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 will do the following:

a) Report the average freezing temperatures and standard deviation for the blanks as well as the warmest observed freezing temperatures of the blanks.
b) Discuss 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) Report 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 will be discussed earlier and in more detail. Specially, we will do the following:

The last sentence of the abstract will be 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 will be deleted and the first part will be 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 will be 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 will be 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 will 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 will add 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 will be 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 will be revised to include 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, a comparison to the Mertes et al. (2007) and Targino et al. (2006) studies on ice crystal residuals size distributions will be added. 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 will also be added to the manuscript to address the referee's comment regarding the work of Welti et al. (2009):

"Laboratory experiments investigating the ice nucleation efficiency of particles as a function of size have also been conducted (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."

[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 will be 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 will be noted in the figure captions.

Technical remarks:

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

[A16] Correction will be made.

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

[A17] Correction will be 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" will be deleted to avoid confusion.

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

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

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

[A20] Correction will be made.

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

[A21] Correction will be 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 will be changed to match that of Figs. 2–5.