Anonymous Referee #1
Received and published: 25 June 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.

Mason et al. present results on ice nucleating particles (INPs) from a coastal site in western Canada during the summer. The INP concentrations strongly correlated with fluorescent terrestrial bioparticles at high temperatures, while particles that were likely mineral dust nucleated ice at lower temperatures. However, predicted INP concentrations using different empirical parameterizations did not corroborate the observations, demonstrating the need for improved modeling of INPs. The paper is overall well written and the methods and interpretation of the results are clear. There are a few needed improvements described below, however, once these are addressed, this paper is suitable for publication in ACP.

We thank the referee for his/her helpful comments!

General remarks:
[1] The abstract could be strengthened by adding a sentence of two of broader implications at the end. What do these results signify and how do they advance our understanding of INPs? Perhaps here, and in general throughout the manuscript, one large motivation for work such as this is that the parameterizations did not corroborate the observations, demonstrating the need for more observations to improve simulated INP concentrations and their subsequent climatic impacts.

[A1] Thank you for the suggestion. The following sentence will be added to the end of the abstract:

"This finding illustrates that additional measurements are needed to improve parameterizations of INPs and their subsequent climatic impacts. "

[2] The introduction would benefit from more background, such as on primary bioparticles versus marine bioparticles. What are some of the sources of these types? What types of bioparticles are marine? Also, the authors conclude that dust was likely observed at the lower temperatures, so some background on mineral and soil dust as IN is warranted. It would be helpful to also include previously documented temperature ranges in which each of the different types of INPs nucleate ice at (use references such as Murray et al. (2012), Conen et al. (2011), DeMott et al. (2003, 2009, 2013), O'Sullivan et al. (2014), etc.).

[A2] In the revised manuscript, we will rewrite the introduction with the referee's comments in mind.

[3] The dates of the sample collection should be provided first thing in the methods. Otherwise, there is only one figure that includes an Aug time period but the exact dates and year should be provided.

> ***[A3]*** The dates of sample collection will be added to the Methods section in the revised manuscript.

[4] In the methods, the DFT measurements were conducted at, "-10 C per minute to -40 C." However, many of the results are presented in -5 C steps. Why are measurements not presented as -10, -20, -30, -40 C? Perhaps the measurements started at -15 C, but this should be explicitly stated. Were measurements acquired at -10 C? That would be an interesting comparison since the focus is on biological particles and these can nucleate ice up to -2 C.

> ***[A4]*** Data was not reported at temperatures above -15 ºC since very few freezing events occurred at these warm temperatures (only 1.3 % of all droplets froze above -15 ºC). Data was not reported below -30 ºC since in some experiments all droplets froze at these temperatures, which prohibits the calculation of INP number concentrations by Eq. (1). To address the referee's comment, the following sentence will be added to Sect. 2.2.
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> "Here we report INP data between -15 and -30 °C as few (1.3 %) of droplets froze at temperatures > -15 C, and at temperatures < -30 C, in some experiments all droplets froze, which prohibited the calculation of INP number concentrations by Eq. (1)."

[5] Can the authors comment on the possible contribution from soil dust? Wouldn't this fluoresce as well with WIBS (as in Gabey, A.M., Stanley, W. R., Gallagher, M. W., Kaye, P.H.: The fluorescence properties of aerosol larger than 0.8 um in urban and tropical rainforest locations, Atmos. Chem. Phys., 11, 5491-5504, doi:10.5194/acp-11-5491-2011, 2011.)?

> ***[A5]*** To address the referee's comment, line 26, page 16282 will be revised to the following:
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> "While some non-biological species such as soot, mineral and soil dusts, polycyclic aromatic hydrocarbons, secondary organic aerosols, and humic-like substances can produce a fluorescent signal (Bones et al., 2010; Gabey et al., 2011; Lee et al., 2013; Pan et al., 1999; Pöhlker et al., 2012; Sivaprakasam et al., 2004), the number of fluorescent particles is generally considered to be a lower limit to the number of primary biological particles (Huffman et al., 2010, 2012; Pöhlker et al., 2012)."

[6] Considering the particle sizes observed and shown in Fig 6. I find it odd that these large sizes are more abundant in number than smaller particles (i.e., 0.5 to 1 um). Wouldn't the authors expect to observe smaller bioparticles, such as bacteria? Perhaps this is due to the transmission efficiency of the WIBS, which could be discussed since this is a relatively new technique.

[A6] To address the referee's comment, at the end of Sect. 2.3 we will add a discussion on the size dependent detection efficiency of the WIBS.

[7] The method for using correlation of wind speed at the site and INPs emitted from the ocean surface may not be the most direct, since the wind speed may be different over the water versus land surface. Have the authors considered estimating the wind speed from the HYSPLIT trajectories? That may lead to a better estimate of wind speeds over the ocean along the transport paths, since most of the trajectories remained fairly low in the marine boundary layer.

[A7] To address the referee's comment, we have determining the average wind speed during each MOUDI sampling period using data collected from a height of 5 m asl by a moored buoy located approximately 35 km WSW of our sampling site (station 46206: http://ndbc.noaa.gov/station_page.php?station=46206). Little difference was found between the two wind speed measurements. Correlations using the buoy data will be added to the revised manuscript.

[8] There should be more broad discussion on the parameterizations in section 3.7. The fact that the parameterizations did not fit the observational data demonstrate the need to improve these parameterizations by conducting more observations in different locations, times of year, and land cover regimes (i.e., arid, vegetation, near BC sources such as fires, etc.).

[A8] Thank you for the suggestion. To address the referee's comment we will add the following text to the end of Sect. 3.7:

"Figure 8 suggests that additional measurements of INPs in other environments, times of year, and altitudes are needed to further test and improve current parameterizations of INPs. The results presented in Fig. 8 also indicate that the application of INP parameterizations to locations dissimilar to that of the original study used to generate the parameterizations should be done with care."

Specific comments:
[9] Page 16275, line 17: Clarify that these are chemical tracers, and if space permits, provide the tracers (i.e., MSA and Na).

[A9] This revision will be made in the final document.

[10] Page 16279, line 4: Briefly define Cfb.

[A10] For clarity, this sentence will be modified to the following in the revised manuscript:

"This region has a temperate maritime climate, characterized by warm summers, mild winters, and relatively high levels of cloud cover and precipitation. According

to the Köppen-Geiger classification scheme (Kottek et al., 2006) the climate type is Cfb which denotes a mild mid-latitude and moist climate (C) with no dry season (f), and a moderate summer where the average hottest-month temperature is < 22 ºC and at least four months have an average temperature > 10 ºC (b)."

[11] Page 16280, line 4: Change "measured" to "collected".

[A11] To address the referee's comment "measured" will be changed to "determined". We feel the use of "determined" is more appropriate than "collected" since the MOUDI-DFT includes sample collection and freezing measurements.

[12] Page 16280, line 18: Spell out DFT on first occurrence.

[A12] This correction will be made.

[13] Page 16288, line 23: Instrument and sampling details for CO, NOx, and SO2 should be briefly provided in the methods.

[A13] In the revised manuscript, this information will be added to the main document.

[14] Section 3.4: Was there any correlation of INPs with wind direction?

[A14] No correlations were found between INPs and local wind direction (R ranged from -0.19 to -0.32). This information will be added to Sect. 3.4 to address the referee's comment.

[15] Section 3.6: In regards to the possible free tropospheric transport of dust, the authors could examine 10-day air mass back trajectories for this particular time period to evaluate the potential sources of the aerosol. For instance, if the trajectories all pass over one of the major arid regions in Asia or Africa, this would support their assumption that mineral dust contributed to the INP concentrations at -30 C.

[A15] In the revised manuscript, ten-day back trajectories will be added to the Supplement. None of the trajectories pass over major arid regions in Asia or Africa; however, this does not rule out mineral dust or soils as a source of INPs in our measurements.

[16] Page 16292, line 15: What are some of the potential sources of INP along the coastal NW that would be larger than 1 um? The vegetation coverage is discussed for the entire region in the first section of the methods, but it could be specified here what is NW of the site.

[A16] To address the referee's comment, the following will be added at the end of section 3.6:

"Vegetation NW of the sampling site closely follows that of the region, and potential sources of supermicron INPs from coastal NW include forests of coastal western hemlock."

[17] Page 16293, line 3: Up until this point, the maximum size for the WIBS used is 10 um, why the change here?

[A17] In Section 3.7 we used data from the WIBS-4A over its full size range (0.5–23.7 μm) to better match the sampling conditions used in D10 and T13, where the parameterizations were developed using total particle and fluorescent bioparticle concentrations over the full size range of the UV-APS (approximately 0.5–20 μm). This information will be added to Page 16293, line 3 for clarity.

[18] Page 16294, line 1: But in the introduction on page 16278, lines 2-3, sea salt is presented as having the ability to serve as INP. Perhaps the authors should clarify that these referenced studies investigated NaCl or sea spray to form ice at very low temperatures (roughly -35 to -58 C), thus sea salt has the potential to form ice, yet is inefficient at temperatures relevant to heterogeneous ice nucleation.

[A18] In the revised manuscript the introduction will be modified to avoid the impression that NaCl can form ice at the temperatures we studied.

[19] Fig 2: It would be useful to, in some way, also show the trajectories colored by source group (similar colors as in Fig 3). Perhaps an additional panel with the same trajectories colored by group would suffice?

[A19] In the revised manuscript we will add an additional figure to the Supplement that will show the trajectories in Fig. 2 colored by source group.

[20] Fig 5: In the manuscript, the authors state that correlations which are insignificant ($p > 0.05$) are not discussed, yet they are shown here and are actually discussed in the manuscript. Perhaps this statement should be removed or revised if the authors choose to keep these data.

[A20] To address the referee's comment the statement "Only correlations with statistical significance (P value < 0.05) are discussed" will be changed to "In the discussion, correlations with statistical significance (P value < 0.05) are emphasized".