Anonymous Referee #3
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[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 will be 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 will add the following to the revised manuscript:

"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 droplets combine as they grow to a final size of 97 ± 42 µm (mean diameter and 1 standard deviation). On average, more than 99 % of particles become incorporated into the droplets as this occurs."

[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 will be 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) will be given 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 will be 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 will be 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 will remove 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 will be made in the revised manuscript.