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Interactive comment on "The role of organic aerosols in homogeneous ice formation" *by* B. Kärcher and T. Koop

Anonymous Referee #2

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General comment: This paper proposes causes for the limiting effects of certain organic containing particles on homogeneous freezing. If one has followed the profusion of literature on modeling homogeneous freezing nucleation for solution droplets under cirrus conditions in recent years, the quantitative impacts of variations in mass fraction/water activity relations and mass accommodation coefficient on the process are intuitively apparent. This well written paper puts a quantitative face on these interactions. The paper is useful in this regard, but can and should be validated by some appropriate laboratory studies. There are a few issues that deserve clarification and justification. For example, it is not clear what the basis was for the chosen size distribution of particles. I note that this choice effectively removes consideration of surface

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tension impacts. I also find odd the statement about the lack of sufficient observations being a "problem". It is only a "problem" when modeling capabilities forge ahead uninhibited in comparison to the state of knowledge of the compositions of atmospheric aerosols and the capabilities for validating their predicted behaviors. I would choose to say that the numerical studies indicate a need for more specific data. Finally, some editorial remarks are presented in the last few pages of the manuscript that seem inappropriate for a scientific article. One editorial remark not there is that the framework designed for the role of organic aerosols does not consider phase state issues that could be very important.

Specific comments:

1. Abstract: The selection of malonic acid as the organic is motivated entirely by the availability of critical data. It should be noted that this dicarboxylic acid is probably not the most representative for such compounds in the atmosphere.

2. Microphysical model: No comment is made with regard to the potential impact of organic composition on solution surface tension. It becomes somewhat clear only later that the impacts of small particles for which surface tension greatly matters are not considered or are somehow judged not to be important for expected typical aerosol size distributions. It seems worth noting that especially in the case SUL/MAL, lowered accommodation coefficient leads to a situation in which surface tension effects could be relatively more important.

3. Section 3.3: The effect of deposition coefficients below 0.1 on homogeneous freezing has been elucidated in a host of studies dating back over many years. Some numerical investigations, such as those by Gierens et al. (2003) have this as their primary topic. Some acknowledgment should probably be given to the fact that the effects of deposition coefficient have been explored previously, if not specifically for an assumption of the impact of organic aerosols.

4. Section 4. A few of the speculations given as alternate explanations for observed

high ice saturation ratios seem dubious at best and advertisements at worst. Is it likely that the action of small-scale temperature oscillations would lead to elevated ice saturation ratios over a widespread region such as those demonstrated in Jensen et al. (2004)? The second item identified is described as differences between vapor pressure parameterizations, although I believe that the authors are actually pointing out the uncertainty in the knowledge of ice and water vapor pressures at low temperatures and how this affects the calculation of saturation ratio. Surely it is possible to determine what relationships were used to estimate ice saturation ratio in a given study of interest and then calculate if uncertainties are large enough to explain the difference between the "observed value" and the value deemed to be required for homogeneous freezing.

5. Conclusions:

a. Partial crystallization and delayed deliquescence are alluded to as possibly changing the freezing mode to heterogeneous ice nucleation, but this is not a given nor the only possible effect of such phase changes on ice formation. For example, a requirement for deliquescence in particles that contain a high organic fraction can place a barrier to water uptake and homogeneous freezing that simply would not otherwise exist for solution droplets.

b. I suggest removing the sentence stating that "we expect the processes outlined here to actually occur in the atmosphere." One would always hope so!

c. This work may provide a framework for future modeling studies, but I would again contend that it is not a comprehensive one if it ignores phase states. Of the remarks made in the last paragraph, the plea for more experimental data rings most true.

Editorial notes: Page 6727, line 18: from spelled "form".

Gierens, K.M., M. Monier, and J.-F. Gayet, The deposition coefficient and its role for cirrus clouds. J. Geophys. Res., 108, d.o.i.:10/1029/2001JD001558, 2004.

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Interactive comment on Atmos. Chem. Phys. Discuss., 4, 6719, 2004.