**Atmospheric
Chemistry
and Physics
Discussions**

# *Interactive comment on* "Dehydration in the tropical tropopause layer estimated from the water vapor match" *by* Y. Inai et al.

**Anonymous Referee #3**

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In general I agree with the two other referees, namely that the paper represents a new approach to an important scientific question, that the data is very interesting, and the analysis is carefully done.

I also agree that 'the text needs careful editing' and that 'in its present form method description, case studies and more general statements are presented in a way that tends to leave the reader confused.'

However, unfortunately I cannot recomment the manuscript for publication in it's present form since I have criticisms with respect to some of the results and methods used in the study. Specifically, I do not agree with the statement (in the abstract and elsewhere):

'Match analysis indicates that ice nucleation starts before the relative humidity with respect to ice (RHice) reaches $207 \pm 81\%$ (1 $\sigma$) and that the air mass is dehy- drated until RHice reaches $83 \pm 30$ % ($1\sigma$).'

I will outline my opinion in the specific comments below.

Specific comments:

1) Page 636, lines 8-10:

'A value of RHice of up to 200 % or more at < 200 K has been reported from studies based on aircraft measurements (Jensen et al., 2005; Krämer et al., 2009).'

The high RHice reported by Jensen et al., 2005 are widely debated to be instrument artifacts and Krämer et al., 2009 observed values up to 200%, but only very sporadically, and never above 200%.

So please reconsider the statement in the conclusions 'The results showed that the estimated upper limit of relative humidity with respect to ice, before the initiation of ice nucleation, is consistent with the super- saturation reported in previous studies.'

For the first part of your sentence, please see also the next comment.

2) Figures 5-7 and respective discussion.

a) In the Figures, I recommend to plot not only the SMR, but also RHice using the SMR of the first measurement. Then you can see the development of the supersatuartion along the air mass trajectory.

b) The conclusion that the supersaturation from the first measurement at SMRmin (124% in Fig. 5, 157% in Fig. 6 and 249% in Fig.7) can be interpreted as 'ice nucleation starts before the relative humidity with respect to ice has reached' those values is not wrong, but somehow useless.

The upper limit of RHice for ice nucleation is the homogeneous freezing threshold

(RHice_hom = Scr_hom * 100), which can be approximated by

Scr_hom = 2.418 - T(K) / 245.68 (Kärcher and Lohmann, 2003, JGR).

The same holds for the mean value of 207%, which is similarly meaningless. If one like to use a mean value of RHice at ice nucleation for temperatures < 200K, one could take 165%, which could be derived from the above formula provided by Kärcher and Lohmann (2003).

I strongly suggest to calculate -and mark in the plot- RHice_hom for the different cases and replace the RHice calculated from SMR_first/SMR_min by these values.

I also suggest to insert RHice_hom in the phrases 'ice nucleation starts (before) latest when the relative humidity with respect to ice has reached XX %' or 'the upper limit of RHice before ice condensation starts' at all places in the paper.

3) a) In the abstract (and elsewhere) you state:

'.. the air mass is dehydrated until RHice reaches 83 ± 30 %..'. Also on

Page 653, line 10 (and elsewhere) you say:

'... dehydration could progress to a RHice state of less than 100 %.'

Though the author's discuss 'possible ... ice growth under unsaturated conditions' on page 653, they could not provide a robust phsical explanation for such a behaviour (I think since there is no ...). Nevertheless, they conclude that dehydration can continue in subsaturated air masses.

I feel that this statement should be removed from the paper. I think that this finding might be caused by temperature biases in the ECMWF data, which are not -but should be- discussed in the paper.

b) Fig. 9 and the respective discussion is connected to the above statement. In this Figure, SMR_second/SMR_min is often < 1. However, this would mean that phase

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transition from gas to ice would occur in subsaturated air masses, which to my knowledge is physically not meaningful. Again, I think it is more likely that biases in the ECMWF temperatures are the reason for this behaviour. This should be discussed in the text.

4) a) Page 653-654: I have problems to understand how you calculate the relaxation time Tau? Could you please explain that in more detail.

b) Page 654, lines 1-2: 'Such calculations are repeated for a given value of RHcri (from 100 % to 250 % at 5 % increments)'.

RHcri could be either the heterogeneous or homogeneous freezing threshold. So it makes no sense to scan it between 100% and 250%. I suggest to do the calculations only for 110% (heterogeneous freezing of efficient ice nuclei), 130% (RHcri of less efficient ice nuclei) and 165% (approximate homogeneous freezing threshold, see above).

c) Page 654, lines 23-24: '... the formation time of ice particles with a mean radius of about 40 $\mu$m (Krämer et al., 2009).'

Krämer et al. (2009) calculated relaxation times between ice formation and the RHice in dynamical equilibrium, not formation time of ice particles... and where you see ice particles with a mean radius of about 40 $\mu$m?

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