

Dear Gabor Vali and co-authors!

We here in Leipzig discussed the new version of your manuscript: “Technical Note – Second draft for ice nucleation Terminology” in a (somewhat smaller) group of people, again, and the final version of this comment was compiled by Frank Stratmann and myself. We acknowledge the improvements that were made, but still have some comments which we list below. – Comments are preceded by the numbering given in the manuscript (using the version put online in February). Technical comments which point out typos and such are added at the end. (Writing of this text was spread over quite a bit of time, and we realize that some of our comments by now were already given by others.)

Before starting with the specific comments, we however would like to note that we are in agreement with most of the other comments and encourage the authors to follow the respective suggestions. This holds especially for the comments of Dennis Niedermeier for the Shaw group, concerning “intensive and extensive properties”. We realize that you already answered to that, but wanted to stress that the differentiation in extensive and intensive parameters (the latter being considered material properties) as given in the comment from the Shaw group might help to clarify things.

Specific comments:

2.3: We suggest to change “ice crystal lattice” to “ice crystal like lattice” or to “cluster of water molecules”. Accordingly, “CLUSTER” could also be mentioned in the title of 2.3, together with “EMBRYO or GERM”.

2.3.2: “Contact angle” is mentioned here, but is not defined anywhere in the manuscript. Also it may/should be noted, that contact angle is only a commonly used technique to “parameterize” the interactions between ice embryo and ice nucleating entity.

3.1.1: Is it necessary to give a definition which is of no meaning for natural systems (as you state, quite correctly)?

3.2 For heterogeneous ice nucleation, definitions for freezing rate and frozen fraction are given. We suggest to give similar definitions for homogeneous nucleation as well. Other concerns we have with here were already covered in the other comments.

4.2: The describing text became somewhat confusing to us after the sentence “This led to confusion”. The concept of a “site” appeared in the literature not necessarily “to expand on the identification of an ice nucleus”, but more to further locate or define the “nucleating substrate” (to use an expression you used earlier in this paragraph). Also, bacteria or other entities “where the nucleation events originate” are not “other than aerosol particles”. A whole bacteria (or pollen or fungal spore), if airborne, also is an aerosol particle. Ice nucleating macromolecules then can be part of an aerosol particle, or might occur separately. This needs to be reformulated, or the respective text could be removed, as the definition is well described already above “This led to confusion.”

4.3: This definition is difficult to read and could be shortened a lot. It might not be necessary to include the background of why sites started to be around as a concept – it might be easier to understand if the first three lines were deleted, and to just say “It is assumed that specific locations on the surface or in cavities exist, which promote ... .” Also, this definition makes it clear that it is still difficult to disentangle an INP (which might carry a site) and an INM (which as a whole might be the site, or of which only some part might be active in ice nucleation).

4.4: It might be necessary to refine the definition, adding “at some value of temperature or supersaturation” at the end of the first defining sentence (“... of the INP, or equivalent.”).

In the text following the initial definition, two different parameters are mentioned: The distribution of sites is different from the integrated site density (the latter, by concept, being the integral of the former). This should be stated more precisely, clearly discriminating between site frequency distribution and cumulative site frequency distribution.

4.4.1: This could become a separate definition for the contact angle, the one we were missing (see what we said concerning 2.3.2. The text would need to be adapted accordingly.

4.4.2: We might miss something here, but in our view, what is described here is the distribution of sites as introduced in 4.4. Heterogeneous freezing in principle is a function of INP surface area (for insoluble INP) or of INP volume/mass (e.g., Snomax, or more general materials that can be suspended), and NOT of the droplet volume. However, some experimental techniques, especially those methods, which were recently named “wet-suspension methods” (i.e., cold stages / freezing arrays), may include a large number of ice nucleating entities in each single examined droplet. This may result in an apparent droplet volume dependence, which needs to be corrected for (e.g., via the equation given here). However, as in our view this volume dependence is artificial, we suggest to only stick to what’s introduced in 4.4 and to remove 4.4.2.

4.7: The first explanatory sentence should be amended by “temperature T”, so that it reads “... of sample units frozen at temperature T and time t:”

The equation given for R does miss this dependence on temperature.

It is said “Freezing rate of any particular sample is dependent on the volumes of the sample units and on the INP contents of the liquid.” Please see our respective comment on 4.4.2.

4.8: The definition given for “singular description” here would also fit to that of a contact angle distribution and may not be specific enough.

4.9: The nucleation rate is not necessarily determined “empirically”, this word should be removed.

4.10: For a contact angle distribution, onto which a stochastic description can well be based, not all sites necessarily have an equal effectiveness (see what you wrote at 4.12). Hence it is not necessary to restrict your description here to a population of sample units, which all have equal probability for nucleation, and sites of equal effectiveness. Please reformulate.

In general, in this part of the text, an alternative sequence of appearance could improve clarity, e.g. swapping 4.8 and 4.9.

4.12: The Soccer Ball Model cited earlier is also a multi-component stochastic model and should be mentioned in the last paragraph of this definition, too. (Niedermeier et al., 2011 & 2014).

4.13: The effect of concentrated solutions on immersion freezing was also examined by Wex et al. (2014): Wex, H., P. J. DeMott, Y. Tobo, S. Hartmann, M. Rösch, T. Clauss, L. Tomsche, D. Niedermeier, and F. Stratmann (2014), Kaolinite particles as ice nuclei: learning from the use of different kaolinite samples and different coatings, *Atmos. Chem. Phys.*, 14, 5529-5546, doi:10.5194/acp-14-5529-2014. It is stated in there, that maybe much of the ice nucleation interpreted as deposition ice nucleation might be immersion freezing in concentrated solutions. The observed ice nucleation below water vapor saturation, where haze droplets formed, could be modeled based on a parameterization of immersion freezing when allowing for a freezing point depression that depended on the concentration of the solution in the haze droplet.

Technical comments:

2.3.2: Describe the acronym CNT (i.e., add “Classical Nucleation Theory”), as this is the first appearance

4.2: “entity” appears twice in the defining lines

4.2: In the second describing paragraph, 3<sup>rd</sup> sentence, the word “particle” should be added: “... with reference to aerosol **particles** that could initiate ice”

4.3: Remove “be” in the last sentence (it is now “... assumed to be occur on the ...”)

4.4.1: The second Niedermeier et al. publication on the Soccer Ball Model appeared online end of 2013, but was officially printed only in 2014. Please correct the year, here and in the literature-list.

4.6: The “or” in the definition could be replaced by “and/or”.

4.7: The sentence given in parenthesis (“It may be noted that for homogeneous nucleation ... .”) would better appear in the section on homogeneous freezing.

4.7: A “third person-s” is missing: “For polydisperse sample volumes the freezing rate needs to be ...”