frozen droplet by vapor deposition and subsequent contact with a neighboring liquid droplet, causing the latter to freeze).
> c) Reported the correction factors for Equation 1 and associated uncertainties.

[2] Also, the atmospheric relevance of the ground-based observations is only discussed very briefly in section 4. Since the manuscript is making a rather strong statement by claiming earlier observations (e.g. by CFDC techniques) underestimated total INP number concentrations, the mentioned wet and dry deposition processes of supermicron particles during atmospheric

transport may be discussed earlier in the manuscript and in more detail.

[A2] To address the referee's comments the differences expected between ground-based observations and observations at high altitude were discussed earlier and in more detail. Specially, we have done the following:

The last sentence of the abstract has been modified to the following: "Further size-resolved studies of INPs as a function of attitude are required since the size distribution of INPs may be different at high altitudes due to size-dependent removal processes of atmospheric particles."

On page 20525, line 16 we will add the following: "The exact proportion of INPs missed may also depend on altitude since the removal of atmospheric particles by wet and dry deposition in the atmosphere is expected to be size dependent. As an example, supermicron particles can have larger dry deposition loss rates than submicron particles."

Specific remarks:

[3] p 20523, l 10 and l 12: What are the standard deviations of the average percentages?

[A3] In the revised manuscript standard deviations will be added to all instances where a concentration or percentile size is given when averaged over all sampling locations.

[4] p 20524 l 29: The causal connection of the first and last part of this sentence is not clear to me (need for INP size distribution for aerial dispersal of fungi?). The sentence should be rephrased and the aerial dispersal part might be deleted.

[A4] To address the referee's comment the last part of the sentence has been deleted and the first part was modified to the following: "Information on the size distributions of INPs are thus needed for accurate modeling of their transport and distribution in the atmosphere."

[5] p 20525 l 125ff: Here, the issue of atmospheric relevance of surface based measurements of INP number concentrations could be addressed.

[A5] See ***[A2]*** above.

[6] p 20531 l 6ff: Here, more details on the uncertainty of the correction factors could be given as well as the average freezing temperatures of blank measurements.

[A6] See ***[A1]*** above.

[7] p 20531 l 11: Quantify what is meant with 'rare', e.g. give a detection limit.

[A7] When averaged over all sampling locations, only 1.3 % of droplets froze at temperatures above -15 ºC. This detail has been added to the revised manuscript.

[8] p 20531 l 21ff: The ice nucleation mode and INP size ranges of the mentioned studies should

be provided.

[A8] For cases where information on the mode and studied particle size range is available, this information has been added to the revised manuscript.

[9] p 20532 l 14: Does it play any role that air flow came off the 'eastern' coasts of 'continents'. Consider deleting 'eastern' and 'of continents'.

[A9] Revision was be made to remove mention of 'eastern' and 'continents'.

[10] p 20536 l 5ff: A comment on the physical plausibility of the differences reported by Rosinski et al. (1986) between condensation and immersion freezing would be interesting and helpful.

[A10] In the revised manuscript we have added a comment on the possible differences reported by Rosinski et al. (1986) between condensation and immersion freezing.

[11] p 20536 l 9: The sentence in brackets is not fully clear to me.

[A11] All of the INPs larger than 0.5 µm were supermicron in size (i.e. no INPs were 0.5–1 µm). The sentence was re-organized to improve clarity.

[12] p 20536 l 22: The Rucklidge (1965) study could be included in the table since the '86 % of INP smaller than 1 µm' are a lower limit considering that always the largest aerosol particle found in the residuals was assumed to be the INP. This means that in this study the majority of INP was clearly found to be below 1 µm which is in contrast to the current observations.

[A12] Table 2 now includes Rucklidge (1965).

[13] p 20536 l 23ff: A comparison to studies of ice crystal residuals size distributions (e.g. Seifert et al. 2003 or Mertes et al. 2007) as well as to laboratory studies of aerosol particle size-dependency of ice nucleation (e.g. Welti et al. 2009) would add to this paragraph.

[A13] To address the referee's comments, the results from Mertes et al. (2007) were added to Table 2. We would prefer not to include the Seifert et al. (2003) study since it was not clear in this study if homogeneous or heterogeneous nucleation was the dominant mode of ice nucleation.

The following has also been added to the manuscript to address the referee's comment regarding the work of Welti et al. (2009):

"The ice nucleation efficiency of particles as a function of size have also been investigated in laboratory experiments (e.g. Archuleta et al. (2005), Welti et al. (2009); Lüönd et al. (2010)). In general this work has shown the ice nucleation efficiency increases as particle size increases."

[14] p 20553 Table 1: The elevation should be given both, a.s.l. and a.g.l., for all locations to be able to compare them.

[A14] This information has been added.

[15] p 20556 Figure 2: The standard error of the mean given in this figure appears in some cases rather small to me compared to other measurements of ambient INP concentrations. Is the calculation done in Poisson statistics or for a normally distributed error? A comment on this could go into the discussion of the uncertainties.

[A15] In using the standard error we have assumed a normal sample distribution. This has now been noted in the figure captions.

Technical remarks:

[16] p 20523 l 17: replace 'ice nuclei' with 'INP'.

[A16] Correction made.

[17] p 20525 l 10 - 11: insert 'D50' before ≤ 2.4 and ≤ 0.75 µm.

[A17] Correction made.

[18] p 20525 l 14: is 'size' really what's meant here? Consider replacing 'size' with 'sites'.

[A18] To address the referee's comment, the last part of this sentence "as well due to the potential dependence of ice-active size on temperature" was deleted to avoid confusion.

[19] p 20532 l 4: replace 'ice nuclei' with 'INP'.

[A19] Correction has been made to "…dust, which can act as efficient INPs at lower temperatures…"

[20] p 20537 l 12: replace 'ice nuclei' with 'INP'

[A20] Correction made.

[21] p 20538 l 20: insert 'would like' before 'to thank'.

[A21] Correction made.

[22] p 20553 Table 1: The ordering of the sampling sites could be the same as in the plots

[A22] The ordering of the sites in Sect. 2.1, Table 1, and Fig. 1 has been changed to match that of Figs. 2–5.

Anonymous Referee #2
Received and published: 1 September 2015

[A0] For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. *[1]*). Authors' responses are offset in blue below each referee statement with matching numbers (e.g. *[A1]*). Page and line numbers refer to online ACPD version.

The paper by Mason et al. presents size-resolved impactor measurements of sub- and super-micron particles collected at seven locations in Canada, the U.S., and France. The samples were analyzed determining the particles' immersion-mode freezing properties, that is, ice nucleating particle (INP) number concentrations as a function of size and temperature.

The main conclusion from the study is that a large fraction of the ice active particles is > 1 μm in diameter. This is particularly important to know for the interpretation of INP concentrations determined with other established on-line measuring instruments, such as the continuous-flow diffusion chambers, which typically miss the super-micron particles in their analysis due to the specific inlet system.
The paper is very well structured, describes the applied methods and discusses the results very nicely. Therefore I can fully recommend the paper for final publication in ACP. I have only very little suggestions for improvement and a few minor questions all listed chronologically in the following:

We thank the referee for his/her helpful comments!

[1] P. 20523, L.7: Here it would be helpful to add one short sentence on the applied measurement principle, the MOUDI-DFT.

[A1] The following is now in the revised manuscript:

"INP number concentrations as a function of size and temperature were determined using the micro-orifice uniform deposit impactor-droplet freezing technique (MOUDI-DFT; Huffman et al., 2013; Mason et al., 2015a). This technique combines aerosol particle collection by a cascade inertial impactor with sharp size-cutoff characteristics (the MOUDI; Marple et al., 1991) with an established droplet freezing apparatus (the DFT) for determining immersion-mode freezing properties (Koop et al., 1998; Iannone et al., 2011; Haga et al., 2013). A similar approach has also been used to study deposition nucleation by particles collected from the atmosphere (Wang et al., 2012; Knopf et al., 2014)."

[2] P. 20525, L. 24-29: I would delete this paragraph at this place because it tells already main results, which not necessarily are part of the introduction section.

[A2] The paragraph was revised to the following to remove the discussion of results from the introduction section:

"Previous studies of INPs as a function of size have been carried out in the field (e.g. Vali,

1966; Rosinski et al., 1986; Mertes et al., 2007; Santachiara et al., 2010), and laboratory (e.g. Welti et al., 2009; O'Sullivan et al., 2015). These and additional studies are further discussed in Sect. 3.2. In the current study, we add to the existing body of size-resolved INP measurements by reporting ground-level INP size distributions from six locations in North America and one in Europe, covering a diverse set of environments and investigating immersion freezing at -15, -20, and -25 °C."

[3] P. 20530: The aerosol particle number size concentration usually varies significantly over the size range of 0.1 to 10 µm. Consequently, I guess the surface coverage must be very different for the individual impactor stages, i.e., small number of particles on upper stages and large number on lower stages. How does that affect the droplet freezing experiments? I could imagine that it is difficult to analyze samples with too high particle load because the growing droplets may run into each other very easily. On the other hand, if there are only few particles on the surface the result might not be statistically significant. How did you handle different surface coverages?

[A3] The surface coverage can affect the freezing temperatures in two ways:

a) If the concentrations are too low, the freezing temperatures will overlap with the "blank" freezing experiments. This was not an issue in the current experiments. To clarify this point, the "blank" freezing experiments have been noted in the revised manuscript.
b) If the surface coverage is too high and there is a significant concentration of INPs on the cover slip, all the droplets will freeze at warm temperatures. We control the latter by controlling the sampling time.

[4] P. 20531 and 20532: I wonder if rounding INP concentrations to two significant digits should be enough, e.g., 3.8 L-1 instead of 3.77 L-1, since I believe your measurements are not more precise than that. Also standard deviations together with the mean values would be interesting to know.

[A4] The reported number of significant figures has been revised as suggested.

[5] P. 20533, L. 25-27: How realistic is the assumption? Did the number size distributions (if available) also show uniformly distributed aerosol particles over this size range?

[A5] Measurements of the total particle size distribution were not available at all locations to fully address the referee's question. Furthermore, the use of such distributions in determining the fraction of stage 4 (1.8–3.2 µm) that belongs in the coarse mode would be contingent on the assumption that the INP size distribution follows the total particle size distribution, which may not be correct in all locations. To address the referee's comment we added the following text at this location of the revised manuscript:

"Measurements of the total particle size distribution were not available at all locations to test this assumption. Furthermore, it is not known if the INP size distribution follows the total particle size distribution *a priori*."

[6] Fig. 3: Why did you not show any error bars for the Labrador Sea results?

[A6] As only one sample (MOUDI set) is available for the Labrador Sea, there is no standard error to report. This point is included in the figure caption. For clarity, we have also repeated this point in the main text when discussing Fig. 3.

Anonymous Referee #3
Received and published: 1 September 2015

[A0] For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. ***[1]***). Authors' responses are offset in blue below each referee statement with matching numbers (e.g. ***[A1]***). Page and line numbers refer to online ACPD version.

The manuscript reports immersion-mode INP number concentrations as a function of aerodynamic size at six ground sites in North America and one in Europe. Size- resolved particle samples were collected using a model 110R or 120R Moudi. The ice-nucleating ability of particles was then determined by a microscope–based immersion freezing apparatus (MOUDI-DFT technique). The authors found that both supermicron and coarse mode aerosol particles were a significant component of the INP population. The paper is well written and of interest. I suggest publication after the following few comments have been addressed.

We thank the referee for his/her helpful comments!

General remarks:
[1] It is known that not all particles colliding with a plate adhere to it. As offline ice nucleation analysis cannot coat (e.g. with vacuum grease) the substrate located on the impaction plate, particle rebound in the Moudi impactor should be discussed. Generally speaking, the rebound increases with aerosol diameter and decreasing air relative humidity. The average relative humidity values (r.h.) in the seven locations range from 48% to 97%. Therefore, the rebound should differ in the sampling sites. At Amphitrite Point, where r.h. during sampling was 97%, the rebound should be much lower with respect to Colby, where the r.h. was 48%. In marine air the bouncing should be lower due to hygroscopic particles.

[A1] Thank you for bringing up this point. In the revised manuscript the issue of particle bounce has been discussed.

[2] The interest of the paper would be enhanced if particle number concentration and size distribution were considered in each site during sampling. This information could allow calculation of the ratio between INP number concentration and the corresponding particle concentration in each size bin, and the correlation between INP number concentration and aerosol.

[A2] We agree that this information would be very useful to report. To carry out these types of calculations we would need the size distribution of the particle population at each site, measured at the same times as the INP measurements. Unfortunately, this data was not

measured at all sites (e.g. UBC), and for the cases were this information was measured, accessing this data and then carryout out these calculations would require a large amount of work. We hope the Editor agrees that these calculations are not required for the current publication. However, we will pursue these types of calculations in future publications where we intend to report INP values from these sites in more detail.

[3] Hygroscopic particles sampled in marine sites form droplets on the examined area sooner than insoluble particles (which are considered efficient ice nuclei). Could this feature influence the INP concentrations measured with the MOUDI-DFT technique?

[A3] Differences in particle hygroscopicity between sampling locations should not influence the droplet freezing technique (DFT), and therefore the measured INP concentrations. The relative humidity of the gas flow in the DFT during droplet growth was held at approximately 120 %. At this relative humidity water will readily condense on both ambient samples containing particles and directly on the hydrophobic glass cover slip (as observed in the blank experiments).

To address the referee's comment we have added the following to the revised manuscript:

"Water droplets were condensed onto the sample and monitored using a CCD camera recording a digital video. Since the relative humidity of the gas flow during droplet condensation was held at approximately 120 %, water condensation occurred uniformly on the cover slip, and growing droplets coagulated as they grew to a final size of 97 ± 42 µm (mean diameter and 1 standard deviation (SD) uncertainty). On average, more than 99 % of particles become incorporated into the droplets based on the projected particle area before water condensation and following droplet growth using the Amphitrite Point samples, with similar observations over the entire dataset."

[4] Paragraph 2.2: Size-resolved INP number concentrations
This paragraph should be broadened by summarizing the most important points of the technique used, reported in the paper of Huffman et al. (2013) and Mason et al. (2015). For instance, the total area of each stage and of the analyzed area should be indicated, and the problem of the non-uniformity of aerosol deposit in each stage of the MOUDI should be addressed. An additional point should be clarified. Huffman et al. (2013) found that the maximum concentration of IN detected for any given slide with the microscope freezing technique is roughly 0.6 -0.9 L-1 (depending on the number of droplets condensed and the total volume of air sampled) and the maximum concentration of IN determined by the microscope technique is small compared to the maximum concentration determined with the CFDC method. The submitted paper reports concentrations up to 10 INPs L-1 (T = -25ºC, size interval 5.6 - 10 µm, at Colby, KS). Which is the maximum number of droplets that can be formed on the area (1.2 mm2) analyzed by the DFT?

[A4] This paragraph was expanded to address the referee's comments, as well as the other referee's comments. For example, values of $A_{deposit}$, f_{nu}, and f_{ne} used in Equation (1) can now be found in the Supplement to provide more information on calculations involving the MOUDI-DFT.

Regarding the difference in the upper limit of the INP concentrations reported here and in Huffman et al. (2013): in the current study we have used shorter sampling times than those of Huffman et al. (2013), allowing us to detect greater INP number concentrations. This information has been added to the revised manuscript. The largest number of droplets condensed in an experiment for this study was 75.

Minor remarks:

[5] Page 20531 - Line 12 and following: "Freezing events were rare at temperatures warmer than -15ºC and are therefore not reported". This statement appears contradict what it is said afterwards, i.e.: - Page 20532, Line 9 and following: " . . .the major source of INPs at Amphitrite Point during the study period was likely biological particles from local vegetation . . ." - Page 20532, Line 20 and following: "...the highest concentrations of INPs at a freezing temperature of -25ºC were found at the Colby, KS sites. . .. aerosol sampling was conducted adjacent to soya and sorghum fields . . .This high concentration of INPs is consistent with previous work of Garcia et al. (2012) . . .. and Bowers et al. (2011). . ."

For instance, Garcia et al. (2012) measured an INP concentration of about 1 L-1 at T = -10ºC. Bowers et al. (2011) found greater INPs downwind of corn fields than in air samples collected from the suburban and forest land-use types, at T> -10ºC. Generally speaking, biological aerosols (bacteria, spores, fungi, pollen) are activated as ice nuclei prevalently at temperatures warmer than -10ºC (Möhler et al., 2007). Therefore, at sites like Amphitrite Point, Colby and Saclay, a fraction of aerosol particles should be activated even at T> -15ºC.

[A5] A small fraction of the droplets did freeze at temperatures above 15 ºC (see response ***[A7]*** for referee 1). This was noted in the revised manuscript.

[6] At Labrador Sea only one sample was available. What does the uncertainty reported in Fig. 2 and Fig. 4 mean?

[A6] As only one sample is available for the Labrador Sea location, the uncertainties reported in Fig. 2 is the uncertainty in the INP concentration from Equation (1). We now realize this has led to confusion. To reduce confusion, we have removed the error bars for the Labrador Sea data in Fig. 2 and indicate in the figure caption that no error bars for the Labrador Sea data are reported since only one sample was available for this location. All other error bars in the figure represent the standard error of the mean.

In Fig. 4, the uncertainty at each location is given as the 25^{th} and 75^{th} percentile INP size as indicated in the figure caption.

[7] We note that the lower concentration of INP was obtained at Alert, where higher air volume was sampled (about 32 m3) and the highest concentration at Colby, where the sampled air volume was the lowest (about 8 m3). Is this a fortuitous event?

[A7] We do not have any reason to expect the INP concentration is related to the sampling volume. Nevertheless we carried out an additional analysis of the Alert data to further

investigate this question. Sampling times at Alert varied from approximately 2.3 to 46.1 hours (with an average of 17.6 h as reported in Table 1). The linear correlation coefficients (R) of INP concentration vs. sample volume were 0.45, 0.04, and -0.39 at freezing temperatures of -15, -20, and -25 ºC, respectively. Furthermore, at temperatures of -15 and -20 ºC the sample with the largest INP concentration had the second largest sample volume. Hence, the Alert data suggests the INP concentration is not related to the sampling volume.

[8] As all graphs report in order: Alert NU, Whistler Mountain, Amphitrite Point, Labrador Sea, Saclay France, UBC Campus, Colby, KS, please follow the same order in the Paragraph: 2.1: Samplings Sites and in: Table 1

[A8] The change in order was made in the revised manuscript.

Referee #4, Gabor Vali
Received and published: 11 September 2015

[A0] For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. ***[1]***). Authors' responses are offset in blue below each referee statement with matching numbers (e.g. ***[A1]***). Page and line numbers refer to online ACPD version.

The problem addressed in this paper - the sizes of atmospheric ice nucleating particles - is important and is far from having been adequately resolved in previous studies.
The paper does leave the impression of a somewhat brief, preliminary study for exploring the potential of the relatively recent MOUDI-DFT technique. That is a legitimate goal and it has been successful. But there are some shortcomings that deserve to be mentioned. No ancillary data are presented, such as aerosol size distributions.
The aerosol were not brought to a standardized humidity so the sizing would be more consistently comparable. Possible aging of the samples was not cotrolled for. Other questions and suggestions are listed in the following sections. Some of the points are perhps marginal, as this paper focuses on the sizes of the INPs not on their concentration; nonetheless, the authors should discuss these problems and reflect on possible implications for the results. Some caveats would be in order and some changes should be included when the paper is revised.

We thank the referee for his helpful comments!

Technical issues:
Since the MOUDI-DFT method employed in this paper has been described in at least two previous publications, and is used here with no changes that would require special attention, the method is taken as established and few details are given. Nonetheless, there are some questions that can be esked regarding some aspects of the method.

[1] Background
Unless I missed it, it seems that the background level, or detection limit, has not been

established. There is mention of sudden freezing at -25C (pg. 20531 line 14) and this may be an indication of the detection limit. But the background levels at other temperatures are still open to question. Tests with the intake air passed through an efficient fiter upstream of the sampler would have yielded blanks for establishing the background. This may have made the condensation of drops difficult but that could have been circumvented by some steps like higher supersaturation, producing drops meachanically, etc. None of this is simple, of course.

[A1] To address the referee's comments the average freezing temperature and standard deviation of the blanks as well as the warmest freezing temperatures of the blanks have been reported.

[2] Sample volume

The volume of the air sample that is evaluated for INP content by Eq. 1 is taken as the ratio of the DFT view area to the area of the glass slip while the total volume is for the entire MOUDI sample. Isn't the ratio of glass slip to MOUDI disk missing here?

[A2] In Equation (1), the number of INPs found using the droplet freezing technique is first multiplied by the fraction of the total sample area to the analyzed area ($A_{deposit}$/ A_{DFT}) and is then divided by the total volume of air sampled. $A_{deposit}$ is the area of the MOUDI disk that the referee is referring to above. To clarify this point, in the revised manuscript $A_{deposit}$ has been referred to as the total area of the sample deposit on the MOUDI impaction plate.

[3] INP content of drops

The assumption of equal drop sizes from Vali (1971) has been maintained in Eq. 1 in spite of the large range of droplet sizes (a factor 20 or more in volume) that form via condensation in these experiments. This can't be simply ignored.

The issue raised in the previous paragraph is linked to the question what is the relevant measure of INP content per drop in these experiments. The Vali (1971) proceedure is formulated for the case when all drops are derived from the same bulk sample of water with a random partitioning of INPs in the drops. In that case, all drops can be assumed to have the same probability of containing any type of INP. With the condensation method employed in these experiments the validity of this assumption needs to be examined carefully, considering what determines where the drops form on the plates and what particles they contain. If condensation is initiated on the INPs themselves, the question reduces to whether there is any dependence of the supersaturation required for condensation and the INP activity of that particle. This in turn depends on what kind of internal mixtures consitute the INPs. If the INPs become included into the drops as those drops grow from a condensation nucleus that is not an INP, than the assumption of random probability of INP content per drop seems to be fulfilled. The main point is that the authors should justify the use of the Vali (1971) formula for these experiments. It is not obvious how the original assumptions apply here. Additional possible problems relate to the adhesion of collected particles on the supporting surface. Perhaps the authors have looked into whether the drops move particles along the surface as they expand during condensation, or cover them up. The same question needs to be asked for the period of evaporation: are particles left behind as the drops shrink? There is a hint that perhaps this is the case in Fig 2(c) of Mason et al. (2015a) with particles(?) seen in circular patterns.

[A3] Droplet growth by water condensation in the DFT occurs in the same manner for samples containing particles and clean hydrophobic glass cover slips (no particles deposited and rinsed with ultrapure water). Therefore, water condenses uniformly on the cover slip, and these droplets then combine as they grow. Particles become incorporated into the droplets as this occurs. Analysis of the Ucluelet samples, with similar observations over the entire dataset, shows that only a small fraction (on average < 1%) of particles visible with the optical microscope remain outside of the droplets once they reach their final size.

We have also investigated binning the measurements based on droplet size and residual inclusion area, using 2–4 bins, to see how this would influence our results. The samples used in the binning analysis are those from the CSU intercomparison study noted in the Mason et al (2015a) instrument paper. We found that the total number of INPs determined both ways (with and without binning) agreed within the experimental uncertainty at freezing temperatures of -25 °C and above. Based on this analysis we conclude that the application of Equation (1), which assumes monodisperse droplets, to the DFT results at -25 °C and above does not lead to large uncertainties. This information was added to the revised manuscript to address the referee's comments.

[4] Cooling rate

The reported cooling rate in the freezing experiments is -10C min^{-1}. Thermal lags throughout the system, specially within the drops but also within the stage, could be a problem with such rapid cooling. Has that been considered? How uniform was the rate of cooling throughout an experiment?

[A4] The possibility of a thermal lag using a cooling rate of -10 ºC min^{-1} was investigated in detail previously using a similar droplet freezing technique (Koop et al., 1998), and the thermal lag in that study was shown to be small at a cooling rate of 10 ºC min^{-1} (the correction was 0.2 ºC at -33 ºC). In addition we tested the possibility of a thermal lag by investigating the freezing of pure water droplets using a cooling rate of -5 and -10 ºCmin^{-1} (unpublished results). The median freezing temperature determined with the two cooling rates of -5 and -10 ºC min^{-1} were -36.3 ± 1.5 and -36.7 ± 2.0 ºC, respectively, with uncertainty given as the standard deviation.

To determine the stability of the cooling rate we cooled the freezing cell from -5 ºC to -37 ºC three times using the set cooling rate of -10 ºCmin^{-1}. The average cooling rate was found to be -9.8 ± 0.4 ºC based on the three experiments. In Fig. 1 of this document (see below) we have plotted the cooling rate from three experiments over the temperature range of -5 to -37 ºC, measured at a frequency of 1 Hz.

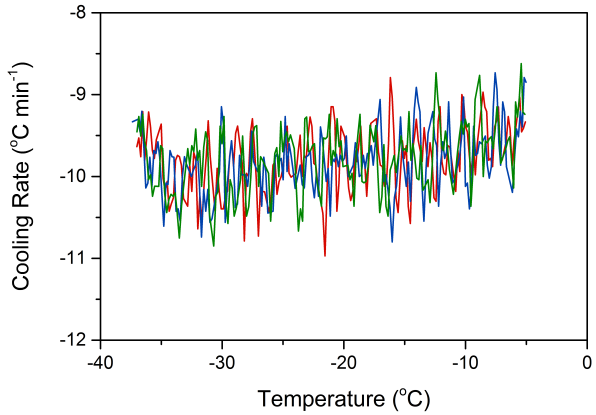

Figure 1. The cooling rate of three experiments measured over the temperature range of -5 to -37 ºC, recorded at a frequency of 1 Hz.

[5] Correction factors

Two corrections factors are included in Eq. (1). The first one, f_{nu} is explained in some detail in the referenced paper (Mason et al. 2015a). The second one, f_{ne} is less clear as the referenced paper (Koop et al. 1997) doesn't show this quantity directly and here it is applied to a formula (Vali 1971) that is differently derived. This requires some explanation. Numerical values for the correction factors should be given.

[A5] Values of these correction factors will be added to the Supplement. In addition, Equation (1) will be separated into two equations; similar to our earlier paper that describes the MOUDI-DFT (Mason et al. 2015a). The correction factors should be clearer when two equations are included. In addition, the Koop correction has been discussed in more detail in the revised manuscript.

Results:

[6] Were the overall average and median concentrations reported calculated with weighting by the number of samples for each location? In light of the sample numbers varying from 1 to 34, giving equal weight to each results in some distortion of the data.

[A6] In our analysis the overall average and median concentrations were calculated by giving each location (i.e. sampling site) equal weight. We felt that weighting by the sample number would skew the average toward the sites of Amphitrite Point, Saclay, and Alert while largely ignoring the remaining locations. This information has been included in the revised manuscript for clarity.

[7] On page 20534 long-range transport is stated to preferentially reduce the numbers of large particles. That is not an obvious result for INPs, specially since the mixture of components that make up the INPs is unknown.

[A7] To address the referee's comments, the discussion on long-range transport was deleted. Specifically, lines 18-27 on page 20534 will be deleted.

[8] There are many facets of the results obtained in this work and the authors have selected meaningful diagrams to present those results. Yet, it would have been useful to see the results also in the form of temperature versus concentration. That would also be a good way to compare present results with other published works.

[A8] To address the referee's comments in the revised manuscript a spectrum of INP concentration vs. temperature for each location has been added to the Supplement.

[9] Results from the Colby site lie at the upper ends of the data both in concentration and in size. It is stated that sampling was during and following combine operations. The range of variability in concentrations and in sizes for these 3 samples is no different from other sites. Is this relative uniformity related to having combine operations in the area, if not directly near the site, throughout the sampling periods? On page 20258 it is indicated that sampling was 3-10 m from the fields. Isn't that an error and kilometers were meant?

[A9] It is possible that additional combine activities in the region also contributed to the measured INP population. However, we don't have information that can confirm this possibility.

The stated proximity of 3–10 m is correct. Equipment was set up in a mobile laboratory parked next to the fields.

[10] There is interesting aspect of the data that the authors might want to explore. The question arises: are the INPs of different sizes of the same composition and have the same surface properties? If so, the probability of a nucleating site being found on any size particle would be the same. A rough estimate of the relative proportions indicated in Fig. 3 of the paper for two sizes indicates a negative answer to the question. Using a ratio of about 1.3 for INPs greater than 1μm to 2.5μm and using those as nominal sizes whose surface areas would differ by a factor of 6.25, the indication is that the larger particles have a much lower probability per unit surface area to contain an ice nucleus. That would be something to ponder. This calculation could be made more precise and extended to the full range of sizes covered by the sampling. There are broad implications of this type of analyses so presentation of the results in this form would make a valuable addition to the paper.

[A10] We assume here that the referee is suggesting that we calculate ice-nucleation active surface site density (n_s) values as a function of size. We certainly agree these calculations would be very interesting. To carry out these types of calculations we would need the size distribution of the particle population at each site, measured at the same times as the INP measurements. Unfortunately, this data was not measured at all sites (e.g. UBC), and for the cases were this information was measured, accessing this data and then carryout out these calculations would require a large amount of work. We hope the Editor agrees that these calculations are not required for the current publication. However, we will pursue these types of calculations in future publications where we intend to report INP values from these

sites in more detail.

[11] It is pointed out in the paper that the size range of INPs examined is restricted to > 0.1μm aerodynamic size and that further studies are needed to examine the contributions of smaller size particles to INP populations. An additional aspect of INP sizing would be worth mentioning. The size distributions of atmospheric particles undergo many changes due to capturing, scavenging and other processes. Few particles are of single composition. The measurements here reported refer to sizes of internally mixed particles and also of aggregates. Thus, small INPs may be detected at larger sizes if they adhere to other particles. This may take place already at the origin (soil especially) but also during atmospheric transport. A difficult issue, no doubt, but one that need to be recognized.

[A11] This is a good point raised by the referee. The last paragraph of the Summary and Conclusions section has been rewritten to address the referee's comment. Specifically the last paragraph will be revised to the following:

"A caveat to this study is that our measurements were confined to aerosol particle sizes greater than either 0.10 or 0.18 μm. If there were a significant contribution from INPs of smaller sizes (Vali, 1966; Schnell and Vali, 1973; Pummer et al., 2012; Augustin et al., 2013; Fröhlich-Nowoisky et al., 2015; O'Sullivan et al., 2015; Tong et al., 2015; Wilson et al., 2015), and these smaller sizes did not coagulate or get scavenged by larger particles, the values presented here would represent upper limits to the contribution of supermicron and coarse mode particles to the total INP population. Additional studies exploring the relative atmospheric abundance of INPs < 0.10 μm are necessary (Hader et al., 2014). Future studies of the size distribution of INPs should also include measurements of particle mixing state to determine if particles are internally or externally mixed at the locations where the size distribution of INPs are being measured."

Supplement:

[12] It is good to have the authors' analyses of the works cited for comparison of size determinations. It helps to illustrate the sparsity of data on this important point.

All but one of the works cited employed aerosol samplers. The exception is the Vali (1966) report which is based on size discrimination by filtering of liquid samples. It would be helpful to readers to point out this difference and its possible implications, namely that soluble components of the INP get removed and particle might break up when immersed in water. Both of these factors introduce a bias in the comparison of sizes, with corresponding aerosol sizes likely to have been considerably larger than the size found in water. Coagulation of particles in the liquid would add a bias in the opposite directions but is less likely to have been a factor.

[A12] These points have now been noted in section S2.2 of the Supplement where the Vali (1966) measurements are discussed. Thank you for the suggestion.

Abstract

Detailed information on the size of ice nucleating particles (INPs) may be useful in source identification, modeling their transport in the atmosphere to improve climate predictions, and determining how effectively or ineffectively instrumentation used for quantifying INPs in the atmosphere captures the full INP population. In this study we report immersion-mode INP number concentrations as a function of size at six ground sites in North America and one in Europe using the micro-orifice uniform deposit impactor-droplet freezing technique (MOUDI-DFT), which combines particle size-segregation by inertial impaction and a microscope-based immersion freezing apparatus. The lowest INP number concentrations were observed at Arctic and alpine locations and the highest at suburban and agricultural locations, consistent with previous studies of INP concentrations in similar environments. We found that 91 ± 9, 79 ± 17, and 63 ± 21 % of INPs had an aerodynamic diameter > 1 µm at ice activation temperatures of -15, -20, and -25 °C, respectively, when averaging over all sampling locations. In addition, 62 ± 20, 55 ± 18, and 42 ± 17 % of INPs were in the coarse mode (> 2.5 µm) at ice activation temperatures of -15, -20, and -25 °C, respectively, when averaging over all sampling locations. These results are consistent with six out of the nine studies in the literature that have focused on the size distribution of INPs in the atmosphere. Taken together, these findings strongly suggest that supermicron and coarse mode aerosol particles are a significant component of the INP population in many different ground-level environments. Further size-resolved studies of INPs as a function of attitude are required since the size distribution of INPs may be different at high altitudes due to size-dependent removal processes of atmospheric particles.

Ryan Mason 10/11/2015 10:01 AM
Deleted: seven

Ryan Mason 10/11/2015 3:24 PM
Deleted: ice nuclei

1 Introduction

Ice nucleating particles (INPs) are a unique class of aerosol particles that catalyze ice

formation under atmospheric conditions. A variety of particle types have been identified as INPs, including mineral dust, black carbon, volcanic ash, glassy aerosols, and primary biological particles such as bacteria, fungal spores, and pollen (see reviews by Szyrmer and Zawadzki, 1997; Möhler et al., 2007; Ariya et al., 2009; Després et al., 2012; Hoose and Möhler, 2012; Murray et al., 2012; Yakobi-Hancock et al., 2013). Although only a small fraction of aerosol particles nucleate ice (e.g. Rogers et al., 1998), INPs are important since they can lead to changes in the properties and lifetimes of mixed-phase and ice clouds, ultimately affecting climate and precipitation (Baker, 1997; Lohmann and Feichter, 2005; Baker and Peter, 2008; DeMott et al., 2010; Creamean et al., 2013).

Vali et al. (2015) describes four modes of heterogeneous ice nucleation: deposition nucleation, where ice forms on the INP directly from the gas phase; condensation freezing, where ice nucleates during the condensing of water onto the INP; immersion freezing, where crystallization is initiated by an INP within a supercooled liquid droplet; and contact freezing, where the freezing of a supercooled liquid droplet is due to impaction by an INP. In this study we focus on freezing via the immersion mode in dilute solution droplets, which is relevant to mixed-phase cloud conditions.

Ryan Mason 10/18/2015 1:09 PM
Deleted: Vali (1985)

Due to the importance of INPs for climate and precipitation, there has been a renewed interest in measuring the concentrations of INPs in the atmosphere (DeMott et al., 2011). While much of this work has focused on measurements of the total number concentration of INPs, there has been less emphasis on determining their size distributions in the atmosphere. Information on airborne INP size distributions may be particularly helpful in identifying the predominant INP sources. For example, information on the size distribution of INPs may help rule out or support the role of fungal spores in atmospheric ice nucleation since they are often in the supermicron

range (Graham et al., 2003; Elbert et al., 2007; Sesartic and Dallafior, 2011; Després et al., 2012; Huffman et al., 2012). A similar approach can be used with black carbon particles, since they are mainly in the submicron range (Clarke et al., 2004; Schwarz et al., 2008, 2013).

Previous modeling studies have shown that the transport and distribution of INPs, and aerosol particles in general, are sensitive to the size of the particles assumed in the models (Burrows et al., 2009; Wilkinson et al., 2011). Information on the size distributions of INPs are thus needed for accurate modeling of their transport and distributions in the atmosphere (Morris et al., 2004; Hoose et al., 2010a, 2010b; Sesartic et al., 2013; Haga et al., 2014; Spracklen and Heald, 2014).

Information on the size distribution of INPs is also needed to determine if techniques used to measure atmospheric INP concentrations capture the entire INP population. For example, the continuous flow diffusion chamber (Rogers et al., 2001b) is often used for measuring INPs (e.g. DeMott et al., 1998; Rogers et al., 2001a; Richardson et al., 2007; Pratt et al., 2009; Prenni et al., 2009; Eidhammer et al., 2010; Chou et al., 2011; Friedman et al., 2011; Hoyle et al., 2011; Corbin et al., 2012; Garcia et al., 2012; Tobo et al., 2013; McCluskey et al., 2014). This type of instrument has the advantage of providing real-time measurements of INPs with the ability to detect very large INP number concentrations, but the aerodynamic diameter of particles measured with it is limited, from $d_{50} \leq 2.4$ µm in some studies (e.g. Garcia et al., 2012) to $d_{50} \leq$ 0.75 µm in others (e.g. DeMott et al., 2003). Such techniques may miss supermicron or coarse mode (i.e. larger than 2.5 µm) INPs. The exact proportion of INPs missed may depend on temperature. Such online instruments have typically focused on measurements below approximately -20 °C as sample volume considerations limit effective sampling of lower INP number concentrations at warmer temperatures. The exact proportion of INPs missed may also

Ryan Mason 11/2/2015 9:37 AM
Deleted: impact on climate and precipitation, as well as for the aerial dispersal of fungi and bacteria

Ryan Mason 10/18/2015 7:23 PM
Deleted: (e.g. DeMott et al., 1998; Rogers et al., 2001a; Richardson et al., 2007; Pratt et al., 2009; Prenni et al., 2009; Eidhammer et al., 2010; Chou et al., 2011; Friedman et al., 2011; Hoyle et al., 2011; Corbin et al., 2012; Garcia et al., 2012; Tobo et al., 2013; McCluskey et al., 2014),

Ryan Mason 11/2/2015 10:18 AM
Deleted: as well due to the potential dependence of ice-active size on temperature

depend on altitude since the removal of atmospheric particles by wet and dry deposition in the atmosphere is expected to be size dependent. As an example, supermicron particles have larger dry deposition loss rates than submicron particles.

Previous studies of INPs as a function of size have been carried out in the field (e.g. Vali, 1966; Rosinski et al., 1986; Mertes et al., 2007; Santachiara et al., 2010) and in the laboratory (e.g. Welti et al., 2009; O'Sullivan et al., 2015). These and additional studies are further discussed in Sect. 3.2. In the current study, we add to the existing body of size-resolved INP measurements by reporting ground-level INP size distributions from six locations in North America and one in Europe, covering a diverse set of environments and investigating immersion freezing at -15, -20, and -25 °C.

Ryan Mason 10/11/2015 4:23 PM
Deleted: from Western Canada (Vali, 1966), the Pacific Ocean (Rosinski et al., 1986), the Gulf of Mexico (Rosinski et al., 1988), Eastern Europe (Berezinski et al., 1988), Italy (Santachiara et al., 2010), and the Central United States (Huffman et al., 2013).

Ryan Mason 10/11/2015 4:23 PM
Deleted: We found that both supermicron and coarse mode aerosol particles were a significant component of the INP population at all surface-based sampling locations. These results indicate that, when averaged over all locations and temperatures, the total INP number concentrations measured in similar ecosystems may be underestimated by a factor of 4.4 and 2.1 if supermicron or coarse mode particles, respectively, are not analyzed.

Ryan Mason 12/15/2015 6:10 PM
Formatted: Normal (Web), Indent: First line: 0 cm, Widow/Orphan control, Adjust space between Latin and Asian text, Adjust space between Asian text and numbers

2 Methods

2.1 Sampling sites

The seven locations used in this study are detailed in Table 1 and shown in Fig. 1. All reported sampling periods are local times. Measurements using the sampling instrumentation described in the next section were made at five locations in Canada: Alert, Nunavut; the Labrador Sea near Newfoundland and Labrador; Whistler Mountain, British Columbia; the University of British Columbia (UBC) campus, British Columbia and Amphitrite Point, British Columbia. Measurements in Canada were conducted as part of the larger NETwork on Climate and Aerosols: addressing key uncertainties in Remote Canadian Environments project (NETCARE; http://netcare-project.ca/). Measurements were also made at Saclay, France and Colby, Kansas, USA.

2.1.1 Alert

Ryan Mason 12/17/2015 9:39 PM
Deleted: , NU, Canada

Arctic sampling was conducted at the Dr. Neil Trivett Global Atmosphere Watch Observatory in Alert, Nunavut, Canada (labeled 1 in Fig. 1; Cobbett et al., 2007) between March 29 and July 23, 2014. This Arctic research station is part of a global network for measuring chemical and physical perturbations of the atmosphere. Aerosol particles were collected through a louvered total suspended particulate (TSP) inlet (Mesa Labs Inc., Butler, NJ, USA) and 0.9 m mast located on the upper level of an outdoor platform free of surrounding obstructions, and were stored in the dark at -15 or 4 °C for a period of 10–112 days prior to analysis.

2.1.2 Whistler Mountain

The Whistler Peak High Elevation Site is located at the summit of Whistler Mountain in Whistler, British Columbia, Canada (labeled 3 in Fig. 1) and operated by Environment Canada (Gallagher et al., 2011; Macdonald et al., 2011). Aerosol particle collection at this alpine site occurred between March 30 and April 23, 2014. The louvered TSP inlet was located approximately 10 m from a chairlift operating station. Although there are no continuous combustion sources at the site, sampled air may have been influenced by engine exhaust for short periods of time due to nearby snowmobile operation. Samples were stored in the dark at 4 °C for a period of 1–4 days prior to analysis.

2.1.3 Amphitrite Point

The coastal site at Amphitrite Point on Vancouver Island, British Columbia, Canada (labeled 5 in Fig. 1) is operated by Environment Canada, the BC Ministry of Environment, and Metro Vancouver for the continuous monitoring of aerosols and trace gases influenced by marine trajectories (McKendry et al., 2014; Yakobi-Hancock et al., 2014; Mason et al., 2015b). The mobile laboratory used during sampling was located approximately 100 m from the high tide line of the Pacific Ocean along a rocky shoreline, separated from the ocean by a narrow row of trees

Ryan Mason 10/11/2015 3:43 PM
Deleted: The Labrador Sea

Ryan Mason 10/11/2015 3:43 PM
Moved (insertion) [1]

Ryan Mason 10/11/2015 3:45 PM
Moved down [3]: The Canadian Coast Guard Service vessel CCGS Amundsen serves as both an icebreaker for shipping lanes and an Arctic research vessel. One set of aerosol particle samples was collected from the top of the bridge of this vessel on July 11, 2014 while in the Labrador Sea off the coast of Newfoundland and Labrador, Canada (labeled 2 in Fig. 1). While sampling was within the marine boundary layer in the presence of sea spray aerosols, back trajectories (not included) calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model of the National Oceanographic and Atmospheric Administration (Draxler and Rolph, 2014) indicate that the sampled air mass spent the majority of the previous 72-hour period over land. Air was passed through a louvered TSP inlet and 1.5 m mast during sampling, and collected aerosol particles were stored in the dark at 4 °C for a period of 45–46 days prior to analysis.

Ryan Mason 10/11/2015 3:44 PM
Deleted: Whistler Mountain

Ryan Mason 10/11/2015 3:44 PM
Moved (insertion) [2]

and shrubs approximately 2–10 m in height. Sampling took place from August 6 to August 27, 2013 using a louvered TSP inlet and 3 m mast. Aerosol particles were stored at room temperature and analyzed within 1 day of collection.

2.1.4 The Labrador Sea

The Canadian Coast Guard Service vessel CCGS Amundsen serves as both an icebreaker for shipping lanes and an Arctic research vessel. One set of aerosol particle samples was collected from the top of the bridge of this vessel on July 11, 2014 while in the Labrador Sea off the coast of Newfoundland and Labrador, Canada (labeled 2 in Fig. 1). While sampling was within the marine boundary layer in the presence of sea spray aerosols, back trajectories (not included) calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model of the National Oceanographic and Atmospheric Administration (Draxler and Rolph, 2014) indicate that the sampled air mass spent the majority of the previous 72-hour period over land. Air was passed through a louvered TSP inlet and 1.5 m mast during sampling, and collected aerosol particles were stored in the dark at 4 °C for a period of 45–46 days prior to analysis.

2.1.5 Saclay, France

Aerosol particle samples were collected at the Commissariat à l'Energie Atomique (CEA) Atmospheric Supersite (AS), CEA l'Orme des Merisiers. The CEA-AS Observatory is a suburban area located 30 km southeast of Paris in Saclay, France (labeled 7 in Fig. 1). The CEA-AS Observatory is surrounded by different sources of bioaerosols such as forest and agricultural fields, and is often influenced by marine or urban air masses (Baisnée et al., 2014). Measurements were made as part of the BIODETECT 2014 intensive campaign, an intercomparison of bioaerosol detection methods (Sarda-Estève et al., 2014). During this study

Ryan Mason 10/11/2015 3:43 PM
Moved up [1]: The Whistler Peak High Elevation Site is located at the summit of Whistler Mountain in Whistler, British Columbia, Canada (labeled 3 in Fig. 1) and operated by Environment Canada (Gallagher et al., 2011; Macdonald et al., 2011). Aerosol particle collection at this alpine site occurred between March 30 and April 23, 2014. The louvered TSP inlet was located approximately 10 m from a chairlift operating station. Although there are no continuous combustion sources at the site, sampled air may have been influenced by engine exhaust for short periods of time due to nearby snowmobile operation. Samples were stored in the dark at 4 °C for a period of 1–4 days prior to analysis.

Ryan Mason 10/11/2015 3:45 PM
Deleted: UBC campus

Ryan Mason 10/11/2015 3:45 PM
Moved (insertion) [3]

Ryan Mason 10/11/2015 3:51 PM
Moved down [6]: Four sets of aerosol particle samples were collected from a weather station on the roof of the five-story Earth Sciences Building on the UBC campus in British Columbia, Canada (labeled 4 in Fig. 1). The UBC campus is located on a peninsula and is surrounded by forest on three sides and ocean on the fourth. The site has been classified as suburban since it is less than 10 km from downtown Vancouver. Samples were collected through a TSP inlet and 0.5 m mast between May 12 and May 16, 2014. The aerosol particles were stored in the dark at 4 °C for a period of 21–23 days prior to analysis.

Ryan Mason 10/11/2015 3:46 PM
Deleted: Amphitrite Point

Ryan Mason 10/11/2015 3:46 PM
Moved (insertion) [4]

period, the site was heavily influenced by urban outflow. A large set of ancillary measurements was done to constrain all the particulate matter sources. Aerosol particles were sampled through a TSP inlet and 10 m mast between July 15 and August 4, 2014, and were stored in the dark at 4 °C for a period of 55–217 days prior to analysis.

2.1.6 UBC Campus

Four sets of aerosol particle samples were collected from a weather station on the roof of the five-story Earth Sciences Building on the UBC campus in British Columbia, Canada (labeled 4 in Fig. 1). The UBC campus is located on a peninsula and is surrounded by forest on three sides and ocean on the fourth. The site has been classified as suburban since it is less than 10 km from downtown Vancouver. Samples were collected through a TSP inlet and 0.5 m mast between May 12 and May 16, 2014. The aerosol particles were stored in the dark at 4 °C for a period of 21–23 days prior to analysis.

2.1.7 Colby, KS

Aerosol particles were collected at the soybean and sorghum fields of the Kansas State University Northwest Research Center in Colby, KS, USA (labeled 6 in Fig. 1). One sample was collected at each location during combine harvesting from a distance approximately 3–10 m downwind of the field. A third sample was also collected at the sorghum field the night following harvest. Sampling took place on October 14 and 15, 2014 and samples were stored in the dark at 4 °C for a period of 41–46 days prior to analysis.

2.2 Size-resolved INP number concentrations

INP number concentrations as a function of size and temperature were determined using the micro-orifice uniform deposit impactor-droplet freezing technique (MOUDI-DFT; Huffman

Ryan Mason 10/11/2015 3:44 PM
Moved up [2]: The coastal site at Amphitrite Point on Vancouver Island, British Columbia, Canada (labeled 5 in Fig. 1) is operated by Environment Canada, the BC Ministry of Environment, and Metro Vancouver for the continuous monitoring of aerosols and trace gases influenced by marine trajectories (McKendry et al., 2014; Yakobi-Hancock et al., 2014; Mason et al., 2015b). The mobile laboratory used during sampling was located approximately 100 m from the high tide line of the Pacific Ocean along a rocky shoreline, separated from the ocean by a narrow row of trees and shrubs approximately 2–10 m in height. Sampling took place from August 6 to August 27, 2013 using a louvered TSP inlet and 3 m mast. Aerosol particles were stored at room temperature and analyzed within 1 day of collection.

Ryan Mason 10/11/2015 3:51 PM
Deleted: Colby, KS

Ryan Mason 10/11/2015 3:46 PM
Moved down [5]: Aerosol particles were collected at the soybean and sorghum fields of the Kansas State University Northwest Research Center in Colby, KS, USA (labeled 6 in Fig. 1). One sample was collected at each location during combine harvesting from a distance approximately 3–10 m downwind of the field. A third sample was also collected at the sorghum field the night following harvest. Sampling took place on October 14 and 15, 2014 and samples were stored in the dark at 4 °C for a period of 41–46 days prior to analysis

Ryan Mason 10/11/2015 3:51 PM
Deleted: .

Ryan Mason 10/11/2015 3:51 PM
Moved (insertion) [6]

Ryan Mason 10/11/2015 3:51 PM
Deleted:

Ryan Mason 10/11/2015 3:46 PM
Deleted: Saclay, France

Ryan Mason 10/11/2015 3:46 PM
Moved (insertion) [5]

Ryan Mason 10/11/2015 3:46 PM
Moved up [4]: . Aerosol particle samples were collected at the Commissariat à l'Energie Atomique (CEA) Atmospheric Supersite (AS), CEA l'Orme des Merisiers. The CEA-AS Observatory is a suburban area located 30 km southeast of Paris in Saclay, France (labeled 7 in Fig. 1). The CEA-AS Observatory is surrounded by different sources of bioaerosols such as forest and agricultural fields, and is often influenced by marine or urban air masses (Baisnée et al., 2014). Measurements were made as part of the BIODETECT 2014 ... [1]

et al., 2013; Mason et al., 2015a). This technique combines aerosol particle collection by a cascade inertial impactor with sharp size-cutoff characteristics (the MOUDI; Marple et al., 1991) with an established droplet freezing apparatus (the DFT) for determining immersion-mode freezing properties (Koop et al., 1998; Iannone et al., 2011; Haga et al., 2013). A similar approach has also been used to study deposition nucleation by particles collected from the atmosphere (Wang et al., 2012; Knopf et al., 2014).

2.2.1 Aerosol particle sampling

Size-fractionated aerosol particle samples were collected onto hydrophobic glass cover slips (HR3-215; Hampton Research, Aliso Viejo, CA, USA) using a model 110R or 120R MOUDI (MSP Corp., Shoreview, MN, USA). Previous work has shown that these hydrophobic glass surfaces do not cause significant heterogeneous ice nucleation (e.g. Haga et al., 2013, 2014; Wheeler et al., 2015). Substrate holders were used on the impaction plates of the MOUDI to reproducibly position the hydrophobic glass cover slips in regions where aerosol deposit particle concentrations did not vary significantly (for details see Mason et al., 2015a). At most locations MOUDI stages 2–9 were used, corresponding to particle size bins of 10–5.6, 5.6–3.2, 3.2–1.8, 1.8–1.0, 1.0–0.56, 0.56–0.32, 0.32–0.18, and 0.18–0.10 µm (50 % cutoff aerodynamic diameter; Marple et al., 1991), respectively. The range in particle size collected at each location is given in Table 1.

Bounce within inertial impactors such as the MOUDI can occur during aerosol sampling, where particles impact the collection substrate but are not retained. This rebounding of particles from the surface could possibly alter the INP number concentrations and size distributions being measured. If composition is held constant, bounce is expected to increase with particle size because of their greater kinetic energy (Dahneke, 1971). Hence, INP number concentrations for

large particle sizes may be underestimated here. Bounce is also expected to increase with decreasing relative humidity. Previous work has shown that having a sample relative humidity of 70 % or greater can be effective in reducing particle bounce (e.g. Winkler, 1974; Fang et al., 1991; Stein et al., 1994; Vasiliou et al., 1999; Chen et al., 2011; Bateman et al., 2014), although its efficacy is dependent on particle type (Winkler, 1974; Lawson, 1980; Saukko et al., 2012). For six out of the seven sites investigated here, the average RH during sampling was 69% or greater. Recently, results from the MOUDI-DFT and the continuous flow diffusion chamber were compared during an ambient field campaign at Colorado State University (Mason et al., 2015a). For particle sizes < 2.4 µm, the INP number concentrations measured by the MOUDI-DFT were within experimental error of those measured by the continuous flow diffusion chamber technique, suggesting that bounce was not an issue during these previous ambient field measurements. Based on these previous measurements, in the current studies we do not consider the issue of particle bounce when calculating INP number concentrations and size distributions. Nevertheless, additional studies are warranted to better quantify the effect of bounce.

Ryan Mason 12/16/2015 12:31 PM
Deleted: on reported INP concentrations and size distributions

2.2.2 Freezing measurements

Samples were analyzed by the DFT to determine the number concentration of particles active in the immersion-freezing mode. Details of the experimental procedure can be found in Mason et al. (2015a). Briefly, samples were transferred to a temperature- and humidity-controlled flow cell coupled to an optical microscope equipped with a 5× magnification objective (Axiolab; Zeiss, Oberkochen, Germany). Water droplets were condensed onto the sample and monitored using a CCD camera recording a digital video. Since the relative humidity of the gas flow during droplet condensation was held at approximately 120 %, water condensation occurred uniformly on the cover slip, and growing droplets coagulated as they grew to a final size of 97 ±

42 µm (mean diameter and 1 standard deviation (SD) uncertainty). The freezing temperature of each droplet was then determined during cooling at a rate of -10 °C min^{-1} using the video timestamp and a resistance temperature detector located within the flow cell. Note that droplet growth and coagulation occurs in the same manner for samples containing particles and clean hydrophobic glass cover slips (no particles deposited). In addition, based on an analysis of samples collected at Amphitrite Point, more than 99 % of particles become incorporated into the droplets prior to the freezing experiments. Here we regard ice nucleation as a singular process (i.e. strictly temperature-dependent) but note that the stochastic (i.e. time-dependent) component to immersion freezing (Vali, 2014) may alter the median freezing temperature of a droplet by 0.5–2 °C per decade change in cooling rate (Murray et al., 2011; Welti et al., 2012; Wright and Petters, 2013; Wright et al., 2013; Wheeler et al., 2015).

A potential issue with the droplet freezing technique is heterogeneous ice nucleation initiated by the hydrophobic glass cover slips used to collect atmospheric particles. To address this issue, experiments were conducted using new hydrophobic glass cover slips that were processed in the same manner as ambient samples except they were not exposed to atmospheric particles drawn into the MOUDI. For the five hydrophobic glass cover slips investigated, which contained 231 droplets generated during the freezing experiments, the average freezing temperature was -36.5 ± 0.5 ºC (1 SD). In addition, none of the droplets froze above -33.7 ºC in these blank experiments. Since we only report INP number concentrations for temperatures from -15 ºC to -25 ºC in this study, heterogeneous ice nucleation by the substrate is unlikely to contribute to the reported INP number concentrations.

2.2.3 Calculating the number concentration of INPs

The number of INPs in the DFT, #INPs(T), was calculated using the following equation

Ryan Mason 11/3/2015 8:10 PM
Deleted: formation

Ryan Mason 12/16/2015 12:36 PM
Deleted: were

which accounts for the possibility of a droplet containing multiple INPs (Vali, 1971):

$$\#\mathrm{INPs}(T) = -\ln\left(\frac{N_u(T)}{N_o}\right) N_o f_{0.25-0.10\mathrm{mm}} f_{ne} \qquad (1)$$

where $N_u(T)$ is the number of unfrozen droplets at temperature T, N_o is the total number of droplets, $f_{nu,0.25-0.10mm}$ is a non-uniformity correction factor that takes into account non-uniformity at the 0.25-1 mm scale, and f_{ne} is a statistical uncertainty derived for a given number of detected nucleation events, with fewer nucleation events leading to greater statistical uncertainty (Koop et al., 1997). For the results reported, here f_{ne} is derived for a confidence level of 0.95.

Equation (1) assumes that droplets in a given freezing experiment have the same volume (Vali, 1971). Using droplet freezing experiments reported in Mason et al. (2015a), which are similar to experiments presented here, we explored if this assumption leads to uncertainties when applied to the DFT experiments. Number of INPs was first calculated from the DFT experiments as described above. Second, number of INPs were calculated by first separating the droplet freezing results into 2-4 bins based on droplet volume. After binning the data by droplet volume, droplets in each bin are more similar in volume. Equation 1 was then used to calculate the number of INPs in each bin. Finally, the total number of INPs was determined by summing the numbers of INPs calculated for each bin. We found that the total number of INPs determined both ways (with and without binning) agreed within the experimental uncertainty at freezing temperatures of -25 °C and above for 97 % of freezing events. We also analyzed the same data by first separating the droplet freezing results into 2-4 bins based on the maximum area the droplets covered. Again, the number of INPs determined with and without binning was in good agreement, being within the experimental uncertainty at freezing temperatures of -25 °C and above for 98 % of freezing events. Based on this analysis we conclude that the application of

Ryan Mason 11/3/2015 9:57 PM
Deleted: [$\#\mathrm{INPs}(T)$]

Ryan Mason 11/3/2015 9:57 PM
Deleted: $\left(\frac{A_{deposit}}{A_{DFT}V}\right) f_{nu}$

Equation (1), which assumes monodisperse droplets, to the DFT results at -25 °C and above does not lead to large uncertainties.

In the DFT, once a droplet freezes it may grow by vapor diffusion and contact a neighboring liquid droplet, causing the latter to freeze as well. To account for these non-immersion freezing events, we calculated the #INPs(T) in the immersion mode using two difference scenarios: (i) we calculated an upper limit by assuming that all droplets which underwent the processes discussed above froze by immersion freezing; and (ii) we calculated a lower limit to the INP concentration by assuming that all droplets which underwent the processes discussed above remained liquid until the homogeneous freezing temperature of approximately -37 °C (Wheeler et al., 2015).

After #INPs(T) were determined for a given freezing experiment, atmospheric INP number concentrations, [INPs(T)], were calculated using the following equation:

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

where A_{deposit} is the total area of the sample deposit on the MOUDI impaction plate, A_{DFT} is the area of the sample analyzed by the DFT (1.2 mm^2 in all samples), V is the volume of air sampled by the MOUDI, and $f_{\mathrm{nu,1mm}}$ is a correction factor to account for non-uniformity in particle concentration of the sample deposit at the 1 mm scale. See Mason et al. (2015a) for details. Values of A_{deposit}, $f_{\mathrm{nu,0.25–0.10mm}}$, $f_{\mathrm{nu,1mm}}$, and f_{ne} are given in Tables S1 and S2 and discussed in Section S1 of the Supplement. Reported INP number concentrations at each location are averaged over all samples and have been adjusted to standard temperature and pressure. In calculating averages over all sampling locations, measurements have not been weighted by sample number.

Ryan Mason 12/16/2015 12:41 PM
Deleted: were

Ryan Mason 10/13/2015 11:45 AM
Deleted: hydrophobic glass cover slips

Allan 12/13/2015 1:23 PM
Deleted: in particle concentration across the entire MOUDI sample deposit as f_{nu} is a correction factor to account for changes in particle concentration across each MOUDI sample (because the DFT analyzes only a fraction of the entire sample)., and f_{ne} is a correction factor

Ryan Mason 11/3/2015 9:59 PM
Deleted: based on the analysis of Koop et al. (1997) to account for the uncertainty associated with the number of nucleation events in each experiment (

Ryan Mason 11/3/2015 10:07 PM
Deleted: s

For the experimental conditions used in the current study the maximum number concentration of INPs that could be detected was roughly 20 L^{-1}. This maximum number concentration is greater than that reported in Huffman et al. (2013) because in the current studies shorter sampling times were used.

3 Results and discussion

3.1 INP number concentrations

The total number concentration of INPs active at -15, -20, and -25 °C are shown for each site in Fig. 2. Freezing events were rare at temperatures warmer than -15 °C, accounting for only 1.3 % of all cases, and are therefore not reported. Some of the DFT experiments proceeded such that all droplets froze at temperatures slightly below -25 °C. Since this scenario prohibits calculation of INP number concentrations, -25 °C is the lowest temperature reported. As expected, INP number concentrations were found to increase with decreasing freezing temperature with the average concentration at -25 °C (3.8 ± 2.9 L^{-1}) being more than an order of magnitude larger than at -15 °C (0.25 ± 0.15 L^{-1}).

INP number concentrations were relatively low at the Alert and Whistler Mountain sites with values of 0.05 and 0.10 L^{-1} at -15 °C, 0.22 and 0.16 L^{-1} at -20 °C, and 0.99 and 1.1 L^{-1} at -25 °C, respectively. These findings are consistent with previous measurements at similar locations. For example, Arctic measurements of Bigg (1996) for particles < 10 µm and Fountain and Ohtake (1985) using filter samples found mean INP concentrations of 0.01 L^{-1} at -15 °C and 0.13 L^{-1} at -20 °C, respectively, with both deposition and condensation modes likely possible, and Prenni et al. (2007) measured an average INP number concentration for particles with and aerodynamic diameter < 1.5 µm of approximately 0.33 L^{-1} between -8 and -28 °C with deposition, condensation, and immersion modes likely possible. At high elevation sites, Bowers

Ryan Mason 12/16/2015 12:42 PM
Deleted: s

Ryan Mason 10/4/2015 9:08 PM
Deleted: As

Ryan Mason 10/12/2015 9:30 AM
Deleted: 77

Ryan Mason 10/10/2015 6:45 PM
Deleted:

Ryan Mason 10/11/2015 3:47 PM
Deleted: , NU

Ryan Mason 10/12/2015 9:37 AM
Deleted: 06

Ryan Mason 10/18/2015 5:16 PM
Deleted: using a diffusion chamber

et al. (2009) at Mt. Werner in Colorado and Conen et al. (2012) at the research station Jungfraujoch in Switzerland measured mean immersion-mode INP number concentrations of approximately 0.02 L^{-1} at -10 and -12 ºC, respectively, using filter samples of 0.2–0.3 µm pore size. High elevation sites can receive large quantities of dust, which can act as efficient INPs at lower temperatures (Chou et al., 2011), but this was unlikely during our measurement period based on the low INP number concentrations.

INP number concentrations at Amphitrite Point were 0.23, 0.94, and 2.15 L^{-1} at droplet freezing temperatures of -15, -20, and -25 °C, respectively. Despite the predominance of marine air masses being sampled, the major source of INPs at Amphitrite Point during the study period was likely biological particles from local vegetation (Mason et al., 2015b). Similar values were measured in the Labrador Sea, where INP number concentrations at -15, -20, and -25 °C were 0.38, 1.3, and 2.8 L^{-1}, respectively. These concentrations are consistent with previous measurements within the marine boundary layer in regions influenced by air flow off of nearby coasts, for instance those of Schnell (1977) in the immersion freezing mode for sizes > 0.45 µm off the coast of Nova Scotia, roughly 1100–1500 km southwest of our sampling site in the Labrador Sea, and Rosinski et al. (1995) over the East China Sea in the deposition and condensation modes for sizes > 0.2 µm. However, INP number concentrations found during marine studies can vary by several orders of magnitude with changing location as summarized by Burrows et al. (2013).

The highest concentrations of INPs at a freezing temperature of -25 °C were found at the Colby sites, where the average number concentration was 8.9 L^{-1}. Aerosol sampling was conducted adjacent to soya and sorghum fields during and following periods of combine operation. This high concentration of INPs is consistent with previous work of Garcia et al.

Ryan Mason 10/18/2015 5:29 PM
Deleted: using freezing assays

Ryan Mason 10/11/2015 3:26 PM
Deleted: be good ice nuclei

Ryan Mason 10/12/2015 9:37 AM
Deleted: 2

Ryan Mason 10/12/2015 9:37 AM
Deleted: 79

Ryan Mason 9/13/2015 8:08 PM
Deleted: the

Ryan Mason 9/13/2015 8:07 PM
Deleted: eastern

Ryan Mason 9/13/2015 8:08 PM
Deleted: of continents

Ryan Mason 10/11/2015 3:48 PM
Deleted: , KS

Ryan Mason 10/12/2015 9:37 AM
Deleted: 89

(2012) that showed elevated concentrations of INPs downwind of corn fields during combine harvesting, and Bowers et al. (2011) who found greater INP concentrations in air above cropland than above suburban or forest sites.

The suburban sites of Saclay and the UBC campus also showed high INP concentrations, being 4.4 and 6.1 L^{-1}, respectively, at a freezing temperature of -25 °C. Both sites were likely influenced by multiple sources of INPs. For example, both are in close proximity to major metropolitan centers and forest vegetation, which are potential sources of anthropogenic INPs (e.g. Hobbs and Locatelli, 1970; Al-Naimi and Saunders, 1985; Knopf et al., 2010, 2014; Ebert et al., 2011; Corbin et al., 2012; Cziczo et al., 2013; Brooks et al., 2014) and biological INPs (e.g. Vali et al., 1976; Kieft and Ruscetti, 1990; Richard et al., 1996; Hirano and Upper, 2000; Diehl et al., 2002; Prenni et al., 2009; Iannone et al., 2011; Pummer et al., 2012; Huffman et al., 2013; Tobo et al., 2013; Haga et al., 2014; Wright et al., 2014), respectively. The sampling site at Saclay was also within 1 km of agricultural fields, an additional source of biological aerosols that may act as INPs (e.g. Lindow et al., 1982; Hirano et al., 1985; Georgakopoulos and Sands, 1992; Möhler et al., 2008; Bowers et al., 2011; Garcia et al., 2012; Haga et al., 2013; Morris et al., 2013; Hiranuma et al., 2015).

Ryan Mason 10/11/2015 3:48 PM
Deleted: , France

Ryan Mason 10/12/2015 9:39 AM
Deleted: 39

Ryan Mason 10/12/2015 9:39 AM
Deleted: 2

Ryan Mason 10/11/2015 3:48 PM
Deleted: , France

3.2 INP size distributions

Figure 3a shows the relative contribution of supermicron aerosol particles to the total measured INP population. Note that the same particle size range was not investigated at all locations with particles in the range of 0.10–0.18 µm not being measured at Whistler Mountain or Amphitrite Point, and no uncertainty is reported for the Labrador Sea measurement as only a single sample was available. Averaging over all sampling locations with a 1 SD uncertainty, 91 ± 9, 79 ± 17, and 63 ± 21 % of INPs had an aerodynamic diameter > 1 µm at ice activation

temperatures of -15, -20, and -25 °C, respectively. At -15 °C, the percentage of supermicron INPs ranged from 78 % at Whistler Mountain up to 100 % at the Labrador Sea and Colby sites. At lower temperatures, there was more variation between samples: at -20 ºC the percentage of supermicron INPs ranged from 52 % at Whistler Mountain to 100 % over the Labrador Sea, and at -25 ºC the percentage of supermicron INPs ranged from 39 % at Whistler Mountain to 95 % over the Labrador Sea.

Figure 3b shows the fraction of INPs that are in the coarse mode, calculated by assuming that half of the INPs found in the 1.8–3.2 µm MOUDI size cut were larger than 2.5 µm (i.e. INPs are uniformly distributed over this size range). Measurements of the total particle size distribution were not available at all locations to test this assumption. Furthermore, it is not known if the INP size distribution follows the total particle size distribution *a priori*. Averaging over all sampling locations, the percentage of INPs in the coarse mode was 62 ± 20, 55 ± 18, and 42 ± 17 % (1 SD) at ice activation temperatures of -15, -20, and -25 °C, respectively. The percentage of INPs in the coarse mode was found to range from 38 % at Saclay to 91 % in Colby at -15 °C, from 26 % at Whistler Mountain to 73 % in Colby at -20 °C, and from 20 % at Alert to 64 % at the Labrador Sea at -25 °C. Despite great diversity in the studied locations, each had a significant contribution from coarse mode particles to the measured INP population.

The median sizes of INPs at ice activation temperatures of -15, -20, and -25 °C are shown in Fig. 4 with the 25th and 75th percentile values. At -15 °C, the median INP size is relatively large at all locations, varying from 2.1 µm at Saclay to 4.7 µm at Colby with an average of 3.2 ± 0.9 µm (1 SD). As droplet freezing temperature decreased, the median INP size also decreased, with the exception of samples from the Labrador Sea and the UBC campus. At -25 °C, the median size of INPs varied from 0.83 µm at Alert to 3.1 µm at the Labrador Sea site with an

Ryan Mason 10/11/2015 3:48 PM
Deleted: , KS

Ryan Mason 10/11/2015 3:47 PM
Deleted: , France

Ryan Mason 10/11/2015 3:47 PM
Deleted: , KS

Ryan Mason 10/11/2015 3:47 PM
Deleted: , KS

Ryan Mason 10/11/2015 3:47 PM
Deleted: , NU

Ryan Mason 10/11/2015 3:48 PM
Deleted: , France

Ryan Mason 10/11/2015 3:48 PM
Deleted: , KS

Ryan Mason 10/11/2015 3:48 PM
Deleted: , NU

average of 1.9 ± 1.0 µm. The median size of the INPs was > 1 µm in all cases with the exception of the Alert and Whistler Mountain sites at a freezing temperature of -25 °C. Alert, in the polar tundra at high latitude, and Whistler Mountain, at high elevation and periodically in the free troposphere, are remote with fewer local sources of aerosols.

In Fig. 4, the difference between the 75th and 25th percentile sizes is relatively small at all locations at -15 °C. A narrow INP size distribution at -15 °C is consistent with a single type or class of particles dominating freezing at this temperature. With decreasing temperature, the interquartile range significantly increased: the 75th and 25th percentile INP sizes decreased by an average factor of 1.2 and 3.7, respectively, between -15 and -25 °C, corresponding to a 63 % increase in the average interquartile range.

INP size distributions are further explored in Fig. 5, where the fraction of the measured INP number concentration found in each MOUDI size bin is shown. Colors on the red end of the scale illustrate that a large fraction of the INPs measured at a particular location belong to that particle size bin. Total INP number concentrations as a function of temperature are given in Fig. S1 and histograms of the INP size distributions are given in Figs. S2–S8 of the Supplement.

Figure 5a shows that most INPs active at -15 °C were 1–10 µm in size. In particular, when averaged over all locations, 72 % of the INPs active at -15 °C were between 1.8 and 5.6 µm. Furthermore, the major mode (i.e. the global maximum in a size distribution) was always larger than 1.8 µm. At lower freezing temperatures the INP size distributions broadened with increased contributions from smaller aerosol particles, evident by the more uniform intensity of Fig. 5b and c. By -25 °C, six of the seven locations had a submicron INP mode, and at Alert and Whistler Mountain this was the major mode. A general broadening of the INP size distribution

Ryan Mason 11/3/2015 12:02 AM
Deleted: .

Ryan Mason 10/11/2015 3:49 PM
Deleted: , NU

Ryan Mason 10/11/2015 3:49 PM
Deleted: , NU

Ryan Mason 11/3/2015 12:02 AM
Deleted: At times, long-range transport may strongly influence atmospheric composition at these two locations (Barrie, 1986; Worthy et al., 1994; Liang et al., 2004; McKendry et al., 2008; Shindell et al., 2008; Macdonald et al., 2011; Takahama et al., 2011; Fan, 2013). If INP sources are not local to these sites, removal of large particles via wet and dry deposition processes during long-range transport may account for the greater contribution from smaller INPs. Similar shifts in size distributions during long-range transport have been observed at other locations for total particles (Mori et al., 2003) and fungal spores (Fröhlich-Nowoisky et al., 2012). Transport losses may have also contributed to the small overall INP number concentrations at the Alert, NU and Whistler Mountain sites as shown in Fig. 2.

Ryan Mason 12/15/2015 8:57 AM
Deleted: H

Ryan Mason 12/15/2015 8:57 AM
Deleted: also available

Ryan Mason 12/15/2015 8:46 AM
Deleted: 1

Ryan Mason 12/15/2015 8:46 AM
Deleted: 7

Ryan Mason 11/3/2015 12:03 AM
Deleted: .

Ryan Mason 10/11/2015 3:49 PM
Deleted: , NU

with decreasing temperature would be expected if there were an increase in the number of particle types exhibiting ice activity with decreasing activation temperature.

Several previous studies have conducted size-resolved INP measurements. In most of these studies, the fraction of INPs larger than a given particle size was not reported, and in some cases the temperatures studied were different than in the current study. To better compare our data with these previous studies, we have used the literature data to calculate the fraction of INPs larger than either 1, 1.2, or 2.5 µm at temperatures as close as possible to the freezing temperatures we used. Details of the calculations are presented in the Supplement, and the results of the calculations are summarized in Table 2.

Table 2 shows that in six out of the nine previous studies, a large fraction (16–100 %) of the INPs was found to be supermicron in size, consistent with the current study. Here the work of Rosinski et al. (1986) is considered as two separate studies given the change in the investigated mode of ice nucleation. Although the condensation and immersion freezing measurements reported in Rosinski et al. (1986) appear contradictory, it is important to note that that the two freezing results for Rosinski et al. (1986) shown in Table 2 correspond to different temperature ranges (compare -5 °C to -6 °C for condensation and -10.8 °C for immersion freezing). One possibility is that small particles dominated the INP population at the warmest temperatures while larger particles dominated the INP population at the colder temperatures investigated by Rosinski et al. (1986).

The ice nucleation efficiency of particles as a function of size have also been investigated in laboratory experiments (e.g. Lüönd et al. (2010), Archuleta et al. (2005), Welti et al. (2009)). In general this work has shown the ice nucleation efficiency increases as particle size increases. Furthermore, several studies have investigated correlations between the concentrations of INPs

Ryan Mason 10/11/2015 10:02 AM
Deleted: seven

Allan 12/13/2015 8:39 PM
Deleted: Kumai (1961) and Rucklidge (1965) also made direct measurements of INP size by measuring aerosol particle residuals found in collected ice crystals using electron microscopy. When multiple particles were present in their samples, the INP was assumed to be the largest particle. Kumai (1961) determined that all INPs were between 0.2 and 8 µm in crystals formed at temperatures of approximately -4 to -21 °C, with the size of maximum frequency ranging from 1–3.5 µm depending on the ice-crystal form. Rucklidge (1965), measuring INPs activated between -12 and -25 °

and aerosol particles above a certain size (Richardson et al., 2007; DeMott et al., 2010; Chou et al., 2011; Field et al., 2012; Huffman et al., 2013; Prenni et al., 2013; Tobo et al., 2013; Ardon-Dryer and Levin, 2014; Jiang et al., 2014, 2015). For example, using data from a variety of field measurements DeMott et al. (2010) observed a correlation between INP concentrations and the concentration of aerosol particles > 0.5 µm, and Ardon-Dryer and Levin (2014) found that INP concentrations in Israel were better correlated to the concentration of aerosol particles 2.5–10 µm in size than those < 2.5 µm. These results are also consistent with INPs being relatively large in size.

4 Summary and conclusions

INP number concentrations in the immersion mode as a function of size and droplet freezing temperature were determined at six locations across North America and one in Europe. INP number concentrations varied by as much as an order of magnitude between locations, and were generally found to be lowest at the remote sites of Alert and Whistler Mountain and highest at the agricultural sites of Colby and the suburban sites of Saclay and the UBC campus, consistent with previous studies. Several key findings indicate the potential importance of large INPs at ground level: (1) 91 ± 9 and 62 ± 20 % of INPs measured at -15 °C across all locations are supermicron or in the coarse mode, respectively; (2) at the lowest temperature analyzed, -25 °C, 63 ± 21 and 42 ± 17 % of INPs across all locations remained in the supermicron regime and coarse mode, respectively; (3) at -15 °C, the median INP size was relatively large at all locations, varying from 2.1 µm at Saclay to 4.7 µm at Colby with an average of 3.2 ± 0.9 µm; and (4) at -25 °C, the median size INP varied from 0.83 µm at Alert to 3.1 µm above the Labrador Sea with an average of 1.9 ± 1.0 µm.

Our measurements indicate that, when averaged over all studied locations and

Ryan Mason 10/11/2015 3:49 PM
Deleted: , NU

Ryan Mason 10/11/2015 3:49 PM
Deleted: , KS

Ryan Mason 10/11/2015 3:50 PM
Deleted: , France

Ryan Mason 10/12/2015 7:28 PM
Deleted: particles as a source of ground-level ice nuclei

Allan 12/13/2015 8:59 PM
Deleted: or

Ryan Mason 10/11/2015 3:50 PM
Deleted: , France

Ryan Mason 10/11/2015 3:49 PM
Deleted: , KS

Ryan Mason 10/11/2015 3:50 PM
Deleted: , NU

temperatures, 78 ± 19 and 53 ± 20 % of immersion-mode INPs may be missed if either supermicron particles or coarse mode particles are not sampled at ground sites. As noted in Sect. 1, some instrumentation for measuring ambient INP number concentrations restricts the upper range of sampled aerosol particles. The data presented may be useful for estimating the fraction of INPs not measured with these instruments at ground sites and in different environments.

All measurements used in this study were conducted at ground level and, apart from those at Whistler Mountain, were also close to sea level. As the large contribution of supermicron and coarse mode INPs to the overall INP population noted here may not necessarily hold for higher altitudes, additional size-resolved INP measurements as a function of altitude are needed. In obtaining such data, careful consideration will be needed toward sampling issues with aerosol inlets and transfer through sample lines on aircraft platforms. It can be anticipated that supermicron particle transfer will be strongly restricted without special inlets or care, and this may especially impact INP sampling.

A caveat to this study is that our measurements were confined to aerosol particle sizes greater than either 0.10 or 0.18 µm. If there were a significant contribution from INPs of smaller sizes (Vali, 1966; Schnell and Vali, 1973; Pummer et al., 2012; Augustin et al., 2013; Fröhlich-Nowoisky et al., 2015; O'Sullivan et al., 2015; Tong et al., 2015; Wilson et al., 2015), and these smaller sizes did not coagulate or get scavenged by larger particles, the values presented here would represent upper limits to the contribution of supermicron and coarse mode particles to the total INP population. Additional studies exploring the relative atmospheric abundance of INPs < 0.10 µm are necessary (Hader et al., 2014). Future studies of the size distribution of INPs should also include measurements of particle mixing state to determine if particles are internally or externally mixed at the locations where the size distribution of INPs are being measured.

Ryan Mason 10/18/2015 11:34 AM
Deleted: , supermicron particle transfer will be strongly restricted

Allan 12/13/2015 9:01 PM
Deleted: One

Ryan Mason 11/12/2015 11:44 PM
Formatted: Indent: First line: 0 cm

Allan 11/1/2015 12:21 PM
Deleted: therefore

Allan 11/1/2015 12:19 PM
Deleted: Furthermore, we have assumed that particles exhibiting ice-nucleating activity were externally mixed.

Allan 11/1/2015 12:19 PM
Deleted: the

Allan 12/13/2015 9:08 PM
Deleted: relative humidity, continuous flow diffusion chamber (Mason et al., 2015a)4continuous flow diffusion chamber

Ryan Mason 11/12/2015 11:44 PM
Deleted:

Allan 11/1/2015 12:20 PM
Deleted: measured aerodynamic diameter gives an accurate representation of INP size or if smaller INPs are components of larger aggregates.

Acknowledgements

The authors would like to thank R. B. Stull and R. Schigas for access to the UBC campus site and associated weather data, and L. A. Miller for assistance coordinating the measurements from the CCGS Amundsen. The authors also wish to thank Juniper Buller and Anton Horvath for access to the Whistler Mountain sampling site, and Freddie Lamm and the agricultural specialists at the Kansas State University Northwest Research Center for access to the Colby, KS sites, advice on harvesting time frames, and their help during harvesting. The sampling site at Amphitrite Point is jointly supported and maintained by Environment Canada, the British Columbia Ministry of Environment, and Metro Vancouver. We thank the Canadian Coast Guard and Department of Fisheries and Oceans staff from the Amphitrite Point site and the CCGS Amundsen for their help. The Natural Sciences and Engineering Research Council of Canada supported this research. K. J. Suski, P. J. DeMott, and T. C. J. Hill acknowledge support under U.S. National Science Foundation grant AGS 1358495, which also provided support for measurements at the Colby, KS site. W. M. Lassar and K. M. Pierce acknowledge funding support through the University of Denver undergraduate research center. J. A. Huffman, W. M. Lassar and K. M. Pierce acknowledge the CEA/DAM/CBRN-E research programs and particularly D. Baisnée (CEA) for support in the intensive campaign and J. Sciare (CNRS) for the ancillary aerosol data.

1449 **References**

1450 Al-Naimi, R. and Saunders, C. P. R.: Measurements of natural deposition and condensation-
1451 freezing ice nuclei with a continuous flow chamber, Atmos. Environ., 19, 1871–1882,
1452 doi:10.1016/0004-6981(85)90012-5, 1985.

1453 Archuleta, C. M., DeMott, P. J. and Kreidenweis, S. M.: Ice nucleation by surrogates for
1454 atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures, Atmos. Chem.
1455 Phys., 5, 2617–2634, doi:10.5194/acp-5-2617-2005, 2005.

1456 Ardon-Dryer, K. and Levin, Z.: Ground-based measurements of immersion freezing in the
1457 eastern Mediterranean, Atmos. Chem. Phys., 14, 5217–5231, doi:10.5194/acp-14-5217-2014,
1458 2014.

1459 Ariya, P. A., Sun, J., Eltouny, N. A., Hudson, E. D., Hayes, C. T. and Kos, G.: Physical and
1460 chemical characterization of bioaerosols - Implications for nucleation processes, Int. Rev. Phys.
1461 Chem., 28, 1–32, doi:10.1080/01442350802597438, 2009.

1462 Augustin, S., Wex, H., Niedermeier, D., Pummer, B., Grothe, H., Hartmann, S., Tomsche, L.,
1463 Clauss, T., Voigtländer, J., Ignatius, K. and Stratmann, F.: Immersion freezing of birch pollen
1464 washing water, Atmos. Chem. Phys., 13, 10989–11003, doi:10.5194/acp-13-10989-2013, 2013.

1465 Baisnée, D., Thibaudon, M., Baumier, R., McMeeking, G., Kok, G., O'Connor, D., Sodeau, J.,
1466 Huffman, J. A., Lassar, W., Pierce, K., Gallagher, M., Crawford, I., Salines, G. and Sarda-
1467 Estève, R.: Simultaneous Real-time Fluorescence and Microscopy Measurements of Bioaerosols
1468 during the BIODETECT 2014 Campaign in Paris Area, AAAR 33rd Annual Conference,
1469 Orlando, Fl, October 20–24, 2014.

1470 Baker, M. B.: Cloud Microphysics and Climate, Science, 276, 1072–1078,
1471 doi:10.1126/science.276.5315.1072, 1997.

1472 Baker, M. B. and Peter, T.: Small-scale cloud processes and climate, Nature, 451, 299–300,
1473 doi:10.1038/nature06594, 2008.

1474 Bateman, A. P., Belassein, H. and Martin, S. T.: Impactor Apparatus for the Study of Particle
1475 Rebound: Relative Humidity and Capillary Forces, Aerosol Sci. Technol., 48, 42–52,
1476 doi:10.1080/02786826.2013.853866, 2014.

1477 Berezinski, N. A., Stepanov, G. V. and Khorguani, V. G.: Ice-forming activity of atmospheric
1478 aerosol particles of different sizes, Lecture Notes in Physics, edited by P. E. Wagner and G. Vali,
1479 309, 709–712, Springer, Heidelberg and Berlin, Germany, 1988.

1480 Bigg, E. K.: Ice forming nuclei in the high Arctic, Tellus B, 48, 223–233,
1481 doi:10.3402/tellusb.v48i2.15888, 1996.

1482 Bowers, R. M., Lauber, C. L., Wiedinmyer, C., Hamady, M., Hallar, A. G., Fall, R., Knight, R.
1483 and Fierer, N.: Characterization of Airborne Microbial Communities at a High-Elevation Site
1484 and Their Potential To Act as Atmospheric Ice Nuclei, Appl. Environ. Microbiol., 75, 5121–
1485 5130, doi:10.1128/AEM.00447-09, 2009.

1486 Bowers, R. M., McLetchie, S., Knight, R. and Fierer, N.: Spatial variability in airborne bacterial
1487 communities across land-use types and their relationship to the bacterial communities of
1488 potential source environments, ISME J., 5, 601–612, doi:10.1038/ismej.2010.167, 2011.

1489 Brooks, S. D., Suter, K. and Olivarez, L.: Effects of Chemical Aging on the Ice Nucleation
1490 Activity of Soot and Polycyclic Aromatic Hydrocarbon Aerosols, J. Phys. Chem. A, 118, 10036–
1491 10047, doi:10.1021/jp508809y, 2014.

1492 Burrows, S. M., Butler, T., Jöckel, P., Tost, H., Kerkweg, A., Pöschl, U. and Lawrence, M. G.:
1493 Bacteria in the global atmosphere - Part 2: Modeling of emissions and transport between
1494 different ecosystems, Atmos. Chem. Phys., 9, 9281–9297, doi:10.5194/acp-9-9281-2009, 2009.

1495 Burrows, S. M., Hoose, C., Pöschl, U. and Lawrence, M. G.: Ice nuclei in marine air: biogenic
1496 particles or dust?, Atmos. Chem. Phys., 13, 245–267, doi:10.5194/acp-13-245-2013, 2013.

1497 Chen, S.-C., Tsai, C.-J., Chen, H.-D., Huang, C.-Y. and Roam, G.-D.: The Influence of Relative
1498 Humidity on Nanoparticle Concentration and Particle Mass Distribution Measurements by the
1499 MOUDI, Aerosol Sci. Technol., 45, 596–603, doi:10.1080/02786826.2010.551557, 2011.

1500 Chou, C., Stetzer, O., Weingartner, E., Jurányi, Z., Kanji, Z. A. and Lohmann, U.: Ice nuclei
1501 properties within a Saharan dust event at the Jungfraujoch in the Swiss Alps, Atmos. Chem.
1502 Phys., 11, 4725–4738, doi:10.5194/acp-11-4725-2011, 2011.

1503 Clarke, A. D., Shinozuka, Y., Kapustin, V. N., Howell, S., Huebert, B., Doherty, S., Anderson,
1504 T., Covert, D., Anderson, J., Hua, X., Moore II, K. G., McNaughton, C., Carmichael, G. and
1505 Weber, R.: Size distributions and mixtures of dust and black carbon aerosol in Asian outflow:
1506 Physiochemistry and optical properties, J. Geophys. Res., 109, D15S09,
1507 doi:10.1029/2003JD004378, 2004.

1508 Cobbett, F. D., Steffen, A., Lawson, G. and Van Heyst, B. J.: GEM fluxes and atmospheric
1509 mercury concentrations (GEM, RGM and Hg^p) in the Canadian Arctic at Alert, Nunavut, Canada
1510 (February-June 2005), Atmos. Environ., 41, 6527–6543, doi:10.1016/j.atmosenv.2007.04.033,
1511 2007.

1512 Conen, F., Henne, S., Morris, C. E. and Alewell, C.: Atmospheric ice nucleators active ≥ -12 °C
1513 can be quantified on PM_{10} filters, Atmos. Meas. Tech., 5, 321–327, doi:10.5194/amt-5-321-2012,
1514 2012.

1515 Corbin, J. C., Rehbein, P. J. G., Evans, G. J. and Abbatt, J. P. D.: Combustion particles as ice
1516 nuclei in an urban environment: Evidence from single-particle mass spectrometry, Atmos.
1517 Environ., 51, 286–292, doi:10.1016/j.atmosenv.2012.01.007, 2012.

1518 Creamean, J. M., Suski, K. J., Rosenfeld, D., Cazorla, A., DeMott, P. J., Sullivan, R. C., White,
1519 A. B., Ralph, F. M., Minnis, P., Comstock, J. M., Tomlinson, J. M. and Prather, K. A.: Dust and
1520 Biological Aerosols from the Sahara and Asia Influence Precipitation in the Western U.S.,
1521 Science, 339, 1572–1578, doi:10.1126/science.1227279, 2013.

1522 Cziczo, D. J., Froyd, K. D., Hoose, C., Jensen, E. J., Diao, M., Zondlo, M. A., Smith, J. B.,
1523 Twohy, C. H. and Murphy, D. M.: Clarifying the Dominant Sources and Mechanisms of Cirrus
1524 Cloud Formation, Science, 340, 1320–1324, doi:10.1126/science.1234145, 2013.

1525 Dahneke, B.: The capture of aerosol particles by surfaces, J. Colloid Interface Sci., 37, 342–353,
1526 doi:10.1016/0021-9797(71)90302-X, 1971.

1527 DeMott, P. J., Cziczo, D. J., Prenni, A. J., Murphy, D. M., Kreidenweis, S. M., Thomson, D. S.,
1528 Borys, R. and Rogers, D. C.: Measurements of the concentration and composition of nuclei for

cirrus formation, P. Natl. Acad. Sci. USA, 100, 14655–14660, doi:10.1073/pnas.2532677100, 2003.

DeMott, P. J., Möhler, O., Stetzer, O., Vali, G., Levin, Z., Petters, M. D., Murakami, M., Leisner, T., Bundke, U., Klein, H., Kanji, Z. A., Cotton, R., Jones, H., Benz, S., Brinkmann, M., Rzesanke, D., Saathoff, H., Nicolet, M., Saito, A., Nillius, B., Bingemer, H., Abbatt, J., Ardon, K., Ganor, E., Georgakopoulos, D. G. and Saunders, C.: Resurgence in ice nuclei measurement research, B. Am. Meteorol. Soc., 92, 1623–1635, doi:10.1175/2011BAMS3119.1, 2011.

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T. and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, P. Natl. Acad. Sci. USA, 107, 11217–11222, doi:10.1073/pnas.0910818107, 2010.

DeMott, P. J., Rogers, D. C., Kreidenweis, S. M., Chen, Y., Twohy, C. H., Baumgardner, D., Heymsfield, A. J., and Chan, K. R.: The role of heterogeneous freezing nucleation in upper tropospheric clouds: Inferences from SUCCESS, Geophys. Res. Lett., 25, doi:10.1029/97GL03779, 1387–1390, 1998.

Després, V. R., Huffman, J. A., Burrows, S. M., Hoose, C., Safatov, A. S., Buryak, G., Fröhlich-Nowoisky, J., Elbert, W., Andreae, M. O., Pöschl, U. and Jaenicke, R.: Primary biological aerosol particles in the atmosphere: a review, Tellus B, 64, 15598, doi:10.3402/tellusb.v64i0.15598, 2012.

Diehl, K., Matthias-Maser, S., Jaenicke, R. and Mitra, S. K.: The ice nucleating ability of pollen: Part II. Laboratory studies in immersion and contact freezing modes, Atmos. Res., 61, 125–133, doi:10.1016/S0169-8095(01)00132-6, 2002.

Draxler, R. R. and Rolph, G. D.: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website, available at: http://ready.arl.noaa.gov/HYSPLIT.php (last access: 27 May 2014), NOAA Air Resources Laboratory, Silver Spring, MD, 2014.

Ebert, M., Worringen, A., Benker, N., Mertes, S., Weingartner, E. and Weinbruch, S.: Chemical composition and mixing-state of ice residuals sampled within mixed phase clouds, Atmos. Chem. Phys., 11, 2805–2816, doi:10.5194/acp-11-2805-2011, 2011.

Eidhammer, T., DeMott, P. J., Prenni, A. J., Petters, M. D., Twohy, C. H., Rogers, D. C., Stith, J., Heymsfield, A., Wang, Z., Pratt, K. A., Prather, K. A., Murphy, S. M., Seinfeld, J. H., Subramanian, R. and Kreidenweis, S. M.: Ice Initiation by Aerosol Particles: Measured and Predicted Ice Nuclei Concentrations versus Measured Ice Crystal Concentrations in an Orographic Wave Cloud, J. Atmos. Sci., 67, 2417–2436, doi:10.1175/2010JAS3266.1, 2010.

Elbert, W., Taylor, P. E., Andreae, M. O. and Pöschl, U.: Contribution of fungi to primary biogenic aerosols in the atmosphere: wet and dry discharged spores, carbohydrates, and inorganic ions, Atmos. Chem. Phys., 7, 4569–4588, doi:10.5194/acp-7-4569-2007, 2007.

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, 266–277, doi:10.1080/02786829108959489, 1991.

1570 Field, P. R., Heymsfield, A. J., Shipway, B. J., DeMott, P. J., Pratt, K. A., Rogers, D. C., Stith, J.
1571 and Prather, K. A.: Ice in Clouds Experiment-Layer Clouds. Part II: Testing Characteristics of
1572 Heterogeneous Ice Formation in Lee Wave Clouds, J. Atmos. Sci., 69, 1066–1079,
1573 doi:10.1175/JAS-D-11-026.1, 2012.

1574 Fountain, A. G. and Ohtake, T.: Concentrations and Source Areas of Ice Nuclei in the Alaskan
1575 Atmosphere, J. Clim. Appl. Meteorol., 24, 377–382, 1985.

1576 Friedman, B., Kulkarni, G., Beránek, J., Zelenyuk, A., Thornton, J. A. and Cziczo, D. J.: Ice
1577 nucleation and droplet formation by bare and coated soot particles, J. Geophys. Res., 116,
1578 D17203, doi:10.1029/2011JD015999, 2011.

1579 Fröhlich-Nowoisky, J., Hill, T. C. J., Pummer, B. G., Yordanova, P., Franc, G. D. and Pöschl,
1580 U.: Ice nucleation activity in the widespread soil fungus *Mortierella alpina*, Biogeosciences, 12,
1581 1057–1071, doi:10.5194/bg-12-1057-2015, 2015.

1582 Gallagher, J. P., McKendry, I. G., Macdonald, A. M. and Leaitch, W. R.: Seasonal and Diurnal
1583 Variations in Aerosol Concentration on Whistler Mountain: Boundary Layer Influence and
1584 Synoptic-Scale Controls, J. Appl. Meteorol. Climatol., 50, 2210–2222, doi:10.1175/JAMC-D-
1585 11-028.1, 2011.

1586 Garcia, E., Hill, T. C. J., Prenni, A. J., DeMott, P. J., Franc, G. D. and Kreidenweis, S. M.:
1587 Biogenic ice nuclei in boundary layer air over two U.S. High Plains agricultural regions, J.
1588 Geophys. Res., 117, D18209, doi:10.1029/2012JD018343, 2012.

1589 Georgakopoulos, D. G. and Sands, D. C.: Epiphytic populations of Pseudomonas syringae on
1590 barley, Can. J. Microbiol., 38, 111–114, doi:10.1139/m92-018, 1992.

1591 Graham, B., Guyon, P., Maenhaut, W., Taylor, P. E., Ebert, M., Matthias-Maser, S., Mayol-
1592 Bracero, O. L., Godoi, R. H. M., Artaxo, P., Meixner, F. X., Moura, M. A. L., Rocha, C. H. E.
1593 D., Van Grieken, R., Glovsky, M. M., Flagan, R. C. and Andreae, M. O.: Composition and
1594 diurnal variability of the natural Amazonian aerosol, J. Geophys. Res., 108, 4765,
1595 doi:10.1029/2003JD004049, 2003.

1596 Hader, J. D., Wright, T. P. and Petters, M. D.: Contribution of pollen to atmospheric ice nuclei
1597 concentrations, Atmos. Chem. Phys., 14, 5433–5449, doi:10.5194/acp-14-5433-2014, 2014.

1598 Haga, D. I., Burrows, S. M., Iannone, R., Wheeler, M. J., Mason, R. H., Chen, J., Polishchuk, E.
1599 A., Pöschl, U. and Bertram, A. K.: Ice nucleation by fungal spores from the classes
1600 *Agaricomycetes*, *Ustilaginomycetes*, and *Eurotiomycetes*, and the effect on the atmospheric
1601 transport of these spores, Atmos. Chem. Phys., 14, 8611–8630, doi:10.5194/acp-14-8611-2014,
1602 2014.

1603 Haga, D. I., Iannone, R., Wheeler, M. J., Mason, R., Polishchuk, E. A., Fetch Jr., T., van der
1604 Kamp, B. J., McKendry, I. G. and Bertram, A. K.: Ice nucleation properties of rust and bunt
1605 fungal spores and their transport to high altitudes, where they can cause heterogeneous freezing,
1606 J. Geophys. Res.-Atmos., 118, 7260–7272, doi:10.1002/jgrd.50556, 2013.

1607 Hirano, S. S., Baker, L. S. and Upper, C. D.: Ice nucleation temperature of individual leaves in
1608 relation to population sizes of ice nucleation active bacteria and frost injury, Plant Physiol., 77,
1609 259–265, doi:10.1104/pp.77.2.259, 1985.

Hirano, S. S. and Upper, C. D.: Bacteria in the leaf ecosystem with emphasis on *Pseudomonas syringae* - a pathogen, ice nucleus, and epiphyte, Microbiol. Mol. Biol. R., 64, 624–653, doi:10.1128/MMBR.64.3.624-653.2000, 2000.

Hiranuma, N., Möhler, O., Yamashita, K., Tajiri, T., Saito, A., Kiselev, A., Hoffmann, N., Hoose, C., Jantsch, E., Koop, T. and Murakami, M.: Ice nucleation by cellulose and its potential contribution to ice formation in clouds, Nat. Geosci., 8, 273–277, doi:10.1038/ngeo2374, 2015.

Hobbs, P. V and Locatelli, J. D.: Ice Nucleus Measurements at Three Sites in Western Washington, J. Atmos. Sci., 27, 90–100, 1970.

Hoose, C., Kristjánsson, J. E. and Burrows, S. M.: How important is biological ice nucleation in clouds on a global scale?, Environ. Res. Lett., 5, 024009, doi:10.1088/1748-9326/5/2/024009, 2010a.

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, 2483–2503, doi:10.1175/2010JAS3425.1, 2010b.

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

Hoyle, C. R., Pinti, V., Welti, A., Zobrist, B., Marcolli, C., Luo, B., Höskuldsson, Á., Mattsson, H. B., Stetzer, O., Thorsteinsson, T., Larsen, G. and Peter, T.: Ice nucleation properties of volcanic ash from Eyjafjallajökull, Atmos. Chem. Phys., 11, 9911–9926, doi:10.5194/acp-11-9911-2011, 2011.

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, 6151–6164, doi:10.5194/acp-13-6151-2013, 2013.

Huffman, J. A., Sinha, B., Garland, R. M., Snee-Pollmann, A., Gunthe, S. S., Artaxo, P., Martin, S. T., Andreae, M. O. and Pöschl, U.: Size distributions and temporal variations of biological aerosol particles in the Amazon rainforest characterized by microscopy and real-time UV-APS fluorescence techniques during AMAZE-08, Atmos. Chem. Phys., 12, 11997–12019, doi:10.5194/acp-12-11997-2012, 2012.

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, 1191–1201, doi:10.5194/acp-11-1191-2011, 2011.

Jiang, H., Yin, Y., Su, H., Shan, Y. and Gao, R.: The characteristics of atmospheric ice nuclei measured at the top of Huangshan (the Yellow Mountains) in Southeast China using a newly built static vacuum water vapor diffusion chamber, Atmos. Res., 153, 200–208, doi:10.1016/j.atmosres.2014.08.015, 2015.

1650 Jiang, H., Yin, Y., Yang, L., Yang, S., Su, H. and Chen, K.: The characteristics of atmospheric
1651 ice nuclei measured at different altitudes in the Huangshan Mountains in Southeast China, Adv.
1652 Atmos. Sci., 31, 396–406, doi:10.1007/s00376-013-3048-5, 2014.

1653 Kieft, T. L. and Ruscetti, T.: Characterization of biological ice nuclei from a lichen, J. Bacteriol.,
1654 172, 3519–3523, 1990.

1655 Knopf, D. A., Alpert, P. A., Wang, B., O'Brien, R. E., Kelly, S. T., Laskin, A., Gilles, M. K. and
1656 Moffet, R. C.: Microspectroscopic imaging and characterization of individually identified ice
1657 nucleating particles from a case field study, J. Geophys. Res.-Atmos., 119, 10365–10381,
1658 doi:10.1002/2014JD021866, 2014.

1659 Knopf, D. A., Wang, B., Laskin, A., Moffet, R. C. and Gilles, M. K.: Heterogeneous nucleation
1660 of ice on anthropogenic organic particles collected in Mexico City, Geophys. Res. Lett., 37,
1661 L11803, doi:10.1029/2010GL043362, 2010.

1662 Koop, T., Luo, B., Biermann, U. M., Crutzen, P. J. and Peter, T.: Freezing of $HNO_3/H_2SO_4/H_2O$
1663 solutions at stratospheric temperatures: Nucleation statistics and experiments, J. Phys. Chem. A,
1664 101, 1117–1133, doi:10.1021/jp9626531, 1997.

1665 Koop, T., Ng, H. P., Molina, L. T. and Molina, M. J.: A New Optical Technique to Study
1666 Aerosol Phase Transitions: The Nucleation of Ice from H_2SO_4 Aerosols, J. Phys. Chem. A, 102,
1667 8924–8931, doi:10.1021/jp9828078, 1998.

1668 Lawson, D. R.: Impaction surface coatings intercomparison and measurements with cascade
1669 impactors, Atmos. Environ., 14, 195–199, doi:10.1016/0004-6981(80)90278-4, 1980.

1670 Lindow, S. E., Arny, D. C. and Upper, C. D.: Bacterial ice nucleation: a factor in frost injury to
1671 plants, Plant Physiol., 70, 1084–1089, doi:10.1104/pp.70.4.1084, 1982.

1672 Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5,
1673 715–737, doi:10.5194/acp-5-715-2005, 2005.

1674 Lüönd, F., Stetzer, O., Welti, A. and Lohmann, U.: Experimental study on the ice nucleation
1675 ability of size-selected kaolinite particles in the immersion mode, J. Geophys. Res., 115,
1676 D14201, doi:10.1029/2009JD012959, 2010.

1677 Macdonald, A. M., Anlauf, K. G., Leaitch, W. R., Chan, E. and Tarasick, D. W.: Interannual
1678 variability of ozone and carbon monoxide at the Whistler high elevation site: 2002-2006, Atmos.
1679 Chem. Phys., 11, 11431–11446, doi:10.5194/acp-11-11431-2011, 2011.

1680 Marple, V. A., Rubow, K. L. and Behm, S. M.: A Microorifice Uniform Deposit Impactor
1681 (MOUDI): Description, Calibration, and Use, Aerosol Sci. Technol., 14, 434–446,
1682 doi:10.1080/02786829108959504, 1991.

1683 Mason, R. H., Chou, C., McCluskey, C. S., Levin, E. J. T., Schiller, C. L., Hill, T. C. J.,
1684 Huffman, J. A., DeMott, P. J. and Bertram, A. K.: The micro-orifice uniform deposit impactor-
1685 droplet freezing technique (MOUDI-DFT) for measuring concentrations of ice nucleating
1686 particles as a function of size: improvements and initial validation, Atmos. Meas. Tech., 8, 2449–
1687 2462, doi:10.5194/amt-8-2449-2015, 2015a.

1688 Mason, R. H., Si, M., Li, J., Chou, C., Dickie, R., Toom-Sauntry, D., Pöhlker, C., Yakobi-
1689 Hancock, J. D., Ladino, L. A., Jones, K., Leaitch, W. R., Schiller, C. L., Abbatt, J. P. D.,
1690 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. Discuss., 15, 16273–16323, doi:10.5194/acpd-8-16273-2015, 2015b.

McCluskey, C. S., DeMott, P. J., Prenni, A. J., Levin, E. J. T., McMeeking, G. R., Sullivan, A. P., Hill, T. C. J., Nakao, S., Carrico, C. M. and Kreidenweis, S. M.: Characteristics of atmospheric ice nucleating particles associated with biomass burning in the US: Prescribed burns and wildfires, J. Geophys. Res.-Atmos., 119, 10458–10470, doi:10.1002/2014JD021980, 2014.

McKendry, I. G., Christensen, E., Schiller, C. L., Vingarzan, R., Macdonald, A. M. and Li, Y.: Low Ozone Episodes at Amphitrite Point Marine Boundary Layer Observatory, British Columbia, Canada, Atmos. Ocean, 52, 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, 848–864, doi:10.1080/02786820701501881, 2007.

Möhler, O., DeMott, P. J., Vali, G. and Levin, Z.: Microbiology and atmospheric processes: the role of biological particles in cloud physics, Biogeosciences, 4, 1059–1071, doi:10.5194/bg-4-1059-2007, 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, 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, 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, 4191–4207, doi:10.5194/acp-11-4191-2011, 2011.

Murray, B. J., O'Sullivan, D., Atkinson, J. D. and Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519–6554, doi:10.1039/c2cs35200a, 2012.

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, 8082, doi:10.1038/srep08082, 2015.

Pratt, K. A., DeMott, P. J., French, J. R., Wang, Z., Westphal, D. L., Heymsfield, A. J., Twohy, C. H., Prenni, A. J. and Prather, K. A.: In situ detection of biological particles in cloud ice-crystals, Nat. Geosci., 2, 398–401, doi:10.1038/ngeo521, 2009.

1730 Prenni, A. J., Harrington, J. Y., Tjernström, M., DeMott, P. J., Avramov, A., Long, C. N.,
1731 Kreidenweis, S. M., Olsson, P. Q. and Verlinde, J.: Can Ice-Nucleating Aerosols Affect Arctic
1732 Seasonal Climate?, B. Am. Meteorol. Soc., 88, 541–550, doi:10.1175/BAMS-88-4-541, 2007.

1733 Prenni, A. J., Petters, M. D., Kreidenweis, S. M., Heald, C. L., Martin, S. T., Artaxo, P., Garland,
1734 R. M., Wollny, A. G. and Pöschl, U.: Relative roles of biogenic emissions and Saharan dust as
1735 ice nuclei in the Amazon basin, Nat. Geosci., 2, 402–405, doi:10.1038/ngeo517, 2009.

1736 Prenni, A. J., Tobo, Y., Garcia, E., DeMott, P. J., Huffman, J. A., McCluskey, C. S.,
1737 Kreidenweis, S. M., Prenni, J. E., Pöhlker, C. and Pöschl, U.: The impact of rain on ice nuclei
1738 populations at a forested site in Colorado, Geophys. Res. Lett., 40, 227–231,
1739 doi:10.1029/2012GL053953, 2013.

1740 Pummer, B. G., Bauer, H., Bernardi, J., Bleicher, S. and Grothe, H.: Suspendable
1741 macromolecules are responsible for ice nucleation activity of birch and conifer pollen, Atmos.
1742 Chem. Phys., 12, 2541–2550, doi:10.5194/acp-12-2541-2012, 2012.

1743 Richard, C., Martin, J.-G. and Pouleur, S.: Ice nucleation activity identified in some
1744 phytopathogenic Fusarium species, Phytoprotection, 77, 83–92, doi:10.7202/706104ar, 1996.

1745 Richardson, M. S., DeMott, P. J., Kreidenweis, S. M., Cziczo, D. J., Dunlea, E. J., Jimenez, J. L.,
1746 Thomson, D. S., Ashbaugh, L. L., Borys, R. D., Westphal, D. L., Casuccio, G. S. and Lersch, T.
1747 L.: Measurements of heterogeneous ice nuclei in the western United States in springtime and
1748 their relation to aerosol characteristics, J. Geophys. Res., 112, D02209,
1749 doi:10.1029/2006JD007500, 2007.

1750 Rogers, D. C., DeMott, P. J. and Kreidenweis, S. M.: Airborne measurements of tropospheric
1751 ice-nucleating aerosol particles in the Arctic spring, J. Geophys. Res., 106, 15053–15063,
1752 doi:10.1029/2000JD900790, 2001a.

1753 Rogers, D. C., DeMott, P. J., Kreidenweis, S. M. and Chen, Y.: Measurements of ice nucleating
1754 aerosols during SUCCESS, Geophys. Res. Lett., 25, 1383–1386, doi:10.1029/97GL03478, 1998.

1755 Rogers, D. C., DeMott, P. J., Kreidenweis, S. M. and Chen, Y.: A continuous-flow diffusion
1756 chamber for airborne measurements of ice nuclei, J. Atmos. Ocean. Technol., 18, 725–741,
1757 2001b.

1758 Rosinski, J., Haagenson, P. L., Nagamoto, C. T. and Parungo, F.: Ice-forming nuclei of maritime
1759 origin, J. Aerosol Sci., 17, 23–46, doi:10.1016/0021-8502(86)90004-2, 1986.

1760 Rosinski, J., Haagenson, P. L., Nagamoto, C. T., Quintana, B., Parungo, F. and Hoyt, S. D.: Ice-
1761 forming nuclei in air masses over the Gulf of Mexico, J. Aerosol Sci., 19, 539–551,
1762 doi:10.1016/0021-8502(88)90206-6, 1988.

1763 Rosinski, J., Nagamoto, C. T. and Zhou, M. Y.: Ice-forming nuclei over the East China Sea,
1764 Atmos. Res., 36, 95–105, doi:10.1016/0169-8095(94)00029-D, 1995.

1765 Rucklidge, J.: The Examination by Electron Microscope of Ice Crystal Nuclei from Cloud
1766 Chamber Experiments, J. Atmos. Sci., 22, 301–308, 1965.

1767 Santachiara, G., Di Matteo, L., Prodi, F. and Belosi, F.: Atmospheric particles acting as Ice
1768 Forming Nuclei in different size ranges, Atmos. Res., 96, 266–272,
1769 doi:10.1016/j.atmosres.2009.08.004, 2010.

1770 Sarda-Estève, R., Huffman, J. A., Gallagher, M., Thibaudon, M., Baisnee, D., Baumier, R.,
1771 McKeening, G., Kok, G., Sodeau, J., O'Connor, D., Crawford, I., Flynn, M., Saari, S., Poeschl,
1772 U., Gros, V., Favez, O., Amodeo, T., Sciare, J., Bonnaire, N., Lassar, W., Pierce, K., Chou, C.,
1773 Bertram, A., Salines, G., Roux, J.-M., Nadal, M. H., Bossuet, C. and Olmedo, L.: BIODETECT
1774 2014 campaign in Paris area: overview of the experimental strategy and preliminary results,
1775 AAAR 33rd Annual Conference, Orlando, Fl, October 20–24, 2014.

1776 Saukko, E., Kuuluvainen, H. and Virtanen, A.: A method to resolve the phase state of aerosol
1777 particles, Atmos. Meas. Tech., 5, 259–265, doi:10.5194/amt-5-259-2012, 2012.

1778 Schnell, R. C.: Ice Nuclei in Seawater, Fog Water and Marine Air off the Coast of Nova Scotia:
1779 Summer 1975, J. Atmos. Sci., 34, 1299–1305, 1977.

1780 Schnell, R. C. and Vali, G.: World-wide source of leaf-derived freezing nuclei, Nature, 246,
1781 212–213, doi:10.1038/246212a0, 1973.

1782 Schwarz, J. P., Gao, R. S., Perring, A. E., Spackman, J. R. and Fahey, D. W.: Black carbon
1783 aerosol size in snow, Sci. Rep., 3, 1356, doi:10.1038/srep01356, 2013.

1784 Schwarz, J. P., Gao, R. S., Spackman, J. R., Watts, L. A., Thomson, D. S., Fahey, D. W.,
1785 Ryerson, T. B., Peischl, J., Holloway, J. S., Trainer, M., Frost, G. J., Baynard, T., Lack, D. A., de
1786 Gouw, J. A., Warneke, C. and Del Negro, L. A.: Measurement of the mixing state, mass, and
1787 optical size of individual black carbon particles in urban and biomass burning emissions,
1788 Geophys. Res. Lett., 35, L13810, doi:10.1029/2008GL033968, 2008.

1789 Sesartic, A. and Dallafior, T. N.: Global fungal spore emissions, review and synthesis of
1790 literature data, Biogeosciences, 8, 1181–1192, doi:10.5194/bg-8-1181-2011, 2011.

1791 Sesartic, A., Lohmann, U. and Storelvmo, T.: Modelling the impact of fungal spore ice nuclei on
1792 clouds and precipitation, Environ. Res. Lett., 8, 014029, doi:10.1088/1748-9326/8/1/014029,
1793 2013.

1794 Spracklen, D. V and Heald, C. L.: The contribution of fungal spores and bacteria to regional and
1795 global aerosol number and ice nucleation immersion freezing rates, Atmos. Chem. Phys., 14,
1796 9051–9059, doi:10.5194/acp-14-9051-2014, 2014.

1797 Stein, S. W., Turpin, B. J., Cai, X., Huang, P.-F. and McMurray, P. H.: Measurements of relative
1798 humidity-dependent bounce and density for atmospheric particles using the DMA-impactor
1799 technique, Atmos. Environ., 28, 1739–1746, doi:10.1016/1352-2310(94)90136-8, 1994.

1800 Szyrmer, W. and Zawadzki, I.: Biogenic and anthropogenic sources of ice-forming nuclei: A
1801 review, B. Am. Meteorol. Soc., 78, 209–228, 1997.

1802 Tobo, Y., Prenni, A. J., DeMott, P. J., Huffman, J. A., McCluskey, C. S., Tian, G., Pöhlker, C.,
1803 Pöschl, U. and Kreidenweis, S. M.: Biological aerosol particles as a key determinant of ice nuclei
1804 populations in a forest ecosystem, J. Geophys. Res., 118, 10100–10110, doi:10.1002/jgrd.50801,
1805 2013.

1806 Tong, H.-J., Ouyang, B., Nikolovski, N., Lienhard, D. M., Pope, F. D. and Kalberer, M.: A new
1807 electrodynamic balance (EDB) design for low-temperature studies: application to immersion
1808 freezing of pollen extract bioaerosols, Atmos. Meas. Tech., 8, 1183–1195, doi:10.5194/amt-8-
1809 1183-2015, 2015.

1810 Vali, G.: Sizes of Atmospheric Ice Nuclei, Nature, 212, 384–385, doi:10.1038/212384a0, 1966.

1811 Vali, G.: Quantitative Evaluation of Experimental Results on the Heterogeneous Freezing
1812 Nucleation of Supercooled Liquids, J. Atmos. Sci., 28, 402–409, 1971.

1813 Vali, G.: Interpretation of freezing nucleation experiments: singular and stochastic; sites and
1814 surfaces, Atmos. Chem. Phys., 14, 5271–5294, doi:10.5194/acp-14-5271-2014, 2014.

1815 Vali, G., Christensen, M., Fresh, R. W., Galyan, E. L., Maki, L. R. and Schnell, R. C.: Biogenic
1816 Ice Nuclei. Part II: Bacterial Sources, J. Atmos. Sci., 33, 1565–1570, 1976.

1817 Vali, G., DeMott, P. J., Möhler, O. and Whale, T. F.: Technical Note: A proposal for ice
1818 nucleation terminology, Atmos. Chem. Phys., 15, 10263–10270, doi:10.5194/acp-15-10263-
1819 2015, 2015.

1820 Vasiliou, J. G., Sorensen, D. and McMurry, P. H.: Sampling at controlled relative humidity with
1821 a cascade impactor, Atmos. Environ., 33, 1049–1056, doi:10.1016/S1352-2310(98)00323-9,
1822 1999.

1823 Wang, B., Laskin, A., Roedel, T., Gilles, M. K., Moffet, R. C., Tivanski, A. V and Knopf, D. A.:
1824 Heterogeneous ice nucleation and water uptake by field-collected atmospheric particles below
1825 273 K, J. Geophys. Res., 117, D00V19, doi:10.1029/2012JD017446, 2012.

1826 Welti, A., Lüönd, F., Kanji, Z. A., Stetzer, O. and Lohmann, U.: Time dependence of immersion
1827 freezing: an experimental study on size selected kaolinite particles, Atmos. Chem. Phys., 12,
1828 9893–9907, doi:10.5194/acp-12-9893-2012, 2012.

1829 Welti, A., Lüönd, F., Stetzer, O. and Lohmann, U.: Influence of particle size on the ice
1830 nucleating ability of mineral dusts, Atmos. Chem. Phys., 9, 6705–6715, doi:10.5194/acp-9-6705-
1831 2009, 2009.

1832 Wheeler, M. J., Mason, R. H., Steunenberg, K., Wagstaff, M., Chou, C. and Bertram, A. K.:
1833 Immersion Freezing of Supermicron Mineral Dust Particles: Freezing Results, Testing Different
1834 Schemes for Describing Ice Nucleation, and Ice Nucleation Active Site Densities, J. Phys. Chem.
1835 A, 119, 4358–4372, doi:10.1021/jp507875q, 2015.

1836 Wilkinson, D. M., Koumoutsaris, S., Mitchell, E. A. D. and Bey, I.: Modelling the effect of size
1837 on the aerial dispersal of microorganisms, J. Biogeogr., 39, 89–97, doi:10.1111/j.1365-
1838 2699.2011.02569.x, 2011.

1839 Wilson, T. W., Ladino, L. A., Alpert, P. A., Breckels, M. N., Brooks, I. M., Browse, J., Burrows,
1840 S. M., Carslaw, K. S., Huffman, J. A., Judd, C., Kilthau, W. P., Mason, R. H., McFiggans, G.,
1841 Miller, L. A., Najera, J., Polishchuk, E., Rae, S., Schiller, C. L., Si, M., Vergara Temprado, J.,
1842 Whale, T. F., Wong, J. P. S., Wurl, O., Yakobi-Hancock, J. D., Abbatt, J. P. D., Aller, J. Y.,
1843 Bertram, A. K., Knopf, D. A. and Murray, B. J.: A marine biogenic source of atmospheric ice
1844 nucleating particles, submitted, 2015.

1845 Winkler, P.: Relative humidity and the adhesion of atmospheric particles to the plates of
1846 impactors, J. Aerosol Sci., 5, 235–240, doi:10.1016/0021-8502(74)90058-5, 1974.

1847 Wright, T. P., Hader, J. D., McMeeking, G. R. and Petters, M. D.: High Relative Humidity as a
1848 Trigger for Widespread Release of Ice Nuclei, Aerosol Sci. Technol., 48, i–v,
1849 doi:10.1080/02786826.2014.968244, 2014.

1850 Wright, T. P. and Petters, M. D.: The role of time in heterogeneous freezing nucleation, J.
1851 Geophys. Res.-Atmos., 118, 3731–3743, doi:10.1002/jgrd.50365, 2013.

Wright, T. P., Petters, M. D., Hader, J. D., Morton, T. and Holder, A. L.: Minimal cooling rate dependence of ice nuclei activity in the immersion mode, J. Geophys. Res.-Atmos., 118, 10535–10543, doi:10.1002/jgrd.50810, 2013.

Yakobi-Hancock, J. D., Ladino, L. A. and Abbattt, J. P. D.: Review of recent developments and shortcomings in the characterization of potential atmospheric ice nuclei: focus on the tropics, Rev. Ciencias, 17, 15–34, 2013.

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, 12307–12317, doi:10.5194/acp-14-12307-2014, 2014.

Table 1. The seven locations used in this study and conditions during sampling.

Location	Environment	Coordinates	Elevation (m)	Sampling period	Number of Samples	Average sampling time (h)	Average temperature (°C)	Average relative humidity (%)	Particle size range (μm)[a]
Alert, NU, Canada	Arctic	82.45° N 62.51° W	12 agl 200 asl	Mar. 29 - Jul. 23, 2014	9	17.6	-17.4	77	0.10–10
Whistler Mountain, BC, Canada	Alpine	50.06° N 122.96° W	2 agl 2182 asl	Mar. 30 - Apr. 23, 2014	4	6.7	0.8	83	0.18–10
Amphitrite Point, BC, Canada	Coastal	48.92° N 125.54° W	5.5 agl 25 asl	Aug. 6–27, 2013	34	7.8	13.8	97	0.18–10
The Labrador Sea, Canada	Marine	54.50° N 55.37° W	2 agl 15 asl	Jul. 11, 2014	1	6.2	10.9	75	0.10–10
CEA, Saclay, France	Suburban	48.70° N 2.14° E	10 agl 168 asl	Jul. 15 - Aug. 4, 2014	15	7.2	20.6	69	0.10–10
UBC campus, BC, Canada	Suburban	49.26° N 123.25° W	24 agl 120 asl	May 12–16, 2014	4	6.3	15.2	70	0.10–10
Colby, KS, USA	Agricultural	39.39° N 101.06° W and 39.39° N 101.08° W	2 agl 968 asl	Oct. 14–15, 2014	3	4.5	17.0	48	0.10–10

[a]Aerodynamic diameter based on the 50 % cutoff of the MOUDI (Marple et al., 1991).

Ryan Mason 11/3/2015 8:04 PM
Deleted: RH

Table 2. Previous size-resolved INP measurements.

Study	Location	Geographical Description	Altitude	Particle sizes investigated (µm)	Mode of ice nucleation	Ice nucleation temperature (°C)[a]	Result
Rucklidge (1965)	West Plains, Missouri	Forest and pasture	4 m agl	TSP	Condensation and/or deposition	-12 to -25	≤ 14 % of INPs > 1 µm
Vali (1966)	Alberta, Canada	Western Canada	Hail melt water	TSP[b]	Immersion	-12.8	16 % of INPs > 1.2 µm[c]
Rosinski et al. (1986)	Central and western South Pacific Ocean	Marine	Near sea level	0.5 to > 8	Immersion	-10.8	100 % of INPs > 1 µm
				< 0.5 to > 8	Condensation	-5 to -6	1 % of INPs > 1 µm
Rosinski et al. (1988)	Gulf of Mexico	Marine	Near sea level	0.1 to > 4.5	Condensation	-15 to -16	45 % of INPs > 1 µm
Berezinski et al. (1988)	European territory of the former Soviet Union	Eastern Europe	100–500 m agl	0.1 to > 100	Condensation	-15 to -20	37 % of INPs > 1 µm
Mertes et al. (2007)	Jungfraujoch, Switzerland	Alpine	3580 m asl	0.02 to 5	Unknown (ice residual)	-17.4	< 1 % of ice residuals > 1 µm[c]
Santachiara et al. (2010)	S. Pietro Capofiume, Italy	Rural Italy	3 m agl	TSP	Condensation	-17 to -19	47 and 30 % of INPs > 1 and 2.5 µm, respectively
Huffman et al. (2013)	Manitou Experimental Forest, CO, USA	Forest during/after rainfall	4 m agl	0.32 to >18	Immersion and deposition	-15 to -20	89 % of INPs > 1 µm
		Forest during dry periods	4 m agl	0.32 to >18	Immersion and deposition	-15 to -20	46 % of INPs > 1 µm

[a]We used data at the temperatures of this study (-15, -20, and -25 °C) when available, otherwise the next closest temperature was used.
[b]TSP = total suspended particulate.
[c]Particle size reported for Vali (1966) is based on filter pore size, and particle size reported in Mertes et al. (2007) is based on electrical mobility and optical measurements. In all other studies, particle size is given as the aerodynamic diameter.

Figure 1. Sampling locations used in this study: (1) Alert, Nunavut, Canada; (2) Whistler Mountain, British Columbia, Canada; (3) Amphitrite Point, British Columbia, Canada; (4) the Labrador Sea, Canada; (5) CEA in Saclay, France; (6) the University of British Columbia campus, British Columbia, Canada; and (7) Colby, Kansas, USA. Site coordinates are given in Table 1 with details in Sect. 2.1. The image was modified from Bing Maps, 2014 (http://bing.com/maps).

Ryan Mason 10/11/2015 4:03 PM
Deleted: the Labrador Sea, Canada;

Ryan Mason 10/11/2015 4:02 PM
Deleted: Whistler Mountain, British Columbia, Canada;

Ryan Mason 10/11/2015 4:04 PM
Deleted: the University of British Columbia campus, British Columbia, Canada;

Ryan Mason 10/11/2015 4:02 PM
Deleted: Amphitrite Point, British Columbia, Canada

Ryan Mason 10/11/2015 4:04 PM
Deleted: Colby, Kansas, USA; and

Ryan Mason 10/11/2015 4:03 PM
Deleted: Saclay, France

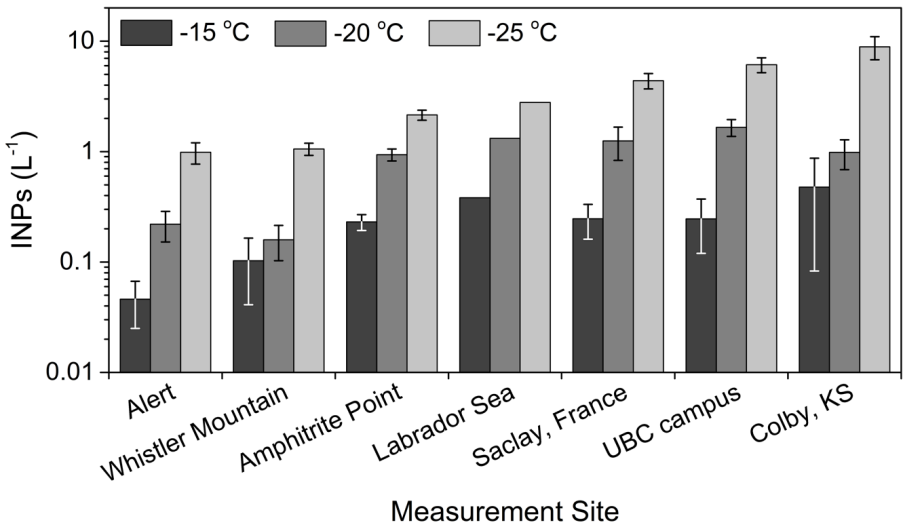

Figure 2. Mean INP number concentrations at droplet freezing temperatures of -15 °C (dark grey), -20 °C (intermediate grey), and -25 °C (light grey). Uncertainty is given as the standard error of the mean, assuming a normal distribution. As only one sample was available from the Labrador Sea, no uncertainty is reported.

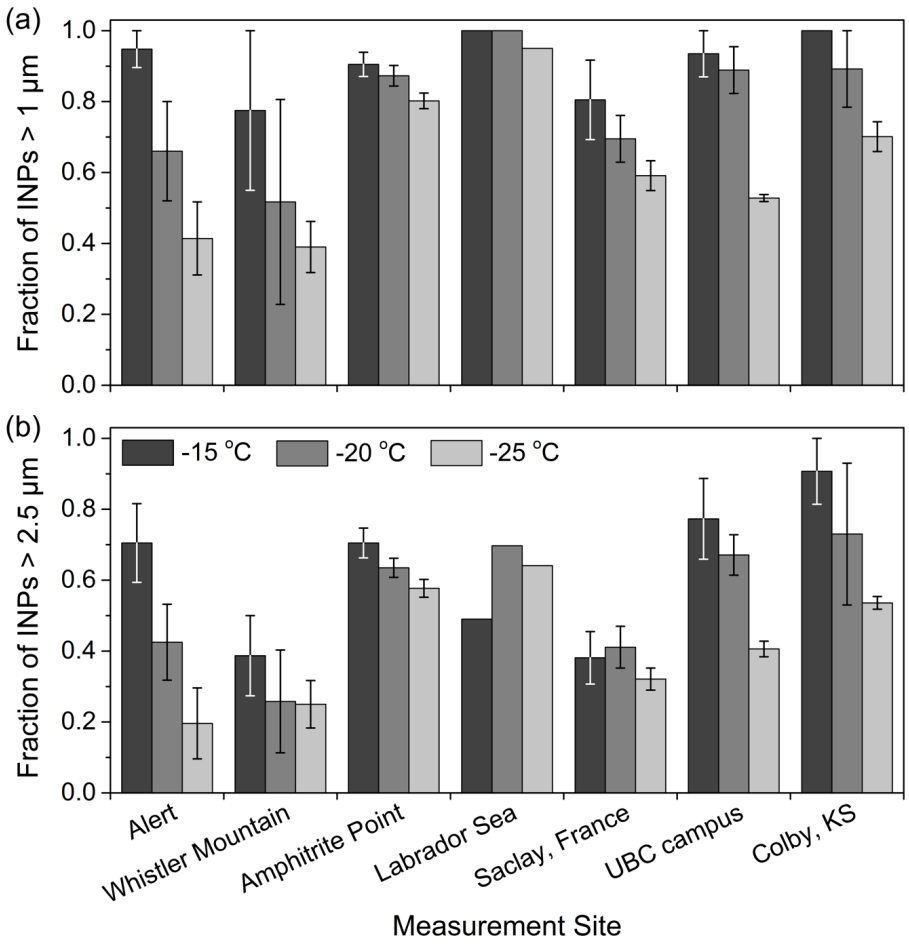

Figure 3. The mean fraction of INPs larger than (a) 1 µm and (b) 2.5 µm. Uncertainty is the standard error of the mean, assuming a normal distribution. Shading in the histogram corresponds to INP activation temperature: -15 °C is dark grey, -20 °C is an intermediate grey, and -25 °C is light grey. As 2.5 µm does not align with the size cut of a MOUDI stage, the fraction of INPs larger than 2.5 µm was found by assuming that number concentration of INPs 1.8–3.2 µm in size was uniformly distributed over that size range. As only one sample was available from the Labrador Sea, no uncertainty is reported.

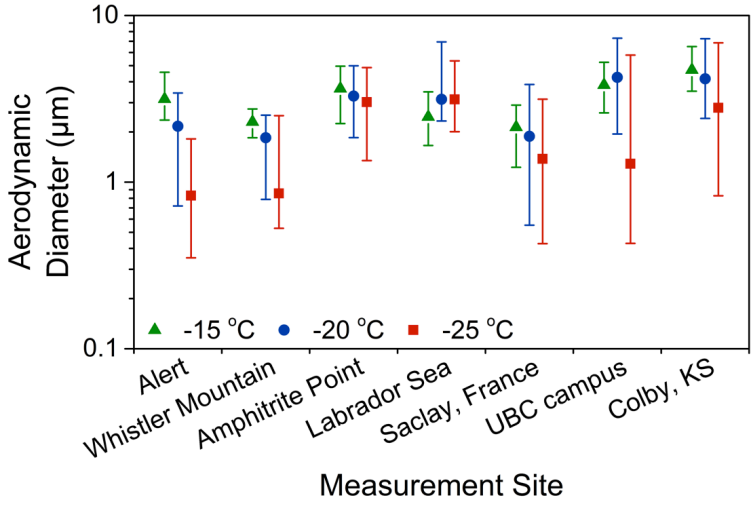

Figure 4. The median size of INPs at ice-activation temperatures of -15 °C (green), -20 °C (blue), and -25 °C (red) when averaged over all analyzed samples. Upper and lower uncertainties are the 75th and 25th percentiles, respectively.

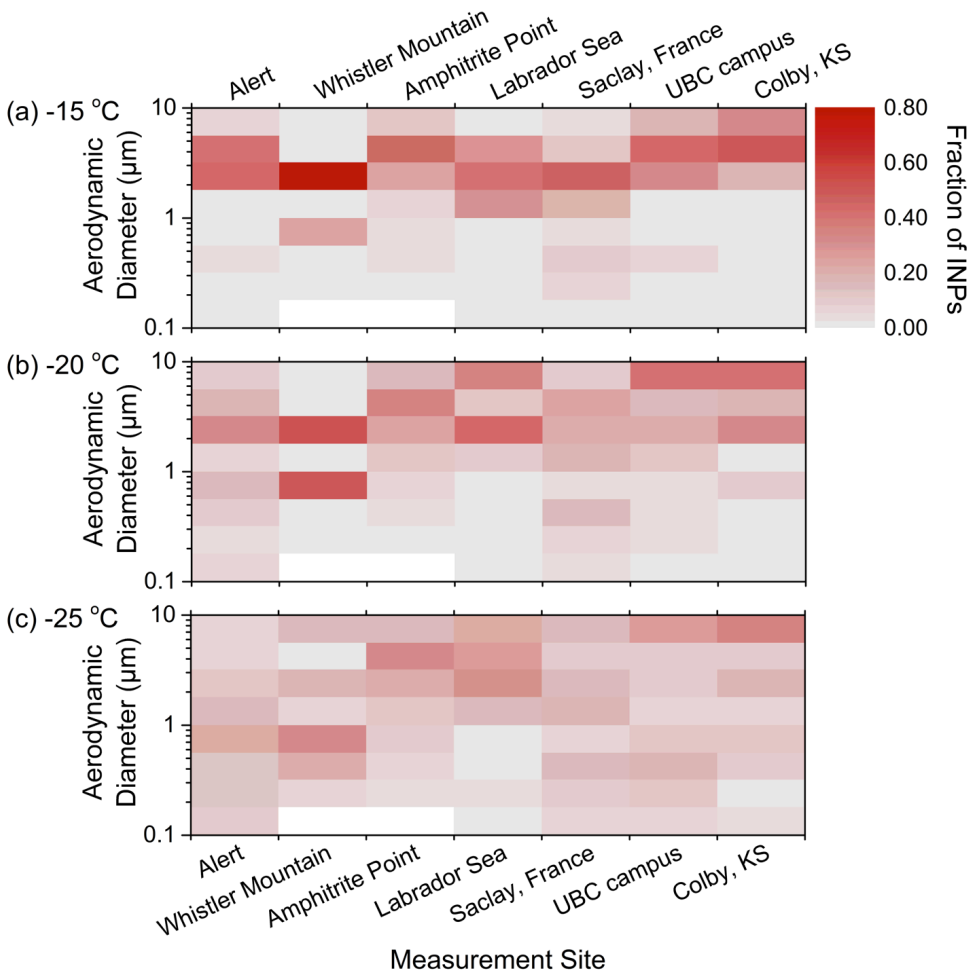

Figure 5. Fractional INP concentrations as a function of aerosol particle size, location, and activation temperature: (a) -15 °C; (b) -20 °C; and (c) -25 °C. The color bar indicates the fraction of INPs measured in each particle size bin. Aerosol particle sizes correspond to the 50 % cutoff aerodynamic diameters of the MOUDI stages (Marple et al., 1991). Missing sizes for the Whistler Mountain and Amphitrite Point sites are uncolored.