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 these critical comments; they will be very helpful for us in our revision of the manuscript. Please find below our point-by-point reply.

1. ***Reviewer*** — The authors carefully set up their simulations so that the vertical velocity changes sign at least once before substantial nucleation rates are reached. This is a clever way to show their expected result but begs the question of how realistic the setup actually is.

   Authors — There is a misunderstanding here regarding the setup of the simulations. The cases in Sect. 4.2 where we designed w to change signs are shown only for illustration purposes. For the simulations with the balloon data we did not impose any constraint on w: it may change sign before substantial nucleation rates are reached (temperature-limit event), or not (vapour-limit event), and we show results for both of these types of events in the manuscript.

2. ***Reviewer*** — As noted by another reviewer, if the filtering of the balloon measurements is applied differently, or if higher temperature fluctuations with periods below 10 min are allowed, the simulations may result in higher number concentration. In fact the authors claim that they can obtain small crystal number even for high vertical velocity, something that is never shown.

   Authors — We have addressed the question about the cut-off frequency used for filtering the data in our response to Reviewer 1. Our study with the balloon time series are specifically performed so as to deal with realistic temperature disturbances in the TTL: larger temperature fluctuations are generally associated with longer-period disturbances, and large temperature fluctuations with periods below 10 min are thus very unlikely.

3. ***Reviewer*** — A related issue, and maybe the most significant one, is the selection of the initial conditions. All runs start on the verge of ice nucleation $S_0 \sim 150\,\%$. It is unrealistic to assume that each parcel starts from a very high supersaturation. One may ask, how do these parcels become such highly supersaturated in the first place? Starting from $100\,\%$ would any of the vertical velocity time series tested result in cloud formation? From the shape of the temperature perturbation profiles

in Fig. 4 it seems that they wouldn't. In reality there must be some underlying vertical movement bringing the supersaturation up to the initial conditions selected by the authors. Such movement (disregarded by the authors) is the actual driver of cloud formation, not the superimposed vertical velocity fluctuations. The analysis based only on the latter is flawed.

Authors — The reviewer noted that the initial saturation is equal to the saturation threshold S_0 and thus is unrealistically high in all of our simulations. This is actually not the case; our calculation begins at $t \leq t_0$ (where t_0 is the time at which nucleation begins) and with $S \leq S_0$ initially. In fact, most of the cases shown in Fig. 4 are with $S = 100\,\%$ initially. This misunderstanding arises perhaps because the evolution of the saturation ratio S is shown only for $t \geq t_0$ in Fig. 4. Please note that the subscript zero (e.g. t_0) denotes the time at which nucleation begins, which may not be the initial time of the parcel calculation.

The initial water vapour content and thus the initial saturation ratio of the air parcels is a free parameter in the simulations, and we have discussed the sensitivity of nucleation to this in Sect. 5.3 of the manuscript. This free parameter is meant to implicitly represent the large-scale ascent of air parcels in the TTL, which is, as noted by the reviewer, a prerequisite to high supersaturation. Lagrangian trajectory calculations show that air detrained from convection at or below saturation level takes typically several days to reach the supersaturation threshold for nucleation. On the other hand, nucleation itself occurs over a time scale of a few minutes to less than half an hour (depending on the chosen nucleation threshold). We exploit this scale separation to study the nucleation event and its sensitivity to high-frequency dynamical fluctuations once nucleation has started. It is implicit that the fluid parcel must previously be led to the verge of nucleation by large-scale motion in cloud free air but there is no point of studying this stage with our model. This will be mentioned in the revised version.

4. ***Reviewer*** — The authors omit important works (and in fact repeat some of the conclusions of those works) that may have helped in their analysis (e.g. Barahona and Nenes, 2011; Jensen et al., 2010, 2012; Cziczo et al., 2013; Murphy, 2014; Shi et al., 2015). For example, just as in this work, other works have shown (e.g. Jensen et al., 2010) that homogeneous nucleation could produce both, low and high ice crystal concentration. Similarly, field campaigns (e.g. Krämer et al., 2009) show high and low number concentration of ice crystals. Any comparison between field campaign data and model results should be done on a statistical basis. A limited set of parcel model simulations over very restricted conditions should not be used to draw conclusions on real clouds. Other aspects of the problem should be evaluated as well. Could the authors setup not only reproduce low crystal numbers but also the sustained clear-sky supersaturation and the small ice crystal size of TTL cirrus?

   Authors — Thank you for mentioning these references. We did cite about half of them and will improve our citation in the revised version. Jensen et al. (2010) and others have indeed shown that homogeneous nucleation could produce both high and low INCs. However, to our understanding, their explanation is based on a one-to-one relationship between cooling rates and INCs. In agreement with Spichtinger and Krämer (2013), we show that non-constant cooling rates break this one-to-one relationship. Our (new) contribution is that we provide the theoretical framework to explain the numerical results for non-constant (and constant) cooling rates, including the classification of the two types of nucleation events, and the analytical relationship between N_i and $\triangle S$ (or $\triangle T$).

Our goal here is not to perform a detailed comparison with observations, which would certainly require much more complex microphysical models. We wish only to stress that some of our main findings, e.g. the large sensitivity of nucleated INCs to initial relative humidity in the presence of high-frequency motions, may give a clue for understanding the observed variability of INC in the TTL.

We are focusing only on ice nucleation process. Clear-sky supersaturation is an issue which is beyond the scope of this paper.

5. ***Reviewer*** — Line 11, page 8771. Such high vertical velocities are not shown.

   Authors — Our results indeed show that low INCs can be obtained for high vertical velocity w if w does not remain consistently high throughout the duration of the nucleation events. As shown in Fig. 1 below, the INC obtained when w decreases with time is much smaller than if w remains constant throughout nucleation. We have also illustrated this concept and shown values of w for the idealised temperature time series in Sect. 4.2. Please see also Fig. 3 in our reply to Reviewer 1 for more values of w in the balloon data.

6. ***Reviewer*** — Line 15, page 8771. This conclusion has been already stated in several papers (e.g. Barahona and Nenes, 2011; Jensen et al., 2010; Murphy, 2014).

   Authors — To our understanding, none of these papers specifically address the issue of time-varying cooling rates during nucleation, or mention temperature-limit events (a concept first developed here).

7. ***Reviewer*** — Line 5–10, page 8773. A concentration of $100\,\mathrm{L}^{-1}$ is just a nominal number, not a threshold that defines a limit between homogeneous and heterogeneous ice nucleation. Further evidence of the predominance of heterogeneous ice nucleation comes from field campaign data (e.g. Cziczo et al., 2013).

   Authors — Indeed, we gave $100\,\mathrm{L}^{-1}$ here as a nominal number, and not a threshold that represents a limit between homogeneous and heterogeneous ice nucleation. We would like to say here that low INCs should not be considered as an argument against homogeneous nucleation. We did not mean to disregard heterogeneous nucleation.

8. ***Reviewer*** — Page 8774–8775. The authors should show a plot of the vertical velocity time series associated with these measurements. Also explain why measurements from only two balloons are assumed as representative of the dynamics of the TTL.

   Authors — Please see Fig. 1 below or Fig. 3 in our reply to Reviewer 1 for values of w.

   The time series of temperature from the balloon measurements are the closest among all observations to the fluctuations experienced by a moving air parcel. At the moment we can only exploit two flights from a limited campaign (more will be available in the future) but they accumulate more than 6 months of flight and travelled around the equator. This is quite a significant sampling of the TTL.

9. ***Reviewer*** — Page 8776, lines 15–20. This is an important issue. Many interesting dynamics occurs from the sedimentation of ice crystals (e.g. Barahona and Nenes, 2011; Murphy, 2014). In particular, sedimentation would allow the build up of enough

supersaturation for homogeneous ice nucleation to occur. Thus the assumption that sedimentation would further decrease ice crystal concentration is erroneous.

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. This sentence causes more confusion than we intended and will be deleted from the revised manuscript.

10. ***Reviewer*** — Page 8777, lines 1–5. Water vapour variability does not necessarily result from temperature fluctuations. In fact field campaigns have shown that temperature fluctuations are only partially responsible for the generation of supersaturation in the TTL (Diao et al., 2014).

    Authors — Here we simply refers to Eq. (4), which indicates that for a chosen/fixed pressure, there is a one-to-one relationship between T_0 and r_0. It is not necessary to discuss the topics of water vapour variability here.

11. ***Reviewer*** — Page 8779, Sect. 4.2. It is not clear how supersaturation can be generated in the first place without some persistent cooling (see general comments).

    Authors — We did allow the air parcels to cool before nucleation begins at t_0. Please see our response to Point 3 above.

12. ***Reviewer*** — Page 8784, Eq. (20). It must be mentioned that this is only true for negligible ice crystal concentrations.

    Authors — Not necessarily, the only requirement is that temperature variations happen much faster than nucleation.

13. ***Reviewer*** — In reality what the authors are defining as "temperature-limit" events is just a low ice crystal concentration regime, and has been introduced before (Kärcher and Lohmann, 2002).

    Authors — Kärcher and Lohmann (2002) studied constant vertical velocities only. They rather distinguish between different types of vapor-limit events by the fast and slow growth regimes (please see their Fig. 2). They did not study temperature-limit events in which the vertical velocities and cooling rates vary with time.

14. ***Reviewer*** — Page 8786, Line 20–25. According to this, the processes bringing up supersaturation to the level used in the initial conditions are the actual control of ice nucleation (see general comments).

    Authors — We disagree with the reviewer. The cooling associated with ascent in the TTL is the first stage leading to cirrus formation. However, we show that the distribution of ice following nucleation depends on the temperature fluctuations during nucleation.

References

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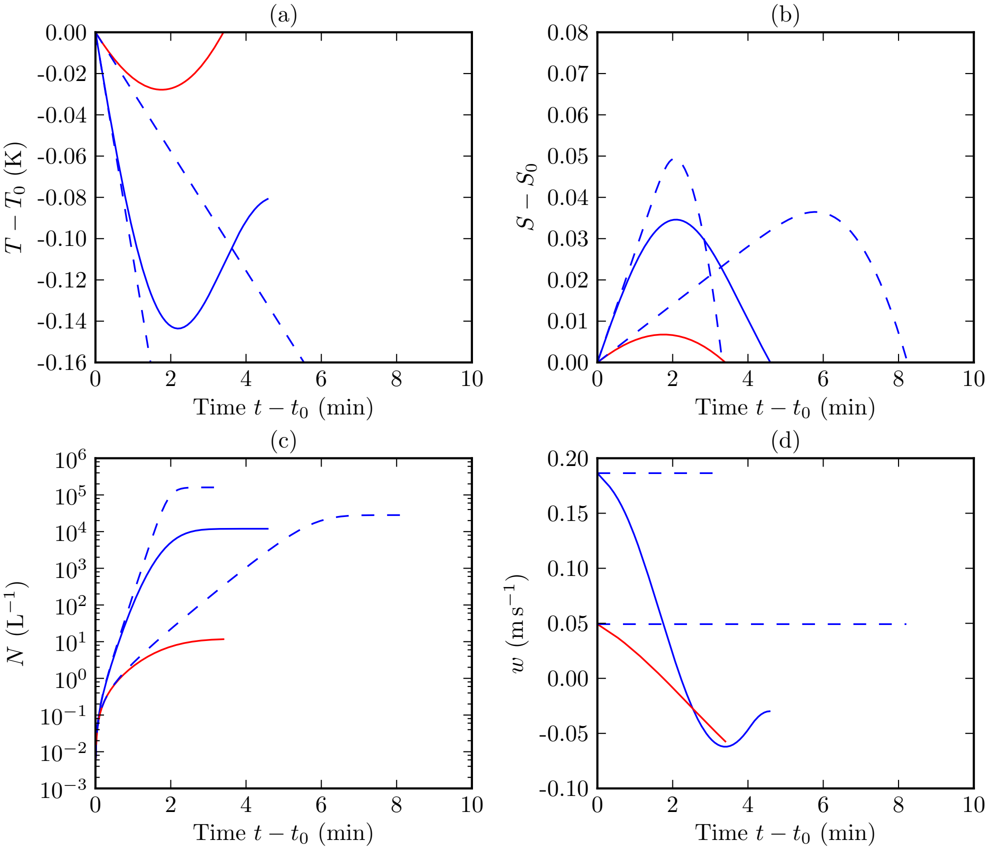

Figure 1: Evolution of temperature, saturation ratio, INC, and vertical velocity during nucleation events simulated using our model. The two solid curves show nucleation events forced by the balloon temperature data, and the two dash curves show the evolution if w remains constant throughout nucleation. Blue curves show vapour-limit events and red curves show temperature-limit events.

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