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Interactive comment on "Classical nucleation theory of immersion freezing: Sensitivity of contact angle schemes to thermodynamic and kinetic parameters" by L. Ickes et al.

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General Comments

The manuscript by Ickes et al. details the ability of different immersion freezing parameterizations based on classical nucleation theory to reproduce laboratory data. The authors target the treatment of the contact angle, α, the interfacial tension between an ice germ and water, σ_{iw}, and the activation energy barrier, $\Delta g^{\#}$. Performance of freezing parameterizations are evaluated in terms of their accuracy in reproducing measured frozen droplet fractions (criteria 1), their consistency with trends in freezing as a function of temperature, T, particle size, and time, t, (criteria 2) and finally if their fit parameters and fitting functions lead to reasonable representations (criteria 3). Comparison of formulations 1), 5) and 7) use the same thermodynamic parameterizations for σ_{iw} (Reinhardt and Doye, 2013) and $\Delta g^{\#}$ (Zobrist et al., 2007), but α is represented by different schemes namely the single-α in 1), the α-PDF in 5) and the $\alpha(T)$ in 7).

The manuscript in question is well written and comprehensive. The overall conclusions are justified. However, a discussion of previous literature that pertains to the performance of freezing parameterizations is lacking and alternative explanations of freezing parameterization should be explored for completeness. The authors cite Marcolli et al. (2007) and Lüönd et al. (2010) on p. 4, l. 17, however, significant advancement in comparing contact angle schemes should also be discussed and attributed to Zobrist et al. (2007), Alpert et al. (2011), Knopf and Forrester (2011), Rigg et al. (2013) and Wheeler et al. (2014). I feel that a concise discussion of the major findings in these previous studies and how the current study in question advances these findings should be presented.

– In Zobrist et al. (2007), a single-α scheme was evaluated for performance, but it could not reproduce the freezing of droplets due to 1-nonadecanol, an organic monolayer coating. Allowing α to be a function of T, i.e. the $\alpha(T)$ scheme, resulted in a good representation of their data (Zobrist et al., 2007). However, here the authors do not mention that the single-α or $\alpha(T)$

scheme was considered in Zobrist et al. (2007). In our recent publication (Alpert and Knopf, 2016), we address the question of the applicability of the single-α scheme to an uneven mineral dust surface, if it does not apply to uniform surface on a molecular level such as a self-assembled organic monolayer coating. In other words, a single-α was never shown to reproduce freezing data for a highly ordered and uniform surface. It has also been argued that α represents the balance of surface tensions which can change as T changes. Thus a single-α scheme is not expected to be physically applicable (Welti et al., 2012; Rigg et al., 2013).

– In the studies by Alpert et al. (2011), Knopf and Forrester (2011) and Rigg et al. (2013) investigating different ice nucleating particle (INP) types, it was shown that the $\alpha(T)$ scheme can be applied and that a linear function may be used. This corroborates findings by Zobrist et al. (2007). However, the authors neglect discussion of these studies and refer only to Welti et al. (2012).

– The authors claim that parameterizations should be consistent with freezing trends of known microphysical processes (critera 3). One easily accessible microphysical process that can be discussed for evaluating freezing parameterizations is the ability to reproduce freezing point depression of aqueous solution droplets. Rigg et al. (2013) evaluated the applicability of the α-PDF and $\alpha(T)$ scheme to describe droplet freezing experiments. In their study, organic particles were immersed in pure water or aqueous ammonium sulfate solution droplets and freezing was observed as a function of T and water activity, a_{w}. The analysis demonstarted that one α-PDF distribution could not reproduce observed freezing data, but the data could be well represented allowing α to be a function of T and a_{w} Rigg et al. (2013). The α-PDF scheme failed as a physical representation of the ice nucleating ability of the particles. If an α-PDF scheme is chosen to be used to describe immersion freezing in future studies, it should be modified to account for changes in a_{w} (Rigg et al., 2013). I suggest to include this discussion adding more detail to the authors' third criteria, how our current knowledge of microphysics can lead to more correct ice nucleation parameterizations and not only just better performance.

– Wheeler et al. (2014) evaluated many different α schemes and found that the α-PDF is not the best performing. Instead, a scheme known as the "active site scheme" (AS) is the best performing. This finding should be discussed in the manuscript on p. 5, l. 28-p. 6, l. 2.

Specific Comments

When introducing the α schemes, there are a few instances where the author claims certainty or implies that INP surfaces have variable ice nucleation efficiency. There is no physical evidence that an INP has variable ice active sites or surfaces with different ice nucleation efficiency. For now, any evidence is a product of circumstance to a conceptual mathematical framework (or fitting procedure with prior assumptions of the existence of active sites). On p. 5, l. 2-3, the authors claim that the single α scheme "assumes" that the surface has one α values for the entire particle surface. However, on p. 5, l. 10, the authors claim that the α-PDF "accounts" for surface heterogeneity. This later statement is incorrect. The α-PDF does not account for anything, but it does *assume* that the surface of particles is covered by sites that have different contact angles. In lack of in situ observations, this is not a better or more accurate assumption, but simply a different conceptual framework. The authors should state the assumptions of all schemes accurately just as they did for the single α scheme.

On p. 5, l. 4-6, the authors claim that the $\alpha(T)$ scheme does not take into account how contact angles are distributed. Then say on l. 6-7 that good IN freeze first, e.g. when performing a cooling rate experiment, which shifts the mean contact angle of the remaining droplet population. These statements are contradictory as it is written. The authors say that the first scheme does not distribute contact angles, but the contact angle distribution shifts? Again, there is no certainty that a contact angle distribution exists in the first place. Rather it is sufficient to say that the $\alpha(T)$ scheme assumes a physical dependence of α on T. To describe this scheme as a compromise is also incorrect as it is different than the single α and α-PDF schemes. It it is based on different assumptions and includes a physical dependence of thermodynamic parameters on α which is neglected in the other two schemes. Similarly, it also does not "reflect a changing α-PDF distribution" (l. 5-6).

The authors emphasize computational efficiency, cost, expense, complexity...as a way of evaluating each scheme or parameterization. Since this is presented as a sort of metric for comparison, it should be a quantifiable metric. As it is presented by the authors here, it is not quantitative. How much time does it take for a computer to calculate σ_{iw} derived by the different parameterizations presented here? What is the extra time it takes to randomly sample from an α-PDF or calculate $\alpha(T)$ before freezing is predicted in a GCM? Understandably, the time it takes to fit various α schemes and other parameters is very different and may take hours is some cases. After finding all parameters and using them in formulations 1-7, how long does each take to predict ice nucleation in GCM's for the same aerosol population and thermodynamic conditions? If the authors choose to not consider the active site scheme and the soccer ball model, they must have some reason and quantitative evidence as to why. For example, p. 5 l. 31-p. 6 l. 32 claims that a scheme is too computationally expensive to be considered, but no quantitative measure is given. In order for this statement and all others like it to remain in the manuscript, the authors must provide quantitative evidence for this. Is it possible to add another column in Table 2 for this purpose?

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