

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

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Dear Gabor Vali, Paul DeMott, Ottmar Möhler and Thomas Whale,

We read and discussed your paper in our weekly “research discussion” and our reactions were so varied and even conflicting within our own group, that felt it would be worthwhile to join the online discussion. The group members who, in the end, contributed to this document are David Ciochetto, Colin Gurganus, Dennis Niedermeier, Raymond Shaw, and Fan Yang. In general, we appreciate your effort in attempting to clarify and unify ice nucleation terminology. Over the years, we too have found nucleation terminology somewhat bewildering, and the feeling is only compounded when reading papers from other disciplines in which nucleation is central: metallurgy, pharmaceuticals, and so on. But as soon we began to discuss the paper in detail, it became painfully clear that nucleation, especially ice nucleation, is so poorly understood that

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different viewpoints make precise definitions unachievable. One of us has been known to show a slide in presentations that depicts the proverbial elephant surrounded by blind scientists, one holding the tusk insisting on contact nucleation, one holding the trunk, ever so sure of immersion, others convinced the tail is stochastic, the leg is singular, and so on. Of course it is meant to be taken partly in jest, and while certainly oversimplified it is provocative because it contains an element of truth: our perceptions of ice nucleation are strongly influenced by the aspects of nucleation touched by our experiments and other studies. Recognizing that there will continue to be different perspectives and that many open questions remain, we offer the following opinions and suggestions regarding definitions and explanations outlined in the paper. We hope these comments can help to improve the paper, but at very least to point out where there still may be some fundamentally ambiguous aspects that perhaps should not be restrained by overly-restrictive terminology.

General comments

Our first comment concerns the general applicability of these definitions. In our view, in spite of the clearly stated intentions in the introduction, some definitions are stated too rigorously. They should be seen more as recommendations (in general the word “definition” might be too strong because it implies that each term is totally understood, which is not the case). For example, in the introduction it is mentioned that most of the definitions are phenomenological, i.e., based on observations. But our concern is that too often we do not really even have the observations to allow for phenomenology: e.g., little is known about ice cluster formation, our “knowledge” is essentially based on classical nucleation theory. Furthermore, investigators often have looked at the same phenomenon with different approaches or from different theoretical perspectives, which has led to (seemingly) contradictory conclusions in the past. Therefore we suggest giving a more explicit statement in the beginning regarding how these “definitions” should be interpreted. First, that they represent the current knowledge and can be expected to evolve as the knowledge about ice nucleation increases, but also to some extent they

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represent the authors' point of view about ice nucleation. Second that they should not be taken as mandatory for use within the field: we do not look forward to receiving a review of a future paper in which we are requested to use terminology we feel is inadequate or misleading, with a terminology publication used as authority. There is value in recommending unified terminology, but the concept of recommendation should be made unequivocal.

Outline: We recommend first introducing the concept of classical nucleation theory (CNT) and on that basis defining the concepts of "embryo", "embryo size" and "critical embryo size." CNT may have some shortcomings, but it has two great advantages: it possesses rigorous connections to thermodynamics, and it is widely used in other disciplines. (Note: We also suggest using the phrase ice cluster or ice nucleus as you recommended later on instead of using the word embryo.) CNT would also be useful for the description of nucleation rate, stochastic nucleation, etc., later in the document.

Equations: It would be great if you could include equations to increase the precision and understanding of the terminology, e.g., the critical cluster size as defined through CNT (within the given assumptions), the connection of nucleation rate to freezing probability or frozen droplet fraction (the quantity we actually measure in most experiments), etc.

Figures: Nucleation theory is very complicated, and it would be useful to include some figures to illustrate the probability of freezing or the frozen fraction of a collection of droplets. You describe the shape of this function as a step function, but it would be useful to provide one or more illustrative examples to make the point.

Definitions of terminology: Our impression is that the paper would be much more readable if you were to provide a succinct definition, and then to explain, elaborate, qualify, or muse in a separate paragraph: Some of the definitions are quite long and tend to diverge from the main topic. We would suggest using one or two sentences to define a term and then separately adding any further explanation.

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References: Please add citations/references. It is difficult to ascertain the origin of many of these definitions, and it would be useful to see how they have been previously specified to compare and contrast with those outlined here. Especially specific references, like the VS66 model, should be at least cited.

Specific comments

"Ice nucleation": The use of the word "metastable" implies the existence of an energy barrier, but we suggest that it should be explicitly stated that ice nucleation is an activated process including the presence of an energy barrier.

"Homogeneous and heterogeneous ice nucleation": We suggest separating this into two separate definitions. We further recommend clarifying that homogeneous ice nucleation can occur in pure liquid as well as in solution droplets. Cantrell and Heymsfield (2005) give two very reasonable explanation/definitions so we recommend building on them, or at least citing them here.

"Deposition nucleation and freezing modes": Firstly, it makes sense to us that all of these modes should be explicitly grouped under the heterogeneous nucleation category. Secondly, do we know enough to define these modes without inadvertently biasing our view of the actual physical processes? E.g.: Can we clearly distinguish between deposition nucleation and condensation freezing? Could there be a link to adsorption and subsequent surface diffusion versus epitaxial growth? Is immersion freezing always induced by a particle (for example, can we look at a biological macromolecule as a particle?). In the case of immersion freezing does the surface have to be wetted, not only be covered (e.g., Leidenfrost droplet)? We were happy to see the reference to water-air-particle triple interface, although we might quibble that it should be referred to as a triple line.

"Substrate": The word "substrate" is somehow ambiguous. Would it be better to call it a "catalyst"?

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“Ice nucleating particle”: In general we appreciate this formulation because the “ice nucleus” corresponds to the ice-like H₂O-clusters. The widespread but probably misleading reference to ice nucleating particles as IN in the atmospheric literature may be difficult to change, but it definitely is one place where a strong recommendation on the terminology would be worthwhile. Where we become less enthusiastic about this definition is with regard to its connection to sites: Can we think about a particle as being a collection of sites, where a site is the location at which ice nucleation can occur? What does such a site look like? This also concerns your definition of site.

“Nucleation rate”: Please state clearly that there are both extensive and intensive nucleation rates. It would also be advisable to add that the nucleation rate corresponds to the probability of nucleation, with an equation to make the meaning precise. For exponentially distributed waiting times the inverse of the nucleation rate corresponds to the mean nucleation time. You might also state that the volume freezing rate refers to “homogenous freezing” and the surface freezing rate refers to “heterogeneous freezing.”

“Stochastic nucleation”: Ice nucleation should be stochastic by default? If it is decided to have a separate definition for stochastic nucleation we would suggest including a definition for singular nucleation (as commonly used in the literature) as well. Again, equations would greatly improve the precision of meaning.

Reference

Cantrell, W. and Heymsfield, A.: Production of ice in tropospheric clouds – A review, *B. Am. Meteorol. Soc.*, 86(6), 795–807, 2005.

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