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Comment

## ***Interactive comment on “Interpretation of freezing nucleation experiments: singular and stochastic; sites and surfaces” by G. Vali***

**Anonymous Referee #2**

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This manuscript reviews experiments on heterogeneous freezing, reviews equations and models used to describe heterogeneous freezing, and provides some clarity on these topics. Since these topics are important for describing ice nucleation in the atmosphere and since these topics are currently rather muddy in the literature, the paper is a valuable addition. I recommend publication after the author has addressed the following comments.

1. In several places the author indicates the focus of the manuscript. Abstract: “The paper focuses on three identifiably separate but interrelated issues: (i) the combination of singular and stochastic factors, (ii) the role of specific surface sites, and (iii) the modeling of heterogeneous ice nucleation.” Page 1712, line 22: “This paper focuses on laboratory experiments of heterogeneous freezing nucleation.” Page 1714,

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line 9-11: “This paper is an examination of how the singular and stochastic aspects of heterogeneous freezing nucleation are evidenced in experiments, the models that have been constructed to describe that behavior, and how the evidence leads to models that combine both aspects.” Page 1718, line 5-6: “The question posed in this paper is: to what extent conditions for valid applications of Eq. (6) have been satisfied in past experiments, and whether interpretations of observation in terms of nucleation rate are justified or not.” To me all these statements are not completely consistent and lead to confusion. Please modify for consistency.

2. Table 1. Table 1 is entitled “Summary of experiments reviewed in the text”; however there are several experiments reviewed in the text that are not included in the table. E.g. Shaw et al. (2005); Marcolli et al. (2007); Broadley et al. (2012); and Hiranuma et al. (2013). Table 1 should be re-labeled to make it clear what experimental data is included and what is not included. Also, I wondered why Shaw et al. was not included when it gave the highest epsilon value ( $> 20$ ).

3. Page 1730, line 25-26. “These two assumptions led to very similar results and reproduced the observations with about the same degree of precision.” This statement seems to contradict slightly the abstract from Marcolli et al.

4. Page 1733, line 26-27. “A test of the distinction between freezing rate and nucleation rate can be found in the influence of cooling rate.” Please elaborate on this statement, since I don’t think it will be completely obvious to most readers how the cooling rate can be used to distinguish between the freezing rate and nucleation rate.

5. Page 1736, lines 1. “Plots of data for  $R(t)$  in Fig. 2a tend to be steeper than those for  $R(T)$  in Fig. 2b.” Is it useful/meaningful to compare these two slopes when they have different units? What should the reader take away from this comparison?

6. Section 5.1. I found it hard to understand the point the author was making in this section. In the second paragraph of this section the author, I think, was trying to show that omega predicted by the Fletcher method is not consistent with experimental data.

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Is this the point the author was trying to make? I think the author's point could be made much clearer if he plotted in Figure 2b several curves predicted with the Fletcher model, anchored at different  $T_c$  values. In the third paragraph the author, I think, is trying to make the point that the temperature dependence of the frequently cited equation for  $J(T)$  cannot reproduce the omega values observed in experiments. I think this point could be made much clearer if the author plotted in Figure 2a several curves predicted with this equation and using several fixed contact angles. Regardless, I think the author should emphasis more clearly the point he is trying to make in this section.

7. Page 1742, line 1-8. I have reread this paragraph several times, and I still don't understand the point the author is trying to make. I understand that the use of equation 12 is an assumption against the existence of quasi- $\infty$  permanent sites. But I don't understand what the author means by the existence of identical sites becoming active spontaneously? Also, what does the author mean by a surface being uniform but different in some way?

8. Section 6, Conclusions. Here the author states several main conclusions. I think the link between the conclusions and the rest of the document could be made stronger by referring to the specific sections in the document where the conclusions are supported.

9. Conclusion 2. This statement seems too strong. I agree that the evidence so far suggests that the static factors dominate. However, there could still be certain types of particles where this is not the case.

10. Conclusion 5. Don't the experiments on single samples with repeated freezing cycles give nucleation rates on the "best" sites? Does the author mean that nucleation rates on all available sites are not accessible by direct measurement?

11. Conclusion 6. "...underscores the weakness of support for the use of CNT nucleation rate expression" I think this should be changed to "... underscores the weakness of support for the use of CNT nucleation rate expression with no temperature-dependent parameters and a single contact angle". Is there significant

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support/evidence against the CNT nucleation rate expression with multiple contact angles (alpha-PDF model for example)?

12. Page 1744, line 1. “Since deposition is thought to be initiated by the formation of a minute amount of liquid followed by freezing,” Please add appropriate references or remove this statement.

13. Page 1719, line 23. I don’t think “ns” was defined in the document. If I missed it, please ignore.

14. Page 1721, line 26. Superscript and subscript issues with “T<sub>c</sub>i”

15. Page 1724, line 2. What does dN/dt represent?

16. In the text and Figure 2, Hartmann et al. (2013) is labelled H13, but the same reference is labelled Ha13 in Table 1, I think.

17. Figure 2. Some of the labels in the legend of Figure 2A are not defined. E.g. SE01b oct and se01b hexa. Please define in the figure caption for clarity.

18. Table 1. The symbol N11 is used in the table but different symbols are used in the main text (N10, N11a and N11b), leading to confusion.

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 1711, 2014.

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