

1 Anonymous Referee #2
2 Received and published: 1 September 2015

3
4 *[A0]* For clarity and visual distinction, the referee comments or questions are listed
5 here in black and are preceded by bracketed, italicized numbers (e.g. *[1]*). Authors'
6 responses are offset in blue below each referee statement with matching numbers
7 (e.g. *[A1]*). Page and line numbers refer to online ACPD version.

8
9 The paper by Mason et al. presents size-resolved impactor measurements of sub- and
10 super-micron particles collected at seven locations in Canada, the U.S., and France. The
11 samples were analyzed determining the particles' immersion-mode freezing properties,
12 that is, ice nucleating particle (INP) number concentrations as a function of size and
13 temperature.

14
15 The main conclusion from the study is that a large fraction of the ice active particles is >
16 1 μm in diameter. This is particularly important to know for the interpretation of INP
17 concentrations determined with other established on-line measuring instruments, such as
18 the continuous-flow diffusion chambers, which typically miss the super-micron particles
19 in their analysis due to the specific inlet system.

20 The paper is very well structured, describes the applied methods and discusses the results
21 very nicely. Therefore I can fully recommend the paper for final publication in ACP. I
22 have only very little suggestions for improvement and a few minor questions all listed
23 chronologically in the following:

24
25 *We thank the referee for his/her helpful comments!*

26
27 *[1]* P. 20523, L.7: Here it would be helpful to add one short sentence on the applied
28 measurement principle, the MOUDI-DFT.

29
30 *[A1]* This sentence will be revised to the following:

31
32 "In this study we report immersion-mode INP number concentrations as a function of
33 size at six ground sites in North America and one in Europe using the micro-orifice
34 uniform deposit impactor-droplet freezing technique (MOUDI-DFT), which
35 combines particle size-segregation by inertial impaction and a microscope-based
36 immersion freezing apparatus."

37
38 *[2]* P. 20525, L. 24-29: I would delete this paragraph at this place because it tells already
39 main results, which not necessarily are part of the introduction section.

40
41 *[A2]* The paragraph will be revised to the following to remove the discussion of
42 results from the introduction section:

43
44 "Previous field studies of INPs as a function of size have been carried out using
45 ambient aerosol samples (e.g. Rosinski et al., 1986; Santachiara et al., 2010), ice-
46 crystal residuals (e.g. Vali, 1966; Mertes et al., 2007), and laboratory experiments

47 (e.g. Welti et al., 2009; O’Sullivan et al., 2015). These and additional studies are
48 further discussed in Sect. 3.2. In the current study, we add to the existing body of
49 size-resolved INP measurements by reporting ground-level INP size distributions
50 from six locations in North America and one in Europe, covering a diverse set of
51 environments and investigating immersion freezing at -15, -20, and -25 °C.”
52

53 **[3]** P. 20530: The aerosol particle number size concentration usually varies significantly
54 over the size range of 0.1 to 10 µm. Consequently, I guess the surface coverage must be
55 very different for the individual impactor stages, i.e., small number of particles on upper
56 stages and large number on lower stages. How does that affect the droplet freezing
57 experiments? I could imagine that it is difficult to analyze samples with too high particle
58 load because the growing droplets may run into each other very easily. On the other hand,
59 if there are only few particles on the surface the result might not be statistically
60 significant. How did you handle different surface coverages?
61

62 **[A3]** The surface coverage can affect the freezing temperatures in two ways:
63 a) If the concentrations are too low, the freezing temperatures will overlap with the
64 “blank” freezing experiments. This was not an issue in the current experiments.
65 To clarify this point, the “blank” freezing experiments will be added to the
66 revised manuscript.
67 b) If the surface coverage is too high and there is a significant concentration of INPs
68 on the cover slip, all the droplets will freeze at warm temperatures. We control
69 the latter by controlling the sampling time.
70

71 **[4]** P. 20531 and 20532: I wonder if rounding INP concentrations to two significant digits
72 should be enough, e.g., 3.8 L-1 instead of 3.77 L-1, since I believe your measurements
73 are not more precise than that. Also standard deviations together with the mean values
74 would be interesting to know.
75

76 **[A4]** The reported number of significant figures will be revised as suggested.
77

78 **[5]** P. 20533, L. 25-27: How realistic is the assumption? Did the number size distributions
79 (if available) also show uniformly distributed aerosol particles over this size range?
80

81 **[A5]** Measurements of the total particle size distribution were not available at all
82 locations to fully address the referee’s question. Furthermore, the use of such
83 distributions in determining the fraction of stage 4 (1.8–3.2 µm) that belongs in the
84 coarse would be contingent on the assumption that the INP size distribution follows
85 the total particle size distribution, which may not be correct in all locations. To
86 address the referee’s comment we will add the following text at this location of the
87 revised manuscript:
88

89 “Measurements of the total particle size distribution were not available at all
90 locations to test this assumption. Furthermore, it is not known if the INP size
91 distribution follows the total particle size distribution *a priori*.”
92

93 **[6]** Fig. 3: Why did you not show any error bars for the Labrador Sea results?

94

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