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Interactive comment on "Ice nucleation terminology" *by* G. Vali et al.

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Interactive Comment

Authors' responses to referee comments and other contributions.

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General

1. First of all we thank all participants of this Discussion for their contributions. The reviews and comments contained varying degrees of support for the idea of formulating the terminology, identified many weak points and errors, made some helpful suggestions and proposed alternative strategies for proceeding. All this can be taken as indication of considerable interest in the issues raised and demonstrate the divergence of views on several points. It may be also be hoped that the discussion has already added to awareness of the language problems that exist, and in the caution needed in publications to avoid misinterpretations.

2. Regarding the viability of this Discussion, several comments argued for the need for broader community input via meetings, workshops, or committee appointments. Clearly those are good suggestions and efforts have been made by a

number of people in the recent past to move in that direction. Practical problems stood in the way. At the same time, ACPD appeared to offer a way forward and the level of participation in the Discussion seems to justify continuing with it. Hence, we provide responses to the main points raised in the reviews and comments and put forth a Second Draft. Editor Ken Carslaw agreed to this process and we thank him for helping to arrange for the continuation of the Discussion.

3. The related question of how this Terminology should be cited in future literature is a vexing one. The three different journal publications of the 1985 Nucleation Terminology (see point #1 in the reply to Dr. Jaenicke , below) were all different in this regard due to journal policies and formatting restrictions. In the Second Draft, authors of Interactive Discussion contributions are acknowledged in the Introduction. We are open to further suggestions.

Responses are listed in the following in order of publication dates of the reviews and comments. The replies center on points of discussions and are taken as an opportunity to expand on the rationale for the way the second draft defines certain terms. Further debates and proposed improvements will undoubtedly follow on some of what is said here.

Dr. Jaenicke:

The Nucleation Terminology published in 1985 in the J. Aerosol Sci., 16, 575 was also published, in the same year, in the Bull. Amer. Meteor. Soc., 66, 1426, and in the J. Rech. Atmos., 19, 333. These were helpful in establishing usage of the terms included, most importantly those of the 'modes' of freezing nucleation. Thanks for the reminder.

The current effort is focused on ice nucleation, and extends rather than replaces the list from 1985. The definitions there given are still accepted, although some doubt is now attached to 'condensation nucleation', and 'contact nucleation' is recognized as

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possibly needing further sub-classification and clarifications.

Anonymous Referee #1:

1. The need to consider aqueous solutions is emphasized. We agree, and included an entry for this in the second draft. Also, 'liquid' rather than 'water' is used in the second draft in order to be more general. This allows for the definitions to be applied to cases where solutes alter the composition and thus the phase boundaries.

2. The focus on what constitute nucleating agents is broadened in the second draft. The entry for 'substrate' has been eliminated. The task remains for authors to clarify what the agent may be in specific instances. We do suggest the use of three capital letters for this purpose with an eye toward ready identification that a nucleating agent or entity is being referred to.

3. A mineral particle suspended in an aqueous solution is still an INP in our view.

4. The +/- designation is widespread in the life sciences referring to assays that yielded positive or negative responses. This is omitted from the second draft.

5. Indeed the same symbol is used both for the function and the rate constant. In our usage, reference to 'nucleation rate' brings to mind the concept, while 'rate constant' is a numerical value for the reaction rate at given conditions.

6. Singular description, surface nucleation rate, etc. are the most controversial terms and the referee is correct in calling for revisions. Hopefully the second draft is clearer.

7. In the partial response (published as Atmos. Chem. Phys. Discuss., 14, C7662–C7663, 2014) we already argued for the importance of retaining the site-specific

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nucleation term and to refer to the time-dependent descriptions. Admittedly these are controversial, but are not arbitrary. Detailed arguments for these concepts were presented in Vali (Atmos. Chem. Phys., 14, 5271–5294, 2014) and in Herbert et al. (Atmos. Chem. Phys., 14, 8501–8520, 2014) although the two publications take different positions. Here too, the second draft is an effort to provide definitions that can facilitate progress in arriving at further clarifications in the future.

Z. Kanji on behalf of the Lohmann Ice Nucleation Group at ETH-Zurich:

1. References were not included in the original draft for the sake of simplicity and focus. The second draft includes references. Authoritative texts like Pruppacher and Klett are cited, even though we disagree in some details. Differences are partly due to developments over the seventeen years since that publication but also because that text is not without errors.

2. The process of formulation of the Terminology is addressed in points #2 and #3 of General.

3. Most of the suggestions for inclusion of terms has been accepted.

4. We disagree with the interpretation of 'germ' as being the critical size embryo.

5. We disagree about the usefulness of 'ice nucleus' and IN. This has been a problem leading to lack of clarity in communication. "Ice nucleator" is not altogether bad, but has the disadvantage of creating a verb that is imprecise , e.g. it could be read as referring to an aircraft spreading AgI in a cloud. We prefer the use of the INP etc. definitions as detailed in the second draft.

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6. The most useful definitions are general enough to describe experimental results and related concepts. We don't see the distinction described in the comment.

7. The definitions of nucleation rate and freezing rate are now stated more clearly. The use of subscripts is suggested as useful ways to define which of several alternative applications of the concept are being addressed. In general, measured rates are not lower limits of some hypothetical rate but direct evaluations of observations, with corresponding error ranges and meanings.

8. Sites are by definition active; use of 'active sites' would imply that there are also inactive sites.

H. Wex on behalf of the TROPOS cloud group:

1. Most of the suggestions, even if not in their entirety, are reflected in the second draft. Thanks for the input. The use of INE (for entity) will likely find broader use. We thought it best to leave it as an alternative to other possibilities, rather than making it an exclusive definition.

2. Condensation-freezing is a tough problem because it filtered into the literature without clear evidence about it. It is conceptually and experimentally nebulous. Cooper and Vali (J. Atmos. Sci., 38, 1244–1259, 1981) saw this as a possible explanation for the rapid appearance of ice particles near the leading edge of mountain cap clouds. DeMott, (Atmos. Res., 38, 63–99, 1985) made the distinction to immersion freezing based on different rate of appearance of ice in cloud chamber studies when clouds were formed at a supercooled temperature versus cooled through the same temperature. Welti et al. (J. Atmos. Sci., 71, 16–36, 2014) similarly distinguished the apparent existence of a distinct condensation

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freezing mechanism when comparing data from different configurations of a continuous flow chamber, one for which droplets were pre-formed on particles prior to freezing and one in which condensation occurred at close to the temperature of freezing. In cloud chamber studies, the evidence for condensation-freezing would have to be made on the basis of minute differences in the time of ice formation.

3. The inclusion of IN in the naming of instruments is a historical fact that is not good precedent and perhaps will be considered archaic if the use of IN is avoided in the future.

4. "Substance" is indeed helpful for talking about heterogeneous nucleation. It is included in the list of alternatives for referring to the agent involved in leading to nucleation.

5. The reference to genes as agents of nucleation was meant to be indirect. The genes are responsible for the expression of the ice nucleating proteins in a very direct relationship.

Anonymous Referee #2:

1. Clearly, we do not believe the proposed terminology to be definitive and to 'remove roadblocks of our collective understanding'. The intention is more modest: to facilitate future discourse. That goal is already being approached with this exchange, we think. Thus, continuation appears to be worthwhile and the referee's suggestions are contributing significantly.

2. The need for authors to state their own versions of the use of common terms, if they differ from the general interpretation, is sound advice. A sentence to that effect is included in the introduction of the second draft.

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3. The use of short definitions followed by more detailed explanations was adopted in the second draft.

4. Of the proposed mechanisms for developing the terminology, a combination of the first and fourth options is being followed. Convening special workshops would need sponsors and organizers. There is an opportunity for discussions during the forthcoming Fifth International Ice Nucleation Workshop (FIN-2) and perhaps the ACPD material will receive some attention there. Achieving approval at a group meeting seems unlikely, so the slower more deliberate exchange on ACPD will perhaps accomplish the best that can be hoped for.

5. Reflections about the more complex meaning of the terms embryo etc. have been removed. Wordings suggested by the referee are reflected in the second draft.

6. We are not in full agreement about the use of 'nucleation rate'. While it is usually observed in terms of frequency, the concept is meaningful in itself. One could speculate that we would be better off if the term had never been introduced, but it is impractical to abolish it. Hence the effort to specify the meaning for given uses of the term.

7. The specification of identical units is needed, in our view, if the nucleation rate is defined with reference to the average size, composition or content of the sample units. If the units are not identical in those characteristics, nucleation rate becomes meaningless. The substitute is to describe observations in terms of the freezing rate R.

8. Regarding the use of 'stochastic' as different from 'random' the intention is to focus on a process not on a distribution. Mathematically there is no difference, but the implication of a process is important. This focus is supported, for example, by the definition given in the Oxford Dictionary of Chemistry: *Any process in which*

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there is a random element. Stochastic processes are important in nonequilibrium statistical mechanics and disordered solids. In a time-dependent stochastic process, a variable that changes with time does so in such a way that there is no correlation between different time intervals. An example of a stochastic process is Brownian movement. Equations, such as the Langevin equation and the Fokker-Planck equation, that describe stochastic processes are called stochastic equations. It is necessary to use statistical methods and the theory of probability to analyze stochastic processes and their equations.

9. The stochastic element in nucleation that arises from molecular motions is basic. The controversy about the use of stochastic descriptions to interpret the results of experiments with distributed samples is discussed in detail in Vali (Atmos. Chem. Phys., 14, 5271–5294, 2014) and with a different interpretation in Herbert et al. (Atmos. Chem. Phys., 14, 8501–8520, 2014).

10. Hopefully the second draft makes clearer the reasons for distinguishing between stochastic and deterministic descriptions. In the context of past literature - mostly the singular description - the use of deterministic is not focused on the single realization of an experiment but on the differences among sample units which determine the nucleation temperatures for each.

11. Site-specific and time-dependent descriptions are important in our view because, together with the singular description, they form the best available basis for modeling ice formation in clouds and other systems that are not restricted to steady cooling or constant temperature assumptions. How the laboratory results are transferred to cloud models is an important practical question and the consequences of which description is used can be significant (Vali and Snider, Atmos. Chem. Phys. Disc. 14, 1–25, 2014). While the debate about the validity of the stochastic description continues, inclusion of the alternative time-dependent description(s), as encompassed by the site-specific description entry, seems to us

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to be well justified.

Referee T. Koop:

1. The paper was accepted by the Editor as a Technical Note and we assumed that it would be identified as such in ACPD. We now include that in the title of the second draft just to be sure.
2. The Terminology is clearly not a review paper. How far to move in that direction was a difficult decision on each entry of the terminology and the intention was to stay as brief as possible. While other reviews and comments also asked for more references and more explanation, we kept those to a minimum. Whether or not we have struck the right balance remains to be seen in the next round of reviews and comments.
3. Point 7 of the referee addresses what we think is the distinction between 'freezing rate' and 'nucleation rate'. While we mention it in the second draft, we are not keen on proposed further use of 'apparent nucleation rate'. That phrase is an amalgam of observed and underlying factors. To us, freezing rate is clearer in referring to measurement results.
4. The other suggestions were incorporated - albeit to varying degrees - in the second draft.

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

1. We agree that the proposed terminology should be viewed as recommendation, not imposed directive. The firmness with which definitions are written is only

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governed by lucidity and evidence. The wording of these issues in this comment is very helpful. Caveats about the use of the terminology are certainly warranted and are given in the Introduction. Hopefully the second draft is more acceptable on this score.

2. Introducing concepts via CNT is a good idea but not very practical for this effort. Clearly, all our basic concepts derive, historically, from CNT, but to go back to those fundamentals is more fit for a textbook than for this exercise. Here, the intention is to give succinct summaries and adaptations to the specifics of ice nucleation, plus to add new terms.

3. The comment about an "activated process" is unclear to us.

4. We agree on the other comments and have incorporated the suggested changes in the second draft. We are unsure about the suggestion to make a distinction between extensive and intensive properties.

B. Murray

1. Yes, the definitions of immersion and condensation freezing have been included in the second draft.

2. Evidence for a metastable form of ice to precede Ice-1h in some cases adds new complexity to many aspects of ice nucleation. However, since the stacking-disordered ice is transient and is not observed in bulk, and thus it changes the requirements and dynamics of nucleation, the definitions here considered appear applicable. Reference to a stable phase forming as a result of nucleation has been omitted from the second draft. As already done in the proposed wording published on page C8256 of this Discussion, ice nucleation is defined as "the first

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appearance of the ice phase ... ". As far as we can judge, this is compatible with the observations in Malkin et al. (2014) and with the use of the term for describing nucleation observed in laboratory and natural systems. We'll await Dr. Murray's assessment of this view.

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