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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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The authors have addressed my initial concerns. I do have a few minor comments the author may want to take into account.

pg. C11893

ICE NUCLEATION suggested wording "The first appearance of a thermodynamically stable ice phase." The reference to critical embryo size imposes an unnecessary mechanistic idea into the definition. References to the type of phase transition is not required to define ice nucleation.

"Aggregate of water molecules in an ice crystal lattice" → do we know the molecular arrangement of the embryo? I suggest: "Thermodynamically unstable aggregate of water molecules resembling the stable phase"

Pg. C11894

“ice structure, the linear dimensions” → insert “or” to ling the two concepts.

“probability of decrease in size” → “probability of decay”

“with respect to the parent phase” → “with respect to the parent and nucleated phase”

“For heterogeneous nucleation, CNT leads to contact angle as the dominant parameter defining critical embryo size” → I suggest to omit this. First CNT isn’t defined at this point. Second, contact angle is a just a parameterization of the free energy of the germ. It therefore doesn’t define the embryo size, it expresses it as a macroscopic quantity”.

C11895 “of ice nucleation in unit volume” → “of ice nucleation events in unit volume”

I suggest to use dimensions rather than units. i.e. change “temperature with units of (cm-3 s-1)” → “temperature with with dimensions of (L-3 t-1)”.

C11897 “on nucleation by other than aerosol” → “on nucleation by entities other than aerosol”

I suggest to remove the bold emphasis for INP.

pg. C11898 SITE FREQUENCY DISTRIBUTION and Modeled site density

The definition of site seems fine to me. The description and review of methods, however, seems to not really clarify the issue. In my opinion, the notion of a site is reasonably trivial and in line with the definition given: “number of sites causing nucleation per unit surface area of the INP”.

Obviously sites are a random variate that has some distribution. The interesting question the community is asking are: (1) what are the natural variables of sites?, and (2) how are the natural variables distributed. To my mind, each site has a nucleation rate Ji(T). The principle variables that determine that rate are the catalytic strength (arbitrarily and interchangeably expressed as Tc, contact angle, or free energy of germ formation), a slope alpha = dJi(T)/dT and a functional form e.g. an exponential dependence on temperature. Different functional forms could be envisioned, although that

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will not change the issue I am raising below. If there is a collection of sites, the collection of site-specific nucleation rate functions Ji(T) = f(Tc, alpha) will be distributed according to some probability density function. Maybe Gaussian, maybe Exponential, maybe Gumbel, maybe non-parameteric. For a heterogeneous population that pdf may vary for example with particle size. The point I would like to raise is that all site models are just special cases of this general view. For example, the ns(T) model simply assumes alpha = infinity, spatially uniform Ji(T) and thus resolves the cumulative distribution function via measurements. Other models make somehow distribute Ji(T, alpha) with some preconceived notion and test whether specific observations can be explained by those assumptions.

Fundamentally, the VS66 model, the INAS model, the alpha-pdf model, the soccer model, and others have their origin in the site frequency distribution and only make different simplifying assumptions. There is nothing special or unique in any particular version of the model, except for the first model that articulated the idea. Many of the debates about “singular vs. stochastic”, “time-dependent vs. time-independent” should ultimately be recast in what of the principle variables are distributed or can be considered as effectively uniform for any given system under investigation.

I suggest to amend the site frequency distribution definition and streamline the discussion of site frequency distribution and stochastic vs. site model.

SITE FREQUENCY DISTRIBUTION “The number of sites causing nucleation per unit surface area of the INP, or equivalent. Each site may have its own nucleation rate function and is characterized by a temperature dependent catalytic strength. ”

Text: Sites are in principle distributed quantities. Different assumption in which of the natural variables are distributed and which ones are prescribed give rise to time-dependent/time-independent and INP size-dependent/size-independent descriptions for a population of particles.

pg. 11900 please add references for “INPs are more active in this mode than when

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immersed in the liquid" and references to each of the "nucleation modes referenced in the literature are electrofreezing ..."

pg. C11904 "the preceding paragraph is." → fix sentence

pg. C11905 "equilibrium boundaries of phases" → I suggested "equilibrium phase boundary"

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 22155, 2014.

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