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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the transition between heterogeneous and homogeneous freezing" *by* K. Gierens

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This paper presents a theoretical study of the relative importance of homogeneous and heterogeneous nucleation for cirrus formation in uplifting air parcels. The author presents interesting modelling results and provides analytical expressions for parametrising the competition between the 2 modes of cirrus formation. I recommend publication with minor comments that the author may wish to consider.

1. The model chosen to describe the number of active sites per unit surface of soot and its dependency on temperature, the parametrisation of DeMott et al. (1997), should be mentioned explicitely in the abstract. Some of the results are likely to be model-dependent.

2. The differences in the levels of pollution might be the most plausible explanation for the hemispheric differences in the thermodynamical control derived from the com-

parisons of SAGE data and ECMWF meteorological analysis [Gierens et al., 2000]. However, other factors could be invoked. The hemispheric differences may also partly reflect differences in the variability of the updraft speed between the tropics, northern and southern hemisphere. Even if the updraft speeds are comparable in both hemispheres, the important factor for a threshlod process such as nucleation is the frequency and magnitude of the small-scale fluctuations. More fluctuations in the northern hemisphere (possibly from the strong orographic activity) would lead to more frequent critical supersaturation, and more homogeneous nucleation events. This is supported by the stronger temperature sugrid (with respect to a 300x300km square) fluctuations seen in the MOZAIC data in the northern hemisphere compared to the tropics [Gierens et al., 1997]. Note that this issue of variability is also important for parametrising nucleation in large-scale models whatever the model chosen to describe the switch between the 2 nucleation modes.

3. In the introduction, the author argues pretty convincingly that our lack of understanding of heterogeneus nucleation makes it extremely difficult to be quantitative. For example, he states that "it must be said that an understanding of the very physical process that induces the freezing at or around the soot particles is still lacking" or "one must admit that it is not possible or justified at the current state of knowledge to define a critical supersaturation at which heterogeneous freezing of soot would commence". Nonetheless, the conclusions are quantitative: critical concentrations of heterogeneous ice nuclei, updraft speed thresholds and so on. I would suggest to highlight the caveats, limititations and uncertainties of the results.

4. page 2348 (paragraph 2): The specific soot surface appears to lie at the lower range of experimental results (60 to 400 m2/g). Interestingly, the author does not find that the number of active sites (and, hence, specific soot surface) is a critical factor for heterogeneous nucleation. It is a bit counter-intuitive. I would suggest to mention this result again in the conclusion.

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