REPLY TO COMMENTS BY REVIEWER #3

We are grateful to the thorough reading and constructive comments on our manuscript. We believe we have incorporated all aspects pointed out. The detailed description on the revision follows:

Interactive comment on “Dehydration in the tropical tropopause layer estimated from the water vapor match” by Y. Inai et al.
Anonymous Referee #3
Received and published: 12 April 2013

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.’

Reply:
We have re-organized Section 3 significantly. The major changes are described as follows:

Section 3.1 describes conservative property of ozone in the TTL.
The conservative property of ozone in the TTL is a key point for the match analysis. Therefore, it has been described in Section 3.1 based on the first paragraph in Section 3.2 of the original manuscript.

Section 3.2 describes the use of trajectories and the effectiveness.
To explain the methodology, the first to the third paragraphs of the revised manuscript have been rewritten on the basis of statements in Section 3.1 of the original manuscript. However, some statements have been rearranged to explain clearly the methodology (See also next reply).

To confirm whether the methodology using trajectories is effective or not, the second paragraph in Section 3.2 of the original manuscript has been moved to this section with some revisions.

Section 3.3 describes all screening procedures.
To make order of screening procedures clear, a statement "To move on screening procedures for the remaining problems, we use the "conservative property of ozone" as the second principle. Note that these screening procedures are examined after the first step." has been inserted at p.642, l.23.
The description for screening procedure for other nonspecific factors by using consistency of ozone concentration between the first and the second observations has been added to the last part of this section.

The terminology has been re-defined as follows:
"match" is defined as a case that sounding some air parcel more than once
"match radius" is defined as a distance of the criterion for the match
"match circle" is defined as a circle with the match radius
"match circular area" is defined as a region inside the match circle
"match air segment" is defined as a segment included in the both match circular areas of the first and the second observations
"match air parcel" is defined as a cluster of match air segments
"preliminary match" is defined as a case that connected by a trajectories, i.e., match air parcel.
Following these re-definitions, all statements in the manuscript relevant to above have been revised.

To improve the text, the manuscript has been English proofread.

In addition to above, the description for case studies in the original manuscript has been divided into individual description of each case (case 1 - case 4).

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 ± 81% (1 σ) and that the air mass is dehydrated until RHice reaches 83 ± 30 % (1σ).'
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; Kraemer et al., 2009).'
The high RHice reported by Jensen et al., 2005 are widely debated to be instrument artifacts and Kraemer et al., 2009 observed values up to 200%, but only very sporadically, and never above 200%.

Reply: The statements "or more" and "Jensen et al., 2005" have been removed.

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 supersaturation reported in previous studies.'
For the first part of your sentence, please see also the next comment.

Reply: The statement in Section 6 "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 supersaturation reported in

previous studies." has been changed to "The results showed that the estimated upper limit of relative humidity with respect to ice, before the initiation of ice nucleation, is approximately 146%."
Please see also the following reply.

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 (Kaercher 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 Kaercher 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.

Reply:
According to these comments (the both a and b), we calculated relative humidity over ice using the water vapour mixing ratio of the first measurement and the SMR on the assumption that the water vapour amount

remains that of the first measurement and homogeneous freezing threshold according to Kaercher and Lohmann (2003).
Following these revisions, Figures 5-7 have been re-made to indicate those values.
p.645, l.25-26, "The histories of pressure, potential temperature, SMR," has been changed to "The histories of RHice, SMR,".
A statement "Here, RHice and SMR are calculated using the water vapour mixing ratio of the first measurement, assuming that the water vapour amount remains that of the first measurement. Figure 5 (a) also shows the homogeneous freezing threshold (RHhom) according to Kaercher and Lohmann (2003), which depends on the temperature of the match air parcel; this threshold is considered as the upper limit of RHice." has been inserted in the first paragraph of Section 4.1.
The caption of Fig. 5: "The figure shows the time evolutions of (a): relative humidity over ice (RHice) on the assumption that the water vapour amount is unchanged from the first measurement, along with the uncertainties (blue) and the homogeneous freezing threshold (RHhom; purple),"
p.647, l.24-28, The statement has been changed to "The RHice shows a maximum value of 249%, far exceeding the RHhom, with an uncertainty of -37%/+38% during the advection. Therefore, the match air parcel is expected to be dehydrated."

According to comment b), we have plotted approximate homogeneous threshold (= 1.65) in Fig. 9 and the value of homogeneous threshold has been taken into account our estimate of the upper limit of RHice before ice condensation starts.
Following these revisions, a statement "For those matches in which the maximum of RHice during the advection exceeds the RHhom, the value of RHhom is used except for RHice because it is considered the upper limit of su-persaturation." has been inserted at p.650, l.4.
"The dashed black lines indicate approximate homogeneous freezing

threshold (= 1.65)." has been added to the caption of Figure 9.

The upper limit of RHice is re-calculated as the value of 146 +/-19% considering the both homogeneous freezing process and ECMWF temperature bias (see also "other revisions").

p.650, l.7, "146 +/-19%"

p.650, l.10, "146 +/-19%"

p.652, l.23, "146 +/-19%"

p.650, l.9, "75 +/-23%"

p.650, l.11, "75 +/-23%"

p.653, l.2, "75 +/-23%"

A statement in the bottom of the Abstract has been changed to "The statistical features of dehydration for the air parcels advected in the lower TTL are derived from the matches. The threshold of nucleation is estimated to be 146 ± 19% (1σ) in relative humidity with respect to ice (RHice), while dehydration continues until RHice reaches about 75 ± 23% (1σ) in the altitude region from 350 to 360 K. The efficiency of dehydration expressed by the relaxation time required for the supersaturated air parcel to approach saturation is empirically determined from the matches. A relaxation time of approximately one hour reproduces the second water vapour observation reasonably well, given the first observed water vapour amount and the history of the saturation mixing ratio during advection in the lower TTL."

A statement in the lower part of the Conclusion has been changed to "The results showed that the estimated upper limit of relative humidity with respect to ice, before the initiation of ice nucleation, is approximately 146%. It is suggested that sub-grid-scale temperature fluctuations (or possibly nitric acid trihydrate or other particles) influence dehydration associated with horizontal advection in the lower TTL. The efficiency of dehydration was defined as the relaxation time taken for a supersaturated air parcel to approach the saturation state by the nucleation and removal of ice particles. It is suggested that the dehydration process associated with horizontal advection is efficiently driven in the lower part of the

TTL. These findings may improve our understanding of cloud-microphysical processes in the TTL."

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 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.

Reply:
Because the significance of this result is not strong enough to claim that ice growth happens at subsaturated condition, a statement "Statistically this level of significance is not large enough to claim that the threshold is below 100%. Therefore, although the derived value is far from 100% RHice, the significance is not strong enough to claim that ice growth happens at subsaturated conditions." has been inserted at p.653, l.4.

Following this revision, a statement at p.658, l.4-6 has been changed to "Given that the dehydration around this level determines the amount of water vapour entering the stratosphere, and to improve on the statistical results from this study, additional observation data and more match events at this level are required."

However, we think that there is a possibility to dehydrate under water saturated state if it is assumed that there are NAT particles in the TTL. Therefore, the statement "In such a case, although the interpretation of our results would become even more complex, it may be possible that the dehydration could progress to a RHice state of less than 100%." has been changed to "Shibata et al. (2012) show that the critical temperature of NAT formation is higher than that in the TTL. In such a case, although the interpretation of our results would become even more complex, it may be possible that the dehydration could progress to a state of RHice less than 100% until the NAT saturation state, due to the absorption of water vapour into NAT particles."

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.

Reply: To make it clear, a sentence "For example, in the case of RHcri = 110%, the air mass starts to be dehydrated, with the rate of dehydration depending on τ when the RHice exceeds 110%." has been inserted.

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).

Reply: Revised as suggested.
Following this revision, Panels (b), (c), and (d) in Fig. 10 have been modified and Fig. 11 has been replaced by Table 2.
p.654, l.1–3: a statement "Such calculations are repeated for a given value of RHcri (from 100% to 250 % at 5 % increments) to identify the value of that is consistent with the second observation of water vapour." has been changed to "Such calculations are repeated for a given value of RHcri (values of 100%, 110%, 130%, and 165%) to identify the value of τ that is consistent with the second observation of water vapour. These values of RHcri correspond to the water saturation, heterogeneous freezing of efficient ice nuclei, heterogeneous freezing of less efficient ice nuclei, and approximate homogeneous freezing thresholds, respectively."

c) Page 654, lines 23-24: '... the formation time of ice particles with a mean radius of about 40 μm (Kraemer et al., 2009).'
Kraemer 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 μm?

Reply: The statement in the original manuscript "If the value of RHcri is assumed to be the RHice values of supersaturation reported previously (e.g. Koop et al., 2000; Gao et al., 2004; Shibata et al., 2007; Jensen et al., 2008; Kraemer et al., 2009; Selkirk et al., 2010; Inai et al., 2012), then the corresponding values of are estimated to be 2-3 h, and these values are nearly equivalent to the formation time of ice particles with a mean radius of about 40 μm (Kraemer et al., 2009)." and a sentence in the bottom of the Abstract "The relaxation time is found to range from 2 to 3 h, which is consistent with previous studies."
has been deleted.

Other revisions:

According to the companion paper, temperature bias of ECMWF has been taken into account our SMR estimates. Following this revision, the statement "In this altitude region, we find that the ECMWF temperature has a cold bias of 2 K on the isentropic surfaces ranging from 355 to 360 K (Hasebe et al., 2013). For all subsequent analyses, this bias is taken into account when estimating SMR along the trajectories in this altitude region." has been inserted at p.646, l.24.

Following this revision in addition to the introduction of RHice and RHhom, p.646, l.24-p.647, l.8, "The time evolution of SMR has small perturbations with an SMRmin value of 8.9 ppmv at about 5 hours before the second observation. At this time, the temperature of the air mass is 197.4 K. This SMRmin value is smaller than the water vapour mixing ratio of the first observation. The RHice during advection indicates a maximum value of RHice of 115% with an uncertainty of ±21%. Because the match air mass is dehydrated, this case indicates that ice nucleation must have started before the RHice reached 115%. As this value is much smaller than RHhom, it might correspond to the heterogeneous freezing threshold. A comparison between the second water vapour observation and SMRmin suggests that dehydration continued until RHice reached 60% with an uncertainty of ±16%. If the dehydration does not proceed to less than 100% of RHice, the temperature of the air mass must have decreased by about 3.2 K from the temperature 197.4 K, when the air mass is coldest, falling to 194.2 K on the 356.4 K potential temperature surface."

Right panel of Figure 3 has been re-made because the dashed lines in the original figure were wrong.

Center panel of Figure 3 has been re-made to be reader-friendly.

As a result of quality recheck of sonde data, the number of matches decreased to 107.

Thus, a statement in p.645, l.4-7 has been changed to "Figure 4 shows scatter plots of the first and second observations of the ozone and water vapour mixing ratios for 107 matches (i.e., all of the matches listed in Appendix C). Note that this number includes matches of observational pairs and potential temperature levels. Among the 107 matches there are 25 different observational pairs."

Fig.3 caption l.1: "right panel" has been corrected to "left panel"

Panel (d) of Fig.7: the error bars have been corrected.

Thank you very much again for your valuable comments and suggestions.