Review of

Modelling the effect of condensed-phase diffusion on the homogeneous nucleation of ice in supercooled water

by Fowler et al.

General:
This study simulates homogeneous ice nucleation by taking into account condensed phase diffusion through individual ultra viscous aerosol particles by means of a new cloud parcel model with bin microphysics. The major findings of the study are that homogeneous ice nucleation is inhibited below 200K due to restricted particle growth and low water volume, while at higher temperatures between 200K and 220K the number of frozen aerosol particles increases because the water molecules are slightly more mobile and a layer of water condenses on the outside of the particle. The topic of the study is interesting and timely, because the representation of especially ice clouds in climate models needs to be improved in order to reduce major uncertainties in the prediction of the future climate.

The paper is well organized and fluently written. I like to mention in particular that the Figures are not only beautiful, but also prepared in a way that makes complex relationships easily accessible for the reader. The methods applied seems sound to me, though I do not feel that I can evaluate the model framework because this is on the edge of my expertise.

My rating of the paper is, however, 'major revisions' for the reason I will explain in the following: unfortunately, the atmopheric conditions framing the paper do not match the presented results. The authors claim to help explain observations of very few ice crystals and high supersaturations in the very cold (< 205 K) tropical tropopause layer (TTL).
Observed conditions in TTL cirrus clouds are: low ice particle concentrations around 0.005 – 0.1 cm-3, vertical velocities most frequently around a few cm/s (or even slower), infrequent waves up to ~2 m/s (the low/high ice concentrations are found in the slow/fast updrafts).
The conditions where suppression of homogeneous ice nucleation is found in the paper (Figure 8) are: ice particle concentrations > ~3 cm-3 at vertical velocities 0.1 – 1 m/s. These vertical velocities and thus ice concentrations represent the atmospheric range of gravity waves, e.g. behind mountains, or convection (see for example Kärcher and Lohmann, 2002), but can not be extrapolated to TTL conditions. The results shown in Figure 8 clearly indicate that for TTL conditions the new and control simulations do not greatly differ, that means that no further understanding of the TTL ice concentrations can be obtained from this study.

Nevertheless, to better understand the processes of homogeneous ice formation is important also in strong updrafts, simply for a correct modelling of high ice concentrations occuring at very low temperatures in many geographical regions (see e.g. Sourdeval et al., 2018 and Gryspeerdt et al., 2018, both ACP) or also for example from the point of view of cirrus seeding. Thus, I would encourage the authors to interpret their interesting findings in relation to the corresponding atmospheric environments.

Specific comments:

Title: From the title I did not have a good idea of the content of the paper. I would suggest to replace ‚supercooled water‘ by ‚ultra viscous particles‘, because the term ‚supercooled water‘ to me implies homogeneous freezing of liquid cloud drops at -38C.

p 2, l 7-9: *‚These observations suggest that our current understanding of the ice formation mechanisms and therefore methods of modelling the formation of low temperature cirrus clouds are incorrect or incomplete (Peter et al., 2006; Krämer et al., 2009 ; Jensen et al., 2010).‘*
Many more publications treating the topic appeared later – in case you mention the TTL in the next version of the manuscript, the most impostant newer studies should also be cited.

p 6, l 20: *‚… extremely high number concentrations of organic aerosol have been observed in the upper troposphere near to regions of deep convective outflow in the tropics (Andreae et al., 2018).‘*
The observations of Andreae et al. (2018) are well below the TTL, so is this statement really relevant for the study ?

P 10, l 5: *‚… up-draft velocities typically found in the tropical tropopause layer from Table 1.‘*
The updraft range used for the study is not typical for the TTL, in particular the large scale updraft is lower, see comments to Table 1.

Figures 4 and 5:
The simulations shown in Figure 4 are (as can be seen from Figure 5) those with almost the largest effect on ice nucleation (Sze distribution 3, 0.6 m/s). The interpretation of the results regarding TTL cirrus are based mainly on this scenario. However, these conditions are untypical for the TTL – especially the updrafts are much slower (see comment on Table 1). For the TTL, the scenario shown at the bottom of the left column of Figure 5 would be most appropriate, though even here the updraft is rather high (only a few cm/s is typical in the TTL).

Figure 8:
This Figure shows that the suppression of homogeneous ice nucleation occurs for ice particle concentrations larger than ~3 cm-3. The low concentrations that are under discussion, however, are in the range of 0.005 – 0.1 cm-3 (Krämer et al, 2009; Jensen et al., 2013; Spichtinger and Krämer, 2013), where no difference between the control and the new cloud parcel model is visible, or this range is not covered by the simulations.

Table 1:

Table 1. Parameter space of atmospheric conditions used to test and compare results from the new and control models.

Variable	Range	Reference
Temperature	185-225 K	Reverdy et al. (2012)
Pressure	150-70 hPa	Fueglistaler et al. (2009)
RH	0.3-0.8	Jensen et al. (2017)
Height	14-18 km	Fueglistaler et al. (2009)
Up-draft velocity	0.1-1 $m.s^{-1}$	Kärcher and Ström (2003)

The parameter spaces for TTL cirrus clouds specified in this table are partly not correct:
Temperature: 185 – 205 K
Updraft: 0.01 – ~2 m/s, or even slower in the TTL
Spichtinger and Krämer (2013), ACP, also
Jensen et al. (2012), JGR