Effect of gravity wave temperature fluctuations on homogeneous ice nucleation in the tropical tropopause layer

August 12, 2015

We would like to thank the reviewer for the insightful evaluation of our work, especially for pointing out the error in Eq. (17), which will be corrected in the revised manuscript. Please note that this error does not affect our conclusions. Below is our point-by-point reply to the reviewer's comments.

1. ***Reviewer*** — There is a fundamental error in Eq. (17). As far as I can see from the original reference, the logarithm of the nucleation rate is represented by a third order polynomial in $\triangle a_{\mathrm{w}} = a_{\mathrm{w}} - a_{\mathrm{w}}^{\mathrm{i}}$ and NOT in $S a_{\mathrm{w}}$ (at least with the implicit definitions of a_{w} and S). It is clear from simple calculations that $\triangle a_{\mathrm{w}} \neq S a_{\mathrm{w}}$, thus the derivation of Eqs. (18)–(20) is incorrect. Probably, it is possible to re-derive similar expressions. However, in the present state the calculations are wrong. I have to express here that the main conclusion of the study remains unchanged, although the theoretical interpretation must be clarified.

   Authors — Thank you very much for pointing out this error in our derivation. Indeed

   $$\triangle a_{\mathrm{w}} = a_{\mathrm{w}} - a_{\mathrm{w}}^{\mathrm{i}} = (S-1) a_{\mathrm{w}}^{\mathrm{i}} \tag{1}$$

   because $S = a_{\mathrm{w}}/a_{\mathrm{w}}^{\mathrm{i}}$. Thus Eq. (17) should be

   $$\log_{10}(J) = P_3((S-1)a_{\mathrm{w}}^{\mathrm{i}}), \tag{2}$$

   and Eq. (18) becomes

   $$\log_{10}(J_{\mathrm{max}}) = P_3((S_{\mathrm{max}}-1)a_{\mathrm{w}}^{\mathrm{i}}(T(t^*))) \approx P_3((S_0 + \triangle S - 1)a_{w}^{\mathrm{i}}(T_0)). \tag{3}$$

   The rest of the derivation and argument remain unchanged.

2. ***Reviewer*** — Although it is stated that beside the reference simulations with $T_0 = 195\,\mathrm{K}$ simulations with $T_0 = 180\,\mathrm{K}$ and $T_0 = 210\,\mathrm{K}$ were carried out, the representation of the results is quite minimalistic; they are just shown in Figs. 5/6. Maybe you should try to present the resulting (low!) ice crystal number concentrations in a kind of statistical matter. In addition you should try to scan the parameter space (T_0, p_0) in a bit finer resolution in order to have a better representation of the realistic cases. You should also try to compare these results at least qualitatively with measurements, e.g. as shown in Spichtinger and Krämer (2013).

Authors — The dependence on T_0 is monotonic, with the theoretical curve for $T_0 = 195\,\mathrm{K}$ lies between the curves for $T_0 = 180\,\mathrm{K}$ and $T_0 = 210\,\mathrm{K}$ (please see Figs. 5 and 6 in the manuscript). Calculations with other values of T_0 confirm this monotonic behaviour. Thus we do not feel it is necessary to add figures with other values of T_0 to the revised manuscript.

We have in fact received several requests from colleagues and readers of the manuscript to provide statistical comparison with observations. After long discussions among ourselves we decided not to do so because this study applies only to the nucleation period (which is very short, on the orders of minutes) at the beginning of the cloud lifetime. Since TTL cirrus may persist over a few days, observations typically do not capture the nucleation period of the clouds. Thus, a direct comparison between our simulations and observations may be somewhat misleading.

3. ***Reviewer*** — A major issue for the formation of low ice crystal number concentrations in the study by Spichtinger and Krämer (2013) was the occurrence of very slow background updrafts on order of $0.01\,\mathrm{m\,s^{-1}}$. You should try to use such background velocity fields for your realistic trajectory simulation.

   Authors — The updrafts used in our simulations are derived directly from the observed balloon temperature time series, and representative values are shown in Fig. 3 in our reply to Reviewer 1. They include both high-frequency motions with periods of several minutes (referred to as the wave component in Spichtinger and Krämer, 2013), and longer timescale motions (periods of several hours) that correspond to the "large-scale" component in Spichtinger and Krämer (2013). In our simulations, low INCs can be obtained even though our updraft velocities are typically much larger than $0.01\,\mathrm{m\,s^{-1}}$ (please see Fig. 3 in our reply to Reviewer 1).

4. ***Reviewer*** — There should be more accurate definitions of the used quantities. For instance, S is never defined although I assume that S is the saturation ratio with respect to ice. Also the scale height is not well-defined, is it the usual value $H \sim 8\,\mathrm{km}$?

   Authors — S is indeed the saturation ratio with respect to ice. The scale height H is typically 6 km in the tropical UT/LS. We will clarify these in the revised manuscript.

5. ***Reviewer*** — Page 8774, line 16: How reasonable is the time resolution of the trajectories in order to get a good representation of the relevant small-scale gravity waves? Please explain this in relation to the frequency of gravity waves, which might be expected.

   Authors — In principle, the balloon sampling rate (30 s, i.e. Nyquist frequency of $1\,\mathrm{min^{-1}}$) is sufficient to resolve all gravity waves (as well as higher frequency turbulence). However, to account for the balloon neutral oscillations at a period of 4 min, we had to filter the data using a high cut-off frequency (f_{high}). The representation of the gravity wave spectrum in the filtered data depends on the cut-off frequency rather than the original sampling rate. Please see our reply to Reviewer 1 for detailed discussions about our choice of f_{high} and the implication for the resolved gravity wave spectrum.

6. ***Reviewer*** — Page 8775, lines 5–7: Which resolution of ECMWF data is used for deriving the background temperature profile? Is it good enough for your considerations?

Authors — We used the highest vertical resolution provided by ECMWF, which is $\lesssim 500\,\mathrm{m}$ in the TTL. This spacing is sufficient to resolve the salient features of the background temperature profile, and thus to provide reasonable values for the lapse rate.

7. ***Reviewer*** — For the background aerosol (heterogeneous IN concentrations, aerosol particles for homogeneous nucleation) you often quote measurement studies, which were carried out mostly in the extratropics; since you want to address tropical tropopause layer, you should make clear that these measurement values are also reasonable for this tropical study.

   Authors — In the revised manuscript we will add another reference for aerosol observations, which included measurements in the tropics (Hermann et al., 2003). The properties of the background aerosols used in our simulations are within the range reported by Hermann et al. (2003).

8. ***Reviewer*** — Sedimentation of ice crystals is not just the effect of removal of ice crystal number concentrations; in combination with other processes (nucleation and diffusional growth) a kind of dynamic equilibrium might occur (see e.g. investigations by Spichtinger and Cziczo, 2010; Wacker, 1995). Probably for your simulations it is okay to omit sedimentation, but you should motivate this in a more convincing way, e.g. arguing about terminal velocities of very small ice crystals.

   Authors — The impact of sedimentation is indeed beyond the scope of this paper. Lagrangian parcel models (such as used in this work) are inadequate to fully address the impact of sedimentation. The sentence on lines 17–19, page 8776 will be deleted as it has caused more confusion than we intended.

9. ***Reviewer*** — The accommodation coefficient for the reference case seems a bit low ($\alpha = 0.05$). Skrotzki et al. (2013) indicate that the usual values are more in the range ($0.1 \leq \alpha \leq 1$). How large is the difference in the simulations between e.g. $\alpha = 0.5$ and simulations with $\alpha = 0.05$?

   Authors — We will use $\alpha = 0.1$ for the reference case in the revised manuscript. The results for the range of α between 0.001 and 1 are shown Figure 4 in our reply to Reviewer 1 and Fig. 7 in the manuscript. Please note that α affects vapour-limit events but not temperature-limit events.

10. ***Reviewer*** — I think your lower boundary of the nucleation rate J_ε is quite small, i.e. the probability of freezing a typical solution droplet at these conditions is probably zero (with respect to machine epsilon).

    Authors — In the revised manuscript we will use $J_\varepsilon = 10^9\,\mathrm{L^{-1}\,s^{-1}}$, compared with the original value of $1\,\mathrm{L^{-1}\,s^{-1}}$. Please see the discussions about J_ε in our reply to Reviewer 1.

11. ***Reviewer*** — It would be nice to add the time evolution of the nucleation rate for the different scenarios in Fig. 3; this would help to understand why the ice crystal number concentration is changed that drastically.

    Authors — The nucleation rate is

$$J = 10^{P_3((S-1)a_\mathrm{w}^\mathrm{i})} \tag{4}$$

(see Eq. 2 above) and thus it changes with time similarly to S. Since we have shown the evolution of S, we think it is not necessary to provide a separate figure for $J(t)$.

12. ***Reviewer*** — Page 8780, lines 14–23: I not understand what you want to say, please explain this in more details.

    Authors — These sensences are indeed confusing and will be removed from the manuscript.

13. ***Reviewer*** — The figures are quite hard to read. Actually, the figure captions could be extended. In Fig. 3 the different curves (all represented with the same colour, i.e. blue or red) should be labelled.

    Authors — Thanks. We will add labels to this figure in the revised manuscript.

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