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Interactive comment on “Ice nucleation terminology” by G. Vali et al.

G. Vali et al.

vali@uwyo.edu

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Authors' responses to second round of reviews and other comments.

G. Vali, P.J. DeMott, O. Möhler, T.F. Whale.

6 May 2015

General

First of all, we thank the referees and other contributors for their comments. It seems to us that the comments reflect a maturing of the terminology. Some comments still touched on fundamental questions and prompted substantial rewrites of a few sections. Where necessary alternative views are identified. The goal remained for definitions to be as general as possible while attempting not to exclude possible processes even if they are not specifically included. This concern arose with respect to a number of terms: deposition, solute effects (liquid versus water), complex modes of nucleation, etc. Also, there is a tricky balance between including references instead of full explanations in a few places and moving toward a critical review paper.

In the following, responses are given first to the referee comments and than to the others in order of the date of publication. The numbering of entries is that used in the Third Draft but where needed the old numbering is also mentioned.

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Anonymous Referee #2:

1. Ice nucleation: the suggested definition has been adopted. References to the parent phases and to the critical embryo size are moved to the explanatory text for this entry.
2. Embryo definition: true, there is no need to assume a strict ice-like structure in the embryos. However, by excluding that simple scenario the definition becomes more difficult. Use of the word “resemble” seems to us insufficiently clear. The solution we propose is to say that with further growth the embryo structure favors development of stable ice. This approach led to additional wording regarding molecular fluctuations of embryos.
3. pg. C11894 - C11897. Changes made as suggested.
4. The designation of dimensions versus units is a valid suggestion, but it was felt useful to keep, in addition to the mention of dimensions, reference to cgs units as this seems to be widespread practice in the ice nucleation literature.
5. Site frequency distributions (SFDs): The reviewer is correct in identifying weaknesses in the explanatory text for this section in the Second Draft. On the other hand, it is good to note that the comment implies support for including this entry in the terminology since this entry focuses on one of the key issues regarding how ice nucleation is described.

The reviewer suggestion for the definition of SFDs contains two basic elements: (i) Site frequency distributions are specifications of the number of sites as a function of effectiveness (catalytic strength), and (ii) Each site is associated with a nucleation rate function. The first part of this definition means viewing ice nucleation as site-specific, i.e. recognizing that sites of different effectiveness exist. This is in agreement with the definition given in entry 3.4 of the draft terminology. The second part of the suggested definition is new and it enters into topics

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elaborated in other parts of the terminology, specially 3.8 and 3.9. Hence, the implications of this proposed definition need to be examined.

If one argues, as I have done in Vali (2014) and before, that, for most materials, site-specificity is the primary factor and that the stochastic element represented by the nucleation rate function contributes variations of nucleation temperatures that are minor compared to the large range of values over which the sites may be active, then the definition as given above has clear advantages. It implies that, for a given sample, the frequency distribution of the most likely temperatures of activity can be determined empirically. The stochasticity described by the nucleation rate adds time dependence to the empirical results.

If, in contrast, the nucleation rate is a relatively slowly varying function of temperature (or supersaturation), or if the range of conditions for all nucleation event is quite narrow, then the frequency distribution of site catalytic strengths and the stochastic element represented by the nucleation rate become intermixed. If the frequency distribution and the relevant values of the nucleation rate cover the same range of temperatures (or supersaturation) the two become practically inseparable. Only by determining the time-dependence of the nucleation events may there be any chance to separate the two factors.

Current evidence supports broad validity of the first of the two scenarios described above. That leads to the situation that time-independent descriptions can be quite useful. First, because temperature-dependent frequencies of INPs are readily measured in experiments. Second, it is very demanding to obtain information on the nucleation rate for all sites of different effectiveness. Third, these approximate descriptions are often derived from measurements that have about the same degree of experimental error as the additional precision that would be introduced by more careful accounting for the time-dependent effects. Thus, it appears that a time-independent, approximate definition of SFDs should be retained. The abundance of papers using this approach to present empirical results

attests to the usefulness of this approach.

At the same time, it is important to recognize that the time-independent descriptions are incomplete and approximate. These time-independent solutions correspond to having the nucleation rate equal to zero at temperatures higher than the characteristic temperature of the site and equal to infinity below that. It behooves authors to recognize this fact and to give as much information about the time scales of the experiments as possible so that some corrections might be attempted when comparing different experiments. An example of this type of empirical correction is given in Vali and Snider (2015) but it is only a first step and other similar avenues will likely emerge in the future.

The definition and the explanatory text for the entry on SFDs has been considerably revised in accord with the arguments presented above. The text is much longer than the concise statement proposed by the referee in order to explain, in some detail, how explicit incorporation of time-dependence, or the contrary, has been implemented in recent literature and how the emphasis on the common elements of the various treatments might help some convergence toward methods of analysis that advance future progress.

6. As requested by the referee, a reference has been added to the discussion of contact nucleation. For other modes, the evidence is much less definitive; a large number of minor works would have to be cited. It seems preferable to not go in that direction and make this paper more like a literature review. If references were to be added for the modes mentioned, the same would be needed for parts like 3.10 and 3.11 and

Referee T. Koop:

1. Thanks for pointing out the error - the wording was changed accordingly to C13109

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metastable.

2. Apologies for misunderstanding the comment regarding apparent nucleation rate.
3. The problem of overlapping uses of the term 'nucleation rate' – both in the literature and in our previous draft of the Terminology – led to a number of changes. "Nucleation rate coefficient" is now used in the definitions of homogeneous nucleation and of stochastic heterogeneous nucleation. In the case of site-specific nucleation, the term "site nucleation rate" is used in order to distinguish it from the other applications of the term. These changes bring greater clarity to the definitions, we hope. Thanks for your emphasis on the need to do so.

D. Niedermeier on behalf of the Michigan Technological University group:

1. The definition of "extensive nucleation rate" offered in the comment is similar to the definition usually applied to homogeneous nucleation where all of the observed volume is fully uniform. For heterogeneous nucleation, it is a directly observable quantity independent of the entities causing the nucleation events. This extensive property of a sample is termed the freezing rate in the proposed terminology. For a given site, this allows nucleation rate to refer - in the same way as it does for an entire sample in the homogeneous case - to the probability per unit time that nucleation will take place. The main advantage of this approach is that it clearly separates characterization of the kinetic process of nucleation on a site from the characterization of the distribution of different sites. Further discussion of this point is given in the response to Referee #1 above, in the reply posted to earlier (February 22, 2015) and in the references cited there.
2. When referring to the nucleation rate as the probability of nucleation the text is corrected to indicate that it is probability per unit time, or that $J \cdot \Delta t$ is the probability for a finite time interval.

3. The differences among stochastic, singular and site-specific descriptions lie in the degree of specificity of sites versus random effects. A stochastic description is not only time-dependent but has the prerequisite of referring to units for which the probability of nucleation is the same at instant. This consideration overrides any differences among sites. Of course, the stochastic equation can be made to cover different types of sites by using different values for the parameters involved. The singular description is time independent. The site-specific description retains the emphasis on different catalytic strength for different sites and adds the time-dependent effects of fluctuating embryo growth through a nucleation rate function specific to each site. The difference between the two time-dependent descriptions - stochastic and site-specific - is that the latter has the nucleation rate applied to sites of given strength, while the former treats all sample units, all sample volumes, or all area of the surface as having the same chance of having nucleation take place. The definitions given on the terminology reflect these concepts. The ideas are further documented in the references cited in the terminology.

A. Bogdan:

1. Indeed, the terminology here proposed originated from studies of ice nucleation in the atmosphere but it is clearly not restricted to treating only atmospheric processes. The concepts dealt with are general. It is true that different communities have different vocabularies but most of those are shorthand usages and survive only because of implicit assumptions made that are valid for particular systems. This is not a desirable situation and a goal of the terminology is to foster broader use of rigorous definitions.
2. Consideration of process of ice growth beyond the formation of a stable ice embryo is not part of this effort of defining a nucleation terminology. This in no way

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implies that process is not of great importance for the atmosphere and for other systems.

3. The effects of dissolved substances are reflected in the vapor pressure and alter the phase boundaries. Thus, the definitions given are not restricted to pure water. Additional effects of dissolved substances are treated in section 3.10.
4. The suggestion to make specific mention of ice in each expression (e.g. "immersion ice nucleation") indeed could be helpful in some situations. However, once 'freezing' is defined, it is appropriate to use that to identify "immersion freezing" while omitting that word would leave it ambiguous as to reference is to deposition or freezing nucleation. Past usage of these terms has established their validity and it would seem unwise to undo that practice. True, some papers may have need to address different disciplines - hopefully the terminology can be used as a base and given further qualifications as the need arises.
5. Context would clarify, we hope, that "fraction frozen" etc. refer to drops, or whatever other sample units (e.g. vials, test tubes, larvae, grains, ...) are being discussed. Thus, including 'drops' in the definition would be unnecessarily restrictive.

B. Murray

1. Altered the text in item 2.1.1 incorporating your suggestion for the definition and indicating the possibility of liquid or amorphous material forming before ice.
2. Note was added regarding the need to test time-dependence experimentally.

H. Wex on behalf of the TROPOS cloud group:

C13112

(References to different entries of the terminology are off in the comments. Responses are given using the correct numbering of items, as they appear in the second draft.)

1. The descriptive text for 1.3 was changed to avoid referring to the embryo as having ice lattice structure.
2. Deleted mention of CNT in the explanatory text for critical embryo size (1.3.2)
3. Based on input from B. Murray, the text for homogeneous deposition nucleation was changed (2.1.1)
4. There is no intention to restrict the use of 'fraction frozen' and 'freezing rate' to heterogeneous nucleation (3.6 and 3.7).
5. The text in 3.2 was shortened.
6. The historical background is included to justify the use of the term 'site' (3.3) even in the face of not having very clear evidence for their characteristics.
7. The text for 3.4 has been thoroughly revised. Discussion of this is given in the response to Referee #2.
8. The sentence about empirical determination of the nucleation rate is correct in our view. That does not mean that theoretical estimates can not also be formulated.
9. We disagree with the change asked for in 3.8 (old 3.10). Hopefully the new text makes the arguments more clearly.
10. In 3.10 additional references were added.
11. Thanks for the other suggested corrections. Most were implemented.

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