Manuscript prepared for Atmos. Chem. Phys.
with version 5.0 of the LaTeX class copernicus.cls.
Date: 1 October 2015

Referee comment

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1 Main points

As summarized in the previous reviews, the paper calls attention to an important aspect of immersion freezing experiments, namely that the size dispersion of particles need to be accounted for. The paper demonstrates this with specific examples. Agreement is shown between selected observations and simulations. The simulations incorporate random variations in INP sizes in a stochastic freezing model. This is a reasonable and worthwhile approach. However, besides demonstrating the importance of size dispersions, the paper also claims to constitute a proof for the general validity of the stochastic model of freezing nucleation. Both aspects, the size effect and the stochastic model, are presented with less scrutiny than would be appropriate.

1.1 Particle size effect

The dispersion of particle sizes is a measurable quantity in principle but has practical limitations which the authors do not mention and thus overemphasize the importance of their point. First, not all immersion freezing studies are made with laboratory preparations of ice nucleating particles (INPs) getting suspended in water. For natural water samples, biological samples, and many others, there is no way to a priori separate INPs from inactive particles and determine their size distribution.

Second, even for suspensions of specific substances prepared in the laboratory, there are technical limitations for determining size distributions over the whole range of potential sizes with sufficient accuracy. Considerable obstacles would be faced in determining the sizes of particles in individual sample drop, as appear to be suggested in lines 180-181 of the manuscript. It is highly likely, that errors in size determinations, and truncations of the measured distributions would add considerable uncertainty to analyses of the freezing experiments.

Third, the assumption that all surfaces of all sizes of particles for a given type of material have equal probabilities of containing ice nucleating sites has little empirical support. When particles of different sizes are produced by mechanical, or other means from a larger mass, or when particles are produced by precipitation or coagulation by chemical processes, the material can be expected to yield to different structural features and thus lead to different surface properties of different size particles. A review of relevant literature should be an important part of the authors' argument.

1.2 Simulations

In Section 2.1, isn't J_{het} determined by trial and error until a fit to experimental data is achieved? If so, this should be made more evident to readers. In any case, how this quantity is determined via the simulations is unclear in the paper.

What values were assumed for a_{w} for each of the experiments?

The significant result here is that by accounting for variable ISA, the slope of the $J(T)$ plot is steeper than for uniform ISA and more in agreement with the predicted ABIFM function. Comparison of the slope values with values derived from other sources would be useful.

1.3 Stochastic model

The paper claims validation of the CNT-based stochastic model of ice nucleation. There are two parts to this. First the stochastic model for interpreting observed freezing nucleation events in a population of samples (drops). Second the agreement with the water activity calculations of the nucleation rate coefficient; the ABIFM model of Knopf and Alpert (2013).

Regarding the first point, the main problem is that the authors claim that the results prove the validity of the stochastic model while it has been argued in Vali (2014) and Herbert et al. (2014) et al. that such a conclusion can not be obtained from experiments at constant temperature or constant cooling alone. Such experiments can be equally well interpreted in terms of site-specific models. Only combinations of various types of experiment, even going beyond the two basic types, can provide distinctions between the stochastic and site-specific models. Thus, analyzing only such experiments eludes the more critical examination of their main claim.

An important part of the paper is Section 3.2 where the empirical cooling-rate dependence is claimed to be a consequence of dispersed INP sizes. This section is very hard to read because of the many turns back and forth between simulation and model results. Improvement of this presentation would be very helpful to readers. Also, in this section, a comparison between the derived value of the nucleation rate coefficient from constant-temperature and from cooling experiments of the Herbert et al. (2014) data is needed in order to fully assess the claimed agreement with CNT.

The second part of the claimed proof, the validity of CNT, is presented without acknowledging that the theory is applied with constants adjusted to fit empirical data. It is not clarified how this was done.

In the "General Response to Reviewers" the authors explain "Why we do not assume uniform ice nucleating efficiency of ISA?” by arguing that the small sizes of the sites allow for the same varieties of sites on all sizes of particles. If that is true for even the smallest particles, then this claim is only one of semantics, since the general interpretation of the stochastic model is that INP surfaces are uniform with respect to finding sites. In arguing the validity of the stochastic model, it would help readers if the authors spelled out this view of theirs in the paper.

The contact angle question raised in the reviews needs to be clarified by the authors by stating what was done in the ABIFM calculations that reflects on this.

In response to the comment on the Discussion paper (C3612 - C3617) the authors’ response evades the argument presented there and the revised paper does not deal with the issue. The site-specific interpretation refers to a variations in expected activity for a given substance. Particle-type specific description is totally different and has never been a point of contention in the ice nucleation literature. It is well established that different substances, or particle types, have different abilities to nucleate ice. ”Particle-type specific” is no substitute, in any sense for ”site-specific”. Hence, the authors’ response doesn’t provide a clarification, as claimed, but side-steps the issue. They do not respond in any way to the point made in the comment that the features of the experiments analyzed in the paper can also be interpreted with site-specific models. Dismissing those models by claiming greater rigor for the CNT-based approach is overly simplistic.

1.4 Summary

The authors are correct in showing that the stochastic model and variations in INP sizes may be sufficient to explain the non-exponential decay of fraction frozen in constant temperature experiments. The emphasis on ”may” is what is missing in the paper, since the explanation depends on two assumptions: (1) the validity of the stochastic model and the (2) the magnitude of the size dispersion parameter. The importance of ”may” is accentuated by the lack of mention of alternative explanations.

The paper is written with an exaggerated valuation of the results by not clearly stating its assumptions and limitations and by not considering or contrasting the methods with alternative approaches. Repeated statements about the method employed being physically based, the use of measurable parameters only, etc. are misleading. The claims of generality, and of ”unifying .. model” could well be toned down in order to correctly reflect the contribution made by the work.

In all, this paper contains a significant contribution to improving analyses of immersion freezing studies with distributed samples. Revisions to provide better valuations of the assumptions and limitations of the work would improve the paper and make it acceptable for publication.

2 Minor points

The cloud model presented is so simple as to make little contribution to the paper. Atmospheric aerosol are heterogeneous and can not be represented by a single type of aerosol. The heterogeneity is almost certain to override the effects here shown.

The dispersion of sizes is taken to be represented by a log-normal distribution (as I understand it) and this is taken as valid for all potential cases. Indeed, distributions are often like that but are also subject to truncations and distortions. The assumption should be specified. Extension of the abscissa in Fig. 9C to 10^3 is quite unrealistic.