

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

**Anonymous Referee #2**

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This 'Comment' addresses issues raised in papers published earlier in ACP concerning the mechanism of homogeneous freezing in aerosol particles and supercooled water droplets. Does the freezing occur at the surface or in the bulk? A focal point is earlier work of Tabazadeh and co-workers which, by analysis of some laboratory studies, has concluded that there is considerable evidence that freezing rates scale better when referenced to particle surface areas rather than volumes, thus indicative of surface-initiated freezing. The 'Comment' is well-written and makes some valuable points, the main one being that not enough is known to conclusively say that freezing of these droplets in air occurs in either the bulk or at the surface. In particular, they point out that bulk properties such as surface tensions lose their meaning at the molecular level, that water may fully wet ice at the freezing point and that many of the laboratory studies considered by Tabazadeh et al. were conducted in oil, i.e. equivalent studies in

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air could significantly affect the freezing mechanism. Plus, they point out that it is very difficult, if not impossible, to accurately infer freezing temperatures from field measurements. Thus, I consider this a valuable perspective to add to the earlier, interesting suggestion of Tabazadeh et al.

Specific points:

1. I am confused in Section 2.1 by the deconvolution of a rate (constant) into a pre-exponential and a free energy term. Usually, the pre-exponential is associated with all entropic factors and the exponential term with energetic factors. Free energy refers to both entropy and energy, and so I believe that the use of "Free Energy" in this perspective is inaccurate.

2. I believe the authors are somewhat misleading in their statement at the end of Section 2.1 that "We are unaware of any convincing evidence for such a large free energy difference". There is plenty of evidence that the chemical composition of a surface can be dramatically different than that of the bulk of a liquid.

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

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