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

We have read and discussed the second draft of your terminology paper and, in the interest of a healthy scientific discussion, wish to take the opportunity to make further comments. The group members who contributed to this document are again David Ciochetto, Colin Gurganus, Dennis Niedermeier, Raymond Shaw, and Fan Yang. In general, we appreciate the changes and improvements to the revised paper. We especially like the new format of first providing the succinct definition, followed by a separate more detailed description of the defined term. We still have concerns and comments, however, particularly regarding some of the equations and the definitions presented in section 4. Although we know that it is unlikely that our viewpoints will converge perfectly we still want to share our suggestions for some of the definitions and terminology. In what follows we outline several comments we would consider of major importance, followed by some minor corrections.

Major comments

- *Definition of nucleation rate.* First, we would like to clarify our suggestion to define extensive and intensive nucleation rates. We intended to use the terms in the sense typically defined in thermodynamics: for example, in two commonly used undergraduate thermodynamics texts (Blundell and Blundell 2009: Concepts in thermal physics, 2nd edition, p. 5; Schroeder 1999: An Introduction to thermal physics, p. 163) variables that scale with system size (e.g., mass and internal energy) are defined as extensive and variables that are independent of system size (e.g., density and temperature) are defined as intensive. It seems reasonable to adopt the terminology widely used in other fields. Second, in many textbooks that cover the nucleation topic (e.g., Adamson 1990: Physical chemistry of surfaces, chapter 9; Lamb and Verlinde, 2011: Physics and chemistry of clouds, chapter 7; Pruppacher and Klett, 1997: Microphysics of clouds and precipitation, chapter 7; Seinfeld and Pandis, 1998: Atmospheric chemistry and physics, chapter 10), the term 'nucleation rate' refers to the intensive property, i.e., with unit [$m^{-3}s^{-1}$] or [$m^{-2}s^{-1}$]. We recommend following this nearly standard usage. Third, we find the current definitions of nucleation rate somewhat ambiguous; we definitely find it misleading to define nucleation rate as equal to the probability of freezing (as in Table 1 of the revised article). Based on your Table 1, we recommend the following concise definitions for 'nucleation rate':

  Extensive (or particle) nucleation rate J(T) [#/s]: Number of critical nuclei formed per unit time in a specified volume of water or on a specified INP surface.
  Surface nucleation rate (J_s(T)) [#/m^{2}s]: Number of critical nuclei formed per unit time, per unit surface area.
  Volume nucleation rate (J_v(T) [#/m^{3}s]): Number of critical nuclei formed per unit time, per unit volume. Both extensive and intensive nucleation rates are as a function of temperature T.

  When no qualification is given, we would interpret nucleation rate as an intensive quantity, and would determine whether it refers to a surface or volume dependent quantity based on context. In any sense, we would suggest that nucleation rate can be interpreted as a nucleation probability only if multiplied by a sufficiently small Δt and an appropriate surface area or volume.

- *Equations.* We appreciate the inclusion of equations, however some of them contain typos or inconsistencies. To avoid confusion we encourage the authors to check each of them. For example:
  - Line 81: The equation given here should read $J_v(T) = 1/(V*N)*dN/dt$ because the volume nucleation rate $J_v(T)$ is given in units $[m^{-3} s^{-1}]$.
  - Line 149-150: Should the V in the equation for $k(T)$ and $K(T)$ be S for heterogeneous nucleation? What is the difference between the cumulative nucleus spectra $K(T)$ and site density $n_s(T)$? $K(T) = n_s(T)*A$ does not make sense, at least the units seem inconsistent.

- *Freezing rate.* The explanation of the freezing rate (section 4.7) seems vague. As it is defined through the given equation it is identical to the extensive heterogeneous nucleation rate $[s^{-1}]$ if each droplet from an investigated droplet population features a single particle and all particles have identical size and nucleation properties. Why should the freezing rate in the heterogeneous case be based on volume of the sample units (line 222)?

- *Stochastic and singular descriptions.* Regarding sections 4.7 – 4.12 we are afraid that students or scientists from other disciplines and research fields will have a hard time understanding and distinguishing between the stochastic, singular and site-specific descriptions; at least we could not agree amongst ourselves as to the meaning. We acknowledge that one's perspective, for example as experimentalist or theoretician, influences one's view on ice nucleation topics and therefore it also influences the way of defining terms. And even within the experimental ice nucleation community there are different points of view on these topics. Below we listed comments to show what confused us and maybe would also confuse others. We suggest that this section should be streamlined and brought into balance with the other sections, i.e., less model specific, more general in nature.
  - Why should the stochastic description be limited to a population of sample units with equal nucleation probability (section 4.10)? We think that this definition is too narrow. The word stochastic implies that ice nucleation is related to a probability, which depends on temperature, surface, etc. Another way to describe the stochastic model is to give some ideal cases to distinguish between the singular and stochastic models. For example, at a constant temperature, whether droplet can freeze or not is deterministic based on the singular description (time independent). In contrast, it might freeze after some time based on stochastic description (time dependent).
  - In contrast, in a strict sense, nucleation in the singular description is viewed as being deterministic. It is mentioned in the definition of singular description that ice nucleation is viewed as independent of time. This is stated again on line 236: "The singular description (3.8) is a time-independent version of site-specific descriptions." However, on line 213 it is stated that the singular description is expressed quantitatively by site density, the latter being based on CNT and therefore explicitly including nucleation time. This is inconsistent. Our sense is that there is little debate in the community that ice nucleation is fundamentally stochastic, and at the same time that a description of

nucleation on realistic particles must account for varying material or defect properties, which may be represented conveniently through singular-type models.

o   Based on these points, it is also not clear to us what the difference is between the stochastic and the site-specific descriptions. Both descriptions are based on the stochastic view, i.e., having a nucleation probability. Why distinguish between the two?

o   In our understanding the MCSM introduced by Broadley et al. (2012) is similar to the alpha-pdf, soccer ball model, etc. i.e., each particle or site is associated with a given nucleation rate but, instead of being based on CNT, they apply a simple functional form for the temperature dependent nucleation rate.

Minor comments

- Page 1, line 8, it should read: "... F. Yang at Michigan Technological University..." (not Technical)

- Page 4, line 90: 'entity' can be deleted here because it is already introduced on line 89.

- Table 1: J(t) should be replaced with J(T).