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Atmospheric Chemistry and Physics Discussions

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

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Comments on the third draft of "Ice nucleation terminology" by G. Vali, et al.

1.1 PHASES OF WATER

The thermodynamically stable phase is defined by the existing water vapor pressure and temperature, as usually depicted in a phase diagram.

Comment for the author, who wrote this subsection: This sentence is incorrect. The thermodynamically stable phase of water is NOT defined by "the existing water vapor pressure and temperature". It is determined by pressure and temperature. See textbooks. In the case of aqueous solutions the third parameter is concentration, as I wrote in my previous comment.

1.3 EMBRYO or GERM

Comment: The author, who wrote this subsection, does not explain what is "GERM"?

2.1.1 Homogeneous deposition nucleation and homogeneous ice nucleation from water vapour

Deposition nucleation is the formation of ice directly from water vapor. Because of the very high supersaturation required for homogeneous deposition nucleation of ice it is not observed in the atmosphere or in other natural systems. However, there is evidence for homogeneous ice nucleation from water vapor via processes that involve the intermediate step of homogeneous condensation of liquid, or an amorphous phase, at supersaturations below that required for deposition (Jensen and Murray, 2010).

Comment 1: What is the difference between "Homogeneous deposition nucleation" and "homogeneous ice nucleation from water vapour"? I do not see a logical connection between "1.2" and "2.1.1". The author, who wrote the subsection of "1.2 ICE NUCLEATION", writes "The ice phase can be initiated in environments of supersaturated vapor (deposition nucleation) or supercooled liquid water (freezing nucleation)." The substitution of "and" for "or" would slightly clarify "2.1.1". However, I think the term of "Homogeneous deposition nucleation" alone would be enough.

Comment 2: In the context of Comment 1, the part of the 3rd sentence of "However, there is evidence for homogeneous ice nucleation from water vapour. . .. " should be discarded. And what is more, I do believe this entire sentence should be removed, because the paper by Murray and Jensen (2010) (by the way, why the order of authors' names has been changed?) does not present any validating evidence. In Abstract, Murray and Jensen state: "It is postulated here that particles of amorphous solid water . . . may nucleate homogeneously in the summer mesosphere." Their postulate is based on the calculations performed "Using classical nucleation theory and a one-dimensional model. . .". However, in science, ′evidence′ is accumulated through observations or laboratory experiments at controlled conditions and usually used for

supporting or rejecting a hypothesis.

Comment 3: In the context of Comments 1 and 2, I think the paper by Murray and Jensen (2010) was cited for the reasons other than scientific.

3.7 FREEZING RATE

Comment: The term of "FREEZING RATE" is misleading for scientists from the outside of atmospheric ice nucleation. (See my previous comment).

3.10 AQUEOUS SOLUTIONS

In many atmospheric and other systems dissolved materials are present in water and alter the conditions for ice nucleation. The magnitudes of the induced changes are determined by the concentration and type of the solute(s) as expressed by water activity and additional factors both for homogeneous nucleation (2.1) and for immersion-freezing nucleation (3.5.2) by different types of INPs (3.2) (Koop et al. 2000; Knopf and Alpert 2013; Wex et al. 2014).

Comment 1: The author, who wrote this subsection, does not understand the water activity criterion (WAC) proposed by Koop et al. (2000). Koop et al. wrote: "the homogeneous nucleation of ice from supercooled aqueous solutions is independent of the nature of the solute, but depends only on the water activity of the solution-that is, the ratio between the water vapour pressures of the solution and of pure water under the same conditions." Further, the author, who wrote this subsection, should explain the expression of "The magnitudes of the induced changes" and inform which physical units are used for their measurement. Eventually, the paper deals with physics and physical chemistry, not with philosophy.

Comment 2: The author, who wrote this subsection, ignores the criticisms of WAC posed in paper by Bogdan and Molina (2010) (see reference in my previous comments), but cited the papers by Knopf and Alpert (2013) and Wex et al. (2014) who also ignored the criticisms of WAC. This fact casts serious doubt on the scientific qual-

ity and credibility of this subsection as well as the papers by Knopf and Alpert (2013) and Wex et al. (2014).

Comment 3: I do believe that taking into account the freeze-induced phase separation, which occurs during the nucleation and growth of ice in aqueous solutions, is necessary to mention in the paper (see my previous comments). The WAC-approach and/or lambda-approach do consider real physical and physical-chemistry processes which occur during freezing of atmospheric aqueous drops. It is very strange and unbelievable to me that the well known fact of the freeze-induced phase separation is stubbornly being ignored by atmospheric scientific community.

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