Additional comment on "Using critical area analysis to deconvolute internal and external particle variability in heterogeneous ice nucleation" by Hassan Baydoun and Ryan C. Sullivan.

Since much of the material presented in the paper depends on it, the meaning of the critical contact angles and of the critical area needs close scrutiny. These terms are defined on pages 11 and 13 of the paper.

The notion underlying these definitions is that the range of activity for any given substance has upper and lower limits other than the melting point and the homogeneous nucleation threshold. These limits are expressed as the smallest and largest contact angles possible (θ_{c1} and θ_{c2}) on the given material. Contact angle is used as a convenient parameter to quantify activity in terms of CNT. The lower end of the range of activities, established by θ_{c2} is less interesting as it corresponds to a high number of possible occurrences, while values near θ_{c1} correspond to rare cases of high activity (freezing temperatures). If this interpretation is correct, the critical area can be stated with Eq. 11, replacing in it *g* by *g*-bar.

The method followed in the paper for determining *g*-bar and the critical contact angles θ_{c1} and θ_{c2} appears to consist of fitting Eq. 15 to he *F(T)* curve for the highest particle loading in Fig. 5. This is unclear in the paper as the integration limits in Eq. 15 are given as 0 to π. It would be useful to have the authors' clarification on this.

The plausibility of the concept of limiting values for θ_{c1} and θ_{c2} can be examined by looking at evidence in terms of spectra of INP concentrations either in terms of $n_s(T)$ or $K(T_c)$[1]. As far as I am aware of, no cases have been reported in the literature with sharp cutoffs in these quantities at either high or low activity values. The corresponding spectra may have steep slopes, but all have monotonic rise (with finite slopes) from the lowest temperatures detectable in given experiments to the maximum concentration values measured. The shape of the *F(T)* curve, or the temperature range it covers is related closely to a segment of the $n_s(T)$ or $K(T_c)$ spectra and a shift of the *F(T)* curve along the temperature axis due to a change in sample volume is indication of the slope of the spectrum remaining constant the temperature interval covered. From the wide variety of spectra reported in the literature, it appears that assuming the existence of limiting values in activity is not justified. Of course, empirical data are subject to sample size and instrumentation limitations. Nonetheless, that is not the explanation given by the authors, so they should explain what a priori reasons they see for upper and lower limits of the contact angle, or of other measures of activity. Specific questions about how the assumption of critical area is supported in the paper, and about how it is used to interpret experiments, are raised in my first set of comments.

In case objections are raised about using $n_s(T)$ or $K(T_c)$ for making the point in the preceding paragraph, it is important to recognize that over the relatively narrow temperature interval involved in the experiments being analyzed, the nucleation rate function $J(\theta)$ does not vary much in shape. Hence the dominant variations in the integral comes from $g(\theta)$ and that quantity is a measure of the frequency of different sites just as $n_s(T)$ or $K(T_c)$ are. Also, such time-independent descriptions are adequate for examining questions like the existence of cutoff values in nucleating ability.

From a practical perspective, there is likely to be a limit to how much material can be suspended in water for nucleation studies, so there is going to be a limit in the highest nucleation temperatures that can de detected in an experiment. However, going to a rather extreme example, it is a common observation that small puddles on soil have ice form on them when the temperature drops ever so little below 0°C. That the temperature didn't drop much below 0°C can be surmised from the fact that there is liquid water below the ice. While this situation is, clearly, a large jump from the laboratory experiments, and it surely involves many different types of INPs, the notion that no upper limit to heterogeneous nucleation exists other than the melting point is perhaps validly illustrated by it. The chance of encountering INP activity in any system decreases rapidly as the temperature approaches 0°C but the decrease is likely to be gradual, not abrupt. Random embryo formation of course also contributes to that fact.

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1 See Vali et al. 2015 (Atmos. Chem. Phys., 15, 10263–10270) for the definitions of the symbols