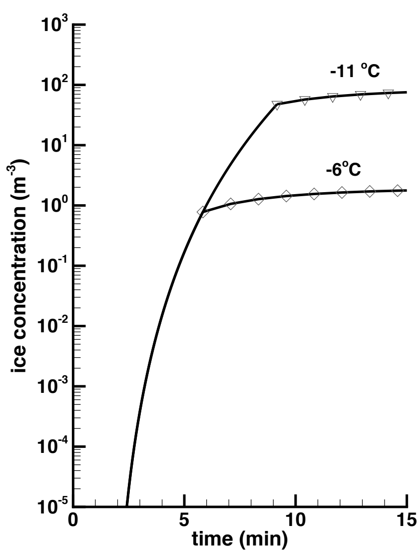
Responses to Referee #1.

Many thanks for the positive comments and suggestions. Point by point responses follow.

1. I fully agree with the author that neither the singular nor stochastic limits are justi able physical models for freezing - reality must be somewhere between these two extremes. However, I am not sure the evidence that you present fully supports the inference that temperature-dependence always dominates over time dependence. In a real cloud, this surely must depend on the magnitude of the cooling rate dT/dt and the lifetime of that cloud. If dT/dt is big and the lifetime is short (eg a cumulus cloud), then one might reasonably expect the temperature dependence to dominate. But in the opposite scenario (eg high-latitude stratus/stratocumulus clouds, or mid-latitude altocumulus clouds), where |dT/dt| is small, and the lifetime can be hours to days (eg McFarquhar et al 2011), one might expect time-dependence could manifest itself more strongly (eg Westbrook and Illingworth 2013). Herbert et al 2014 make a similar argument - the residence time of the drop is critical to the significance of time-dependent effects. I worry that points 4 and 7 in your conclusions lead the reader to the assumption that time-dependence is almost negligible in all physical situations, and I don't think that is justified based on the evidence presented. Likewise the statement on page 1738 "the cooling rate dependence and freezing after cooling stops are relatively small effects in comparison to the strong temperature dependence found for almost all types of INPs   - again this seems too strong to me, whether this is true must depend on the residence time / cooling rate.

The reviewer contrasts the cases of active convection with significant updraft velocities and cooling rates versus clouds with slow development and long lifetime. Clearly, the latter type of cloud has the potential to have more ice form via immersion freezing as time goes on even without further cooling. However, the main determinant for how much ice forms is the temperature to which that cloud has cooled. An estimate of the relative magnitudes involved is shown in the figure below, based on the Time-dependent Freezing Rate (TDFR) parcel model (Vali and Snider, 2013). Details of the model and the assumed abundance of freezing nuclei are described in the reference.



The plot shows ice development in a parcel rising with 4 m s^{-1} updraft velocity and then stopping at -6ºC in one case and at -11ºC in another. Line segments with symbols show the increase in ice concentration after the parcel ceased to rise. While far from negligible, the additional ice formation is smaller than what additional cooling produces.

While this example, and other similar calculations, confirm that temperature is the principal factor to consider, the reviewer's comments indicate that more precision was needed in stating that fact. Changes were made in the revised manuscript to correct this in the Conclusions and in the sentence on page 1738 of the earlier version.

Reference: Vali, G. and J. R. Snider, 2013: Time and temperature dependence of freezing nucleation in a cloud parcel model. Nucleation and Atmospheric Aerosols, 19th International Conference, Ed. P.J.DeMott and C.D.O'Dowd, AIP Publishing, Melville, New York, pp. 914-917

2. The relationship of the present paper with the paper by Herbert et al 2014, included as supplementary material in the discussion paper should of course be incorporated into the main manuscript. Please include a full reference to the paper - this was not included in the supplement

The supplementary material re the Herbert et al. 2014 paper was incorporated into the paper. This led to a fairly large number of additions in the text to refer to the new data on time-dependence and to discuss the implications of those results.

3. Page 1724, line 5 - Heneghen et al experiments. It may be worth clarifying that to the reader that these experiments did not seem to suffer from the same systematic variations in time to freezing as the Baldwin and Vonnegut experiments, and I recall they performed some statistical tests to demonstrate the random variation in time to freezing from run to run (which I think you mention later on).

This is correct. I did mention the statistical tests but by mistake did that when discussing the Heneghen et al. (2002) paper. The statistical tests were in fact reported in Heneghen et al. (2001). The sentence referring to these tests was moved to the correct location in the text.

4. A minor point, but for consistency can you settle on a single unit for the size of the drop being frozen. This varies through the paper from μL to cm^3 to μm diameter. It is a trivial point to rectify, but makes it easier for the reader to understand how the sample size is changing across the various experiments.

A good point. All volumes are now given in microliters.

5. Page 1527 line 25 - mention the type of IN immersed in the drops in Vali 2008

It was a suspension of a soil sample' It was used after the larger particles settled out. This description is now included in the text.

6. On page 1734, line 5 you discuss the dependence of freezing rate on the applied cooling rate, and suggest that very little variation is found as cooling rate is varied. Is this inconsistent with Figure 4 in Heneghen and Haymet (2002) who find that the time to freezing of a single sample is very strongly dependent on cooling rate?

The data shown in Table 1 of Heneghen and Haymet (2002) shows a lowering of the mid-point of the survival curve for increased values of the cooling rate and this can be seen to some extent in their Fig. 4 too. However, the lowering of freezing temperatures is attributed to thermal lag in the sensor used (their Appendix A) and the "survival curves" (fraction frozen) after correction overlap fully, as shown in their Fig. 6. The conclusion drawn in the paper is that "... we have verified explicitly, we believe for the first time, and with minimal analysis, that the "survival curve" is independent of cooling rate ..." (third paragraph of Section B of the paper). This work is not cited in my paper because of the uncertainty associated with the temperature measurement: the correction was worked out from the measured nucleation temperatures which is then the data presented as the final result.

7. Section 4.1, item 3: "narrow range" - this is a matter of opinion - to me a factor of 10 is not a narrow range! "limited" might be more accurate. Similarly item 7 "different" experimental approaches produce comparable results - this should be made more specific - the reader could interpret this as contradictory to item 6 in this list!

Thanks for identifying the need for clarification; the text was changed accordingly with the use of more definitive words.

8. At a number of points you refer to drops containing nuclei which are externally identical. Can you define this a bit more clearly, and be clearer about how easily realised this is in practice?

A footnote was added with the following text: "The phrase "externally identical" refers to a set of sample units of the same volume, drawn from the same bulk sample."

9. Conclusions, item 2: again I think this is a bit strong: Most recent publications attest to the dominance of static factors . I don't think you can make a general statement saying that static or dynamic factors are dominant - and certainly I didn't see the evidence from this clearly in the rest of the paper. Again it surely depends on whether the conditions the drop is placed in favour the dominance of one or other factor (ie cooling rate and residence time).

The conclusions were re-organized in response to this comment and those of Referee #2. The specific point here raised led to new wording which is introduced after points referring to time dependence: "The dominance of the static factors allows meaningful use of the singular model as a pragmatic tool (cf. next item) but time dependence has to be accounted for in certain experiments and in the application of nucleation models under certain conditions."

Responses to Referee #2.

Many thanks for the positive comments and for the suggestions. Some of the questions raised effectively identified weak points in the text Point by point responses follow.

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 combina- tion 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 fo- cuses on laboratory experiments of heterogeneous freezing nucleation.” Page 1714, C530
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 mod- els 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.
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The wording at several places in the text using 'this paper' were in fact intended to introduce specific analyses rather than to re-state the objectives of the entire paper. Corrections were made to make this more evident.

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).

The title of Table 1 will be modified. There are various reasons for not including in Table 1 all the experiments discussed in the text. In the case of Shaw et al. (2005) the value of ε given in the text is only a rough estimate because of the narrow temperature range of the data in the original paper. Even so, it is now included in Table 1. The Hiranuma et al. (2013) paper does not contain detailed data that would allow determination of ω. The Marcolli et al. (2007) paper presents the results as smooth histograms making it vary laborious and error prone to extract the data needed for inclusion in the figures and in Table 1. Data from Broadley et al. (2012) was included.

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.

To improve my description of the results presented by Marcolli et al (2007) the text was changed to the following: " The results did not match the predictions of a stochastic model. Instead,two variants of the singular model approach were constructed using contact angle as a proxy for effectiveness. In one version each particle was assumed to be characterized by a single value of the contact angle, assigned from a distribution of values. In the other version, particle surfaces were characterized by a distribution of sites of different effectiveness (defined by contact angle) in proportion to their surface area. These two versions of the singular model reproduced the observations with about the same degree of precision." The final sentence is based on the statement found in the last paragraph of Sect. 4 of the Marcolli et al. (2007) paper. It is also consistent with Figs. 4 and 5 of the paper.

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.

This material is clarified in the revised paper by a nearly complete re-write of the section on time-dependence. Results from Herbert et al. (2014) were incorporated and discussed. The section now starts with the statement: "While time dependence is a factor in all the experimental methods already discussed, two types of measurements are of special relevance in this regard: (i) observing the evolution of the frozen fraction with time at a constant temperature for populations of externally identical sample units, and (ii) varying the rate of cooling for such samples." Statements regarding the cooling-rate dependence based on the stochastic and singular models follow.

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?

Since the slopes of the plots are defined in terms of ln(*R*) versus *T*, not *R* versus *T*, the absolute values of *R* do not impact the comparison. This point is now stated explicitly in the revised text after the definitions of omega and epsilon: "Both quantities are independent of the absolute values of R and have the same value for temperature scales in Celsius or Kelvin."

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. 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 Tc 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.

Indeed this section was sketchy. Full details would have diverted the flow of the paper but what was included was unclear. It is now stated that the discussion is aimed at "the discrepancy between values derived from CNT and the observed values" and the material is reduced to comparisons of the predictions of omega at -10°C and of its temperature trend versus the observed values.

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 quasiâĂŤpermanent 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?

"... identical sites becoming active spontaneously ..." was meant to describe the stochastic assumption reconciled with the idea of sites. The phrase was removed.

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.

This was implemented for as many of the conclusions as possible .

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.

This is a fundamental issue which was raised chiefly by data for KGa-1b kaolinite, silver iodide and perhaps volcanic ash which shows agreement with the stochastic model. However, as discussed in the revised paper, it is not resolved whether sites on these materials arise at random locations or that numbers of similar specific sites have the same probability function describe their potential for activity at a given temperature. The importance of the dynamic factor, in any case, comes into play when the temperatures is constant or when cooling rate is varied.

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?

The conclusions have been changed extensively. The old #5 is replaced by this: "The most direct measurements of nucleation rate are many repetitions of taking a single sample to the same temperature and observing the time to freezing (Sect.~3.1.1). However, even these experiments have problems such as accounting for the time during cooling and possible alterations of the sample with time. Interpretation empirical methods in terms of nucleation rate need detailed justification." The reviewer is right, that these measurements refer to the 'best' site in the sample.

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

The re-written conclusions make no reference to CNT. The CNT-based soccer-ball and alpha-pdf models are described in Sect. 4.2, and the derived results in terms of site densities are emphasized.

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.

Bryant, Hallett and Mason (1959), Roberts and Hallett (1968, Quart. J. Roy. Meteor. Soc., 94, 25-34) and others have shown that saturation with respect to water is required (above a specific temperature) for ice to form at specific locations on substrate surfaces. The statement is also consistent with Ostwald's rule of stages. Still, the phrase was removed for the sake of simplicity.

13 - 18. Thanks for catching these errors. Corrections made.