

1 Anonymous Referee #2  
2 Received and published: 31 August 2015

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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 In this manuscript, Mason and co-authors present an experimental study on the  
10 abundance, the nature and the origin of ice nucleating particles (INPs) measured at a  
11 coastal site in British Columbia. It was a pleasure to read this article for various reasons:  
12 it is clearly structured and well written, the applied methods are well described or cited,  
13 the measurements well documented, the data evaluation and interpretation is solid and  
14 appropriate, and the conclusions and atmospheric implications are clear and carefully  
15 formulated which I prefer versus over interpretation. In summary, this is an excellent  
16 manuscript with new and relevant results which in principle can be accepted for  
17 publication in ACP as is.

18  
19 We thank the referee for his/her helpful comments!

20  
21 Referee comments:

22 *[1]* I just have a few minor comments the authors may consider for the final manuscript  
23 version. My first comment refers to the mixing state of the different compounds  
24 (biological, mineral, soot, etc.) in the aerosol size distribution. This mixing state was not  
25 measured in this study, but may have an influence on some of the conclusions, like the  
26 contributions of biological particles to the INP abundance concluded from the size  
27 distributions as shown in Figure 6. What if the larger particles are just more likely to  
28 carry a fluorescent biological particle but the ice nucleation activity is related to some  
29 other particle component? The same can happen with soot or other smaller particles that  
30 have been collected by larger particles and thus still may contribute to the ice nucleation  
31 activity of the apparently larger particles. This possibility or limitation may be mentioned  
32 somewhere in the manuscript and also in the conclusion section. This comment also  
33 refers to the need of particle mixing state information in future atmospheric INP studies.  
34 Would it e.g. be possible in future studies to co-locate the INPs with fluorescent  
35 signatures (or other particle compound or property signatures) on the same substrate?

36  
37 *[A1]* To address the referee's comments, the following text will be added to the  
38 conclusions:

39  
40 "In this paper we assumed that particles were externally mixed. In future studies it  
41 would be useful to include mixing state measurements together with studies similar  
42 to those presented here to quantify the extent of external versus internal mixing. In  
43 addition, studies that identify INPs followed by chemical composition measurements  
44 of these particles by electron microscopy (e.g. Knopf et al., 2014) or fluorescence  
45 microscopy would be useful."  
46

47 [2] I agree to referee 1 that the abstract could be strengthened. I also recommend  
48 extending the conclusion section for the most important findings and atmospheric  
49 implications.

50

51 [A2] See response to Question 1 from Referee 1. In addition, the conclusion section  
52 will be extended slightly.

53

54 [3] In the abstract line 12 the correlation between INPs at -30°C and total particles larger  
55 than 0.5 µm is mentioned. I think Figure S1 shows INP at -30°C being equally well  
56 correlated with fluorescent and total particles. How can then be concluded for an extra  
57 contribution of non-biological particles to INPs? I recommend moving Figure S1 to the  
58 main manuscript.

59

60 [A3] At a droplet freezing temperature of -30 °C, fluorescent bioparticles and total  
61 particles have the same linear correlation coefficient to INPs ( $R = 0.66$ ). Here we are  
62 interested in the trends in  $R$  values from previous temperatures as given in Table 1  
63 when discussing possible changes in the composition of INPs. The decrease in  $R$   
64 from 0.83 to 0.66 between -25 and -30 °C for fluorescent bioparticles suggests that  
65 the relative contribution of fluorescent bioparticles to the overall INP population is  
66 decreasing at temperatures below -25 °C. On the other hand, an increase in  $R$  from  
67 0.49 to 0.66 between -25 and -30 °C for total particles also suggests that non-  
68 biological particles are becoming an increasingly important source of INPs at lower  
69 temperatures (given that biological particles are more poorly correlated).

70

71 We would prefer to keep Fig. S1 in the supplement since the some of the panels in  
72 Fig. S1 are the same as some of the panels in Fig. 5 in the main text. However, if the  
73 editor prefers we can move Fig. S1.

74

75 [4] The first sentence of the introduction reads as if there is no contribution of  
76 heterogeneous ice nucleation in cirrus clouds, which certainly is not the case.

77

78 [A4] The first sentence will be rewritten to address the referee's comments.

79

80 [5] When discussion the various ice nucleation modes you may also cite Vali et al.,  
81 Atmos. Chem. Phys. Discuss., 14, 22155-22162, 2014.

82

83 [A5] A reference to Vali et al. (now published in ACP) will be added to the revised  
84 manuscript.