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## Ice nucleation terminology

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## Abstract

Progress in the understanding of ice nucleation is being hampered by the lack of uniformity in how some terms are used in the literature. This even extends to some ambiguity of meanings attached to some terms. Suggestions are put forward here for common use of terms. Some are already well established and clear of ambiguities. Others are less engrained and will need a conscious effort in adoption. Evolution in the range of systems where ice nucleation is being studied enhances the need for a clear nomenclature. The ultimate limit in the clarity of definitions is, of course, the limited degree to which ice nucleation processes are understood.

## 1 Introduction

The phase transformations of water are of great scientific and practical interest. However, many uncertainties exist, especially with respect to the initiation of ice from water vapor and from liquid water. The goal of this document is to facilitate further discoveries by proposing the use of a common terminology. Special consideration was given to the need to bridge the types of expressions and the language used in the physical and in the life sciences. The definitions listed have various degrees of firmness; some are well understood and uniformly applied in the literature, others are based on partial knowledge and almost certain to involve simplifications that will eventually be cleared up. One could say that the definitions are, for the most part, phenomenological, i.e. descriptive of observations. Furthermore, there is a fundamental limitation of the observations in that they are restricted to detecting the emerging bulk phases of ice, since possibilities are very limited for observations on the atomic and molecular scales where ice formation is initiated and is subject to the most important controlling conditions and influences.



## 2 Terminology

*Ice nucleation*: Formation of a thermodynamically stable body of ice from a metastable phase (vapor or liquid) of water.

5 *Embryo* (also referred to as *germ*): Aggregate of water molecules in an ice crystal lattice at a sub-critical size. This definition of an embryo is based on the building-block view of nucleation, where the critical size is reached through the successive addition of molecules either singly or in multiples. It is possible that the sub-critical embryo is not really in an ice-like structure but is bonded differently and that the precise ice  
10 structure arises only at the point of becoming stable.

*Embryo size*: Imprecise term for describing the extent of an ice embryo in one of various possible ways, such as the number of water molecules making up the ice structure, the linear dimension of the embryo or the radius of curvature of its surface  
15 toward the metastable phase. The term is a substitute for a more detailed description of the embryo, since we know little about the forms and internal structures ice embryos take in different situations.

*Critical embryo size*: The size at which the probability of growth of an embryo becomes equal to the probability of decrease in size. This size is the minimum that can lead to ice nucleation because the embryo at this size is in stable equilibrium with respect to the metastable phase and if this size is exceeded then the probability of growth is greater than that of decay. We do not know quantitatively how the probability increases with additional growth beyond this point but it is assumed to be very rapid. Conversely,  
20 the probability to fall back below the critical size becomes negligible. The concept of critical size is useful even though shape and other factors may be found in future studies to lead to definitions of the critical embryo that involve more than a single  
25 quantity. Using the classical nucleation theory calculations of critical embryo size

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provides a rough idea of the magnitudes involved. More elaborate molecular dynamics calculations may lead to better estimates but are still uncertain.

*Homogeneous and heterogeneous ice nucleation:* Ice nucleation without and with any foreign substance aiding critical embryo formation.

*Deposition:* Nucleation of ice from water vapor without the formation of bulk liquid water prior to ice nucleation. Since it is not clear how “bulk” liquid can be defined or diagnosed, there is some ambiguity in the definition and indicates that caution is needed in the use of the term.

*Freezing:* Nucleation of ice from supercooled liquid water. One can distinguish between (i) immersion-freezing: initiated by a particle inside a drop or by a surface fully covered by water, and (ii) contact-freezing: initiated by a particle just coming in contact with a supercooled drop or forming a water-air-particle triple interface. In addition, in the atmosphere (or in simulations thereof), one can recognize as different the case in which the ice nucleating particle (INP – see below) is also a condensation nucleus (CCN). Differences in effectiveness among these three sequences, or modes, are not well established and need further research. Details of the contact-freezing and condensation-freezing processes also need clarification.

*Substrate:* Solid (exceptionally liquid) material of specific physico-chemical characteristics on which ice embryos form. Other terms used for this in the literature are “nucleant”, “host”, “nucleation active entity”, “nucleating particle”, “nucleator” or “ice nucleus”. In a generic sense, *substrate* is a reference to the entity that serves as the promotor of heterogeneous nucleation. This is a general reference to the agent of heterogeneous nucleation, without naming the material it is made of, or the form it takes. More specific identification of the substrate can be expressed using the terms

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that follow.

*INP – ice nucleating particle:* In a very high proportion of cases, the substrate is a small entity – a particle – so a specific acronym is reserved for those cases. This still does not specify the composition of the particle, but refers to the unit that carries the nucleating substrate. A number of different terms have been used in the literature for this. For decades, the terms “ice nucleus” or “ice nuclei” were used almost exclusively with reference to atmospheric aerosol that could initiate ice, that is individual particles, each of which resulted in the formation of one ice crystal. While it was recognized that only a specific location on the particle surface is actually where an ice embryo becomes stable, the entire particle was referred to as the ice nucleus. This led to confusion. Although the concept of a “site” appeared in the literature to expand on the identification of an ice nucleus, this term was used only restrictively. With the entry of ice nucleation studies to systems other than clouds and also involving biological substances (bacteria, fungi, etc.) usage became more confusing because focus fell rapidly on nucleation by other than aerosol particles and on details like what part of the bacteria, or other entity was where the nucleation events originated. In all, the term “ice nucleus” has become both overused and vague. For atmospheric applications, or in more general when dealing with many separate entities, it would be more appropriate to use *ice nucleating particle*, *INP* to refer to individual units or *INPs* to refer to a collection of them. Since ice nucleation is more complicated than condensation due to the different modes it can follow, using “ice nuclei” in the general sense similar to “condensation nuclei” is overly ambiguous and can be misleading.

*IN<sub>x</sub> – specific class of ice nucleating substance:* The first part of the acronym, IN has been used to stand for “ice nucleating” or “ice nucleus” or other versions. To simplify usage, it is recommended that IN stand for ice nucleating (in the sense of a qualifier, not a noun) and that it be followed by a lower case word or letter identifying the specific agent being referred to. Examples: *INprotein*, *INgene*, *INcrystal*, *INmaterial*,

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*INblob* (not really), *INgroup*, *INstep*, *INdislocation* etc. are possible terms. In any case, if in a given work further shortening is desired and the intended usage is clearly declared, the abbreviation can be shortened to *INp* or *INg*, etc. The addition of the letter *A* to form abbreviations like *INA*. is superfluous, since “nucleation” is meant to indicate activity by itself. The  $\pm$  shorthand notation is useful in some cases and can be preserved as *IN+ gene* etc.

**Nucleation rate:** Number of nucleation events per unit time, designated by  $J(t)$ . It represents the probability of nucleation within a time period for a collection of identical units. Nucleation rate can refer to the probability per unit volume of the vapor or liquid,  $J_v(t)$ , or it can refer to the probability of nucleation per unit surface area of the INPs,  $J_s(t)$ . For invariant conditions, the inverse of the nucleation rate is the time constant of the exponential decrease in the fraction of sample units that are not nucleated.

**Freezing rate:** Fraction of sample units (drops, vials, etc.) of sample volume  $V$  freezing per unit time,  $R(t)$ , or per unit temperature interval,  $R^*(T)$ . For homogeneous nucleation  $R(t) = J_v(t) \cdot V$ . For polydisperse sample volumes, the freezing rate need to be specified by volume range. More precisely, if needed, “freezing nucleation rate” can be used to avoid ambiguity with the rate of crystal growth.

**Stochastic nucleation:** Commonly used to refer to equal probability of nucleation in each time step for a given system, independently of previous history of arriving at the current state. In that sense, it means a random process in which the exact time of nucleation is not predicted, only its probability. Application of the term to nucleation is based on the analogy to radioactive decay where each atom has the same probability of undergoing decay at any given time. In nucleation, equivalents to radioactive atoms may be separate but otherwise identical volumes of the metastable phase (homogeneous nucleation), or areas of a substrate/nucleant surface if the areas can be considered comparable in the same sense as atoms of a radioactive

substance. In this latter case, the nucleation rate (probability of nucleation) is defined with reference to a unit surface area of the INPs,  $J_s(t)$ , or with reference to unit mass of the suspended INPs,  $J_m(t)$ . These definitions can equally apply to deposition or to freezing nucleation.

5 *Site*: Some distinct location on a substrate/nucleant surface where ice nucleation takes place. There is controversy about what constitutes sites and how to diagnose them. Some evidence points to crystal dislocations, steps, impurities, molecular configurations, other crystallographic or morphological features on the nucleant surface as preferred locations for critical embryo formation. The concept of sites is most meaningful if it is used, either in a predictive or diagnostic sense, to refer to some distinguishing characteristic, say  $X$  so that the density of sites (per unit area of the surface) can be expressed by a function  $n_s(X)$ . Ideally the parameter  $X$  would be some microscopic property of the surface and its value would be predicted by theory. Lack of knowledge about the properties of sites leaves that ideal to be reached only in the future. In the absence of other identifying parameters for  $X$ , the site density is most readily derived from observations of nucleation temperatures; the result is expressed by the integrated site density  $n_s(T)$  (see below).

10 In terms of the stochastic description (see above)  $n_s(X)$  is assumed to exhibit a step function increase to a very high value at some  $X$  so that all sites can be considered identical. The spread of observed nucleation temperatures is then accounted for by the temperature dependence of the  $J_s(t)$  function.

15 *Site-specific nucleation*: The simplest form of site-specific description is the *singular model* which assumes that an observed freezing event can be ascribed to nucleation at a site whose characteristics determined the temperature of freezing. This view is only an approximation because it ignores time-dependence associated with fluctuations in embryo size. It is useful for descriptions of the ice nucleating abilities of different materials for comparative purposes. The activity of some substrate, or INP, per unit volume of

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liquid can be expressed with the differential or integrated site density functions (spectra),  $k(T)$  or  $K(T)$ . The numbers of nucleation sites active above temperature  $T$  per unit surface area of the substrate,  $n_s(T)$  is termed the integrated surface site density. With these definitions  $K(T) \cdot V = n_s(T) \cdot A$ , where  $A$  is the substrate area present per unit volume of the liquid. The quantities  $k(T)$ ,  $K(T)$  and  $n_s(T)$  can refer to freezing or to deposition depending on context. The singular approach is equivalent to assuming that the probability of nucleation changes from zero to unity as the sample is cooled to the characteristic temperature,  $T_c$ .

If the probability of nucleation is not assumed to be a step function at  $T_c$ , but is described, more realistically, by a nucleation rate function (probability of nucleation)  $J_{T_c}(t)$ , as in the VS66 model, the freezing rate or deposition rate can be described by the  $J_{T_c}(t)$  function in combination with  $n_s(T_c)$ . This description allows for site-specific characteristics to be significant but also incorporates time-dependence via the stochastic effect of the nucleation rate function.

In practice, if a system is steadily cooled so that  $T(t)$  is a monotonically decreasing function, it may be difficult to distinguish between the relative importance of the two factors in the product  $J_{T_c}(t) \cdot n_s(T_c)$  and hence may be difficult to establish if  $J_{T_c}(t)$  is different from  $J_s(t)$ , the stochastic description. The distinction is more clearly evidenced with a system at  $T = \text{const.}$  but is best examined by a combination of different testing methods.

### 3 The process

We suggest that support for the definitions given above, or proposals for altering the terminology, be submitted as comments to this article, or emailed to the authors. The result may be a revised version of the terminology, again submitted for further comments, or if a consensus seems to be forming, a final publication of the results.