Response to Reviewer I

Reviewer's comments: The nucleation mechanism of polar mesospheric clouds has been a longstanding problem. At least since the late 1960s it has been debated whether homogeneous or heterogeneous nucleation is the dominant nucleation mechanism leading to the formation of ice particles under the extreme conditions of the polar summer mesopause (e.g., Witt 1969). While homogeneous nucleation had been deemed very unlikely given the contemporary knowledge on temperatures, water vapor mixing ratios and the (at that time only conceived) occurrence of meteoric smoke particles, observations of extreme gravity wave-induced temperature perturbations by Lübken et al. (2009) triggered Murray and Jensen (2010) to reinvestigate the problem. Based on (slightly modified) classical nucleation theory they concluded that homogeneous nucleation could indeed lead to the formation of amorphous solid water particles in the mesopause region if such extremely strong gravity wave-induced temperature perturbations (and hence cooling rates) occurred. However, they also found that if homogeneous nucleation had to compete with heterogeneous nucleation on meteoric smoke particles, the latter was more efficient and homogeneous nucleation became negligible.

In their current manuscript Tanaka and coauthors reconsider this problem based on the fact that the classical nucleation theory used in the work of Murray and Jensen is known to strongly disagree with laboratory observations for the case of water. Hence, Tanaka et al. apply a semi-phenomenological model which is known to be in much better agreement with observations. This model shows a much higher free energy barrier for nucleation such that homogeneous nucleation of ice particles in the mesopause region would require unrealistically low temperatures, i.e., well below 100K. Hence, this nucleation pathway can be ruled out (because it contradicts observed temperatures) while heterogenous nucleation is found to be feasible (in agreement with the recent groundbreaking laboratory measurements by Duft et al. 2019).In all this is a sound study that contributes to the important fundamental problem of ice nucleation in the mesopause region. While the study of Murray and Jensen predicted homogeneous nucleation to possibly occur under extreme, but still conceivable conditions (extreme cooling rates, no competing meteoric smoke) the work by Tanaka et al. now clarifies that even under such extreme conditions homogeneous nucleation cannot be expected. This result certainly warrants publication.

My recommendation is hence to publish this work provided that the following mostly minor issues are properly addressed before publication:

The referencing in the introduction could be improved by referring to the original papers for the statements made. Here are my suggestions:

Thank you for your valuable comments. We also appreciate many references that you provided. All the comments were for the improvement of the paper. We have added all references listed by the reviewer and revise our paper according to the reviewer's comments.

- line 16/17: original reference for noctilucent clouds: Jesse 1885; maybe also Vestine 1934

We have added two references to the paper.

- line 20: the original reference for satellite-based PMC observations is Donahue et al 1972; a very good review until 2006 is DeLand et al., 2006.

We have added the references.

- line 21: reference for particle sizes: Thomas and MacKay, 1985, von Cossart et al., 1999;

We have added the references.

- line 23/24: well, this statement is not correct as it stands here: the ground based visual sightings of NLC actually do not show a unique trend as shown in Kirkwood and Stebel (2003); however, a trend is observed in the brightness of satellite-based PMC observations as presented in Thomas et al (2003) and updated in DeLand and Thomas (2015).

Thank you for your useful comments. According to the reviewer's comment, we have added the references and rewritten the description.

- line 25: while the reference to Lübken et al. (2018) is good, the original paper posing this hypothesis should also be mentioned, i.e., Thomas et al., Nature 1989.

We added the reference.

- line 27: original reference on gravity wave-NLC-interaction: Witt, 1962.

We added the reference.

- line 28: to my knowledge temperatures as low as 100K (and even lower) have only been reported in Lübken et al. (2009). Lübken 1999 is a climatology for mean temperatures at 69°N (from falling sphere measurements); Rapp et al. (2002) do show gravity wave perturbed temperature measurements in NLC but with minimum temperatures of 110K.

We thank for the useful comment. As the reviewer pointed out, the value of the minimum temperature due to gravitational waves is 110 K, so we revised the text. We also cited Lübken et al. (2009) showing an observed value of 100K.

- line 33: homogeneous nucleation has only been considered feasible again (after many years during which it was regarded extremely unlikely) after Lübken et al. (2009) reported enormous temperature variability due to gravity waves (see their figures 9, 10 and 11). Until then the consensus in the community was that it was rather heterogenous nucleation on meteoric smoke (see e.g., Rapp and Thomas 2006 for a discussion).

Thank you for your comments on the consensus of the community so far. It is an important point, so we include the statement in our paper.

- line 39 and 40: The authors are mixing two things here: as reviewed in Rapp and Thomas, the stated species have been suggested in the literature as potential nuclei for mesospheric ice particle formation. However, not all of the stated species are candidates for the composition of meteoric smoke (e.g., proton hydrates, soot are independent of meteoric origin). Meteoric smoke composition is indeed discussed in Plane (2015). I recommend to have a look at this paper and change the sentence accordingly.

Thank you for pointing this out. According to the comment, the description of meteor smoke was not accurate, so we revised the description.

- Section 3: in order to put the results in perspective, it would be useful if the authors included a short section describing typical ranges of mesospheric variables like observed temperatures, water vapor mixing ratios or partial pressures, concentrations of meteoric smoke particles (e.g. from rocket borne observations), and cooling rates due to tides and gravity waves. This will help assessing the assumptions made and results achieved in the paper. In this context, the authors should clearly state if derived or used values are way outside of observed ranges.

Accodring to the reviewer’s comment, we included a short subsection describing typical ranges of mesospheric variables in Section 3.1.

- line 238/239: the authors should point out that cooling rates as low as 10^{-6} Ks^{-1} at initial temperature of 135K also corresponds to a completely unrealistic time that the nucleation would take. However, observations do show that PMSE (which are also evidence for ice particles, but already at times when they have not yet grown large enough to be optically detectable) form rapidly for example in updrafts of gravity waves (i.e., within minutes).

As the reviewer pointed out, the cooling rate of 10^{-6} Ks^{-1} is unrealistic in the mesosphere. So we added the description in Section 3.

- line 235/236: These formulations are misleading. "the amount of water vapor present was 20 times higher at 145K than at 135K" – this certainly doesn't have anything to do with the atmosphere. In the atmosphere, the water vapor mixing ratio in the mesosphere is determined by transport across the tropical tropopause and oxidation of methane in the stratosphere (roughly at a ratio 50:50) and does not depend on the local temperature. Please clarify what you mean.

**As the reviewer suggested, it was a misleading expression, so we rewrote it.
We meant that for the equilibrium vapor pressure, the number density at 145 K is 20 times greater than 135 K. We have changed it to such an expression.
On the other hand, as the reviewer pointed out, the number density of water molecules depends on various other factors, so we included the description in Section 3.1.**

- Figures 6 and 7: please give "dust density" in number densities and not mass densities for easier interpretation in terms of known values from previous models and observations.

According to the reviewer's suggestion, we rewrote the vertical axis of the graph as the number density. We have also revised the derivation of the conditional equation in the text to reflect this change in Section 2.

- Section 3.3: these are important results. The authors should maybe also state that measurements with SOFIE on AIM can only then be properly explained if the refractive index for crystalline ice is used, but not for amorphous ice. I remember that this was presented by Mark Hervig at several meetings. The authors might like to check back with him where this is published.

Thank you for your useful comment. We contacted him and cited the reference in our paper. We also added the description about the measurements with SOFIE in Section 4:

The phase of ice particles in polar mesospheric clouds (PMCs) was determined using observations of the infrared extinction of the mesosphere from the Solar Occultation for Ice Experiment (SOFIE) on the AIM satellite (Hervig and Gordley, 2010). The observations could be explained using refractive indices of crystalline ice as opposed to amorphous ice; hence suggesting that not amorphous ice particles but rather particles of cubic ice existed near the mesopause (Hervig and Gordley, 2010). This observational result is consistent with our theoretical results that the nucleation leads to the formation of crystalline ice.