Atmos. Chem. Phys. Discuss., 3, S1374–S1376, 2003
www.atmos-chem-phys.org/acpd/3/S1374/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Comment on evidence for surface-initiated homogenous nucleation" *by* J. E. Kay et al.

J. E. Kay et al.

Received and published: 24 August 2003

We thank AT for her comment. We address her concerns below:

1) **Thermodynamic criterion for surface-initiated nucleation (section 2)**
We use the same thermodynamic criterion for surface-initiated nucleation as Tabazadeh et al. (2002a, 2002b): the imperfect wetting criterion (see section 2.2). This criterion takes into account the free energy difference between the initial and the final state of the system. It also incorporates the free energy gain from losing the water-air interface. We note that this criterion is discussed throughout the literature (e.g., Introduction to Wetting Phenomena (N. Holland) 1990, Ch. 2, many theoretical papers in journals such as the Physical Review, the Journal of Low Temperature Physics, and applied papers on nucleation in material sciences journals). In our opinion, the Djikaev et al. (2002) discussion of the imperfect wetting criterion does not add any new un-

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derstanding to this body of literature. Thus, we cannot understand the concerns raised paragraphs 1 and 2 of AT's comment.

In our comment, we discuss limitations of the imperfect wetting criterion and the data available to evaluate it. We conclude that it is impossible to evaluate this criterion at -40 °C with current data. Even if it the imperfect wetting criterion favored surface nucleation on one facet as suggested by AT, a large free energy difference is required to offset the smaller attack frequency (see section 2.1). In addition, any individual facet would be created so far from equilibrium that the surface energy should strongly depend on the individual conditions for every embryo. Regardless, it is hard for us to imagine that facets could be defined in bodies as small as ice embryos. Given the above, we still conclude there is no demonstrated thermodynamic advantage for surface-initiated nucleation over volume-initiated nucleation.

2) **Atmospheric relevance of experiments completed in oil (section 3)**
We clearly state a main conclusion from the PNAS paper (Tabazadeh, 2002a) in section 3: "The fact that the measured nucleation rates varied by orders of magnitude when the ambient phases were changed was used to provide support for nucleation occurring at the ambient-water interface."

While the Tabazadeh et al. (2002a, 2002b) observations are interesting; we must await further laboratory experiments and analysis to understand the role of surface-initiated nucleation for an air-water interface. The cited laboratory data were not measured in an air ambient and thus, are not directly relevant for our atmosphere. Surfactants and oils may interfere with the nucleation process and their role in changing nucleation rates is unknown.

3) **Cahn and Elbaum**
As we have explained, Cahn (1977) and Elbaum et al. (1993) are not relevant for evaluating the imperfect wetting criterion at -40 °C.

4) **Basis for comment**

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We wrote this comment to encourage further discussion about the thermodynamics and atmospheric relevance of surface-initiated nucleation. Indeed, we submitted to ACPD/ACP because we wanted to promote interactive discussion and an open review process. We believe that we bring new perspective and ideas to the discussion of surface-initiated nucleation (and the referees agree).

5) **Typographic errors**
We have corrected the spelling of 'homogeneous' and other typographic errors.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3361, 2003.