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

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Technical Note – Second draft for ice nucleation terminology

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The first draft (ACPD-14-22155-2014) of this ice nucleation terminology received three referee comments and several comments from scientists. Authors of Interactive Comments were (in order of the date of posting): two Anonymous Referees, Dr. R. Jaenicke, Dr. Z. Kanji on behalf of the Lohmann Ice Nucleation Group at ETH-Zurich, Dr. H. Wex on behalf of S. Augustin-Bauditz, H. Bieligk, T. Clauss, S. Hartmann, K. Ignatius, L. Schenk, F. Stratmann, J. Voigtländer (members of the Cloud-group at the Institute for Troposphereic Research, TROPOS), Dr. T. Koop, Dr. D. Niedermeier on behalf of D. Ciochetto, C. Gurganus, R. Shaw and Y Wang at Michigan Technical University, and Dr. B. Murray.

What follows here is a major modification of the original text. Reviews and comments pointed to some basic agreements, but also to major differences in views with respect to the meaning of several items. Perhaps most debated were the definitions of sites, nucleation rate, stochastic nucleation and site-specific nucleation. It became clear that the state of knowledge is not sufficiently settled to be unequivocal about what these terms mean. The situation in 1985, when the first Terminology was published (Vali 1985) was much simpler. That terminology focused on atmospheric processes; it was

timely and helped to solidify the ways the three major "modes" of freezing nucleation were defined.

It is to be emphasized that the terms here listed do not constitute an exhaustive list but cover only some of the most frequently used terms. Neither are the descriptions here given meant to be full explanations. Focus is on characteristics that are essential to the meaning of the terms. Fuller explanations can be found in Pruppacher and Klett (1985), Vali (1999), Cantrell and Heymsfield (2005), Murray et al. (2012), Khvorostyanov and Curry (2014) among others. Important differences exist among these references on some points, in the sense similar to that indicated in the preceding paragraph, reflecting the unsettled nature of these topics.

This second draft incorporates several suggestions made by the reviewers and in the comments. Nonetheless, it cannot pretend to reflect consensus. Emphasis has been placed on making definitions as general as possible, and also to be clear about doubts and about differences in views. As noted in the Introduction of the first draft, the aim of the terminology is to facilitate communication and to remind authors of the need to clearly define their use of terms whenever those terms are not uniformly understood to mean the same thing.

This second draft is a continuation of the Discussion. Perhaps further comments will help to better illuminate the possibilities and limitations of the terms here listed. Hopefully, a third version will go forward for publication in ACP.

Following the naming of entries in capital letters, a brief definition is given in italics. Additional details follow in the paragraph(s) that follow in normal font. References to other entries are given in bold-face numbers.

1 General

1.1 PHASES OF WATER

Within the range of normal atmospheric conditions water can exist in three different phases, namely vapor, liquid and ice.

The thermodynamically stable phase is defined by the existing vapor pressure and temperature, as usually depicted in a phase diagram. A metastable state (supersaturation, supercooling, superheating, etc.) arises when conditions change from those corresponding to one stable phase to those corresponding to another. The first formation of the new stable phase from the metastable state is a nucleation event.

1.2 ICE NUCLEATION

The first appearance of the ice phase exceeding the critical embryo size, either from supersaturated vapor (deposition nucleation) or from supercooled liquid water (freezing nucleation).

1.3 EMBRYO or GERM

Aggregate of water molecules in an ice crystal lattice at a sub-critical size.

1.3.1 Embryo size:

The size of an ice embryo expressed 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 toward the metastable phase.

1.3.2 Critical embryo size:

The size at which the probability of growth of an embryo becomes equal to the probability of decrease in size.

The critical size is the point of unstable equilibrium with respect to the parent phase. With minimal additional increase in size (perhaps just one molecule) nucleation takes place. For heterogeneous nucleation, CNT leads to contact angle as the dominant parameter defining critical embryo size.

2 Homogeneous ice nucleation

2.1 HOMOGENEOUS ICE NUCLEATION

Ice nucleation without any foreign substance aiding the process.

2.1.1 Homogeneous deposition nucleation

Ice nucleation from supersaturated vapor, without apparent prior formation of liquid.

Because of the very high supersaturation required for homogeneous deposition nucleation of ice it is not observed in the atmosphere or in other natural systems.

2.1.2 Homogeneous freezing nucleation

Ice nucleation within a body of supercooled liquid without any foreign substance aiding the process.

2.2 Nucleation rate

The probability, or observed frequency, of ice nucleation in unit volume of supercooled liquid or supersaturated vapor within a unit of time.

Homogeneous nucleation rate (for freezing) has been usually denoted as $J(T)$. To help distinguish this from the heterogeneous case, and to focus on the fact that homogeneous nucleation rate refers to a volume of liquid, it is recommended to apply a subscript v, thus using $J_v(T)$. The value of J_v at a given temperature is the homogeneous nucleation rate coefficient for that temperature with units of $(\mathrm{cm}^{-3}\mathrm{sec}^{-1})$.

Homogeneous nucleation rate is observed as the frequency of freezing events in a collection of sample units of the same volume: $J_v(T) = \frac{1}{N} \cdot \frac{\mathrm{d}N}{\mathrm{d}t}$. CNT relates $J_\mathrm{v}(T)$ to the properties of the liquid and the net rate at which molecules are added to the ice embryos. Additional factors, like dissolved substances, electric fields, etc. may influence the homogeneous nucleation rate.

3 Heterogeneous ice nucleation

3.1 HETEROGENEOUS ICE NUCLEATION

Ice nucleation aided by the presence of a foreign substance so that nucleation takes place at lesser supersaturation or supercooling than is required for homogeneous ice

nucleation.

3.2 INP, INM, INE, etc.

Ice nucleating particle (INP), molecule (INM), entity (INE), material, matter, substance, object, item, entity, unit, or else, that is assumed to be the agent responsible for observed heterogeneous nucleation.

Because of the variety of materials and forms that can be responsible for heterogeneous ice nucleation, it is impractical to have a single designation that covers all possibilities and is sufficiently informative. It is preferred that authors refer to what is the nucleating agent in each particular case in the manner most appropriate for the system studied. The form of the designation, IN plus a third letter, may be helpful enough for effective communication. The term "nucleator" is also used as a general reference to the object whose presence is responsible for observed ice nucleation.

Reference to an INP (or equivalent) does not, in general, 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 ice begins to form, 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 for a long time was rarely used. 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 is more appropriate to use **ice nucleating particle**, **INP**, or the other forms listed in the definition, to refer to individual units or **INPs**, or other forms, to refer to a collection of them. Since ice nucleation is more complicated than condensation nucleation, 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.

3.3 SITE

Preferred location for ice nucleation on an INP, or equivalent.

Direct experimental evidence for deposition nucleation (e.g. Mason 1957; Anderson and Hallett 1976) and expectations that derive from non-uniform surface properties of INPs, and the associated differences in binding energies to water molecules and incipient ice structure, point to the role of specific locations on the surfaces or in cavities of particles, molecules, etc., which promote freezing with greater effectiveness than other locations. Thus, sites are considered important for both deposition and freezing nucleation. Observed nucleation on, or within, a sample is taken as being due to the most effective site found in it. Sites of various effectiveness are assumed to be occur on the surfaces of most materials.

3.4 SITE FREQUENCY DISTRIBUTION

The number of sites causing nucleation per unit surface area of the INP, or equivalent. Quantitative measure of the abundance of sites of different ice nucleating effectiveness.

The distribution of sites, or integrated site density (Connolly et al. 2009; Niemand et al.

2012) expresses the number of sites per unit surface area of INPs that cause nucleation by some value of temperature or supersaturation. Hoose and Möhler (2012) used INAS (Ice Nucleation Active Site) density to refer to this quantity. Site density is a time-independent description of nucleating activity. The quantity is designated as $n_s(T)$, or $n_s(S)$, with units of cm^{-2}. Values of n_s can be derived either from model assumptions, usually in terms of a contact angle, or can be taken from observed nucleation temperatures using the singular description (**4.8**).

3.4.1 Modeled site density

Use of the contact angle as a surrogate to define effectiveness is achieved using CNT, and is expressed as a nucleation rate function. This approach has been therefore linked to the stochastic description (**4.6**) but the characterization of sites can be considered on its own. Examples of this approach are: Marcolli et al (2007) used contact angle as a surrogate to express site effectiveness. That idea was further developed by Welti et al. (2012) in the α-pdf model. Niedermeier et al. (2011, 2013) constructed the Soccer Ball Model to describe the distribution of sites of different effectiveness. Hartmann et al. (2013) modeled the distribution of sites among sample units.

3.4.2 Nucleus spectra

Sites of different effectiveness can be diagnosed via the temperature or supersaturation at which they produce nucleation. With reference to unit volume of the samples, the number concentration of sites is derived from the number of sample units in which nucleation is observed. The differential and cumulative nucleus spectra defined in Vali (1971) are, for freezing: $k(T) = \frac{1}{V \cdot (N_0 - N_F(T))} \cdot \frac{\mathrm{d}N_F(T)}{\mathrm{d}T}$ and $K(T) = -\frac{1}{V} \cdot ln(1 - \frac{N_F(T)}{N_0})$. These definitions rely on the singular description (**4.8**) but are valid more generally if T_c replaces the observed freezing temperature T (**4.11**).

Site density with reference to surface area and the cumulative nucleus spectrum, for freezing, are related as $K(T) = n_s(T) \cdot A$.

3.5 MODES OF HETEROGENEOUS ICE NUCLEATION

Distinctions in the mode of nucleation are made on the basis of the process envisaged to lead to nucleation.

Definitions of nucleation modes was given by Vali (1985) with a focus on atmospheric processes. The definitions are given below with some changes.

3.5.1 Deposition nucleation

Ice nucleation from supersaturated vapor on an INP or equivalent, without apparent prior formation of liquid.

3.5.2 Freezing nucleation

Ice nucleation within a body of supercooled liquid ascribed to the presence of an INP, or equivalent.

Further specifications of modes are:

Immersion-freezing refers to ice nucleation initiated by an INP, or equivalent, located within the body of liquid.

Contact-freezing is initiated by an INP, or equivalent, at the air-water interface as the INP comes into contact with the liquid, or forms at an air-liquid-

particle triple interface.

This process is defined as separate from immersion freezing because of empirical evidence that some INPs are more effective in this mode than when immersed in the liquid. There is as yet no definite clarification for how to distinguish this mode from immersion freezing. Some laboratory evidence points to a difference depending on whether the particle is inside the liquid or outside it; these are described as inside-out versus outside-in nucleation. In the atmosphere, preactivated particles may cause freezing when coming into contact with supercooled liquid droplets.

Condensation-freezing is thought to take place when freezing is initiated concurrently with the initial formation of liquid on a cloud condensation nucleus (CCN) at temperatures below the melting point of ice. Whether condensation-freezing is truly different from deposition nucleation, or distinct from immersion-freezing, is not fully established. Hence, reference to this mode of nucleation requires added circumspection.

Other modes of freezing nucleation reported in the literature are electro-freezing, evaporation-freezing, mechanical shock freezing and collision-freezing. Evidence available at this time does not permit general definitions to be established for these processes.

3.6 FRACTION FROZEN

The ratio of the cumulative number of sample units frozen at T to the original number N_0: $f = \frac{N_F}{N_0}$ with N_F given as either a function of time or of temperature.

Frozen fraction represents the results of experiments with sample units drawn from the same original volume. It can be used when the sample units are gradually cooled or

when held at a fixed temperature. Similar quantities can be readily defined for nucleation modes other than freezing.

3.7 FREEZING RATE

Feezing rate expresses the results obtained from an experiment in which the freezing of a number of sample units is observed.

Freezing rate is expressed as a function of the number of sample units frozen at time t: $R = \frac{1}{N_0 - N_F(t)} \cdot \frac{\mathrm{d}N_F(t)}{\mathrm{d}t}$. The freezing rate coefficient for given T has units of inverse time, e.g. (sec^{-1}). The freezing rate function is a direct description of empirical observations with distributed samples and can be used both for experiments in which the samples are steadily cooled or others in which the temperature is held constant. Freezing rate is related to the time derivative of the frozen fraction (**4.8**): $R = \frac{1}{1-f(t)} \cdot \frac{\mathrm{d}f(t)}{\mathrm{d}t}$. Freezing rate for any particular sample is dependent on the volumes of the sample units and on the INP contents (site density or nucleus spectrum) of the liquid. It is also influenced by dissolved substances. For polydisperse sample volumes the freezing rate need to be specified by volume range. (It may be noted that for homogeneous nucleation (no INP), the freezing rate is directly proportional to the nucleation rate and the volume of the sample units: $R = J_v \cdot V$.)

Some authors use "apparent nucleation rate" to refer to the same quantity, but this term carries with it some undesirable ambiguity.

3.8 SINGULAR DESCRIPTION

Description/model of observed nucleation events for a population of sample units containing INPs (or equivalents) and assuming that the preferred sites have a spectrum of different nucleating abilities. Also referred to as the deterministic description of ice

nucleation.

This description is based on evidence that points to sites having well-defined, albeit not perfectly stable, potentials for promoting nucleation. Each site is then characterized by the temperature, or supersaturation, at which it is observed to nucleate ice for a given mode, with the time history of the sample not taken into account. In that sense, the singular description is often referred to as deterministic. The singular description is expressed quantitatively by site density (**4.4.1**) or nucleus spectra (**4.4.2**).

3.9 NUCLEATION RATE

In analogy with homogeneous nucleation, nucleation rate is envisioned to express the probability that nucleation takes place at some site on the INP (or other) involved. It is determined empirically from the frequency of nucleation events per unit time. It is a function of temperature (for freezing), time, and other factors reflecting the nature of the nucleating site.

Application of the term is different for the stochastic description (**4.10**) and for the site-specific description (**4.11**), as detailed in the following. It is recommended to identify the interpretation of J with appropriate subscripts.

3.10 STOCHASTIC DESCRIPTION/MODEL

Description/model of the observed frequency of nucleation events in a population of sample units which all have equal probability for nucleation to take place in them within a period of time.

This description assumes that there are large numbers of sites of equal effectiveness on the surfaces of INPs and interpret observed frequencies of freezing in terms of

nucleation rate per unit surface area $J_s(T)$ with units of $(\mathrm{cm}^{-2}\mathrm{sec}^{-1})$ or per unit mass $J_m(T)$ with units of $(\mathrm{g}^{-1}\mathrm{sec}^{-1})$. If the number of sites of the same effectiveness per unit surface area is $n_S(T)$ then $J_s(T) = n_s(T) \cdot J(T)$. Empirical values of $J_s(T)$ are obtained from $R(T)$ via $J_s(T) = \frac{R(T)}{A}$. From these values of $J_s(T)$, the site density n_s can be derived if $J(T)$ is taken from theory (e.g. CNT) or is independently determined.

3.11 SITE-SPECIFIC DESCRIPTION/MODEL

*Descriptions that encompass definitions of site density distributions (**4.4**) and account for the time-dependence of freezing nucleation.*

The singular description (**4.8**) is a time-independent version of site-specific descriptions.

In the VS66 model, Vali and Stansbury (1966) view nucleation rate as reflecting the kinetics of embryo formation on an individual nucleation site and define a characteristic temperature T_c for each site (**4.4.2**) by the temperature at which the nucleation rate J_{site} (a new notation) has a specific value C. The form of the nucleation rate function may vary with the value of T_c and is thus designated as $J(T_c, T)$; it has units of (sec^{-1}). Definition of T_c is from $J_{\mathrm{site}}(T_c, T_c) = C$. Determination of the J_{site} function has only been approached indirectly (e.g. Vali 2008) since multiple examples of sites with the same T_c are not identifiable a priori.

3.12 Comparison of stochastic and site-specific descriptions

While the concept of nucleation rate appears to be similar in the stochastic description (**4.10**) and in the site-specific description of VS66 (**4.11**), different underlying assumptions are incorporated in that function in the two cases. To apply the stochastic description to a given data set, the function J_s is taken in most cases, to extend over the range

of observed freezing temperatures in the sample. For the site-specific description the function J_{site} is assumed to rise very rapidly over a narrow range of temperatures.

A consequence of this difference is that the spread of freezing temperatures observed in an experiment is ascribed primarily to the presence of a variety of sites of different effectiveness in the site-specific description, while the stochastic description ascribes that spread to random effects represented by the $J_s(T)$ function as well as the distribution of sites. The two descriptions lead to divergent predictions about the time-dependence of nucleation (Vali, 2014).

The multi-component stochastic model (MCSM)of Broadley et al. (2012) and Herbert et al. (2014) combines elements of both views described in the preceding paragraph is. A number of species of nucleating surfaces are each defined by a different nucleation rate function $J_{\text{s,i}}$ and a given assumed abundance.

3.13 AQUEOUS SOLUTIONS

Dissolved substances in water change the equilibrium boundaries of phases and influence ice nucleation.

In many atmospheric and other systems dissolved materials are present in water and alter the conditions for ice nucleation. The magnitudes of the induced changes are determined by the concentration and type of the solute(s) as expressed by water activity and additional factors both for homogeneous nucleation (**3.1**) and for immersion-freezing nucleation (**4.5.2**) by different types of INPs (**4.2**) (Koop et al. 2000; Knopf and Alpert 2013).

3.14 PRE-ACTIVATION AND MEMORY EFFECTS

Mode or efficacy of observed nucleation influenced or altered by the pervious temperature/humidity history of the INP, or equivalent.

Experiments have shown that prior exposure to low temperature or high humidity, or a combination of both leads to enhanced activity in comparison with what the INP, or equivalent, would exhibit otherwise. Such effects may introduce ambiguity in diagnoses of the mode of activity (**4.5**) in laboratory experiments or in atmospheric or other natural systems. Certain INP characteristics (composition, configuration, surface properties) may favor such effects. Explanations of the effects focus on the potential for cracks, pores and other features on surfaces to hold ice even under conditions where bulk ice would be unstable.

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Table 1. Nomenclature (with CGS units indicated).

A	total surface area of INPs in a sample unit [cm^2]
f	fraction of samples frozen (**4.6**)
$J(T)$	nucleation rate (probability of freezing) per unit time as a function of temperature [s^{-1}]
$J_s(t)$	nucleation rate per unit time and per unit surface area of INPs [$\mathsf{cm}^{-2}\ \mathsf{s}^{-1}$]
$J_m(t)$	nucleation rate per unit time and per unit mass of INPs [$\mathsf{g}^{-1}\ \mathsf{s}^{-1}$]
$J_v(t)$	nucleation rate per unit time and per unit sample volume (**3.2**) [$\mathsf{cm}^{-3}\ \mathsf{s}^{-1}$]
J_{site}	nucleation rate on a specific site (**4.11**) [s^{-1}]
$k(T)$	differential nucleus spectrum; number of sites active within a 1C interval at T per unit sample volume [$\mathsf{cm}^{-3}\ \mathsf{C}^{-1}$]
$K(T)$	cumulative specgtrum, or integrated volume density of active sites : number of sites active above T per unit sample volume [cm^{-3}]
$n_s(T)$	surface density of sites (number per unit surface area of INPs) active above T [cm^{-2}]
N_F	number of samples frozen
N_0	total number of samples in an experiment
R	freezing rate per unit time [s^{-1}]
T	temperature [C]
T_c	characteristic temperature for a nucleating site [C]
V	volume of sample unit [cm^3]
CNT	classical nucleation theory