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

June 10, 2015

We would like to thank the reviewer for the insightful evaluation of our work, which will guide us to revise and improve the manuscript. Please find below our point-by-point reply.

1. ***Reviewer*** — Measurements of ice crystal number concentrations in the TTL cirrus usually yield values that are substantially lower than a theory based on the assumption of homogeneous nucleation would predict. This so called "ice nucleation puzzle" (Spichtinger and Krämer, 2013) can be solved by assuming temperature fluctuations (caused by fluctuations of the vertical wind) with time scales similar to the nucleation time scale (e.g. triggered by gravity waves). So far, simulations using idealistic temperature time series have been used to demonstrate this. The present authors want to go a step further and use measured time series of temperature. I endorse this goal.

   The balloon measurements from which the time series are obtained, must be filtered at the high frequency (short period) end, at a period of 10 min. That is, processes that are faster, cannot be treated with this method. Unfortunately, homogeneous nucleation is such a quick process and to my opinion the authors miss their goal. It seems, however, that the authors found a trick to circumvent this problem, namely to choose an extremely low nucleation threshold. This trick works insofar as it extends the nucleation time scales to a few minutes up to an hour (Sect. 4.3.1). However, this is achieved only for a high price. Usually the threshold is chosen in a way that the nucleation rate is practically zero below the threshold and many orders of magnitude larger above it. In this paper the nucleation rate at the threshold and some percent above is practically zero as well (see Fig. 3). It seems that this makes results differing from corresponding results from other papers, qualitatively and quantitatively. This choice of threshold and the consequent differences from results from other papers are not discussed at all; instead the authors claim consistency with other results, a view that I cannot support.

   My recommendation is therefore to accept the paper only after a major revision (addition) where the authors demonstrate either that their nucleation results are similar and consistent with those of other authors (e.g. Kärcher and Lohmann, 2002; Spichtinger and Gierens, 2009) or that those other results are wrong. This is a pity, because the paper does contain an interesting concept, i.e. the distinction between vapour- and temperature-limit nucleation events. I like also the analytical derivation in Sect. 5.

Authors — Our cut-off frequency choice is driven by the search of a compromise between incorporating the gravity wave disturbances as thoroughly as possible while avoiding motions associated with the balloon flight mechanics. In particular, the balloon neutral oscillations have periods $\sim 4\,\mathrm{min}$, and that is why we used a safe ($f_{\mathrm{high}} = (10\,\mathrm{min})^{-1} = 0.1\,\mathrm{min}^{-1}$) cut-off frequency. As stated in the manuscript, the chosen cut-off frequency enables us to virtually resolve the whole spectrum of gravity-waves in most of the TTL. Higher frequency motions are typically associated with turbulence past the Brunt-Väisälä frequency. Nevertheless, we compare here the microphysical simulations with filtered data at cut-off frequencies of $0.2\,\mathrm{min}^{-1}$, and $0.1\,\mathrm{min}^{-1}$ (original value). For the data set with $f_{\mathrm{high}} = 0.2\,\mathrm{min}^{-1}$, we additionally apply a butterworth filter to eliminate the residual balloon-induced motions (see Fig. 1 below). Figure 2 shows that the number of ice crystals as a function of $\triangle S$ (or $\triangle T$) is not sensitive to the cut-off frequency.

The nucleation threshold is a parameter associated with the numerics that allows us to precisely define the beginning and end of the nucleation events. The reviewer is concerned that the threshold that we chose for our simulations was too small. To address this, we are now using a threshold of $J_\epsilon = 10^9\,\mathrm{L}^{-1}\,\mathrm{s}^{-1}$, compared with the original value of $1\,\mathrm{L}^{-1}\,\mathrm{s}^{-1}$. In general, the duration of nucleation is shorter with larger J_ϵ (compare Fig. 3 here with Fig. 4 in the manuscript). However, the specific choice of the threshold does not usually affect the total number of ice crystals N_{i}. This is because N_{i} depends largely on the maximum nucleation rate J_{max} (and not on J_ϵ), as long as $J_{\mathrm{max}} \gg J_\epsilon$.

In summary, the statistics of N_{i} as a function of $\triangle S$ (or $\triangle T$) is independent of the parameters f_{high} and J_ϵ. Our conclusions are based on this statistics, and are confirmed by the analytical derivation in Sect. 5.1 of the manuscript. Please note that the mathematics in this section applies regardless of the chosen threshold of nucleation as well as the nature of the waves in the temperature time series.

As pointed out by the reviewer, the INCs shown in Fig. 2 of the manuscript are indeed larger than in other papers, specifically Kärcher and Lohmann (2002, Fig. 3), and Spichtinger and Gierens (2009, Fig. 7). This difference is because we set the deposition coefficient to be $\alpha = 0.05$ for the calculation in this figure, while Kärcher and Lohmann (2002) used $\alpha = 0.5$. We are able to obtain consistent numbers as in these previous work with larger values of α (see Fig. 4). As mentioned in the manuscript, the deposition coefficient is poorly constrained by experimental data. The sensitivity of the INCs to the deposition coefficient ranging between 0.001 and 1 is discussed further in Sects. 4.3.3 and 5.2 of the manuscript.

2. ***Reviewer*** — Page 8777, line 20: I am surprised of the low critical saturation that you assume at 195 K. Looking at Fig. 3 of Koop et al. (2000) it seems that the critical supersaturation at 195 K is much higher. Using Eq. (4) from Kärcher and Lohmann (2002) I calculate $S_0 = 1.645$.

   Authors — The saturation ratio at the threshold of nucleation increases with the chosen threshold J_ϵ. We now have $S_0 = 1.553$ for $J_\epsilon = 10^9\,\mathrm{L}^{-1}\,\mathrm{s}^{-1}$. For an aerosol radius of $r_{\mathrm{a}} = 0.25\,\mu\mathrm{m}$ and aerosol number concentration of $N_{\mathrm{a}} = 200\,\mathrm{cm}^{-3}$, this corresponds to a production rate of $\frac{\mathrm{d}N}{\mathrm{d}t} = 0.013\,\mathrm{L}^{-1}\,\mathrm{s}^{-1}$ which is in the same order as that used in Spichtinger and Krämer (2013). Please note that Koop et al. (2000) plot the nucleation threshold S_0 for a freezing probability of $1\,\mathrm{min}^{-1}$, which corresponds to two-thirds of the aerosol population being frozen in 1 min. They do not claim that their formula should only be applied above this threshold. To our understanding, the lower the threshold the more accurate the result.

Please also note that we have updated the formula for the water activity based on Koop and Zobrist (2009, Appendix), which is different from the original formula in Koop et al. (2000). In addition, we now use the formula for the saturation water vapour pressure given in Murphy and Koop (2005), instead of the Goff-Gratch formula used in the original manuscript. These updates also change the value of S_0 slightly.

3. ***Reviewer*** — Figure 2: It might be that the low critical supersaturation or your assumption of a monodisperse aerosol leads to a much higher sensitivity N_i vs. w. From Kärcher and Lohmann (2002) I assume that $N_\mathrm{i} \propto w^{3/2}$ in most cases. Figure 2 shows a relation that is rather $N_\mathrm{i} \propto w^{5/2}$ for low w. Also the number of ice crystals is much (factor 30 or so) larger in your model than for instance in Kärcher and Lohmann (2002) or Spichtinger and Gierens (2009, Fig. 7). These differences require an explanation.

   Authors — Please see the INCs as a function of w for the different values of the deposition coefficient in Fig. 4 below. For $\alpha = 0.5$ our calculation gives very similar numbers as in these previous work, as well as in Spichtinger and Krämer (2013, Fig. 2).

4. ***Reviewer*** — Figure 3: To my opinion we see here another strange result of the choice of an extremely low nucleation threshold. As the top right panel shows, we are above the nucleation threshold from t_0 on, but it needs 12–13 min before the curve in the bottom panel indicates an Ni of 0.001 per litre, and it takes still 10 and more minutes until all ice crystals are formed. The simulations suggest that ice formation occurs on a time scale of half an hour or so. Compare this to Spichtinger and Krämer (2013, Fig. 1) where a time scale of 140 s is indicated. How can you state that these results are consistent?

   Authors — We are now using a larger threshold and thus the duration of nucleation is shorter. As shown in Fig. 3, we also have nucleation events that last for only one or two minutes. Please note that the threshold does not affect the INCs obtained after the nucleation events, as explained above.

5. ***Reviewer*** — Page 8774, line 8–9: "whole equatorial area" sounds exaggerated considering that there are only 2 balloons.

   Authors — We agree and will rewrite this as "whole equatorial circle."

6. ***Reviewer*** — Eq. (1): R should be R_a.

   Authors — Thanks. We will fix this typo.

7. ***Reviewer*** — Section 3, par. 4: Please explain why sedimentation would reduce INC. If crystals get lost from the parcel by sedimentation, another nucleation event could occur earlier than without sedimentation. Why should this not happen?

   Authors — We meant that sedimentation reduces the INC strictly within each nucleation event. If nucleation occurs following ice sedimentation, then we will count this occurrence as a new event.

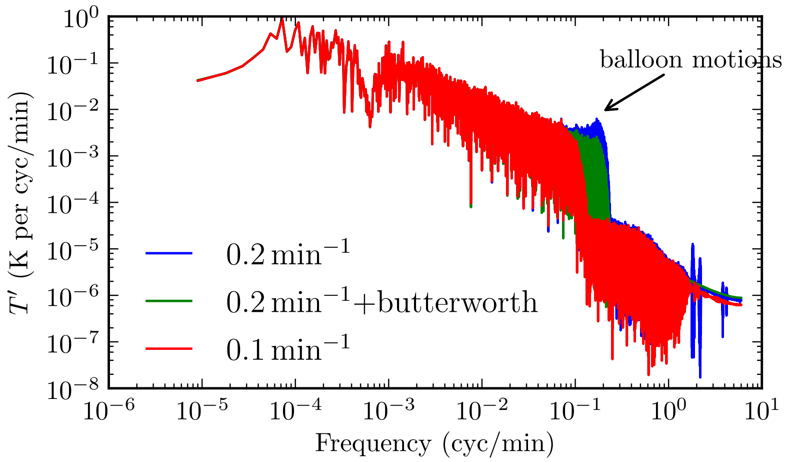

Figure 1: Power spectrum of the balloon temperature perturbations at cut-off frequencies of $0.2\,\mathrm{min}^{-1}$ and $0.1\,\mathrm{min}^{-1}$.

8. ***Reviewer*** — Beginning of Sect. 4: I do not understand how you can mention adiabatic motion and pressure variations in the first sentence, and assume constant air pressure in the second. Does "constant pressure" just mean that your parcels are sufficiently flat?

   Authors — The air parcels experience adiabatic motions, for which there are both pressure and temperature variations. However, the contribution associated with temperature variations to the variations in water vapour mixing ratio is much larger than that due to pressure variations. Thus, to calculate the water vapour mixing ratio of the air parcels we can assume constant pressure.

9. ***Reviewer*** — Figure 6 and Sect. 4.3.2: It is not easy to understand why $N_\mathrm{i}(210\,\mathrm{K})$ is higher than $N_\mathrm{i}(180\,\mathrm{K})$ as a function of $S_\mathrm{max} - S_0$ and vice versa as a function of $T_\mathrm{min} - T_0$. A more detailed explanation would be welcome.

   Authors — Indeed. We will add explanation to the revised manuscript. This comes from the dependence of a_w on T_0 (Eq. 18): a_w increases with increasing temperature, hence the different slopes of the functions $N_\mathrm{i}(S_\mathrm{max} - S_0)$ and $N_\mathrm{i}(T_\mathrm{min} - T_0)$ at different temperatures.

10. ***Reviewer*** — Eq. (19) and following text: if t^* is the point in time where $J = J_\mathrm{max}$ and $S = S_\mathrm{max}$, then $\mathrm{d}S/\mathrm{d}t$ should be zero.

    Authors — Yes, thank you for pointing this out. This eliminates the second term in Eq. (19).

References

Kärcher, B. and Lohmann, U.: A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols, J. Geophys. Res., 107, D2, doi:10.1029/2001JD000470, 2002.

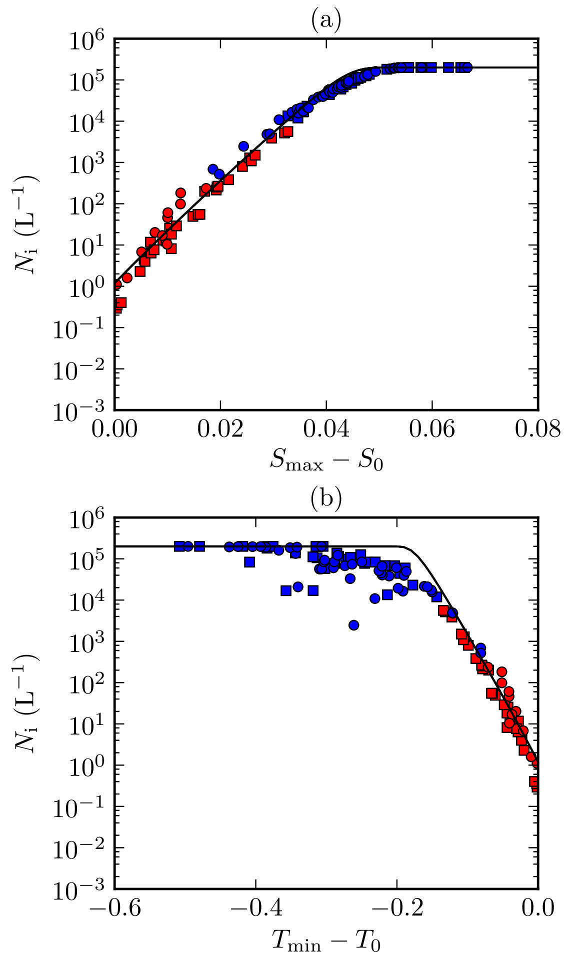

Figure 2: Number of ice crystals nucleated using the balloon temperature data, which has been filtered at high cut-off frequencies of $0.2\,\mathrm{min}^{-1}$ (squares) and $0.1\,\mathrm{min}^{-1}$ (circles). Blue markers show vapour-limit events and red markers show temperature-limit events. The threshold of nucleation is $J_\epsilon = 10^9\,\mathrm{L}^{-1}\,\mathrm{s}^{-1}$.

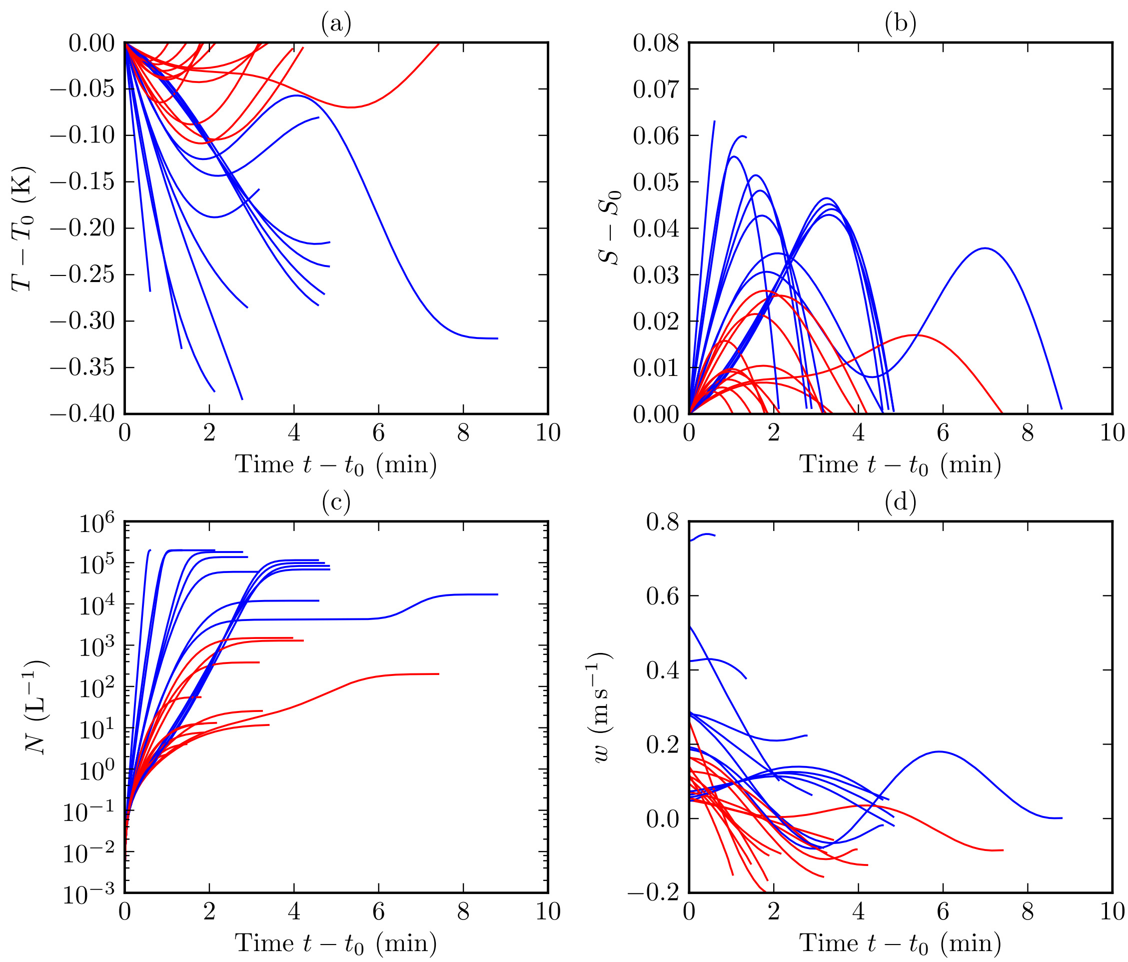

Figure 3: Evolution of temperature, saturation ratio, INC and vertical velocity during representative nucleation events forced by the balloon temperature data, which has been filtered at a high cut-off frequency of $0.2\,\mathrm{min^{-1}}$. Blue curves show vapour-limit events and red curves show temperature-limit events. The threshold of nucleation is $J_\epsilon = 10^9\,\mathrm{L^{-1}\,s^{-1}}$.

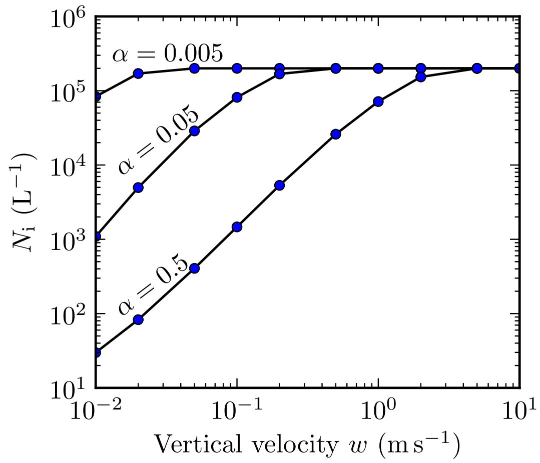

Figure 4: Number of ice crystals obtained for nucleation events forced by constant vertical velocity w for different values of the deposition coefficient α.

Koop, T. and Zobrist, B.: Parameterizations for ice nucleation in biological and atmospheric systems., Physical chemistry chemical physics : PCCP, 11, 10 839–10 850, doi: 10.1039/b914289d, 2009.

Koop, T., Luo, B., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406, 611–614, doi:10.1038/35020537, 2000.

Murphy, D. M. and Koop, T.: Review of the vapour pressures of ice and supercooled water for atmospheric applications, Q. J. R. Meteorol. Soc., 131, 1539–1565, doi: 10.1256/qj.04.94, 2005.

Spichtinger, P. and Gierens, K. M.: Modelling of cirrus clouds - Part 1a: Model description and validation, Atmos. Chem. Phys., 9, 685–706, doi:10.5194/acp-9-685-2009, 2009.

Spichtinger, P. and Krämer, M.: Tropical tropopause ice clouds: a dynamic approach to the mystery of low crystal numbers, Atmos. Chem. Phys., 13, 9801–9818, doi:10.5194/acp-13-9801-2013, 2013.