

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

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General comments: This paper comments on the strength of evidence for surface-initiated versus volumetric homogeneous freezing nucleation. A new point introduced in this paper is the thermodynamic argument against the likely existence of surface-initiated nucleation, at least as currently hypothesized by Tabazadeh and colleagues. While the comment also evaluates laboratory and field evidence for the mechanism of homogeneous ice nucleation, much of this information is apparent from existing literature or was either discussed or acknowledged in the original articles on surface versus volume nucleation rates. The material reiterates and supports the need for more critical data over a range of drop sizes in air that was mentioned by Tabazadeh and colleagues. Nevertheless, I do believe that it is important that scientists active in the field register their input on the concept of surface-initiated nucleation and this paper

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is certainly a valuable contribution. More study is definitely needed before relevance to the full range of atmospheric ice formation processes can be confirmed or denied. I believe that this comment would be most effective if made more succinct and focused on the theoretical and thermodynamic questions that it raises. I list a few additional specific comments.

Specific Comments:

Last paragraph of section 3: The experiments of DeMott (please note slight misspelling in manuscript) and Rogers (1990) actually examined a polydisperse population of drops, although it is probably an accurate statement to say they could be approximated as nearly monodisperse at the point of freezing.

Second paragraph of Section 4: These comparisons to atmospheric observations, as well as the ones by Tabazadeh and colleagues, are somewhat unsatisfying in light of the very accurate statements made by the authors in the last sentence of this paragraph. The temperature of glaciation in the atmosphere is often kinetically controlled by the combination of freezing rates, droplet size, updraft and ice mass growth. I think that this comment needs to be brought forward in the discussion, so that it is clear to readers before the comparisons are shown. Difficulty detecting small ice crystals adds to the uncertainty of freezing temperature. I also note that the updrafts in the cases described by Sassen and Dodd (1988) or Heymsfield and Milosevich (1993; 1995) are probably an order of magnitude less than in the Rosenfeld and Woodley (2000) case. It would be possible for someone to more carefully consider these cases in the micro-physical context of a cloud parcel and instrumental detection limitations for ice crystals. There also exists a growing body of literature on the freezing of haze droplets in a variety of experimental devices, conclusively documenting the atmospheric phenomenon noted by Heymsfield and Milosevich (1993) that is mentioned here, including recent realistic cloud simulation experiments (Möhler et al. 2003) and numerical modeling studies of these (Haag et al. 2003). While constraint of solution drop composition is the most critical issue and other issues exist in computing nucleation rates from the lab-

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oratory studies, it should be noted that the opportunity exists for validation of surface versus volumetric nucleation theories via existing and new laboratory studies.

Table 1: There appears some inconsistency between the temperature noted for observation 2 and what is plotted in Figure 2.

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