Atmos. Chem. Phys. Discuss., 11, C395–C398, 2011
www.atmos-chem-phys-discuss.net/11/C395/2011/


Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Heterogeneous ice nucleation: bridging stochastic and singular freezing behavior" *by* D. Niedermeier et al.

**Anonymous Referee #1**

Received and published: 28 February 2011

General comments

This manuscript addresses a topic that is important for the treatment of heterogeneous ice nucleation, namely, whether this process should be described using a stochastic or a singular approach. The answer of the authors to this question is that ice nucleation is fundamentally a stochastic process but that for realistic atmospheric particle populations this process can be masked by the heterogeneity of surface properties. The authors present a "soccer ball" model that they use in idealized simulations to illustrate how the experimental conditions and the properties of the ice nuclei can lead to seemingly singular or stochastic behavior. This part of the paper is very convincing. However, it remains unclear what the principle difference and advantage of this model is compared with the modified singular description and the models developed by Marcolli et al. (2007) and Lüönd et al. (2010).

The “soccer ball” model and the model of Marcolli and Lüönd both assume that the surface of each particle can be divided into a number of surface sites with each site having well-defined properties. The nucleation rate on individual sites is described by Classical Nucleation Theory with contact angles that can vary between surface sites and consequently between particles, too. In both approaches, the contact angles are drawn from a contact angle distribution function. The main difference that I can find between the two approaches is, that the “soccer ball” model assumes that the whole surface of the particle is active as IN while the approaches by Marcolli and Lüönd limit IN activity to active sites. The authors should therefore more explicitly discuss the new features and the advantages of their “soccer ball” model. Having to divide the particle surface into an arbitrary number of patches with different surface properties might indeed become a disadvantage when the model is applied to polydisperse aerosols.

Moreover, in this paper the model is just used to exemplify how the time and temperature dependence of the nucleated fraction depends on the standard deviation of the error function and the number of different surface sites on each particle. The discussion of the datasets by Shaw et al. (2005) and Niedermeier et al. (2010) is only qualitative. The authors did not attempt a quantitative fitting of the results because the system is under-determined. Nevertheless, a more rigorous discussion of literature data would add to the quality and value of the paper. Immersion freezing of ATD has been measured by several groups including Niedermeier et al. (2010), Marcolli et al. (2007), and Connolly et al. (2009). A thorough discussion of at least this IN should be attempted. In principle, all ATD immersion freezing data from different studies should be describable by the same contact angle distribution function. The authors should try to formulate such a function for their “soccer ball” model. It would also be interesting to see whether the contact angle distribution function of Marcolli et al. (2007) can be applied to the Niedermeier et al. (2010) data.

Specific comments



ACPD

11, C395–C398, 2011

Interactive Comment

Page 3164, lines 16-18: Here the authors claim that the experiments are sufficiently controlled so as to allow interpretation with a simple model. However, I could not find an interpretation of the results of these experiments in the manuscript that makes real use of the model.

Page 3164, lines 24-25: Why can the modified singular hypothesis not explain these results? One active site on a particle that is described by Classical Nucleation Theory with a specific contact angle would lead to the observed fluctuations in temperature.

The nomenclature in equations (1) and (2) has to be improved: In equation (1), P is a function of T and the contact angle, in equation (2), however, only a function of T. Obviously, P is also a function of t. The frozen fraction should also be a function of T and t. The meaning of N0 is not explicitly stated.

Page 3170, lines 19 – 21: The authors assume that the curves become steeper with increasing number of sites because the stochastic behavior is recovered. However, the curves become also steeper because the contact angles of the best sites are becoming smaller and more similar with increasing number of sites. This explains also the shift of freezing temperature to higher values. It would be interesting to investigate how many of the best sites are responsible for freezing. This could be done by shutting off (1) all but the best site (2) all but the two best sites(3) all but the three best sites and so on, and then compare the frozen fraction for the different cases.

Page 3171, lines 14-15: where in Niedermeier et al. (2010) is the missing time dependence for freezing of ATD shown? This missing time dependence should be shown and discussed in this paper in more detail.

Page 3171, lines 23-27: It would be interesting to see how different combinations of the fit parameters in the “soccer ball” model that are all able to describe the frozen fraction of the ATD particles in Niedermeier et al. (2010) influence the time dependencies of the frozen fraction. The authors should present such calculations in this manuscript.



ACPD

11, C395–C398, 2011

Interactive Comment

Figure 4: the y-axis in this Figure could be chosen better. Ln(Nu/N0) = -3 corresponds to an unfrozen fraction of 0.05. In the atmosphere and in the lab, such low unfrozen fractions are difficult to measure and are not very interesting. However, small frozen fractions are especially important for mixed phase clouds. This region of the Figure should be enlarged, especially in the panels (c) and (d).

---

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3161, 2011.