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

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We very much appreciate the reviewer's insightful and careful evaluation of our work. The reviewer's comments have helped us to further clarify and improve the manuscript. Please find below our reply, which we hope will adequately address the points raised by the reviewer.

1. ***Reviewer*** — In their response the authors assert that all simulations started at $S = 100\,\%$, and the system is brought to $S \sim 150\,\%$ by large scale movement. The only way I see this happening in Fig. 6 is that the vertical velocity is constant for hours until the selected saturation threshold is achieved, then fluctuations appear resulting in ice nucleation, and after a few minutes, both fluctuations and the large scale vertical movement disappear. If this is the case it must be shown explicitly and justify why it is representative of the conditions in cirrrus. Such highly idealized velocity evolution does not seem particularly realistic. Also, are Figs. 7–9 produced using the same vertical velocity series as in Fig. 6? Please explain.

   In some of the answers to earlier comments the authors stated that: "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))." This must be shown explicitly. Show that the average vertical velocity of your simulations is indeed positive and not additional background ascent is required. My fear is that extended for a longer period of time the blue lines in Fig. 6a would indeed result in net large scale ascent, but the red lines in a large scale downdraft. In such case the temperature limit would apply where both the fluctuating component of the velocity and the large scale ascent change sign during ice nucleation (since net ascent is required to generate $S \sim 150\,\%$ at t_0); a rather improbable situation.

   Authors — We would like to correct that *most* (not all) simulations that we showed in the first version of the manuscript were started at $S = 1.0$. In addition to an initial saturation of $S = 1.0$, some of the simulations were started at $S = 0.8$, 0.9, 1.1, and 1.2 (and also with different initial base temperatures). This has been restated more clearly in the manuscript (please see the last paragraph immediately before Sect. 4.1). For different initial S values, we obtain different nucleation events and correspondingly different ice number concentrations N_i, but the relationship between N_i and $\triangle S$ (or between N_i and $\triangle T$) is invariant among these different nucleation events. Each nucleation event is represented by a data point in Figs. 7–9. For the purpose of illustration, Fig. 6 shows only a subset of the data points in Fig. 7. All data for Figs. 6–9 are produced using the balloon temperature time series and the corresponding vertical velocities associated with the balloon temperature time series. The reason that we repeated the simulations for slightly different initial S is to obtain a larger sample of events to demonstrate that the relationship between N_i and $\triangle S$ (or between N_i and $\triangle T$) is statistically significant. In principle, one can repeat the simulations with any initial S to obtain more data points for these figures. However, if the air parcel is initially too dry ($S \to 0$), then nucleation may not occur at all.

For S in the range stated above, the parcel is indeed brought to the threshold of nucleation by the large-scale motions. The presence of large-scale waves is demonstrated in the spectrum of the temperature time series in Fig. 1 in the manuscript. These large-scale motions are located towards the top left corner of the figure (low frequency, large amplitude perturbations). Further, we extended the plots of Fig. 6 (please see the attached latest version of the manuscript) to show the behaviours of the system both before and during nucleation. The extended plots help to clarify the background condition leading to nucleation. However, please note that the ice number concentration obtained by nucleation depends on J_max, which is sensitive to the fluctuations during the nucleation period only (and not to the fluctuations before t_0). Recall that a nucleation period is defined to start when J exceeds J_ε (at $t = t_0$) and to end when $J < J_\varepsilon$ (at $t = t_0 + \tau$). The nucleation events may appear to be longer if J_ε is chosen to be very small (as in the first version of the manuscript). However, the total ice number concentration obtained is not sensitive to the choice of J_ε as long as $J_\varepsilon \ll J_\mathrm{max}$ (please see the extended discussions about J_ε in the previous revisions).

Finally, we would like to note that the main point of the paper is not whether the air parcel may be brought to the threshold of nucleation, but rather, once it is at the threshold of nucleation, how the fluctuations in water vapour and temperature during the short time period during which the ice number is changing rapidly govern the total number of crystals that are nucleated. Both our numerical simulations and theoretical consideration have shown that the ice number concentration obtained by nucleation (N_i) is a function of $\triangle S$ (or a function of $\triangle T$ for temperature-limit events). These two quantities ($\triangle S$ and $\triangle T$) are characterization of the fluctuations in water vapour and temperature during nucleation; they are defined exclusively within the period of nucleation and are independent of any information before nucleation.

2. ***Reviewer*** — Why do the authors stop their simulations after a few minutes (indeed some of the lines extent for 30 s only)? In most of temperature limited events in Fig. 6 further nucleation events would take place since the ice crystal concentration is small. If that is not the case it must be shown explicitly, i.e., extend the simulation period to show that no further nucleation events take place.

   Authors — The plots in Fig. 6 show each nucleation event only until the end of the nucleation period for that event (as defined formally when $J < J_\varepsilon$). The model calculation does not stop there. Indeed, further nucleation events take place downstream of all nucleation events (until we reach the end of the temperature time series of course). We count these downstream events as new nucleation events (additional curves and data points for Figs. 6–9). However, please note that we count only nucleation events that take place in ice-free air at t_0 (i.e. if all ice crystals from previous events have sublimated). In other words, we do not consider the effect of pre-existing ice on nucleation (please see Sect. 3).

3. ***Reviewer*** — The assumptions behind the theoretical development should be better stated. The theoretical results only apply to cases where the ice crystals are not efficient at removing supersaturation. This can be seen in the cases where the growth rates are substantial ($\alpha = 1$, higher temperature, high N_i) and the numerical model and the theoretical results diverge. It is clear in the development of Eq. (13) (the symbol in that equation must be $\approx$ instead of $=$) that both μ and J depend on S which can be reduced by growing ice crystals. Therefore, they are constant only if the ice crystals do not reduce S (J changes several orders of magnitude over very small $\triangle S$ so any factor affecting S would have a big incidence on N_i). Later in section 5.2 it is mentioned that the relationship N_i versus $\triangle T$ is independent of α which is of course not the case since α indirectly affects S.

   Authors — Equation (13) is the definition for μ. We rechecked the equation but don't think that there is any approximation involved in this particular equation. Please note that the derivation in Sect. 5.1 does not rely on the assumption of constant μ. In fact, the dependence of μ on $\triangle S$ (and thus on the deposition coefficient α) is shown explicitly in Eq. (17). We assume constant μ only to plot the analytic curves in Figs. 7–9.

   If μ is assumed constant, the relationship between N_i and $\triangle T$ is independent of α. Figs. 7–9 show that the assumption of constant μ is appropriate for temperature-limit events, but not for vapour-limit events. We have rewritten Sect. 5.2 to clarify these points. Please see the highlighted blue text in the attached PDF (latest version of the manuscript) for the revisions.

References

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.