Atmos. Chem. Phys. Discuss., 11, C1170–C1173, 2011
www.atmos-chem-phys-discuss.net/11/C1170/2011/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #3

Received and published: 24 March 2011

Niedermeier et al. present a conceptual semi-quantitative model that attempts to explain observations of the ice nucleation ability of an aerosol made by two different techniques. This model works on the premise of dividing the surface into a number of equal-area sites, each being parameterized having a fixed and randomly chosen contact angle. Classical nucleation theory is used to compute the fraction of particles that is frozen as a function of the thermodynamic state and time.

The topic of the paper is appropriate for Atmospheric Chemistry and Physics and of general interest to the Atmospheric Science community. However, I find that the manuscript is not sufficiently worked out to unequivocally endorse it for publication at this time. Specifically, I find that the authors have not taken the time and effort to really test their ideas but that they want to publish some untested ideas that will be elaborated

upon in future papers. For example, the authors write "It is a separate question whether such an ensemble view reasonably captures the features of natural aerosol systems, and we leave detailed evaluation of that question for future work." As I argue below, I disagree that this is a separate question since the ideas inherent in the conceptual model are not really novel.

As succinctly outlined by the comments by Gabor Vali, the model presented here is not really new. The model is simply an implementation of ideas that pervaded the ice nucleation literature for the last six decades. Clearly heterogeneous ice nucleation cannot be completely stochastic since that would obviate the need of an ice nucleus. Equally clearly, ice nucleation is not completely deterministic as experiments of repeated drop freezing have demonstrated. The stochastic vs. singular debate is therefore more like the nature vs. nurture debate; both are important and their relative importance depends on the actual case considered. No convincing experiment or data is presented that sheds light on the actual mechanism of the nucleation process for ATD or aerosol samples in general. The manuscript simply posits a model and computes the implications without attempting to constrain the model with data. For this reason, the claim that this paper "bridges stochastic and singular behavior" is incorrect. The model simply highlights what behavior follows from what assumption.

This in itself does not merit rejection of the manuscript. It implies however, that the authors need to validate their model and present data and/or calculations that go beyond a mathematical implementation of a conceptual description of the ice nucleation process. For example, can the model be used to fit meaningful parameters to actual data? If this is done, is it consistent with all data collected to date? Do these parameters then make specific predictions that can be tested in further laboratory experiments or observational studies to verify or falsify the model? In my opinion the authors need to very explicitly point out how the model can be applied.

A second major criticism of the manuscript is the cherry picking of data. The authors qualitatively contrast studies by Niedermeier and Shaw (two of the authors), ignoring a vast body of ice nucleation literature on the same aerosol type. To have merit, a model/theory must be quantitatively consistent with all of the data, or it must be argued why some of the data are flawed or why it cannot be considered by the model. Presumably there is enough diversity in the techniques to provide a span in nucleation time scales, specific aerosol surface area used and other important parameters to test if the model can be used to parameterize data and/or quantitatively explain the range of observations as the authors set out to do.

To achieve such a description it is necessary to carefully evaluate the technique by which the ice nucleation activity was observed. There is significant uncertainty regarding the measured IN activity by different instruments and techniques; see special issue on the ice nucleation workshop in ACP (http://www.atmos-chem-phys.org/special_issue139.html which also contains more ATD data for the authors to consider). Thus before the data presented here can be used to support theoretical claims, a self-consistent quality-controlled and validated dataset must be put together by the authors. Uncertainties must be included and potential biases must be discussed for all of the techniques.

In summary, factors speaking in favor for publication of this paper are: (1) that it is written clearly, (2) that the points made are readily understood, (3) that the calculations clearly show the consequence of the conceptual assumptions, (4) that the work is technically sound, and (5) that the results are novel in the sense that the results shown in Figures 3-5 have not been discussed in the literature. Factors speaking against publications of the paper are: (1) that the model is highly speculative and it is unclear how to apply it, (2) that the conceptual underpinning is not novel, (3) that the model is unconstrained by data, (4) that the dataset used to qualitatively discuss the merits of the model is handpicked and limited, and (5) that the uncertainties in the data are ignored. To this referee the factors against outweigh the factors for publication. However, a significantly revised manuscript that mitigates these criticisms could be reconsidered for publication in ACP.

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