

1 Anonymous Referee #2  
2 Received and published: 3 April 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 AMTD version.  
8

9 The manuscript reports on investigations of the non - uniformity of size - dependent  
10 aerosol deposit in a rotating MOUDI. Non – uniformity was studied at three different  
11 optical resolutions of a microscope. In the second part, the paper discusses ice nucleation  
12 experiments done on selected areas of some of the MOUDI stages using a droplet  
13 freezing technique (DFT). It introduces a method to correct ice nucleating particle  
14 concentrations for the aerosol non - uniformity found in the first part. The MOUDI –  
15 DFT results are compared to measurements done simultaneously with a continuous flow  
16 diffusion chamber. The comparison shows reasonable agreement of the two different  
17 techniques at the chosen conditions. The manuscript presents a detailed investigation and  
18 an improvement of the recently introduced MOUDI-DFT technique for measuring ice  
19 nucleating particle concentrations as a function of size. It is well written, the study is put  
20 into context and relevant earlier work is referred to. The manuscript is of interest to the  
21 readers of Atmospheric Measurement Techniques. I recommend it for publication after  
22 the few following comments have been addressed.  
23

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

26 *[1]* General remarks: Similar to the 1st referee's comment, it is not clear to me from the  
27 manuscript if Fig. 4-6 are averages over all analyzed glass cover slips and if so, what the  
28 variation between the slips is.  
29

30 *[A1]* Data presented in Figs. 4-6 are indeed averages over all analyzed hydrophobic  
31 glass cover slips with the error bars representing the 95 % confidence interval, which  
32 is related to the sample-to-sample variation. In the revised manuscript, instead of the  
33 95% confidence intervals we report the standard deviation, which illustrates the  
34 sample-to-sample variability. In addition, the text has been modified to clarify that  
35 we are reporting the average from the different cover slips and the standard  
36 deviation. Specifically, the sentences of 2235/10-16 have been revised to the  
37 following:  
38

39 "The normalized particle concentration, which is the quotient of the particle  
40 concentration of a given step divided by the maximum particle concentration, was  
41 calculated as a function of distance from the center of the MOUDI aerosol deposit  
42 for each hydrophobic glass cover slip at spatial resolutions of 1 and 0.25 mm. Visual  
43 inspection of aerosol deposits showed that there was spatial variability of the particle  
44 concentrations at a spatial resolution as low as 0.10 mm for MOUDI stages 6-8, so  
45 these stages were also analyzed at this spatial resolution. A total of three

46 hydrophobic glass cover slips were analyzed for stages 2 and 8 and four hydrophobic  
47 glass cover slips for stages 3-7.”

48  
49 The statement at the beginning of Sect. 3.1 (2238/3-6) has also been revised for  
50 improved clarity:

51  
52 “Shown in Figs. 4, 5, and 6 are the normalized concentrations of aerosol particles as  
53 a function of distance from the center of the MOUDI aerosol deposit at spatial  
54 resolutions of 1, 0.25, and 0.10 mm, respectively, when averaged over all analyzed  
55 samples. The uncertainty in Figs. 4-6 is the standard deviation of these samples.”  
56

57 **[2]** In addition, have you looked into the effect on the normalized particle concentration  
58 of the 0.5 mm uncertainty in centering the microscope viewing area and the hydrophobic  
59 glass cover slip to the deposit center? Your results show that there are non-uniformities at  
60 the 0.1mm scale, hence they might as well exist in the y-direction (perpendicular to the  
61 line through the center).

62  
63 **[A2]** Here we think the referee is pointing out that we have only taken into account a  
64 0.5 mm uncertainty in the x-direction, but there also may be an effect in the y-  
65 direction. In response, in the regions where we are analyzing the DFT, we are far  
66 enough from the center of the aerosol deposit that an uncertainty of 0.5 mm in the y-  
67 direction does not significantly influence the measured particle concentrations in the  
68 x-direction or the derived correction factors. The 0.5 mm uncertainty in the y-  
69 direction only changes the correction factors by approximately 1 % at most. This is  
70 because particles are concentrated in rings, and the radii of the rings are relatively  
71 large where we are analyzing the DFT (radius of the rings varies from 2.4 to 10.1  
72 mm).  
73

74 **[3]** The comparison to the CFDC measurements are a good first step to validate the  
75 MOUDI-DFT method. However, only two data points with some uncertainty in both  
76 instrument’s measurements is not enough to proof the accuracy of the new method. Thus,  
77 as promised by the authors, further comparison studies are necessary in the future.  
78

79 **[A3]** We agree with the referee.  
80

81 **[4]** Are the results expected to be affected by different aerosol types?  
82

83 **[A4]** At this point we do not know if the intercomparison results depend on the  
84 aerosol type. Different levels of agreement have been observed in past  
85 intercomparison studies depending on the aerosol type studied. For example, more  
86 disagreement between instrumentation was found when investigating the immersion  
87 freezing properties of illite NX (Hiranuma et al., 2015) than when using Snomax  
88 (Wex et al., 2015). Hence additional intercomparison studies are needed with  
89 different aerosol types. To address the referee’s comments this discussion has been  
90 added to the revised manuscript.  
91

92 **[5]** Specific Remarks Page 2227, Line 7: Please clarify at this point already if size is  
93 (aerodynamic, geometric, mobility) diameter or radius?

94  
95 **[A5]** This line has been revised with additional details:

96  
97 “For example, instruments based on the continuous flow diffusion chamber (CFDC)  
98 design of Rogers et al. (2001) limit the size of particles analyzed to those with an  
99 aerodynamic diameter  $\leq 0.75 \mu\text{m}$  in some cases (DeMott et al., 2003) and  $\leq 2.4 \mu\text{m}$   
100 in others (Garcia et al., 2012).”

101  
102 **[6]** Page 2237, Line 3: What are the standard deviations of temperature and  $SS_w$  of the  
103 CFDC during each of the two experiments?

104  
105 **[A6]** This sentence has been revised to include the requested information:

106  
107 “In sample CSU-1 the average CFDC temperature and  $SS_w$  with 1 SD uncertainty  
108 were  $-21.7 \pm 0.3 \text{ }^\circ\text{C}$  and  $5.5 \pm 0.6 \%$ , respectively, while in CSU-2 the CFDC  
109 conditions were  $-26.6 \pm 0.2 \text{ }^\circ\text{C}$  and  $5.8 \pm 0.6 \%$   $SS_w$ .”

110  
111 The uncertainties in the CFDC temperature and  $SS_w$  have also been added to Table 1.  
112

113 **[7]** Page 2237, Line 4: If it cannot be ruled out that dust was a major component of the  
114 samples, the factor 3 should be treated as an uncertainty in the CFDC data.

115  
116 **[A7]** The recent work of DeMott et al. (2015) showed that the CFDC may  
117 underestimate the INP concentration in some samples of natural mineral dust by a  
118 factor of 3, but more work is needed to determine if ambient aerosols in general are  
119 similarly underestimated. Therefore, we feel that it would be premature to apply this  
120 correction to the samples of this intercomparison for the data presented in Fig. 9.  
121 However, to address the referee’s comment the paragraph of 2237/9-17 has been  
122 revised to the following:

123  
124 “DeMott et al. (2015) found that CFDC measurements of natural mineral dust where  
125 particles were exposed to an  $SS_w$  of approximately 5 %, as was used in this study,  
126 resulted in an under-prediction of INP concentrations by a factor of 3 when  
127 compared to the use of a higher  $SS_w$  (approximately 9 %). It was therefore suggested  
128 that a correction factor of 3 be applied to INP concentrations of mineral dust samples  
129 determined by the CFDC using an  $SS_w$  of 5 %. More work is needed to determine if  
130 INP concentrations are similarly underestimated in general ambient aerosol samples  
131 such as those of this study, but the potential impact of this factor of 3 on the  
132 intercomparison results is discussed in Sect. 3.5.”

133  
134 A new sentence has also been added to Sect. 3.5 beginning at 2243/5:

135  
136 “If we apply a correction factor of 3 to the CFDC data due to this technique  
137 underestimating the INP concentration (DeMott et al., 2015), a possibility noted in

138 Sect. 2.6 although not established for our sampling conditions, the average INP  
139 concentration found by the CFDC would be greater than that of the MOUDI-DFT by  
140 a factor of 11.5 in sample CSU-1 and 2.6 in sample CSU-2.”

141

142 **[8]** Page 2237, Line 22: Are representative particle size distribution measurements  
143 available for this location which could be used to validate or constrain this factor?

144

145 **[A8]** Total aerosol particle size distributions are available, but during measurements  
146 at both CSU and other locations (e.g. Mason et al., 2015) we found that INP and total  
147 particle size distributions do not match. As our data does not suggest that using the  
148 total particle size distribution to constrain this factor would improve its accuracy, we  
149 therefore prefer to keep the current method.

150

151 **[9]** Page 2241, Line 24: Does  $f_{nu,0.25-0.10mm}$  vary with the number of sections used in  
152 the described calculations? Have you looked into that and could comment on it?

153

154 **[A9]** The non-uniformity correction factor  $f_{nu,0.25-0.10mm}$  does change with the number  
155 of sections used. If too few sections are used such that aerosol deposit non-  
156 uniformity is not sufficiently captured, the value of  $f_{nu,0.25-0.10mm}$  will be too small. We  
157 used four sections for stages 2–5 as non-uniformity in these stages was not found at  
158 spatial scales below 0.25 mm, and we used ten sections for stages 6–8 as non-  
159 uniformity in these stages was found down to spatial scales of 0.10 mm. Using more  
160 steps would not change the value of  $f_{nu,0.25-0.10mm}$ . The following sentences have been  
161 added to 2241/25 for clarity:

162

163 “The number of sections used to divide the microscope viewing area was selected for  
164 each MOUDI stage such that the section width was smaller than or equal to the  
165 spatial scale of non-uniformity. If fewer (i.e. wider) sections are used, non-  
166 uniformity is not sufficiently captured and  $f_{nu,0.25-0.10mm}$  is under-estimated. However,  
167 using more (i.e. narrower) sections does not change  $f_{nu,0.25-0.10mm}$ .”

168

169 **[10]** Technical corrections Page 2226, Line 7: change ‘can be’ to ‘is.’

170

171 **[A10]** Correction made.

172

173 **[11]** Page 2231, Line 19: Replace ‘video images (. . .) were’ by ‘video recording (. . .)  
174 was’ or ‘image recording (. . .) was’, depending on what has been done.

175

176 **[A11]** This sentence has been changed to the following:

177

178 “In the DFT, a hydrophobic glass cover slip that contained particles collected with  
179 the MOUDI was placed on the base of the flow cell, the rest of the components of the  
180 flow cell were then assembled, and a video recording of the particles was initiated  
181 (Fig. 2a).”

182

183 **[12]** Page 2232, Line 7-8: insert a minus sign before ‘1°C’.

184

185 *[A12] Correction made.*

186

187 *[13] Page 2237, Line 11: replace ‘under prediction’ by ‘underprediction.’*

188

189 *[A13] Correction made to “under-prediction.”*

190

191 *[14] Page 2238, Line 22: change ‘three SDs’ to ‘three standard deviations (SDs)’s.*

192

193 *[A14] The use of “SD” was chosen by the editor of the manuscript.*

194

195 *[15] Page 2261, Line 6: delete ‘in’ before ‘found.’*

196

197 *[A15] Correction made.*

198

199

## 200 **References:**

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223 particles using Snomax<sup>®</sup> as test substance, *Atmos. Chem. Phys.*, 15, 1463–1485,  
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