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Atmospheric Chemistry and Physics Discussions

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

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Vali et al. propose a common terminology for adoption by the ice nucleation community. The motivation for this proposed terminology is the authors' perceived need that lack of uniformity and ambiguity of the meaning of certain terms hinders progress in our collective understanding of the ice nucleation process.

Comments:

I preface my comments by stating that this is not a classical research article that establishes new facts and interprets these facts in the context of a theory. Instead the article represents a (well-informed) opinion that solicits community input and ultimately community approval via the open peer-review process in ACPD. There are debatable pros and cons to the adopted approach and venue for this undertaking.

There is a clear benefit to work with well-defined terms. The terms immersion-freezing,

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contact-freezing, etc. that were defined in Vali (1985) have served the community well. Availability of these broadly understood and agreed upon terms/concepts allows for shorter papers and focused community discussion.

Ambiguity of meaning in manuscripts can be frustrating and difficult to deal with for individual readers. However, this is really a failing of the authors that write such papers. Those authors either have difficulty in articulating their ideas precisely or it may just as well be problematic to precisely define the problem in the first place. The proposed terminology here could be of some help if it helps clarify concepts. Lack of uniformity, however, is not a problem as long as authors define what they mean and if there is not an obvious uniform word choice for the concept. As I argue further below, I do not fully subscribe to the notion that this article will remove the roadblocks of our collective understanding.

Major concerns

I believe that the choice of terminology is somewhat a matter of preference. I therefore do not oppose publication even though I have some issues with the proposed approach. Nonetheless, I feel the need to caution against hasty publication.

First, one can legitimately disagree with this or that definition given. Some members in the community might feel the need to follow convention and adopt the proposed terminology not because the terms describe reality best, but because they are from a perceived authoritative source. That authority may be needed to be appealed to in order to circumvent problems during the necessary peer-review of unpublished manuscript. This conformity problem can prevent people from expressing their preferred (and potentially better) terminology, thus limiting the free exchange of ideas. This is especially a concern when the terminology is controversial. While this concern is anathema to classically trained scientists beholden to an idealized scientific process, it is a real concern with science as practiced.

Second, some of the proposed terms/concepts (e.g. site specific nucleation model,

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singular model, stochastic nucleation) are not definitions to clear up ambiguity. These really are expressions of a particular paradigm in which ice formation is observed and interpreted. Those interpretations are still evolving and it may seem premature to enshrine these concepts within a rigid terminology framework. This is a particular concern for the later definitions of "site-specific" nucleation which are framed in terms of the VS66 viewpoint and this or that time dependence model. Good terminology should enable researchers to express their ideas more precisely but should not have build a particular viewpoint into the definitions.

Third, terminology works best when a crisp, unambiguous glossary style definition can be provided. Explanations of the term should be provided separately from the definition, as is done in the American Meteorological Society glossary (http://glossary.ametsoc.org/wiki/Ice_nucleus).

In their current form the editorializing of the concepts is a mix of literature review (without citations to any work except to VS66, which isn't cited after all) and description which is insufficient convince me to adoption one or another term, mainly because it mixes the viewpoint of the writers with the definition. I would like to contrast the manuscript under consideration with the work of Murphy et al. (2014) who propose a naming convention for atmospheric organic aerosol. The Murphy et al. (2014) paper could serve as an example model for the scope and depth required to argue for a more universally accepted naming convention.

Finally, the contributions of the Lohmann IN group and the TROPOS cloud group to the open discussion, and additional comments regarding the terminology below, clearly demonstrate that there is some disagreement on the proper terminology to be used. It may be challenging to unify these viewpoints into a coherent and broadly accepted set of definitions. Since this is an unusual and unique manuscript and process, I will make a highly unusual proposal to facilitate the articulation of terminology that will find widespread acceptance in the community.

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0. The authors agree/disagree with the proposed process below. If they disagree the manuscript review will proceed as decided by the editor. If they agree the contact the editor and request an indefinite extension of the open discussion.

1. The editor grants an indefinite extension of the open discussion. Simultaneously, the editor posts a editor comment explaining the further handling of the manuscript.

2. The authors select a venue where the various groups can openly discuss the nucleation terminology. For example, one could reserve a block of time at one of the upcoming ice nucleation workshop activities at AIDA where many members of the community will be present. Interested researchers that cannot be present should be alerted of that activity via the open interactive discussion forum. Those individuals should be given opportunity to provide input via webex or a similar service.

3. The authors post a revised manuscript on the interactive discussion. Contributions from the workshop and online discussion will be acknowledged in the revised manuscript in a form that is amenable to the contributing participants (added authors, acknowledgement, or description of the activities leading to the revised paper in the main text).

4. Once the revised manuscript is posted, the editor will allow for one month of additional open discussion and the paper will undergo a second round of formal peer-review.

5. Assuming that a community consensus will have emerged, and critical issues raised during the review process have been addressed, the terminology paper will be published in ACP.

Additional comments on the terminology

These comments are structured as follows. First I provide how I would define the term with obvious overlap from the original text. Below that definition I enumerate reasons why I disagree with the definition as provided in the original manuscript or provide

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related comments. As I conceded above, some of these are a matter of personal preference so my points below should be seen as my contribution to the debate rather than requests for changes that must be adhered to by the letter in order to get the manuscript published.

Embryo (also referred to as germ): Thermodynamically unstable aggregate or cluster of water molecules.

(1) I prefer the above definition. There is no need to invoke the ice crystal lattice since nobody has observed an embryo.

(2) The description really attempts to explain nucleation theory. I do not believe that this belongs here. Perhaps it would be suitable in a supplement explaining the origin and example use of the term.

Critical embryo: Cluster of water molecules that upon addition of a single molecule has probability of growth that is greater than that of decay.

(1) In reference to the explanation in the text: perhaps I am misunderstanding nucleation theory? Is the critical embryo really in stable equilibrium with respect to the metastable phase? It is my understanding that critical embryo has $dG > 0$ when referenced against the pure liquid (metastable phase). It should therefore be thermodynamically unstable with respect to the metastable phase. Either addition or subtraction of a molecule to the critical embryo reduces dG relative to that of the critical embryo. If a molecule is added to the critical embryo the probability of growth that becomes greater than that of decay and the embryo can be considered as activated, although it still has a finite probability to shrink to sub-critical size.

(2) Again the description really attempts to explain nucleation theory. It does not belong into a definition.

Embryo size: The number of water molecules making up the aggregate, the linear dimension of the embryo, or the radius of curvature of its surface toward the metastable

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phase

(1) Should the term aggregate (alternatively cluster) be defined somewhere?

(2) Are embryo, germ, aggregate, and cluster ultimately the same thing? If so, should this be clarified?

Deposition nucleation: ice nucleation from water vapor without apparent prior formation of liquid water.

(1) I believe that nucleation should be added to the term.

(2) The addition of 'apparent' formation of liquid should obviate the need to specify if a liquid intermediate is involved.

Should there be a difference between freezing and nucleation? I thought the distinction was that nucleation refers to the physical process of embryo/germ activation and freezing to the observation in an experiment. The way it is defined in the manuscript it refers to nucleation of ice from supercooled water. It is unclear then why nucleation and freezing should mean the same thing.

I propose to define (deposition, immersion, condensation, contact, contact-inside out, evaporation, electro) -nucleation: a precise definition for each mode. A distinction between freezing and nucleation should be stated separately. The additional modes that have been defined since the Vali (1985) manuscript and are given in Baumgardner et al. (2012) should be included in an updated terminology paper.

INP – ice nucleating particle: a particle that serves as substrate for ice nucleation.

(1) I like this definition and it helps to clearly distinguish the nucleus from the agent that carries it.

(2) However, again I propose to move the rationale to a supplement or separate section. The proposed definition (or something similar) is sufficient.

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Nucleation rate: Frequency of nucleation events per unit (surface area, volume) of the liquid or per unit (number, surface area, mass) of a substrate.

(1) I propose a revised definition above. First, I believe that it is important to include number and mass based nucleation rates since those may be experimentally better defined than surface area. Also surface-area based homogeneous nucleation is also a possibility.

(2) I suggest to use frequency instead of probability since frequency is observed, probability is inferred.

(3) I don't believe that "a collection of identical units" is the required to define a nucleation rate. In a statistical and practical sense 'identical' means that the sample is representative of a known distribution and that the sample size is large enough to determine the frequency experimentally.

Related, I have issues regarding the use of the term stochastic. According to Wolfram MathWorld (http://mathworld.wolfram.com/Stochastic.html): "Stochastic is synonymous with "random." The word is of Greek origin and means "pertaining to chance" (Parzen 1962, p. 7). It is used to indicate that a particular subject is seen from point of view of randomness.". Thus there is nothing special in the word. It is in the concept how it is applied.

Stochastic nucleation: in the context of ice nucleation experimentation there are several stochastic elements that sometimes muddy the understanding.

(1) The growth and decay of aggregates/clusters is a kinetic rate phenomenon. Aggregates/cluster sizes are distributed and thus a random variate. The observed frequency of nucleation depends on the frequency of clusters having size > x. As such nucleation is a fundamentally a random/stochastic process.

(2) My reading of the literature strongly suggests that heterogeneous nucleation proceeds on preferred sites. Without dwelling on all of the implications of that, it is clear

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that sites are distributed in their catalytic strength (e.g. expressed as contact angle or critical temperature), space (i.e. among particle number, surface or volume), and perhaps time (i.e. particle aging). This randomness will always prevent the sampling of "a collection of identical units" and necessitate the characterization of the salient underlying distributions with an effort to characterize and control them as best as possible. For example, when working with monodisperse particles and a mostly pure dust sample. On the other hand, such controls may neither be desirable nor necessary when investigating actual atmospheric nuclei.

(3) Any practical experiment is stochastic (random) since each particle, drop, or experiment represents an instant of a stochastic experiment where properties are sampled from a the salient multivariate distribution. If the number of particles/drops sampled is large enough then the theorem of the law of large numbers states that the observed frequency will have little error and the result appear as a well characterized statistical distribution with deterministic moments (e.g. a stable mean freezing temperature emerges and can be related to the moments of the sample properties [e.g. mean number, size, surface area, drop volume...]).

The term stochastic and deterministic nucleation debate seems really meaningless to me. There is no doubt that most processes in nature and experiments in laboratories are realizations of stochastic processes. There is no doubt that with sufficient sample size the moments of the observed outcome will approach deterministic observables. What is needed is a clarification on the sample size needed for the the theorem of large numbers to apply, and how to properly interpret and extrapolate experimental data when a large sample size is not feasible.

The proposed definition for "stochastic nucleation", i.e. the "probability of nucleation in each time step for a given system" is not the probability of nucleation as the word suggests but the but includes processes (2) and (3) also.

Site: Some distinct location on a substrate/nucleant where ice nucleation takes place

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(1) This is sufficient. I propose to remove 'surface' since it could occur within the substrate volume, e.g. pores.

(2) I suggest to remove the description. It could go into a review part with current research on the topic. Terminology should not state that the concept of sites is most useful if used one way or another. Future research may or may not clarify what a site is. It should also not be necessary to reconcile the idea of a site with stochastic nucleation.

Site-specific nucleation and singular model:

(1) I do not believe this belongs into a terminology paper. Terminology may emerge if the authors want to review various site-specific nucleation models (e.g. pdf-alpha, CHESS, ...) and feel the need to define terms that facilitates the discussion.

(2) Discussion on time-dependence and how to best examine it does not belong in a terminology paper. These are topics for a thorough review article. A definition of the time-dependence may an observable metric for either modelers or experimentalists.

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