Atmos. Chem. Phys. Discuss., 14, C12473–C12477, 2015
www.atmos-chem-phys-discuss.net/14/C12473/2015/

Atmospheric Chemistry and Physics Discussions

ACPD

14, C12473–C12477, 2015

Interactive Comment

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

A. Bogdan

anatoli.bogdan@uibk.ac.at

Received and published: 28 February 2015

Comment for Introduction:

Authors write: The phase transformations of water are of great scientific and practical interest. However, many uncertainties exist, especially with respect to the initiation of ice from water vapor and from liquid water. The goal of this document is to facilitate further discoveries by proposing the use of a common terminology. Special consideration was given to the need to bridge the types of expressions and the language used in the physical and in the life sciences.

Comment/suggestion: I would suggest instead of "the physical and the life sciences" to write only 'the atmospheric sciences', because the discussion of paper and all interactive comments are focused on ice nucleation in the atmosphere.

I would also suggest instead of "liquid water" to write 'aqueous solutions'. In the atmosphere, as in general on the Earth, liquid water is not in the pure state, but a component of various aqueous solutions. Since ice is highly intolerant to impurities, the ice nucleation and subsequent freezing of atmospheric sub/micrometer-scaled drops results in the formation of not pure ice but mixed-phase particles: an ice core enveloped with a freeze-concentrated solution (FCS). This freeze-induced separation, which is of great importance not only for the atmospheric physics and chemistry but also for biotechnology, biopharmaceutics, food industry, life sciences etc., was described and shown in pictures and videos in our papers: 1. Bogdan, A. and Molina, M. J.,"Aqueous aerosol may build up an elevated upper tropospheric ice supersaturation and form mixed-phase particles after freezing." J. Phys. Chem. A, 114, 2821-2829 (2010); 2. Bogdan, A. and Molina, M. J. "Why does large relative humidity with respect to ice persist in cirrus ice clouds?" J. Phys. Chem. A, 113, 14123-14130 (2009); 3. Bogdan, A., M. J. Molina, K. Sassen, and M. Kulmala, "Formation of low-temperature cirrus from H2SO4/H2O aerosol droplets." J. Phys. Chem. A, 110, 12541-12542 (2006); 4. A. Bogdan, M. J. Molina, H. Tenhu, E. Bertel, N, Bogdan, T. Loerting, "Visualization of freezing process in situ upon cooling and warming of aqueous solutions." Scientific reports 4: 7414, DOI: 10.1038/srep07414, (2014). 5. A. Bogdan, M. J. Molina, M. Kulmala, H. Tenhu, T. Loerting. "Solution coating around ice particles of incipient cirrus clouds". PNAS, July 2, vol.110, no.27, E2439 (2013). 6. A. Bogdan, M. J. Molina, H. Tenhu,T. Loerting, "Multiple glass transitions and freezing events of aqueous citric acid", http://pubs.acs.org/doi/pdf/10.1021/jp510331h (2014).

Comments/suggestions for text:

p. C11893: The thermodynamically stable phase is defined by the existing vapor pressure and temperature, as usually depicted in a phase diagram.

Comment: This is true only for pure liquid/substance, including water. But in the case of aqueous solutions a third parameter, namely, composition should be considered. As I mentioned above, atmospheric drops does not consist only of pure water. In the

ACPD
14, C12473–C12477, 2015

upper troposphere and lower stratosphere, the weight fraction of solutes can reach as much as 30 wt% (some authors believe that organic mass fraction can be even larger than 50 wt%!).

pp. C11895 and C11900: 2.1.2 Homogeneous freezing nucleation and 3.5.2 Freezing nucleation

Comment/suggestion: I think the proposed by the authors expressions of “Homogeneous freezing nucleation”, “Freezing nucleation”, “Immersion-freezing”, “Contact-freezing”, and “Condensation-freezing” will be understandable only for scientists dealing with ice nucleation in the atmosphere. For scientists dealing with freezing in biopharmaceutics, biotechnology, tissue engineering, food industry they may be misleading. Therefore, I would suggest to use, for example, the expressions of Homogeneous ice nucleation, Heterogeneous ice nucleation, Immersion ice nucleation, and Contact ice nucleation. I suggest to give up the historical expression of “Condensation-freezing” because it bears much uncertainty, as the authors write themselves.

A reason why I suggest changing the above expressions is that the paper is focused only on ice nucleation, but not on freezing process. The ice nucleation event is only the very onset of freezing process which can easily be observed both upon cooling and warming (see, for example, supplementary videos in my paper: A. Bogdan, et al., “Visualization of freezing process in situ upon cooling and warming of aqueous solutions.” Scientific reports 4: 7414, DOI: 10.1038/srep07414, (2014). In this connection, I would also suggest to include some expressions which would reflect the most recent findings in freezing aqueous solutions. We after all are interested in how cloud frozen-drops/ice-crystals impact on atmospheric physics and chemistry, but not only in ice nucleation itself. I do believe that the expressions such as ‘Freeze-induced phase separation’, ‘Freeze-concentrated solution’, etc. should be included in text.

p. C11901: 3.6 FRACTION FROZEN and 3.7 FREEZING RATE.

Comment/suggestion: Similarly, these expressions can be misleading for scientists

ACPD

14, C12473–C12477, 2015

not dealing with ice nucleation in the atmosphere. I would suggest, for example, the expressions of Fraction of frozen drops and Freezing rate of drop ensemble. In the case of aqueous solution drops, a fraction of drops will remain liquid (FCS) or glassy at atmospheric temperatures. Freezing rate is a velocity of the propagation of ice/liquid interface. The fraction of frozen liquid and freezing rate can be seen in pictures and videos in our above mentioned papers.

As an example, I send you a picture (attached) which is a snapshot of ice/liquid interface propagating during the freezing of aqueous solution relevant for the atmosphere. In the picture, FCS is in between ice crystals composing a continuous ice framework (IF) and over IF/FCS body.

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

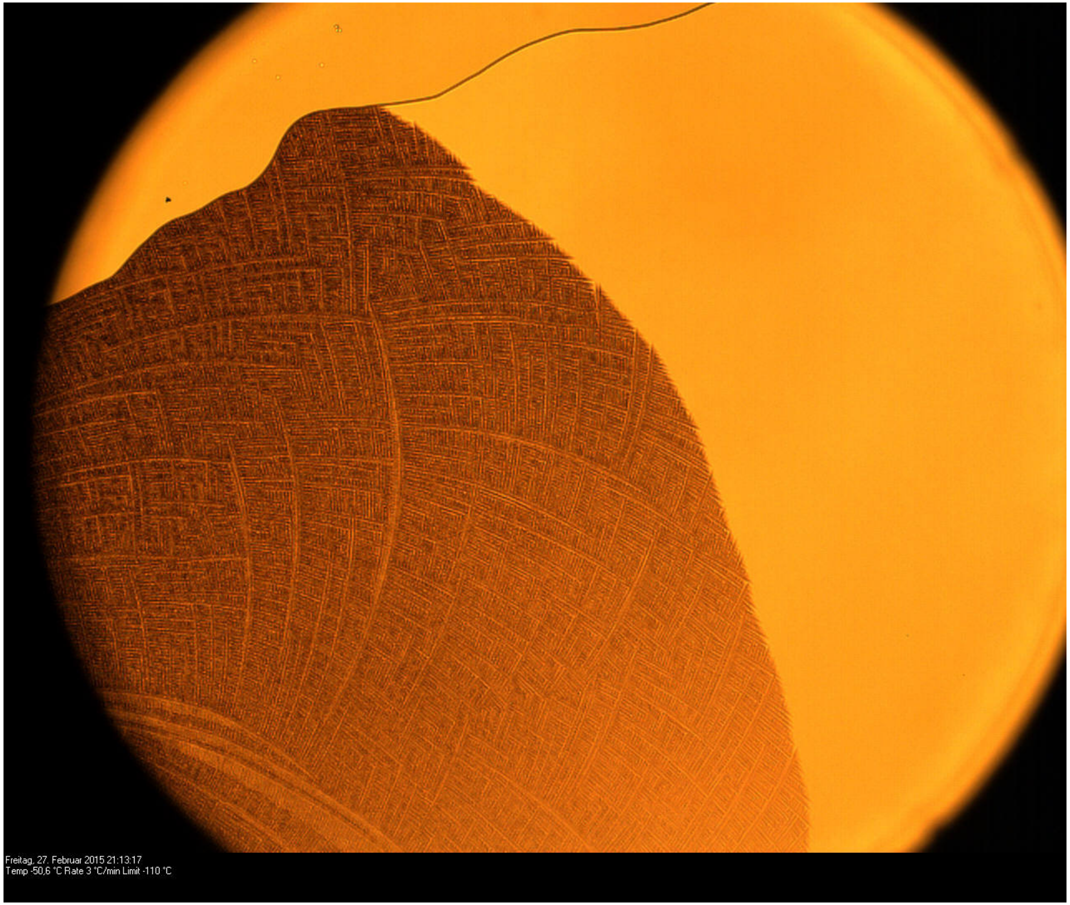

Fig. 1.

ACPD
14, C12473–C12477, 2015

Interactive Comment