Review of Vali on the interpretation of freezing nucleation experiments

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Overall, this is an interesting and useful paper. The author clarifies some key issues surrounding how ice nucleation of supercooled drops should be represented mathematically and conceptually. This is not a trivial problem - getting a clear, and general, description of the problem is surprisingly subtle. I found that this clarified some of my own thinking on this subject. The author also analyses the results from a number of past experimental works quantitatively in a novel manner, and attempts to draw some general conclusions from that analysis. This is a valuable thing to do. I would support publication of this manuscript, after the following minor corrections are taken into account:

1   Comments

1. I fully agree with the author that neither the singular nor stochastic limits are justifiable 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 $\frac{dT}{dt}$, and the lifetime of that cloud. If $\frac{dT}{dt}$ 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 $\left|\frac{dT}{dt}\right|$ 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.

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

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).

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.

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

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 freezing rate?

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!

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?

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).

2 Typos / tiny points

1. A bit of nomenclature - you talk about "freezing nucleation". Most people talk about immersion, condensation, contact, or deposition freezing (eg Pruppacher and Klett 1997). Best to be clear at the start of the article which of these 4 comes under "freezing nucleation" (first one definately, but other 3 are not obvious to me)

2. Equation (6) need to define A in the text at this point for clarity

3. page 1721, line 13 "a reasonable range of w values" - this is vague, please be specific.

4. line 25 of same page $T^c i \rightarrow T_i^c$

5. Many of the experiments reviewed here (in sections 3.1.1, 3.1.2 and 3.2.2) are also reviewed in section 4 of Westbrook and Illingworth (2013) in the context of time-dependence. Your focus here is different, but it may be worth mentioning.

6. section 4.1, item 2: "linear increase" is vague here - "linear dependence of lnR with T" would be more accurate

7. section 4.1, item 9: "smaller (less negative)" -> "smaller in magnitude"

3 References

- Herbert et al 2014 Atmospheric Chemistry and Physics Discussion

- Heneghen and Haymet (2002) Liquid-to-crystal nucleation: A new generation lag-time apparatus J. Chem. Phys. 117, 5319 (2002); http://dx.doi.org/10.1063/1.1497635

- McFarquhar et al 2011: Indirect and Semi-direct Aerosol Campaign. Bull. Amer. Meteor. Soc., 92, 183–201. doi: http://dx.doi.org/10.1175/2010BAMS2935.1

- Westbrook and Illingworth (2013) The formation of ice in a long-lived supercooled layer cloud. Q.J.R. Meteorol. Soc., 139: 2209–2221. doi: 10.1002/qj.2096