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We thank both anonymous reviewers and P. A. Alpert for their positive review and the detailed comments on the manuscript. We have revised the manuscript accordingly (see track-changes in the manuscript). Our replies to your comments are given below in blue after the specific comment.

1 Interactive comment 1 by Peter A. Alpert

1.1 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 α(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.

Thanks for that comment. We added a section on previous literature to point out how this study conplements the earlier findings. The aim of the current manuscript is to evaluate how suitable four different CNT based schemes are for implementation in a GCM. A broad review on ice nucleation parameterizations is not within the scope of the current work.

– 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 α(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 α(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).

Differently to Zobrist et al. 2007, the schemes are evaluated to reproduce ice nucleation measurements on solid surfaces of mineral dust particles. As stated in Zobrist et al. 2007: "The experimentally determined heterogeneous ice nucleation rate coefficient (of a nondecanol monolayer) shows a much weaker temperature dependence than homogeneous ice nucleation and heterogeneous freezing in the presence of a solid ice nucleus such as Al_2O_3". Early experiments on AgI and MD simulations e.g. Cabriolu and Li (2015) support the applicability of single-alpha in certain cases. We added the references mentioned above and some further sentences about the single-α scheme in the manuscript.

– 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 α(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 change in contact angle with RH_{i} and temperature can be interpreted either as a result of the temperature dependence of the interfacial tensions (σ_{is} and σ_{iw}, where the index i stands for ice, w for water and s for the aerosol surface) or as the apparent contact angle of an ensemble with a diversity of contact angles from particle to particle. In contrast to the mentioned references we follow the second interpretation as was done in Welti et al. (2012). Therefore we only referenced this paper. We added some further explanation to the manuscript and added the references.

– 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 α(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.

Thanks, this is added in the discussion.

- 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.

  This is added in the section refering to other studies from literature.

1.2 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.

You are right, all asumptions should be stated. We changed this accordingly.

On p. 5, l. 4-6, the authors claim that the α(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 α(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).

We rephrased the explanation of the α(T) scheme used and hope that the additional explanation leads to less confusion. In our case the α(T) scheme is a simplified temperature dependent α-PDF scheme and not a scheme based on the physical dependence of α on T. Accounting for a physical dependence of α on T as a result of the temperature dependence of the interfacial tensions leads to a decrease of α, which is contradictory to the assumption we made here.

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 α(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?

We agree that a quantification of computational efficiency would be a nice additional information to know. However, that is not easy to derive and was therefore not mentioned explicitily. The computational time for certain freezing schemes depend on many aspects, amongst others the GCM used (treatment of aerosol particles, microphysics etc.) and can therefore not be generalised. A quantitative number in Table 2 would be meaningless.

There are some general thoughts, which can be made to assess the computational complexity of schemes before implementing them, which is the number and kind of variables neededed for the scheme. In that sense an α(T) scheme is not different from a single-α scheme (same number of variables, the same number of equation have to be solved). The α-PDF scheme on the other hand is therefore more difficult as it requires an extratracer (variable, which is stored in the model longer than for one timestep) if it is implemented physically correctly. That is because the model has to memorize which contact angles from the distribution were already used in the timestep before. Otherwise the same "good" IN would be used over and over again. Using extratracer for the contact angle of mineral dust particles would approximately lead to an increase of computational costs of 21% in the GCM ECHAM6-HAM2. The depletion of contact angles can be ignored if one assumes that the aerosol particles are replenished within one time step, so that there are always the same contact angles available (if aerosol particles are available). If the time evolution of the contact angle distribution is not taken into account, the α-pdf scheme becomes computationally similarly expensive as the single-α scheme. However, the integral of the contact angle distribution can not be solved analytically. Therefore, to minimize computational costs, a look-up table could be used instead of discretized finite sums. Using look-up tables is depending on the size and format of the look-up table more expensive compared to solving an equation with simple constants as in the case of the single-α scheme. In the case of the soccer ball model it might be extensive work to create look-up tables. The computational costs are higher for the active sites scheme. It requires a memory of used contact angles in dependence of time and therefore at least one tracer variable. In many GCMs with explicit microphysics and aerosol dynamics, tracers are one of the major sources for computational time, which makes the code costly compared to other GCMs.

We added this discussion in the revised manuscript.

1.3 References

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