Atmos. Chem. Phys. Discuss., 14, C12361–C12363, 2015
www.atmos-chem-phys-discuss.net/14/C12361/2015/

Atmospheric Chemistry and Physics Discussions
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Interactive comment on "Ice nucleation terminology" *by* G. Vali et al.

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Received and published: 23 February 2015

General comments:

The revised manuscript has been improved and the authors have addressed most of my comments appropriately. I am not satisfied, however, with the response and changes to my initial comment 7 as outlined below.

In summary, I think the paper is suitable for publication in after the following points are considered appropriately.

Specific comments:

(1) Page C11894, "The critical size is the point of unstable equilibrium with respect to the parent phase". This is not entirely correct. In the thermodynamic definition of

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the Gibbs free energy of the embryo, all embryos smaller than the critical size are metastable and all embryos larger than the critical size are unstable, i.e. the critical size is the point where the equilibrium between the embryo and the supercooled liquid changes from metastable to unstable.

(2) Page C11901-11902: It appears that the authors have misunderstood point (7) in my original comment on the first draft. They now added the following sentence “Some authors use ‘apparent nucleation rate’ to refer to the same quantity, but this term carries with it some undesirable ambiguity.” which is actually quite confusing, because the undesirable ambiguity remains unexplained, and thus the statement is not very helpful. In their answer the stated “Point 7 of the referee addresses what we think is the distinction between ’freezing rate’ and ’nucleation rate’.” In fact, this is not entirely what I meant. It is okay for me to define nucleation rate on page C11895 as “The probability, or observed frequency, of ice nucleation in unit volume of supercooled liquid or supersaturated vapor within a unit of time.” instead of an apparent nucleation rate. However, in the next sentence it is written “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 Jv(T).” This is also fine. But my point is that for most types of experiments it not 100% guaranteed that the observed nucleation rate actually is entirely due to homogeneous nucleation. Thus it is easy to state, but not necessarily correct, that the homogeneous nucleation rate Jv(T) is simply J(T), when J(T) = R(T) / V. (The same applies to heterogeneous nucleation rates.)

Along the same line I propose to use the term rate COEFFICIENT only when one really means the material property, i.e. when one is convinced that the observed Jv(T) REALLY IS due to homogeneous nucleation, and not just simply the nucleation rate per unit volume. (The latter can be calculated even if the observed nucleation process was due to heterogeneous nucleation.) But if one follows this true, then Jv(T) actually has two meanings, which I think is not very helpful either. I strongly recommend removing this ambiguity.

(3) Page C11901, last paragraph: I do not understand the difference between freezing rate and freezing rate coefficient as outlines here. The rate R in the first equation is already scaled to the number of samples (No-Nf(t)) and hence should be constant throughout the entire experiment at constant temperature T. What is the freezing rate coefficient then? I suggest removing the term rate coefficient here. If not, the authors must clearly state the difference between rate and rate coefficient, because at present the text is confusing and ambiguous.

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

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