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Atmospheric Chemistry and Physics Discussions

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

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General comments:

The manuscript by Vali et al. proposes and provides a consistent terminology for ice nucleation research. This aim is particularly useful because scientists from different disciplines often use different wordings for the same processes. In that sense I think the manuscript may become a very helpful piece of work harmonizing the sometimes Babylonian slang used in that area. Of course, the manuscript is not a regular scientific paper because it does not provide any scientific results or conclusions and, thus, in my opinion falls into the category of a "technical comment". Therefore, I suggest that the term "technical comment" is also reflected in the manuscript title as is the norm for such type of papers in ACP.

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Discussion Paper

However, I think the paper could be even more useful if it included more details in some places. For example, I find it quite disturbing that no references to any previous work is given, not even to previous papers on terminology, some by the same author(s). In some cases the discussion of why to use or to avoid a certain term could also profit from citing appropriate papers that discuss the matter or provide examples to some of the relevant processes. Moreover, I also think that in some places a figure may be very helpful in defining some of the terms and distinctions between different terms much more clearly than by just providing a few sentences.

After reading the paper I am not sure about the future usage and citation by other authors to the current manuscript. Do I have to reference the paper just because I use the term ice nucleating particle (INP)? Or nucleation rate? Probably not. In my subjective view the previous paper by Gabor Vali "Ice nucleation theory – a tutorial" is very often cited because authors felt that most of the relevant definitions and processes are described and explained in that paper, not just because of terminology. Therefore I would ask the authors to reconsider whether they also want to include a section outlining the fundamentals of the ice nucleation processes in general. Then the basic processes and explanations as well as the most up-to-date terminology would be in one document which I guess would not only be highly cited, but would also draw more attention to some of the important subtleties of ice nucleation terminology.

In the last paragraph (page 22162, lines 21-25) the authors briefly outline their vision for improving the manuscript under discussion with which I fully agree. Reaching a consensus interactively through one or several further rounds of revisions is a very good and constructive idea. But this procedure may require extending the open discussion of the paper also towards revised version(s) of the manuscript. I suggest this to be considered by the editor of the manuscript.

In summary, I think the paper is suitable for ACP but it requires changes before it can be published. Below I am listing a number of specific comments, of which I consider comment (7) to be the most important one.

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Specific comments:

(1) Page 22157, lines 3-4: Maybe this definition of ice nucleation should also include nucleation from amorphous solid water as well as from aqueous solutions (instead of just water vapor and liquid water).

(2) Page 22157, lines 5-10: I suggest including a statement that the usage of “critical nucleus” is discouraged because of the ambiguity with the term “ice nucleus”.

(3) Page 22157, lines 19 to Page 22158, lines 2: I suggest that it is noted somewhere that in CNT the critical embryo size is defined as an equilibrium property, that is derived from the maximum in the deltaG for cluster formation.

(4) Page 22158, line 12: “from supercooled liquid water”. The term “and aqueous solutions” might be added, see comment above.

(5) Page 22158, line 22: There are examples of ice nucleation triggered by single molecules or molecular aggregates in solution. It appears that the definition of substrate given here does not include such ice nucleation molecules, does it?

(6) Page 22160, line 4: INA is not defined yet at this point. The argument might be explained in more detail thus providing a better description of why it is superfluous. Also remove period after the term INA.

(7) Page 22160, lines 8-19: To me, these are two very important paragraphs. In line 8 it does not become clear whether J is used here as an observed rate or as a material property. Personally, I think this is a very delicate but important point that is used ambiguously in many papers on ice nucleation (including some of my own). To bring this point across more clearly, I use the example of the authors of freezing nucleation, for which I assume that both homogeneous and heterogeneous nucleation are stochastic processes. Consider a certain number of identical water droplets that are investigated in freezing experiments. Each of the droplets contains the same concentration of identical INP (with a homogeneous surface) and each droplet sits on the same clean and

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identical substrate. In constant temperature experiments, the freezing rate per unit time R(t) of these droplets is then determined experimentally. In many cases (but not always) each of these freezing events is triggered by a single nucleation event, implying that the observed nucleation rate, which I here tentatively term NR(t), is identical to the freezing rate, i.e. NR(t) = R(t) which both have the unit of inverse time. In very many cases it is not entirely clear whether the observed nucleation rate corresponds to a homogeneous nucleation process in the droplet volume, a heterogeneous nucleation process triggered at the surface of the substrate or a heterogeneous nucleation process triggered at the surface of the INP, or a mixture of several of them. Tentatively, we can assign the observed NR(t) to each of the processes by scaling it to the droplet volume V or substrate surface area As or total particle surface area Ap. This procedure will result in an apparent homogeneous nucleation rate $NR_v(t)$ in units of per time and volume, an apparent heterogeneous nucleation rate $NR_{As}(t)$, and an apparent heterogeneous nucleation rate $NR_{Ap}(t)$ both in units of per time and surface area. In the worst case, none of these apparent nucleation rates may actually represent the usually sought after "real" nucleation rate of water under these conditions; in the best case it is equal to ONLY ONE of the apparent nucleation rates. With "real" nucleation rate I mean the material property of water that is usually termed $J_v(t)$, $J_{As}(t)$, or $J_{Ap}(t)$. The fact that I am struggling with a name for these and so use the term "real" nucleation rate indicates to me another terminology problem that I think should be resolved within this manuscript. In many papers it is not distinguished between apparent nucleation rates and the "real" nucleation rates: in most cases both of them are termed simply "nucleation rate". If we consider that in the above example the actual nucleation was triggered by the INP, then $NR_{Ap}(t) = J_{Ap}(t)$. But for the other two it is very likely that $NR_v(t) \gg J_v(t)$ and $NR_{As}(t) > J_{As}(t)$.

Therefore, I strongly suggest terming the material properties $J_v(t)$, $J_{As}(t)$, or $J_{Ap}(t)$ either "nucleation rate coefficients" or "nucleation rate constants" to better distinguish it from the apparent rates that are observed. (Note that this terminology would be in line with that used in chemical kinetics: "reaction rate" for the observable that depends on

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reactant concentrations, and “reaction rate constant or coefficient”, which is the constant specific to a particular elemental reaction and listed in handbooks.) I suggest that another variable is also given for the apparent rates, not J.

(8) Page 22160, line 16: What does the asterisk indicate? When is it to be used?

(9) Page 22160, line 18-19: Similarly, an even more problematic ambiguity may arise for “deposition rate” instead of “deposition nucleation rate”. It may be mentioned that in this case, “deposition rate” should be strictly avoided.

(10) Page 22161, line 21-22: I am not sure what is meant by this sentence. Can you be more explanatory here?

(11) Page 22161, line 28-29: As I understand this sentence the term “substrates” should be used rather than “materials” according to the definition for substrate. Given earlier in the paper.

(12) Page 22161, lines 17/18 and page 22162, line 3: there is an ambiguity here for the term $n_s(T)$, which is termed “integrated site density” in the first location and “integrated surface site density” in the latter.

(13) Page 22162, lines 11: The “VS66 model” has not been defined up to this place in the manuscript, and the corresponding paper should be referenced.

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