The authors thank reviewer #1 for the many fruitful comments which were helpful to improve the paper. All changes to the paper are highlighted in red color. Point by point answers to your comments are reported below.

Major comments

- - *I think it is necessary to include more background in the introduction. This may include, but not limited to, characteristics of the monsoon anticyclone and why the transport in and near the Asian monsoon anticyclone is important in global scale circulation.*
  We included more information about the Asian monsoon and to transport pathways to better introduce the background and to motivate this study. For example, we now discuss the papers by Randel and Jensen, 2013, Ungermann et al. 2016.
- - *It is not clear what the scientific goals are. Why is the quantitative analysis of changes in the water vapor amount in the extra-tropical lower stratosphere important? Is 10*
  We added more information about water vapor and motivate why the knowledge about small changes of water vapor concentration in the UTLS are important for global climate like the study by Solomon et al. 2010.
- - *Also, explain why in-situ measurements of chemical species are necessary and unique in this study compared to the previous studies. What are the limitations of using satellite measurements of water vapor in this type of study?*
  We already explained that especially satellites, like limb sounders, have a coarse vertical resolution. We added a sentence to the introduction to stronger motivate the use of in-situ instrumentation: Especially in the tropopause region this coarse resolution can smooth the strong vertical gradient of water vapor at the tropopause and may lead to both an over- or underestimation of the water vapor concentration in the lower stratosphere. Here, we present high resolution and precise in-situ water vapor and methane measurements in the northern lower stratosphere performed in August and September 2012.
- - *Explain why water vapor and methane are used and what we can learn from those. What are the lifetimes of water vapor and methane in the troposphere and stratosphere? What other species were measured during the field campaign?*
  First, water vapor is a major green house gas and the variability of water vapor at tropopause altitudes has a strong impact on surface climate (Solomon et al. 2010, Riese et al. 2012). This is a major motivation of our study which is now included in the introduction. Second, Methane has a long lifetime of around 8.9 years in the troposphere and even longer in the lower stratosphere (Wuebbles et al. 2002), whereas water vapor has a very short life time of about 9 days (Hartmann 2015) due to its strong temperature dependence. Therefore water vapor is very variable with respect to transport history, in contrast to methane. This is also the reason why methane shows a stronger correlation to the MON tracer compared

to water vapor. Both correlation coefficients are now included in the text. In addition, both trace gases are important green house gases and small changes in their concentration in the UTLS have strong climatic relevance. We included now the life times and more motivation concerning the choice of trace gases in the introduction and section 3.3.

- *- Why is CLaMS used here? What is unique about this model? And are there any modeling studies on the similar subject?*
  For the scientific issues addressed here, a model is required that provides a good resolution in the UTLS and, in particularly, describes well quasi-isentropic transport in the UTLS as well as transport in the presence of strong tracer gradients. Due to its Lagragian transport on isentropic surfaces with a physically based mixing scheme (that avoids the classic problem of numerical diffusion in Eulerian transport schemes) CLaMS is indeed very well suited for the scientific questions addressed in our paper. CLaMS is indeed widely used like Vogel et al., ACP, 2015, Vogel et al., ACP, 2016, Ploeger et al., JGR, 2013, Pommrich, GMD, 2011, Pommrich, GMD , 2014, Vogel et al., JGR, 2012, Konopka et al., ACP, 2010, Hoppe et al., GMD, 2014. Some (not all) of these studies are now cited in the text.

- *- Writing can be improved. Careful selection of words should help clarifying some of the ideas. I think simple grammatical errors can easily be avoided by putting more effort into proof reading.*
  We agree with the reviewer and thank her/him for the many good and helpful suggestions. We carefully checked the writing and sent this manuscript to the language office at our institution for proof reading by a native speaker.

Specific comments

- *- P1, L1 – from Asia -> locally*
  Changed.

- *- P1, L2 water vapor (H2O) of about 0.5 ppmv (11*
  Changed.

- *- P1, L4 by in-situ instrumentation in the northern hemisphere is redundant*
  Changed.

- *- P1, L4 What are the full names of TACTS and ESMVal?*
  We added the full names in the text of Section 2 and in the abstract.

- *- P1, L5 this water vapor and methane increase -> the increased water vapor and methane, with the help of -> using*
  Changed.

- *- P1, L6 transport model (CLaMS)*
  Changed.

- *- P1, L9 influence of air. . .region. -> influence due to the Asian monsoon anticyclone.*
  Changed.
- *- P1, L10 remove between*
  Changed.
- *- P1, L15 gases like water vapor and methane -> gases, such as, water vapor and methane,*
  Changed.
- *- P1, L19 with a low -> with low*
  Changed.
- *- P2, L3 higher values. . .temperature The meaning of this is not clear.*
  Changed.
- *- P2, L10 by a potential -> by strong potential*
  We did not add the word strong, because the transport barrier is often rather leaky as shown in Ploeger et al. 2015 and the gradient is much lower than at the polar vortex for example.
- *- P2, L15 (and others) Eddy shedding -> eddy shedding*
  Changed.
- *- P2, L18 AURA-MLS -> the Aura Microwave Limb Sounder (MLS)*
  Changed.
- *- P2, L22 . . .altitude range -> add a few references here.*
  Changed.
- *- P2, L27 This study bases on -> This study is based on the (or In this study, the data collected. . . are analyzed. . .)*
  Changed.
- *- P2, L27 Also what other species were measured during TACTS and ESMVal?*
  There were other species like CO, N2O, and O3 measured during TACTS and ESMVal. The results are published in Müller et al. 2016. In this study, we particularly highlight the importance of the water vapor transport associated to the AMA.
- *- P2, L28 (and others) northern hemisphere -> Northern Hemisphere*
  Changed.
- *- P2, L33 A reference for CLaMS is necessary here. Also, why is CLaMS perfect for this type of research?*
  We included a reference in the text and put more information as well as a brief motivation in the sub section about CLaMS. See also the answer to your general comment #5.

- *- P3, L1 location -> locations*
  Changed.

- *- P3, L5 bases -> is based*
  Changed.

- *- P3, L11 The full name for TRIHOP should be given here. Also bases -> is based*
  Both is changed.

- *- P3, L20 is driven by ERA-Interim -> is driven by the European Reanalysis (ERA)- Interim*
  Changed.

- *- P3, L22 What are the definitions of emission tracers?*
  The "emission tracers" are artificial tracers with no chemical depletion, but are transported within the atmosphere. During this transport mixing is applied, thus these tracers can be diluted by emission tracers from different source regions. More information can be found in the cited literature of Vogel et al. 2015 and 2016.

- *- P3, L24 concentrations -> concentration*
  Changed.

- *- P3, L27 I do not think PV is measured by the MLS.*
  You are right. We changed the text according to your suggestion.

- *- P3, L27 proxy -> proxies, allow for transport -> allow transport*
  Changed.

- *- P3, L29-30 Either add references here or explain how this can be done.*
  Beside the Asian monsoon e.g. typhoons can uplift air masses from the ground into the upper troposphere in the vicinity of the monsoon system. These air masses are dragged by the AMA and loop around the the outer part. A reference is added to the text.

- *- P3, L30 regions are -> regions considered are*
  Not changed. Because this are the important source regions found in Vogel et al. 2016.

- *- P4, Table 1 A map can be added here. Or this table can be replaced by a map.*
  We decided to skip the map, because it is already published in the Vogel et al. 2015 and Vogel et al. 2016 and would not provide more information than the table.

- *- P4, L3-4 that these. . .stratosphere -> that these regions do not make a significant contribution to the lower stratosphere*
  Changed.

- *- P4, L7-8 In contract to three -> In contrast to the three, temperature development -> changes in the temperature*
  Changed.

- - *P4, L8 A reference is needed at the end of this sentence.*
  We mitigated the formulation of this sentence to: However, the position of an air parcel and the changes in the temperature along the trajectory can be tracked with sufficient accuracy over several days or even weeks. In Addition, we included two studies of Vogel et al. 2014 and Rolf et al. 2015 which successfully used long trajectories. c
- - *P4, L10 remove of the ECMWF*
  Changed.
- - *P4, L22 the tropical Pacific I am not sure if this has been mentioned previously.*
  It is mentioned in the Section 2.3, where the contributing artificial tracer source regions are stated. Nothing changed here.
- - *P5, L1 reinforce the hypotheses What does this mean?*
  We rephrased the text to make it more clear.
- - *P5, L6 Figures 1a and b -> Figures 1a and 1b (also P9, L8)*
  Changed.
- - *P5, L7 along equivalent latitude -> along the equivalent latitude. Also explain how equivalent latitude is defined.*
  Changed and we added "PV based" as definition for equivalent latitude.
- - *P5, L8 What does tropospheric influenced air mean? Does this mean the air is mixed with tropospheric air?*
  Yes, exactly. In this case we mean stratospheric air masses which are influenced by tropospheric air masses by mixing. We rephrased the text to make it more clear.
- - *P5, L11 have a similar. . .extent. . . -> have similar vertical and horizontal extents*
  Changed.
- - *P5, L16 at a PV value -> at PV values, remove in the time*
  Changed.
- - *P5, L19 All air -> All the air*
  Changed.
- - *P5, L22 The distribution -> The frequency distribution*
  Changed.
- - *P5, L25 vapor concentration -> vapor concentration between the two phases*
  Changed.
- - *P5, L28 It might be easier to refer a figure in Vogel at al., instead of a page number.*
  You are right, we changed the reference to the Figure instead of the page number.

- *- P5, L32 Figures 2a, b -> Figures 2a and 2b*
  Changed.

- *- P5, L33 remove relatively*
  Changed.

- *- P6, Figure 1 a)-d) can be written on the top left instead of bottom left (also in other figures).*
  Changed.

- *- P6, L3 Figure 2d) -> Figure 2d*
  Changed.

- *- P6, L4 from phase 2 to phase 1 -> from phase 1 to phase 2*
  Changed.

- *- P8, L2 enhancements -> enhancement*
  Changed.

- *- P8, L5 Only few -> Only a few*
  Changed.

- *- P8, L7 Explain what core region means.*
  The core region of the scatter plot is defined by the 75% percentile of the frequency distribution (black contour). It is now written in the text.

- *- P8, L10 the slope of A linear regression line can be added in Fig. 3a.*
  We added a linear regression to the plot to better highlight the slope of the correlation.

- *- P8, L18 What does the imprint on water vapor mean here?*
  It is directly connected to the sentence before. "The amount of water vapor which is transported from the troposphere into the stratosphere is strongly coupled with the Lagrangian cold point (LCP), where typically the water vapor is dehydrated close to the saturation mixing ratio by ice crystal formation and subsequent sedimentation. Thus, the amount of water vapor in these air masses is nearly preserved after passing the LCP in the tropical, sub-tropical and mid-latitude stratosphere." The imprint means the minimum saturation mixing ratio along the temperature. We make it clear by changing the wording to "these imprint".

- *- P9, L1 that Eddy shedding -> that the eddy shedding*
  Changed.

- *- P9, L5 A reference can be added at the end of this sentence.*
  We added two references which address this point.

- *- P10, L2 between both phases -> from phase 1 (Aug) to phase 2 (Sep)*
  Changed.

- *- P10, L7 model study of -> modeling study of*
  Changed.

- *- P10, L20 gives observational -> gives an observational*
  Changed.

References:

- Hartmann, Dennis: Global Physical Climatology (Second Edition), Elsevir, ISBN: 978-0-12-328531-7,p. 399, 2015

- Hoppe, C. M., Hoffmann, L., Konopka, P., Grooß, J.-U., Ploeger, F., Günther, G., Jöckel, P., and Müller, R.: The implementation of the CLaMS Lagrangian transport core into the chemistry climate model EMAC 2.40.1: application on age of air and transport of long-lived trace species, Geosci. Model Dev., 7, 2639-2651, https://doi.org/10.5194/gmd-7-2639-2014, 2014.

- Konopka, P., Grooß, J. U., Günther, G., Ploeger, F., Pommrich, R., Müller, R., and Livesey, N.: Annual cycle of ozone at and above the tropical tropopause: observations versus simulations with the Chemical Lagrangian Model of the Stratosphere (CLaMS), Atmospheric Chemistry and Physics, 10, 121132, https://doi.org/10.5194/acp-10-121-2010, 2010.

- Müller, S., Hoor, P., Bozem, H., Gute, E., Vogel, B., Zahn, A., Bönisch, H., Keber, T., Krämer, M., Rolf, C., Riese, M., Schlager, H., and Engel, A.: Impact of the Asian monsoon on the extratropical lower stratosphere: trace gas observations during TACTS over Europe 2012, Atmospheric Chemistry and Physics, 16, 10 57310 589, https://doi.org/10.5194/acp-16-10573-2016, 2016.

- Ploeger, F., Günther, G., Konopka, P., Fueglistaler, S., Müller, R., Hoppe, C., Kunz, A., Spang, R., Grooß, J. U., and Riese, M.: Horizontal water vapor transport in the lower stratosphere from subtropics to high latitudes during boreal summer, Journal of Geophysical Research- atmospheres, 118, 81118127, https://doi.org/10.1002/jgrd.50636, 2013.

- Ploeger, F., Gottschling, C., Griessbach, S., Grooß, J. U., Günther, G., Konopka, P., Müller, R., Riese, M., Stroh, F., Tao, M., Ungermann, J., Vogel, B., and von Hobe, M.: A potential vorticity-based determination of the transport barrier in the Asian summer monsoon anticyclone, Atmospheric Chemistry and Physics, 15, 13 14513 159, https://doi.org/10.5194/acp-15-13145-2015, 2015.

- Pommrich, R., Müller, R., Grooß, J.-U., Konopka, P., Günther, G., Pumphrey, H.-C., Viciani, S., D'Amato, F., and Riese, M.: Carbon monoxide as a tracer for tropical troposphere to stratosphere transport in the Chemical Lagrangian Model of the Stratosphere (CLaMS), Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmdd-4-1185-2011, 2011.

- Pommrich, R., Müller, R., Grooß, J. U., Konopka, P., Ploeger, F., Vogel, B., Tao, M., Hoppe, C. M., Günther, G., Spelten, N., Hoffmann, L., Pumphrey, H. C., Viciani, S., DAmato, F., Volk, C. M., Hoor, P., Schlager, H., and Riese, M.: Tropical troposphere to stratosphere transport of carbon monoxide and long-lived trace species in the Chemical Lagrangian Model of the Stratosphere (CLaMS), Geoscientific Model Development, 7, 28952916, https://doi.org/10.5194/gmd-7-2895-2014, 2014.

- Randel, W. J. and Jensen, E. J.: Physical processes in the tropical tropopause layer and their roles in a changing climate, Nature Geoscience, 6, 169176, https://doi.org/10.1038/ngeo1733, 2013.

- Riese, M., Ploeger, F., Rap, A., Vogel, B., Konopka, P., Dameris, M., and Forster, P.: Impact of uncertainties in atmospheric mix- ing on simulated UTLS composition and related radiative effects, Journal of Geophysical Research-atmospheres, 117, D16 305, https://doi.org/10.1029/2012JD017751, 2012.

- Rolf, C., Afchine, A., Bozem, H., Buchholz, B., Ebert, V., Guggenmoser, T., Hoor, P., Konopka, P., Kretschmer, E., Müller, S., Schlager, H., Spelten, N., Suminska-Ebersoldt, O., Ungermann, J., Zahn, A., and Krämer, M.: Transport of Antarctic stratospheric strongly dehy- drated air into the troposphere observed during the HALO-ESMVal campaign 2012, Atmospheric Chemistry and Physics, 15, 91439158, https://doi.org/10.5194/acp-15-9143-2015, 2015.

- Solomon, S., Rosenlof, K. H., Portmann, R. W., Daniel, J. S., Davis, S. M., Sanford, T. J., and Plattner, G. K.: Contributions of Stratospheric Water Vapor to Decadal Changes in the Rate of Global Warming, Science, 327, 12191223, https://doi.org/10.1126/science.1182488, 2010.

- Ungermann, J., Ern, M., Kaufmann, M., Muller, R., Spang, R., Ploeger, F., Vogel, B., and Riese, M.: Observations of PAN and its con- finement in the Asian summer monsoon anticyclone in high spatial resolution, Atmospheric Chemistry and Physics, 16, 83898403, https://doi.org/10.5194/acp-16-8389-2016, 2016.

- Vogel, B., Günther, G., Müller, R., Grooß, J. U., Afchine, A., Bozem, H., Hoor, P., Krämer, M., Müller, S., Riese, M., Rolf, C., Spel- ten, N., Stiller, G. P., Ungermann, J., and Zahn, A.: Long-range transport pathways of tropospheric source gases originating in Asia into the northern lower stratosphere during the Asian monsoon season 2012, Atmospheric Chemistry and Physics, 16, 15 30115 325, https://doi.org/10.5194/acp-16-15301-2016, 2016.

- Vogel, B., Günther, G., Müller, R., Grooß, J. U., and Riese, M.: Impact of different Asian source regions on the composition of the Asian monsoon anticyclone and of the extratropical lowermost stratosphere, Atmospheric Chemistry and Physics, 15, 13 69913 716, https://doi.org/10.5194/acp-15-13699-2015, 2015.

- Vogel, B., Günther, G., Müller, R., Grooß, J. U., Hoor, P., Krämer, M., Müller, S., Zahn, A., and Riese, M.: Fast transport from Southeast Asia boundary layer sources to northern Europe: rapid uplift in typhoons and eastward eddy shedding of the Asian monsoon anticyclone, Atmospheric Chemistry and Physics, 14, 12 74512 762, https://doi.org/10.5194/acp-14-12745-2014, 2014.

- Wuebbles, D. J. and Hayhoe, K.: Atmospheric methane and global change, Earth-science Reviews, 57, PII S00128252(01)00 0629, https://doi.org/10.1016/S0012-8252(01)00062-9, 2002.

The authors thank the reviewer #2 for the many comments which were very helpful to improve the paper. All changes of the paper are highlighted in red color. Point by point answers to your comments are reported below.

Major comments

- *(1)My main concern is the novelty or substance of the paper. Moistening of the Ex-LS via the AMA has been reported before (Randel and Jensen, 2013), even based on in-situ data from the same 2012 campaign and a similar methodology (Müller et al., 2016, Vogel et al., 2016). Vogel et al. (2016) also already quantified the moistening for the entire monsoon season of 2012. Although Rolf et al. provide additional and detailed analyses, new findings should be brought out better in the paper.*
  We agree that the paper Vogel et al. (2016) / Müller et al. (2016) and our present study are strongly connected. However, the major contribution of the present study is the investigation of in-situ measurements of water vapor with a *high vertical resolution*. Clearly, vertical resolution is the key in UTLS processes and remote sensing instruments like MLS have clear deficits in this respect (Santee, JGR, 2017). Moreover, water vapor and changes in its distribution in the UTLS impacts the radiation budget and thus climate. Therefore, the knowledge about water vapor transport pathways, change rates, and sources is highly important. We added more information about this in the introduction.

  Further, with this study we connect the in-situ measurement directly to the CLaMS model simulations and can partially confirm the moistening found by Vogel et al. 2016 with the model and with the MLS satellite data. Therefore, highly precise in-situ measurements are a justification and constitute a different method in estimating the moistening. The study by Müller et al., 2016 is focused in detailed on other trace gases like CO, O3, and N2O, which are important as well, but the study of Müller et al. 2016 is not complete in investigating transport pathways concerning water vapor.

Methodology:

- *(1) I dont really see the point in using tracers of airmass origin here. The tracer(s) described in the paper do not seem to be reliable indicators of AMA air, and do not provide additional relevant information as compared to back-trajectories.*
  It is important to note here that CLaMS transport is Lagrangian (i.e. by trajectories) so that any CLaMS tracer (including air mass origin) will be transported mainly Lagrangian (i.e. by trajectories) with an *additional* impact of mixing. Thus, back-trajectories show "only" the history and the origin of an airmass with the underlying advection. The origin tracer are constantly emitted at the ground, are chemical inert, and are mixed in the atmosphere, which is an important difference to trajectories. Especially mixing processes like eddy shedding, which is one of the crucial transport pathways of monsoon air, can be better quantified.

This is already shown by Vogel et al. 2015, 2016 where they showed that the origin tracers provide information of airmass composition of different source regions, which can be better associated with the true airmass composition compared to back-trajectories alone. In addition, with the combination of origin tracer we can better confirm the studies of Vogel et al. 2016 and Ploeger et al. 2013 and directly associate the hydration found in the measurements with the increase of airmass contribution from the Asia.

- *(2) The choice of the two time periods for detecting changes to H2O and CH4 seems to be rather arbitrary. The delta should be discussed in the context of the transport time scales involved, which might be available from back trajectories. Transport time scales also seem to be the key for relating the results of this paper to the numbers found by Vogel et al. (2016).*
  The periods and the two week break in between are the result of the campaign planning and are in this way arbitrary. But they fit perfectly to the period of the strongest increase in water vapor in the Ex-LS found by Vogel et al 2016 in CLaMS water vapor and in the MLS observation at 380 K and 400 K. Between the two phases, the strongest increase of Ex-LS water vapor is the result of a pronounced eddy-shedding event which occurred on 20 September 2012 (Vogel et al., 2014, 2016) and matches perfectly the separation of the two phases. We added more text in the discussion part (Sec. 3.1) and in the conclusion section of the paper, to relate the increase observed by Vogel et al. 2016 to the increase found with the in-situ measurements.

- *(3) Convection is an important process when it comes to the AMA, but is notoriously hard to capture in large-scale models. Please discuss, how you consider convection.*
  Convection is the major processes to transport air masses vertically from the ground into the upper troposphere in the Asian monsoon region. In the model simulations shown here, vertical transport is taken into account as represented in ERA-Interim including latent heat related transport (for pressure less than ≈300 hPa). Thus, the very small-scale rapid uplift in convective cores cannot included in CLaMS simulations. However, for the questions addressed here, which are concerned about large scale uplift in the monsoon, these small-scale features might be less relevant. In particular, previous studies using 3-D CLaMS simulations or trajectory calculations (e.g., Ploeger et al., 2010, 2015; Pommrich et al., 2014; Vogel et al., 2014, 2015; Müller et al., 2016; Ungermann et al., 2016; Konopka et al., 2016), in comparison with satellite or in situ measurements, show that ERA-Interim data are well suited for studying transport processes in the vicinity of the Asian monsoon anticyclone and in the tropical tropopause layer.

- *(4) The term statistic seems to have been added to the corresponding terms in response to the initial review, but I still do not see actual statistical calculations. Please provide details on the statistical methods used for those obviously non-Gaussian distributions, mark significant points/ranges where applicable, provide*

correlation coefficients, confidence intervals etc. This might not apply to each and every use of statistical terms (correlation, significance, . . .), but you should make clear where it is normal language rather than backed by maths.
You are right, this is an important information. We added correlation coefficients and p-values to show the significance at the important parts in the text. In the Section 3.1 we added: The difference in the mean water vapor mixing ratio between the two phases indicates a statistical significant (Mann-Whitney-U-Test with p-value $< 10^{-100}$) moistening of the Ex-LS of about 0.5 ppmv within a time period of less than a month.

In Section 3.3 we provide correlation coefficients and further discuss them: The associated correlation coefficients for water vapor and methane are 0.79 and 0.67 for phase 1 and phase 2, respectively. The correlation coefficients for measured water vapor and modeled MON tracer are 0.27 in phase 1 and 0.40 in phase 2 and for methane and MON tracer 0.41 for phase 1 and 0.63 for phase 2. All correlation coefficient (calculated according to Pearson) have a very high significance with p-values of nearly zero ($< 10^{-18}$) rejecting the null hypothesis of an uncorrelated dataset. In fact, the correlation for water vapor and methane is obvious higher compared to the correlation with the MON tracer, which corroborates the consistency between both in-situ measurements. But also shows the limitations of CLaMS to reproduced every small scale feature in the measurements. However, the correlation for the model based tracer and the measurements is still quite satisfying and strongly supports the hypothesis that the increase in water vapor is correlated to airmass affected by the AMA.

- *(5) Flight paths determine the region of study and should be shown. Citing Müller et al. (2016) only helps partially, because they analyse slightly different periods.*
  We see the point and included a map with the flight paths and attribution to the respective phase into the manuscript.

- *(6) Please distinguish between "concentration" and "mixing ratio" throughout the paper. For instance, "ppbv" is a unit for mixing ratios (better: nmol/mol). The SI unit for molar concentration is mol/m3, for number concentration 1/m3.*
  Yes, that is totally right. We carefully scanned the manuscript and exchanged the wording according to the suggestion.

Specific comments

- *P1L8: Are those the exact numbers? Otherwise please use about*
  We added "about" in the text.

- *P1L13: exclusively -> mostly/predominantly*
  Changed to predominantly.

- *P2L9: AMA is leaky*
  We rephrased the text to make it more clear that the AMA constitute as more a weak transport barrier.

- *P2L29: ESMVal went around Africa*
  This is true. The campaign was conducted to get a full meridional cross section of the earth. So there were also measurements in the northern hemisphere. And for the for the total number of measurements the most part of both campaigns took place above Europe. Therefore we keep the current wording with "mainly over Europe and the Atlantic Ocean".

- *P2L30: Why those dates? The Monsoon starts in June/July, but your approach seems to be based on the assumption that less trajectories from the AMA have reached the flight paths at the beginning of Sep compared to the end of Sep. Please discuss the time scales for transport from the AMA to the measurements.*
  We choose this combination of dates because of the clear difference in their water vapor distribution in the Ex-LS. You are right, that the monsoon season starts earlier but concerning transport into the Ex-LS the later phase between August and September seems to be more important. This is also visible in Fig 15. of Vogel et al. 2016. The water vapor mixing ratio increases most strongly in the time between August and September. This study confirm the CLaMS modeling from Vogel et al. 2016 results quite nicely! We included this brief motivation for the selection if dates in the text.

- *P3L12: explain acronym*
  Changed.

- *P3L28: Does that mean that this type of emission tracer preferentially ends up in the AMA? For using MON as a proxy for the AMA, it is not enough to show that MON is the main source of AMA air. Additionally, MON must not end up in significant amounts outside the AMA.*
  In this study, we focus on all air masses affected by the AMA. This includes air masses within the anticyclone as well as air masses which are lifted to this altitudes and circulate around the AMA at the outer edge. Vogel et al. 2016 explains this in more detail: Maximum percentages for the emission tracer for south-east Asia/tropical Pacific Ocean are found at the edge of the Asian monsoon anticyclone, indicating the transport of these air masses around the outer edge of the Asian monsoon anticyclone outside of a PV-based transport barrier (Ploeger et al., 2015) as discussed by Vogel et al. (2015). Thus, air masses from southeast Asia/tropical Pacific Ocean are found within a widespread area around the anticyclone caused by the large-scale anticyclonic flow in this region acting as a large-scale stirrer. Moreover, large contributions of these emission tracers are found in the tropics associated with deep uplift outside of the monsoon region.

  Also note that the composition of the AMA is not static, rather the composition is influenced by a steady upward flux of boundary layer air, which leads to a different composition of the AMA air over the monsoon period (see Fig. 8 in Vogel et al. 2015 and the related discussion).

- *P4L17: Does possible supersaturation play a role for these analyses?*

Supersaturation is needed to initiate ice formation, but is reduced to the saturation mixing ratio within short time. This occurs typically at the coldest point along the trajectory. With additional ice crystal sedimentation the water vapor mixing ratio ends up at the saturation mixing ratio without ice in a first order approximation. Especially, with the long transport times of several days in the lower stratosphere supersaturation and ice formation don't play a role. Thus, for this type of analysis we don't see a need to account for supersaturation.

- *P4L20: Consider rewording to make clear that both conditions must be met.*
  It is now rephrased.

- *P5L9: The motivation for choosing potential temperature difference to the local thermal tropopause is not clear to me. Water vapour depends on temperature, but the thermal tropopause is based on a lapse rate. Also, potential temperature is good for characterizing isentropic transport, but not necessarily temperature history.*
  The thermal tropopause in the sub-tropics and mid-latitude is found to be a good indicator to mark the critical level of temperature gradient and sharp relative humidity change as shown by e.g. Pan and Munchak 2011. Thus separates the moist tropospheric regime from the dry stratospheric regime. Therefore the thermal tropopause seems to be a good estimate concerning the water vapor gradient. It is true, that isentrophes do not show the temperature history, but we decided to use this coordinate mostly associate the water vapor distribution to the underlying isentropic transport. We added this information to the text to make it clearer.

- *P5L10: Please briefly motivate using equivalent latitude*
  We included a short motivation in the text.

- *P5L11: consider choosing the contours in Fig. 1 to display the threshold used in the text*
  We tried to put contour lines into these figures, but they are getting confusing and messy because the water vapor distribution is not that smooth. Therefore we decided to skip additional contour lines in these plots. It is better visible with just the colors from the colorbar.

- *P5L15: Fig. 1 only shows equivalent latitude as vertical coordinate. Were similar longitudinal regions sampled during both phases?*
  We show equivalent latitude as horizontal coordinate. The most data were collected in an longitude range between 15W and 20E during both phases (phase1 86% and phase2 75% of the measurements). In phase 1 also measurements more in westerly direction up to 24W and in phase 2 more in easterly direction up to 60E where conducted. So we think that both phases are comparable. We also think that the exact longitudinal coverage is of minor importance because of the variability in the Ex-LS at least above 8 PVU is low and the increase of water vapor in the Ex-LS is a general feature in the entire northern Ex-LS in this time period.

- *P5L16: This sentence is not clear to me. Do you mean that below 8PVU some "pixels" in your eqLat-DTTP-space show larger differences than others of H2O between the two phases? How does the thermal tropopause affect local variability? Fig. 1c shows local differences in water vapour. It sounds a bit odd to attribute local differences of water vapour to local variability of water vapour. Consider revising.*
The thermal tropopause used here is derived from ECMWF data and does not always match perfectly the true tropopause height. In this case you would attribute moist tropospheric air masses to the stratospheric regime and vice versa. So in the end, it is a mismatch of the applied criterion directly around the tropopause. You are right, that it is a odd argument to attribute local differences of water vapor to local variability of water vapor. We revised the text concerning this point.

- *P5L26: You point out that the distribution is compact for period 1. This gives the impression that most of the moistening happens between the two periods. However, period2 is already at the end of the monsoon season. Why is 0.5ppmv consistent with 1-1.5ppmv then? What are the transport time scales involved?*
The moistening begins already in the beginning of July and continue until beginning of October. The gradient of increasing water vapor mixing ratio is strongest between both phase. This is already shown by Vogel et al. 2016. and clearly visible in Figure 15, which shows the increase of water vapor from CLaMS as well as MLS satellite measurements. In fact, our high precise in-situ measurements are a strong confirmation of the MLS and CLaMS results. We included one additional sentence to make the connection between both papers more clear.

- *P5L28: How did you test significance for those non-Gaussian distributions?*
We tested the difference between bot distributions with the Mann-Whitney-U-Test. We found very small p-values of 1e-120 which indicates a significant difference between both distribution. This we did also for the monsoon tracer and for the methane distribution and found similar results.

- *P5L28: Are your measurements representative of the entire Ex-LS?*
The measurements during TACTS and ESMVAL were focused both to get a meridional cross-section at the subtropical jet and from equator to the pole. The flight paths were not planed to catch explicitly air from the Asian monsoon, but rather to get a climatological representation of the Ex-LS. Also the agreement of between CLaMS and MLS, mentioned in the previous comment and the correlation of Asian monsoon surface tracer indicate the representativeness of the water vapor measurements presented in this study.

- *P5L33: Please show CH_4. It might support your claim that the signature originates in the AMA.*
We included the PDF for both phases of methane for PV larger than 8 in the manuscript. In addition, we added also the correlation coefficient for water vapor and methane in the text to corroborate the increase of water vapor to the AMA.

- *P6L4: 100*
  We are not sure what is meant here.

- *P6L5: Do you just want to say increasing H2O corresponds to increasing MON? How- ever, providing some correlation coefficients for the region above 8PVU would be good. To me the pattern above 8PVU looks different between Figs. 1c and 2c.*
  The pattern of Fig. 1c and 2c does not match perfectly, this is correct. We have to keep in mind however that we compare here a model with measurements, which includes always some deviations. The correlation coefficients for water vapor and MON tracer are 0.27 in phase1 and 0.40 in phase2. The correlation coefficient for methane and MON tracer are 0.41 for phase1 and 0.63 for phase2. All correlation coefficient have a very high significance with p-values of nearly zero ($<$ 1e-20). In fact, methane shows a better correlation. This is because methane has a long life time of around 8.9 years in the troposphere and even longer in the lower stratosphere (Wuebbles et al. 2002), whereas water vapor has a very short life time of about 9 days (Hartmann 2015) due to its ability to condense and change phase in response to temperature. Therefore we can expect a more reduced variability of water vapor in comparison to methane, due to the damping effect of the Lagrangian cold point (LCP). This reduces the correlation in the case of water vapor. This is also the reason, why we performed the trajectory analysis concerning the LCP to show the origin of the significant increase in the water vapor concentration. We included the numbers and part of this explanation in Sec. 3.3, were the correlations of water vapor, methane and MON tracer are discussed.

- *P7L8: "Thus" and "because" in one sentence is confusing. Please clearly express why you consider CH4 to be a monsoon tracer. Please provide references for each argument.*
  The text is now rephrased.

- *P8L4: What is the statistically significant region in this plot? Please provide numbers for the background mixing ratios.*
  The statistical significant region are represented by the water vapor mixing ratio below the background and show that the influence of air masses from the AMA is of minor importance for the first phase. We included also the background mixing ratio of 5 ppmv in text, which fits to the limits of the core region.

- *P8L5: The grammar of this sentence is odd. However, no matter how I interpret it, the definition of the core region remains unclear. If the core region is defined by the 75*
  We rephrased the text to make it clearer.

- *P8L11: sounds odd, consider rewording*
  Rephrased.

- *P8L12: This sentence is not clear to me. Did you test statistical significance? If so: How exactly did you do that? The scatter plots show H2O mixing ratio ver-*

sus CH4 mixing ratio. If you want to discuss the statistics of deltas ("increase"), please show deltas.
You are right. We tested the methane and water vapor concentration to the MON tracer and not directly the increase. We changed the text and show some correlation coefficient and an explanation.

- *P8L12: slope CH4/H2O -> tilted towards a higher ratio or higher CH4, but not towards higher H2O. Please revise. Also, you discussed that contributions from the ASM in- crease H2O and CH4. Fig. 3 looks like the ratio CH4/H2O changes. Is that also a tell-tale sign of ASM origin?*
  We revised the text according to your suggestion. We also recognize the change in the ratio and found with the help of the emission tracer the slope is dependent on the source region. Low methane high water vapor seems to come from the tropical Pacific, whereas the high methane in phase 2 is more from the South east Asia. But we found this result to be to speculative, therefore we excluded it from the text.

- *P8L17: 1. "Water vapor is dehydrated" sounds odd; 2. Do you mean that dehydration typically happens close to the saturation mixing ratio, or that air masses typically get close to saturation at the LCP? (Only wet air might produce ice crystals). Please revise.*
  That's right. We revised the text. Ice crystals can only form in supersaturation therefore it is unlikely that air masses can be moister than the LCP. It can happen, if not all ice crystals sediment out of the air mass. The LCP is also the coldest point along the airmass history and therefore it is unlikely that you find dryer tropospheric air masses than the saturation mixing ratio at the LCP.

- *P9L2: Are you considering only trajectories that originate in the troposphere here? The LCP that determined water vapor might lie further back than the reach of the backward trajectory.*
  Yes, we took only trajectories, which were in the troposphere during the last 50 days. It might be that the LCP for some of the stratospheric trajectories lie further back in time, but these trajectories cannot be related to the AMA. This is just because the it would be before the onset of the Asian monsoon. Other mixing processes seems to be week and unimportant for water vapor, because in the first phase we observe most mixing ratios up to the stratospheric background.

- *P9L10: remove "notably" or "interestingly"*
  Changed.

- *P9L20: ... and region of the flights?*
  We clam, that the whole northern Ex-LS is affected by this water vapor enhancement and this includes of course also the region of the flights.

- *P9L23: Fig. 5 can be removed. The next sentence contains all the information.*
  You are right, that all information can also be found in the text. However, we

decided to keep the Figure because it supports the reader to better recognize the proportions of tropospheric and stratospheric air masses within the two phases in comparison to numbers in the text only.

- *P9L25: amount -> fraction or percentage*
  Changed to fraction.

- *P9L25: in phase -> contributing to phase (Please have in mind that transport from the AMA needs some time.)*
  This is true. We changed the text.

- *P10L5: How did you calculate the correlation? Please provide numbers.*
  This is true, we did not calculated the statistical significance of the increase itself. We calculated the correlation between the different tracers. We revised the text to clarify this point.

- *P10L6: distinguish between AMA and the respective surface tracers*
  We changed the text to "region of the Asian monsoon". This is more specific in terms of the origin tracers.

- *P10L11: I am not convinced. Please elaborate on how you relate your results to Vogel et al. (2016). Is water vapour transport from the AMA to the ex-LS constant throughout the monsoon season? How do you relate your phase1/gap/phase2 periods to the monsoon period? Are the diagnostics comparable?*
  See answer to Methodology part 2. We included also more explanation in the conclusion section.

- *P10L19: crossing is displaced OR crossings are displaced; What is combined with quasi-isentropic transport? Please revise and consider splitting it into two sentences.*
  We revised the text according to your suggestions.

- *P10L21: "picture of influencing" sounds odd*
  Changed.

- *P10L25: This should be shown, at least via a rough estimate.*
  This is a very good suggestion, but beyond the scope of this paper. There are several studies showing the climatic impact of water vapor changes in the UT/LS. For example Riese et al. 2012 showed that a 5-10% increase of water vapor in the Ex-LS produced a top of the atmosphere radiative forcing of 100-500 mWm-2. We included this information also in the conclusion.

References:

- Hartmann, Dennis: Global Physical Climatology (Second Edition), Elsevir, ISBN: 978-0-12-328531-7,p. 399, 2015.

- Konopka, P., F. Ploeger, M. Tao, and M. Riese (2016), Zonally resolved impact of ENSO on the stratospheric circulation and water vapor entry values, J. Geophys. Res. Atmos., 121, 11,48611,501, doi:10.1002/2015JD024698.

- Müller, S., Hoor, P., Bozem, H., Gute, E., Vogel, B., Zahn, A., Bönisch, H., Keber, T., Krämer, M., Rolf, C., Riese, M., Schlager, H., and Engel, A.: Impact of the Asian monsoon on the extratropical lower stratosphere: trace gas observations during TACTS over Europe 2012, Atmospheric Chemistry and Physics, 16, 10 57310 589, https://doi.org/10.5194/acp-16-10573-2016, 2016.

- Pan, L. L., and L. A. Munchak (2011), Relationship of cloud top to the tropopause and jet structure from CALIPSO data, J. Geophys. Res., 116, D12201, doi:10.1029/2010JD015462.

- Ploeger, F., Konopka, P., Günther, G., Grooß, J.-U. and Müller, R.: Impact of the vertical velocity scheme on modeling transport in the tropical tropopause layer. Journal of Geophysical Research 115: doi: 10.1029/2009JD012023. issn: 0148-0227, 2010.

- Ploeger, F., Günther, G., Konopka, P., Fueglistaler, S., Müller, R., Hoppe, C., Kunz, A., Spang, R., Grooß, J. U., and Riese, M.: Horizontal water vapor transport in the lower stratosphere from subtropics to high latitudes during boreal summer, Journal of Geophysical Research- atmospheres, 118, 81118127, https://doi.org/10.1002/jgrd.50636, 2013.

- Ploeger, F., Gottschling, C., Griessbach, S., Grooß, J. U., Günther, G., Konopka, P., Müller, R., Riese, M., Stroh, F., Tao, M., Ungermann, J., Vogel, B., and von Hobe, M.: A potential vorticity-based determination of the transport barrier in the Asian summer monsoon anticyclone, Atmospheric Chemistry and Physics, 15, 13 14513 159, https://doi.org/10.5194/acp-15-13145-2015, 2015.

- Pommrich, R., Müller, R., Grooß, J. U., Konopka, P., Ploeger, F., Vogel, B., Tao, M., Hoppe, C. M., Günther, G., Spelten, N., Hoffmann, L., Pumphrey, H. C., Viciani, S., DAmato, F., Volk, C. M., Hoor, P., Schlager, H., and Riese, M.: Tropical troposphere to stratosphere transport of carbon monoxide and long-lived trace species in the Chemical Lagrangian Model of the Stratosphere (CLaMS), Geoscientific Model Development, 7, 28952916, https://doi.org/10.5194/gmd-7-2895-2014, 2014.

- Randel, W. J. and Jensen, E. J.: Physical processes in the tropical tropopause layer and their roles in a changing climate, Nature Geoscience, 6, 169176, https://doi.org/10.1038/ngeo1733, 2013.

- Santee, M. L., Manney, G. L., Livesey, N. J., Schwartz, M. J., Neu, J. L., and Read, W. G.: A comprehensive overview of the climatological composition of the Asian summer monsoon anticyclone based on 10 years of Aura Microwave Limb Sounder measurements, Journal of Geophysical Research: Atmospheres, 122, 54915514, https://doi.org/10.1002/2016JD026408, http://dx.doi.org/10.1002/2016JD026408, 2016JD026408, 2017.

- Vogel, B., Günther, G., Müller, R., Grooß, J. U., Afchine, A., Bozem, H., Hoor, P., Krämer, M., Müller, S., Riese, M., Rolf, C., Spel- ten, N., Stiller, G. P., Ungermann, J., and Zahn, A.: Long-range transport pathways of tropospheric source gases originating in Asia into the northern lower stratosphere during the Asian monsoon season 2012, Atmospheric Chemistry and Physics, 16, 15 30115 325, https://doi.org/10.5194/acp-16-15301-2016, 2016.

- Vogel, B., Günther, G., Müller, R., Grooß, J. U., and Riese, M.: Impact of different Asian source regions on the composition of the Asian monsoon anticyclone and of the extratropical lowermost stratosphere, Atmospheric Chemistry and Physics, 15, 13 69913 716, https://doi.org/10.5194/acp-15-13699-2015, 2015.

- Vogel, B., Günther, G., Müller, R., Grooß, J. U., Hoor, P., Kräer, M., Müller, S., Zahn, A., and Riese, M.: Fast transport from Southeast Asia boundary layer sources to northern Europe: rapid uplift in typhoons and eastward eddy shedding of the Asian monsoon anticyclone, Atmospheric Chemistry and Physics, 14, 12 74512 762, https://doi.org/10.5194/acp-14-12745-2014, 2014.

- Ungermann, J., Ern, M., Kaufmann, M., Müller, R., Spang, R., Ploeger, F., Vogel, B., and Riese, M.: Observations of PAN and its con- finement in the Asian summer monsoon anticyclone in high spatial resolution, Atmospheric Chemistry and Physics, 16, 83898403, https://doi.org/10.5194/acp-16-8389-2016, 2016.

- Wuebbles, D. J. and Hayhoe, K.: Atmospheric methane and global change, Earth-science Reviews, 57, PII S00128252(01)00 0629, https://doi.org/10.1016/S0012-8252(01)00062-9, 2002.

Water vapor increase in the lower stratosphere of the northern hemisphere due to the Asian monsoon anticyclone observed during the TACTS/ESMVal campaigns

Christian Rolf[1], Bärbel Vogel[1], Peter Hoor[2], Armin Afchine[1], Gebhard Günther[1], Martina Krämer[1], Rolf Müller[1], Stefan Müller[2,3], Nicole Spelten[1], and Martin Riese[1]

[1]Institute for Energy and Climate Research (IEK-7), Forschungszentrum Jülich GmbH, Jülich, Germany.
[2]Institute for Atmospheric Physics, Johannes Gutenberg University, Mainz, Germany.
[3]now at: Enviscope GmbH, 60489 Frankfurt, Germany.

Correspondence to: C. Rolf (c.rolf@fz-juelich.de)

Abstract. The impact of air masses originating in Asia and influenced by the Asian monsoon anticyclone on the northern hemisphere stratosphere is investigated based on in situ measurements. A statistically significant increase in water vapor (H_2O) of about 0.5 ppmv (11 %) and methane (CH_4) of up to 20 ppbv (1.2 %) in the extra-tropical stratosphere above a potential temperature of 380 K was detected between August and September 2012 during the HALO aircraft missions TACTS (Transport and Composition in the UT/LMS) and ESMVal (Earth System Model Validation). We investigate the origin of the increased water vapor and methane using the three-dimensional Chemical Lagrangian Model of the Stratosphere (CLaMS). We assign the source of the moist air masses in the Asian region (North and South India, East China, South East Asia, and the tropical Pacific) based on tracers of air mass origin used in CLaMS. The water vapor increase is correlated with an increase of the simulated Asian monsoon air mass contribution from about 10 % in August to about 20 % in September, which corresponds to a doubling of the influence from the Asian monsoon region. Additionally, back trajectories starting at the aircraft flight paths are used to differentiate transport from the Asian monsoon anticyclone and other source regions by calculating the Lagrangian cold point (LCP). The geographic location of the LCPs, which indicates the region where the imprint of water vapor mixing ratio along these trajectories occurs, can be predominantly attributed to the Asian monsoon region.

1 Introduction

Radiatively active trace gases, such as water vapor and methane, play a key role in determining the radiative balance in the upper troposphere and lower stratosphere (UTLS) and thus have an impact on the surface climate of the Earth (Forster and Shine, 2002; Riese et al., 2012). In particular, only small increases/decreases of stratospheric water vapor in the order of 10 % have a significant influence on the radiative forcing and thus constitute a warming/cooling potential for global surface temperature in the order of 25-30 % (Solomon et al., 2010). In this paper, we focus on transport pathways and exchange processes between troposphere and stratosphere to better understand the variability of water vapor and methane in this climatically sensitive region of the atmosphere.

In general, freeze-dried air masses with a low amount of water vapor enter the stratosphere in the tropical tropopause layer (TTL) and are transported vertically via the Brewer–Dobson circulation (BDC) deep into the stratosphere and quasi-horizontally into the extra-tropical lower stratosphere (Ex-LS) (e.g. Gettelman et al., 2011). Thus, the air in the Ex-LS constitutes a mixture of young air masses originating from quasi-horizontal transport out of the TTL and old stratospheric air in the downwelling branch of the BDC (e.g. Bönisch et al., 2009; Ploeger et al., 2013; Vogel et al., 2016). As a rule, an increasing inflow of tropospheric air masses from the tropics in combination with higher temperatures in the tropopause region causes a moistening of the Ex-LS in summer compared to winter months (e.g. Hoor et al., 2005; Krebsbach et al., 2006; Ploeger et al., 2013; Randel This moistening is partly due to the Asian summer monsoon which facilitates the transport of water vapor into the northern lowermost stratosphere (e.g. Dethof et al., 1999; Randel and Jensen, 2013). The monsoon system is linked to rapid vertical transport of surface air from India and east Asia in the summer months (July to October). These young and moist tropospheric air masses from

close to the surface are to some degree horizontally confined in the upper troposhere by the Asian monsoon anticyclone (AMA). The edge of the AMA constitutes a transport barrier characterized by a potential vorticity (PV) gradient (Ploeger et al., 2015). The transport barrier is rather leaky due to the strong dynamic variability of the anticyclone consisting of meridional displacements, splits, and eddy-shedding events (e.g. Vogel et al., 2014, 2015; Ungermann et al., 2016). However, this confinement with reduced cross-gradient transport is responsible for an enrichment of tropospheric constituents and higher amounts of water vapor or methane in the upper troposphere (e.g. Park et al., 2004, 2007; Schuck et al., 2010; Patra et al., 2011; Baker et al., 2012; Bia A horizontal transport pathway out of the anticyclone is directed westwards into the tropical tropopause layer (TTL) (Popovic and Plumb, 20 A second pathway of moist and polluted air masses exists, where air from the AMA is transported eastwards along the subtropical jet and subsequently into the northern lower stratosphere via eddy shedding followed by tropopause crossing (e.g. Dethof et al., 1999; Hsu and Plumb, 2000; Vogel et al., 2014, 2016). These frequently occurring eddy-shedding events triggered by Rossby wave activity flood the northern lower stratosphere with tropospheric air masses from the Asian subcontinent and cause a moistening of the Ex-LS (e.g. Müller et al., 2016; Vogel et al., 2014, 2016). In addition, Aura Microwave Limb Sounder (MLS) satellite observations showed that the Asian monsoon is hydrating the northern stratosphere (Uma et al., 2014).

From an observational point of view, the influence of the Asian monsoon on the Ex-LS is mostly analyzed based on satellite limb sounder measurements, which only have a limited vertical resolution, typically larger than 1-2 km in the lowermost stratosphere (e.g. Hegglin et al., 2013; Santee et al., 2017). Especially in the tropopause region, this coarse resolution can smooth the strong vertical gradient of water vapor at the tropopause and may lead to both an over- or underestimation of the water vapor concentration in the lower stratosphere. Here, we present high-resolution and precise in situ water vapor and methane measurements in the northern lower stratosphere performed in August and September 2012. We investigate the changes of these trace gases during the observational time period and attribute them to transport from the Asian monsoon region by using Lagrangian back-trajectory analysis in combination with artificial tracers of air mass origin.

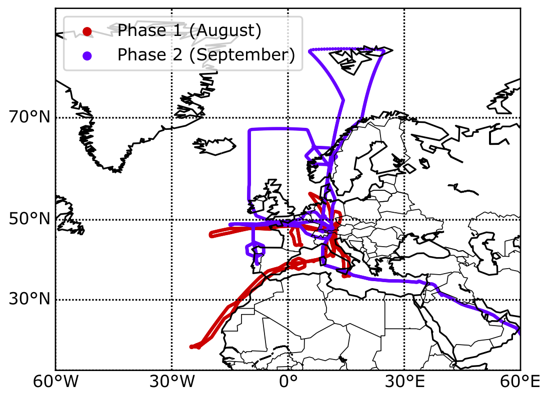

Figure 1. Flight paths of all analyzed flights during the TACTS and ESMVal campaigns. The flight paths of phase 1 and phase 2 are plotted in red and blue, respectively.

2 Methods and Instruments

This study is based on the data collected during the two aircraft campaigns TACTS (Transport and Composition in the UT/LMS) and ESMVal (Earth System Model Validation). These campaigns were conducted between August and October 2012 mainly in the northern hemisphere in the region from the Cape Verde Islands to the Arctic, mainly over Europe and the Atlantic Ocean. The data are split into two time periods. These two periods, referred to as phase 1 and phase 2 hereafter, cover the time period from August 28 to September 5 and from September 18 until September 26, 2012, respectively. Figure 1 shows the map of all considered flight paths and the respective phases they are attributed to. Vogel et al. (2016) provided the motivation for this selection, showing that the strongest transport of young air masses from the AMA into the Ex-LS occurred in the period between August and the end of September, mainly associated with a pronounced eddy-shedding event on 20 September 2012 (Vogel et al., 2014). Thus, the selection of these dates reveals a clear difference in the water vapor distribution in the Ex-LS, as shown in Sec. 3.1. More information about the data coverage can be found in Müller et al. (2016). The separation allows the temporal evolution of the trace gas composition of the Ex-LS between the two phases to be investigated. In the following, the water vapor and methane instruments aboard the HALO aircraft are briefly described. In addition, we used CLaMS (Chemical Lagrangian Model for the Stratosphere) (McKenna et al., 2002b) results interpolated onto the flight path, and also CLaMS backward-trajectory calculations from the measurement locations for identification of the air mass origin as described further below.

2.1 Water vapor measurements

The water vapor data used in this study were measured using the FISH (Fast In situ Stratospheric Hygrometer) instrument. The measurement technique is based on Lyman-α photofragment fluorescence and is suitable for measuring water vapor in the range of 1 ppmv to 1000 ppmv (Zöger et al., 1999). FISH is a well-established closed-path hygrometer with a long history in aircraft measurements and instrument intercomparisons (Meyer et al., 2015). Calibrations are performed during the campaigns with a MBW DP30 frost point hygrometer achieving accuracy of 6 % $\pm$ 0.4 ppmv at a time resolution of 1 Hz during TACTS/ESMVal.

2.2 Methane measurements

The methane data are obtained using the TRIHOP (TRacer In situ QCL and HydrOgen Peroxide monitor) instrument. The measurement technique is based on infrared absorption spectroscopy with a three channel quantum cascade laser spectrometer capable of measuring CO, CO_2, N_2O and, CH_4. An in situ calibration against a secondary standard traceable to the NOAA scale was applied during the campaign. TRIHOP methane data are available every eight seconds with an integration time of 1.5 seconds, precision of 9.5 ppbv (2 σ), and an uncertainty of 13.5 ppbv relative to the standard. For more details, see Müller et al. (2016).

2.3 CLaMS simulations with artificial air mass origin tracers

In this study, we used the three-dimensional chemistry transport model CLaMS (McKenna et al., 2002b, a; Pommrich et al., 2014, and references therein) according to the setup described in Vogel et al. (2015). The air parcels within the CLaMS model are represented by an ensemble of air parcels on a time-dependent irregular grid and transported in a Lagragian way using trajectories including a physical representation of mixing (Konopka et al., 2004, 2010). This allows tracer gradients to be represented in a particularly accurate manner in CLaMS model simulations, making the model simulations well-suited for investigating transport and troposphere/stratosphere interaction (e.g. Konopka et al., 2010; Pommrich et al., 2014; Vogel et al., 2015, 2016).

The CLaMS simulation used here covers the time period from May 1 to October 31, 2012, and captures the Asian monsoon season in 2012.

The simulation is driven by the meteorological fields of the European Reanalysis (ERA)-Interim (Dee et al., 2011) from the ECMWF (European Centre for Medium-Range Weather Forecasts) and covers the atmosphere from the surface up to 900 K potential temperature ($\approx$ 37 km altitude) with a vertical resolution of 400 m around the tropopause. In the simulation, "emission tracers" are released in the boundary layer ($\approx$ 2-3 km above surface) which globally marks all regions of the Earth's surface. These different artificial tracers are freshly emitted every 24 h in the boundary layer and their concentration in the atmosphere represents the air mass contributions from the respective source regions. For more details, see Vogel et al. (2015, 2016).

Vogel et al. (2015) showed that the emission tracers for India and eastern China are correlated with high values of potential vorticity (PV) and AURA-MLS (Livesey et al., 2011) satellite measurements of O_3 and CO. Thus, they can be used as proxies for the location and shape of the AMA and allow transport studies of air masses from the AMA into the northern extra-tropical

Table 1. Latitude and longitude range of artificial boundary layer sources in the CLaMS model, which are used to define the "Asian monsoon surface tracer" (MON).

Emission tracer	Latitude	Longitude
Northern India (NIN)	20-40°N	55-90°E
Southern India (SIN)	0-20°N	55-90°E
Eastern China (ECH)	20-40°N	90-125°E
Southeast Asia (SEA)	12-20°N	90-155°E
Tropical Pacific Ocean (TPO)	20°S-20°N	55°E-80°W

lower stratosphere. In addition, the emission tracers from southeast Asia and the Pacific Ocean contribute to the air mass composition of the outer-edge of the AMA (Vogel et al., 2015, 2016). Thus, to study the transport of air masses affected by the Asian monsoon, the most important emission regions are India/China, southeast Asia, and the tropical Pacific Ocean. The sum of all these model boundary layer tracers is referred to as "Asian monsoon surface tracer" (MON) (see Table 1). This tracer marks young air masses (younger than 5 months) that are confined during summer in the AMA and air masses from southeast Asia and the tropical Pacific Ocean influenced by the large-scale flow around the anticyclone in the UTLS. All other source regions can be neglected, since it was shown in Vogel et al. (2016) that these regions did not make a significant contribution to the transport of young tropospheric air masses into the lower stratosphere in summer and autumn 2012.

2.4 Air mass trajectory calculations

In contrast to the three-dimensional CLaMS simulations, pure trajectory calculations do not account for mixing. However, the position of an air parcel and the changes in temperature along the trajectory can be tracked with sufficient accuracy over several days or even weeks (e.g. Vogel et al., 2014; Rolf et al., 2015). The air mass trajectories are calculated by means of the CLaMS trajectory module for the last 50 days and from every 10th second from the location of the flight path using ERA-Interim data (Dee et al., 2011). The trajectories are based on the horizontal wind fields and diabatic heating rates with a resolution of 1° x 1° with 60 vertical levels from the surface up to 0.1 hPa. Additional parameters such as temperature, pressure, and potential vorticity (PV) are interpolated from the ERA-Interim data onto the trajectories. In addition, the local distance to the thermal tropopause (WMO) is calculated for each trajectory time step and added to the trajectory data (e.g. Krebsbach et al., 2006). The water vapor saturation mixing ratio with respect to ice (hereafter: saturation mixing ratio) along the trajectories is calculated based on the actual trajectory temperature. The Lagrangian cold point (LCP) is defined by the minimum of the temperature along the trajectory and thus the minimum saturation mixing ratio. With these parameters, pure stratospheric trajectories can be separated from trajectories with a tropospheric origin within the last 50 days by comparing the distance to the tropopause before and after the LCP. All trajectories with a negative distance to the thermal tropopause before the LCP and a negative afterwards passed the tropopause from the troposphere into the stratosphere and represent air masses which originate in the troposphere.

3 Results

The main purpose of this study is to corroborate the hypothesis that air masses from Asia and the tropical Pacific affected by the AMA are the main source of the moistening found in the Ex-LS in late summer and fall in the northern hemisphere. Firstly, we present in situ water vapor measurements of the two phases (August and September) and support the hypothesis based on water vapor - methane correlations. Secondly, we combine the measurements with CLaMS model results and the 50-day backward trajectory analysis to ultimately confirm the hypothesis.

3.1 Moistening the Ex-LS

Water vapor is strongly coupled to the temperature. The thermal tropopause in the sub-tropics and mid-latitude is found to be a good indicator for marking the critical level of the temperature gradient and sharp relative humidity change, as shown by e.g. Pan and Munchak (2011), and thus separates the moist tropospheric regime from the dry stratospheric regime. In addition, isentropic transport is found to be the main pathway from the AMA into the Ex-LS (Vogel et al., 2016). Therefore, we choose the distance to the local thermal tropopause as the vertical coordinate in units of potential temperature. In Figures 2 a and 2 b, we display the mean water vapor distributions of both phases (phase 1 and phase 2) along the PV-based equivalent latitude (eqLat). The use of EqLat allows us to combine observations performed at different times and locations by neglecting short-term dynamical variability (e.g. planetary wave activity at the sub-tropical jet). High water vapor mixing ratios (> 10 ppmv), representing moist tropospheric air, can be found below and up to 20 K above the thermal tropopause during both phases. Drier air masses down to the stratospheric background level of 4-5 ppmv are observed 30 K to 90 K above the thermal tropopause. In phase 1, the drier air masses are observed closer to the thermal tropopause, while in phase 2 such dry air masses are found at potential temperatures 10 K higher. Both distributions have similar vertical and horizontal extents and thus the difference between both phases reveals local changes in water vapor mixing ratios over the time period (see Fig. 2c). Air masses with PV < 8 PVU reveal a large variability between phase 2 and phase 1, most likely due to the local variability of the thermal tropopause not captured by the ECMWF data and planetary wave activity which distort the location of the sub-tropical jet core. Kunz et al. (2015) showed that the climatological transport barrier between tropospheric and stratospheric air masses is located at PV values of 7-8 PVU between June and November in the northern hemisphere. Thus, we will focus on air masses with PV > 8 PVU in the following analysis, since they are not affected by small-scale mixing processes directly at the tropopause.

All the air masses with PV values larger than 8 PVU show a distinct 5-10 % increase of water vapor in phase 2 compared to phase 1. The occurrence of these air masses ranges from 40°N to 75°N in equivalent latitude horizontally and corresponds to potential temperatures larger than 380 K. The frequency distributions of the water vapor mixing ratios with PV values larger than 8 PVU allow us to quantify the strength of moistening between both phases (see Fig. 2d). The frequency distribution of phase 1 is very compact with a mean water vapor mixing ratio of 4.5 ppmv, while the distribution of phase 2 is shifted to higher water vapor mixing ratios with a mean value of 5 ppmv and shows a tail towards higher water vapor mixing ratios. The difference in the mean water vapor mixing ratio between the two phases indicates a statistically significant (Mann-Whitney-U-Test with p-value $< 10^{-100}$) moistening of the Ex-LS of about 0.5 ppmv within a time period of less than a month. This finding is in accordance with CLaMS model simulations and MLS satellite measurements which show a total increase of mean water vapor mixing ratio in the northern Ex-LS by 1.5 ppmv (1.0 ppmv) at 380 K (400 K) in the time period from May to November due to the transport of air masses originating from source regions in Asia (see Fig. 15 in Vogel et al., 2016). The gradient of this increase was found to be of its strongest between the end of August and the end of September, which is the result of a pronounced eddy-shedding event on 20 September 2012 (Vogel et al., 2014, 2016). This strong increase perfectly matches the separation of the two phases and is in agreement with the increase observed in this study here. In addition to the measured increase in mean water vapor of 0.5 ppmv, a mean increase of 20 ppbv methane is observed in the same time period (see Fig. 4).

3.2 Tracer of air mass origin

Our simulations of corresponding artificial air mass origin tracers (Figures 3 a and 3 b) indicate that the moistening observed during phase 2 is the result of an increase in air mass contribution from the Asian monsoon region. MON increased by about 20 % to 100 % from phase 1 to phase 2 above the 8 PVU level, which roughly corresponds to a doubling (see Fig. 3 c). The increase of MON in this altitude range is correlated with the observed increase in water vapor between both phases. The frequency distributions in Figure 3 d reveals the clear shift of MON to higher values, where the mean values show an increase of 8.6 % from phase 1 to phase 2. In particular, there is only a slight increase in surface tracers ($\sim$1 %) from other locations (residual surface without the monsoon region, MON) in the same time period. Therefore, the moistening of 0.5 ppmv can be linked to the 8.6 % increase of MON.

3.3 Water vapor - methane correlation

The correlation of observed water vapor with observed methane provides further evidence of the large air mass contribution of the Asian monsoon region. Methane is sourced at the Earth's surface and thus has the highest concentrations in the troposphere. In the stratosphere, the methane mixing ratio decreases with altitude due to oxidization. Methane concentrations are especially high in regions with large industrial activity or natural out-gassing from wetlands such as rice cultivation in India/Southeast Asia (Park et al., 2004; Xiong et al., 2009). Due to the high emissions on the ground, strongly enhanced mixing ratios of methane are found in the AMA compared to the free troposphere, as shown by the CARIBIC measurements (Schuck et al., 2010; Patra et al., 2011; Baker et al., 2012). Thus, methane is suitable as a monsoon-specific tropospheric tracer, in particular for the Asian monsoon regions. The difference in the methane distribution between both phases is visible in Figure 4. The frequency distributions look very similar to the ones found for water vapor and MON (Sec.3.1 and 3.2) and reveal a mean increase of 24 ppbv methane between both phases. This provides further evidence to combine water vapor and methane measurements together with the artificial origin tracer MON.

Figure 5 shows the correlations of measured water vapor and methane mixing ratios in the lower stratosphere (PV > 8 PVU) for both phases. The core region (black contour) of the scatter plot encompasses 75 % of the data points of the underlying frequency distribution and ranges from around 3.8 ppmv to 5.1 ppmv in water vapor and from 1700 ppbv to 1730 ppbv in methane. The correlation in phase 1 is compact with nearly no significant enhancement in methane and water vapor compared

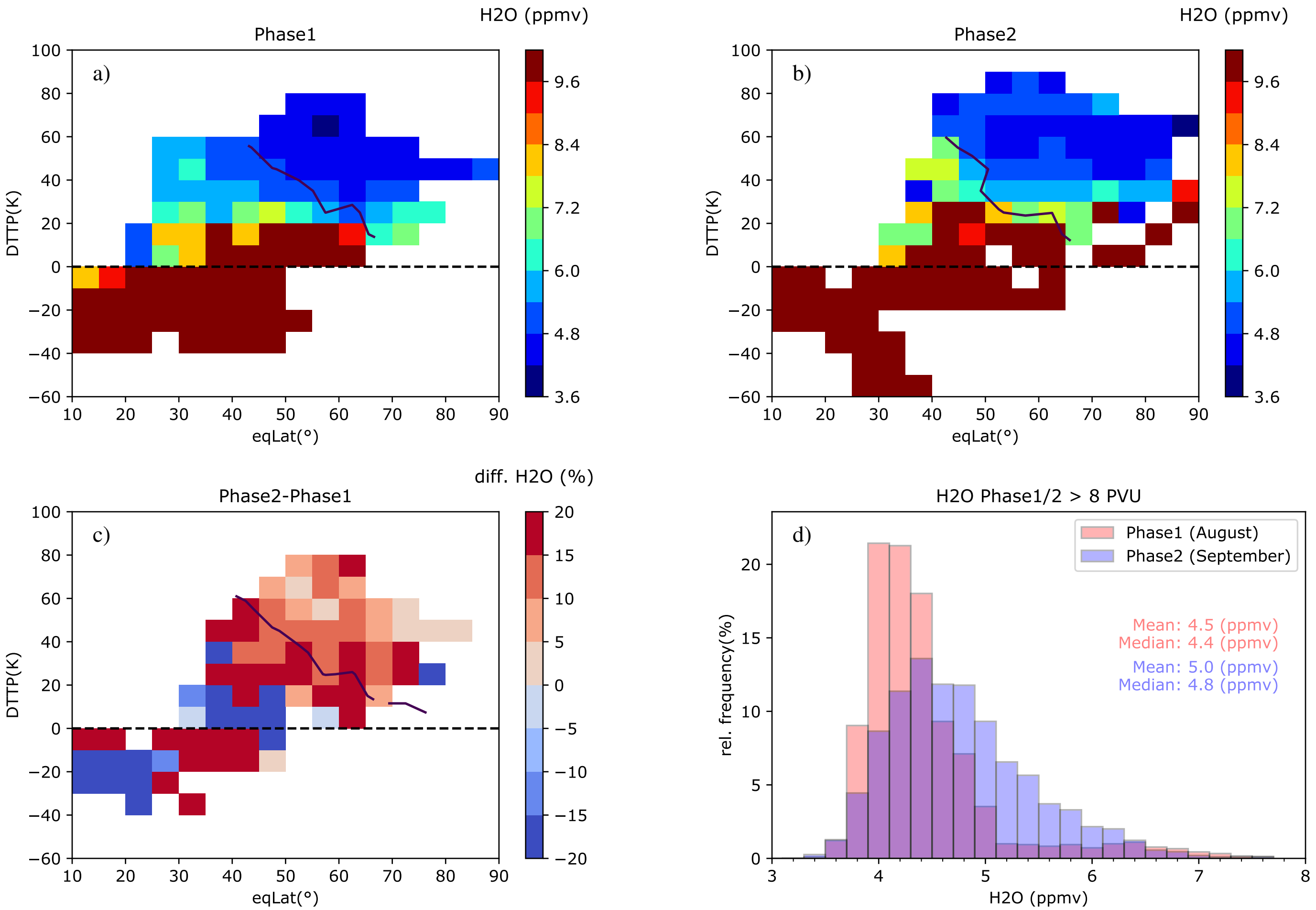

Figure 2. Mean distribution of H_2O as a function of equivalent latitude and distance to thermal tropopause (DTTP) in potential temperature units: a) phase and 1 b) phase 2. c) Relative differences of H_2O between phase 2 and phase 1. The dashed/solid black lines represent the thermal tropopause and the 8 PVU isoline, respectively. d) Frequency distribution of H_2O for the lower stratosphere (PV > 8 PVU) for phase 1 (red) and phase 2 (blue). The mean and median values of each distribution are given in red and blue, respectively.

to the water vapor background mixing ratio of 5 ppmv in the lower stratosphere (Zahn et al., 2014). The amount of MON is relatively low with values around 12 % in the core region. Only a few measurements (around 10% of the data) with high MON values of up to 30 % are found, showing the highest water vapor and methane values in phase 1. This indicates the moistening of the Ex-LS from the earlier phase of the monsoon before the end of August. In contrast, the correlation in phase 2 is dominated by measurements with higher methane and water vapor mixing ratios. The core region of the data ranges from 1700 ppbv to 1790 ppbv in methane and 3.7 ppmv to 6.2 ppmv in water vapor. The amount of MON in phase 2 is 30 % and thus significantly higher compared to phase 1. The water vapor and methane mixing ratio is statistically correlated with the MON tracer. In fact, the slope of the correlation is tilted towards higher methane mixing ratios in phase 2, which confirms the influence of methane-

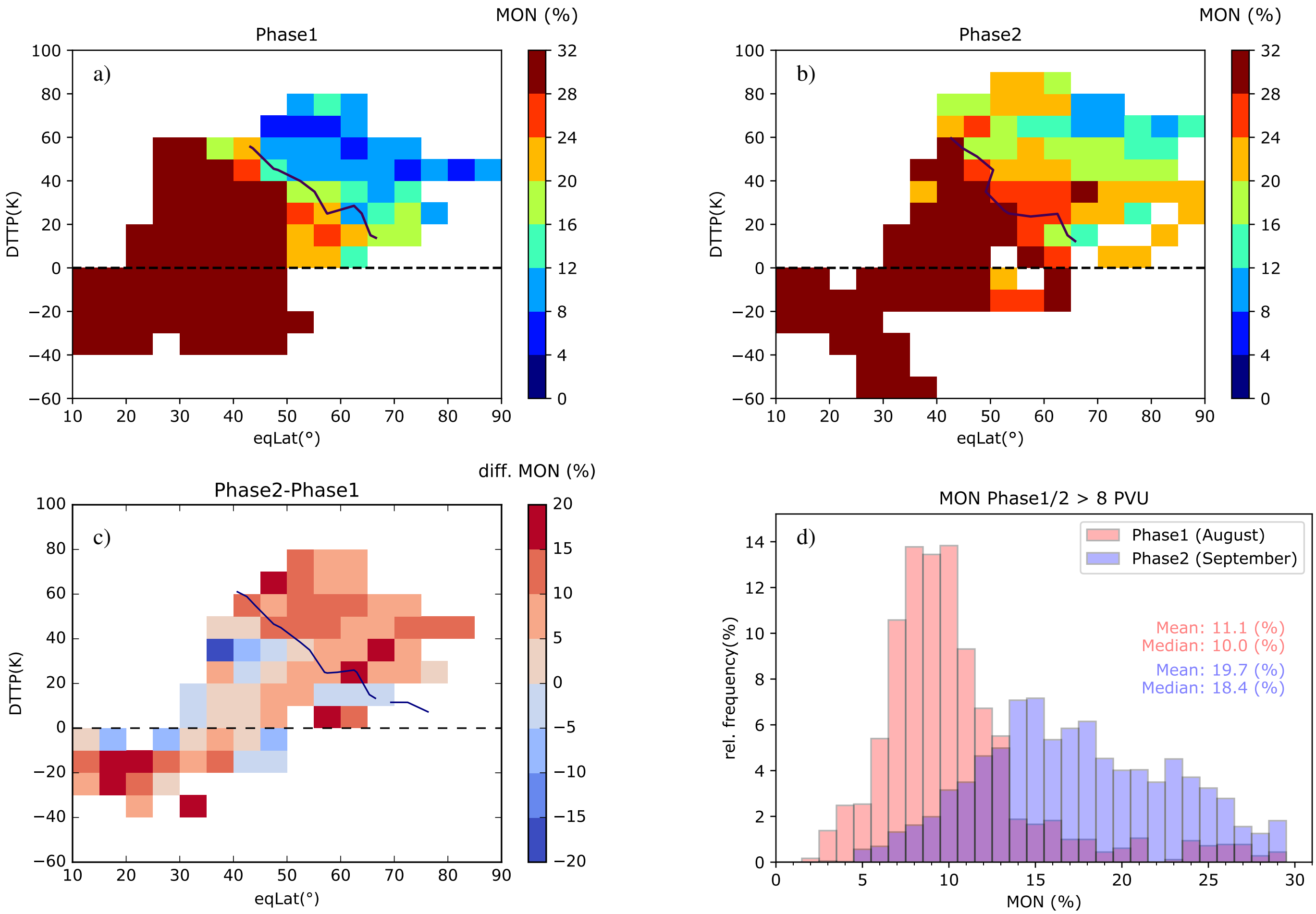

Figure 3. Same as Figure 2 but for the simulated Asian monsoon surface tracer MON.

rich tropospheric air masses from the Asian monsoon region in the Ex-LS. The associated correlation coefficients for water vapor and methane are 0.79 and 0.67 for phase 1 and phase 2, respectively. The correlation coefficients for the measured water vapor and modeled MON tracer are 0.27 in phase 1 and 0.40 in phase 2, whilst for methane and MON tracer they are 0.41 for phase 1 and 0.63 for phase 2. All correlation coefficients (calculated according to Pearson) are highly significant with p-values of nearly zero ($< 10^{-18}$), thus rejecting the null hypothesis of an uncorrelated dataset. In fact, the correlation for water vapor and methane is clearly higher compared to the correlation with the MON tracer, which corroborates the consistency between both in situ measurements. It also shows the limitations of CLaMS to reproduce every small-scale feature in the measurements. However, the correlation for the model-based tracer and the measurements is still quite satisfactory and strongly supports the hypothesis that the increase in water vapor is correlated to the air mass affected by the AMA.

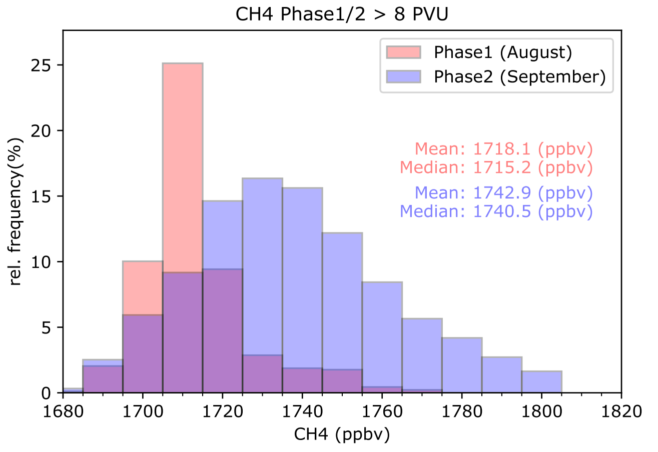

Figure 4. Frequency distribution of CH_4 for the lower stratosphere (PV > 8 PVU) for phase 1 (red) and phase 2 (blue). The mean and median values of each distribution are given in red and blue, respectively.

In addition, methane shows a better correlation to MON due to its long life time of around 9 years in the troposphere and even longer life time in the lower stratosphere (Wuebbles and Hayhoe, 2002). It also reacts similarly to the ideal model tracer MON. In contrast, water vapor has a very short life time of about 9 days (Hartmann, 2015) due to its strong temperature dependency. Therefore, we can expect a reduced variability of water vapor in comparison to methane and thus a better correlation of methane with the MON tracer. This provides the motivation for showing the connection of measured water vapor mixing ratios to their air mass temperature history in the next section.

3.4 Lagrangian cold point from backward trajectories

The amount of water vapor transported from the troposphere into the stratosphere is strongly coupled with the Lagrangian cold point (LCP), where typically the air masses are dehydrated close to the saturation mixing ratio by ice crystal formation and subsequent sedimentation (e.g. Schoeberl and Dessler, 2011). Thus, the amount of water vapor in these air masses is nearly preserved after passing the LCP in the tropical, sub-tropical, and mid-latitude stratosphere. The geographic location of the LCP along the backward trajectory marks the location of these imprints on the water vapor mixing ratio.

Vogel et al. (2016) showed that the eddy shedding and in-mixing of tropospheric air masses can occur at potential temperatures of around 380 K directly between double tropopause features, passing the transport barrier at the climatological jet core. However, the locations of LCPs in the case of eddy shedding events do not implicitly coincide with the point of in-mixing into the Ex-LS (Schoeberl and Dessler, 2011; Vogel et al., 2016). In this case, the locations of LCPs are typically at the top of the upwelling in the AMA region, whereas in-mixing into the Ex-LS can be found at locations where Rossby wave breaking occurs eastwards or westwards of the AMA along the subtropical jet.

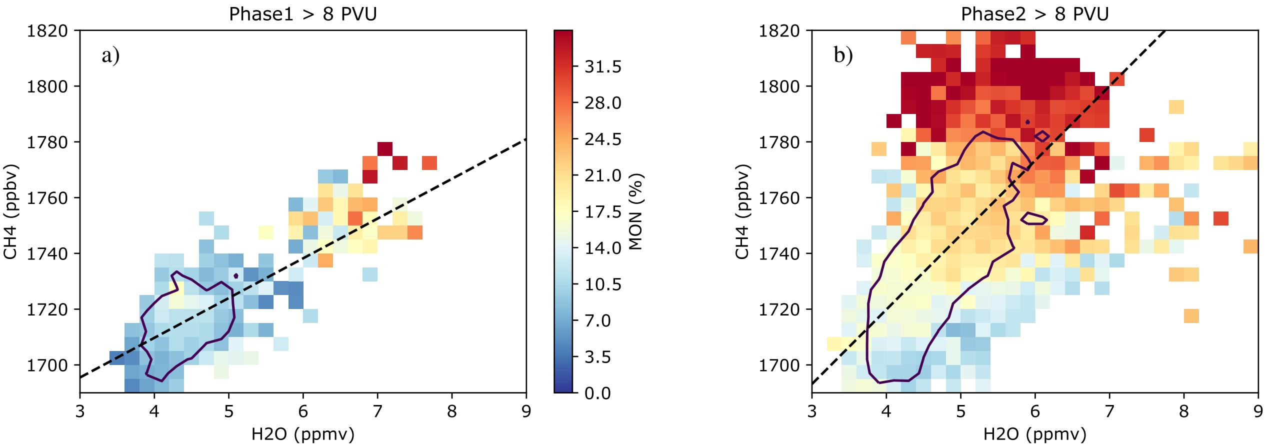

Figure 5. Correlation of water vapor and methane for PV > 8 PVU, color-coded by the Asian monsoon surface tracer MON. The solid black line marks the core region, which encompasses 75 % of the data. The black dashed line represents a robust linear regression fitting curve (Theil–Sen regression).

In the following, all backward trajectories for air parcels with PV > 8 PVU according to Section 3.1 are considered (i.e. 2909 out of 11333 (25.6 %) trajectories for phase 1 and 7301 out of 14673 (49.8 %) trajectories for phase 2). The LCPs (see Figure 6 a and 6 b) are mostly located in the subtropics with equivalent latitudes ranging from 0° N to 50°N. They cluster meridionally above India/China/Indonesia, North America, and West/North Africa in both phases. Interestingly, all these regions can be assigned to respective monsoon systems over Asia, America, and Africa. In phase 1, only 158 out of 2909 (5.4 %) trajectories with PV > 8 PVU are classified as tropospheric with the LCP criteria above, i.e. more trajectories are entirely in the stratosphere. Furthermore, phase 2 reveals more frequent LCP occurrence with 846 out of 7301 (11.6 %) trajectories with PV > 8 PVU in these regions. In addition, phase 2 also shows a clear increase of LCPs in the region of India/China/Indonesia compared to phase 1. This is further confirmed by the relative difference between phase 2 and phase 1, normalized to all trajectories (tropospheric and stratospheric, see Figure 6c). The signal of the two other monsoon circulations, i.e. the American monsoon and the African Monsoon, disappears in this analysis. The dominant appearance of LCPs in the Asian monsoon region is evident. Therefore, the Asian monsoon has the most frequent LCP occurrence, the largest extent, and thus the largest impact on stratospheric water vapor enhancements in the observed time period.

The selection criterion for trajectories described in Section 2.4 can be used to quantify the amount of trajectories which remained in the stratosphere for at least 50 days (stratospheric origin) or passed the LCP from the troposphere into the stratosphere (tropospheric origin), as shown in Figure 7. In phase 1, the fraction of flight path back-trajectories with tropospheric origin ($p1t$) is 5.4 % (94.6 % stratospheric, $p1s$), while in phase 2, 11.6 % ($p2t$) had a tropospheric origin (88.4 % stratospheric, $p2s$). The larger fraction of tropospheric origin trajectories corroborate the existence of stronger troposphere to stratosphere exchange contributing to phase 2. The increase in mean water vapor from phase 1 (4.5 ppmv) to phase 2 (5.0 ppmv) by the Asian monsoon trajectories can be calculated with a system of linear equations:

$$\begin{pmatrix} p1t & p1s \\ p2t & p2s \end{pmatrix} \begin{pmatrix} wv_1 \\ wv_2 \end{pmatrix} = \begin{pmatrix} 4.5\,\text{ppmv} \\ 5.0\,\text{ppmv} \end{pmatrix} \quad \Rightarrow \quad wv_1 = 4.1\,\text{ppmv}, wv_2 = 11.9\,\text{ppmv} \tag{1}$$

Here, wv_1 denotes the water vapor background mixing ratio and wv_2 the water vapor contribution from the monsoon trajectories. From this simple calculation, it can be concluded that the observed increase of 0.5 ppmv in the mean water vapor mixing ratio between both phases (see Figure 2d) can be attributed to the transport of moist tropospheric air masses with a mean water vapor mixing ratio of about 12 ppmv compared to a stratospheric background of 4.1 ppmv. These values fit quite well with the CLaMS model simulations in Vogel et al. (2016), who determined a stratospheric background of around 4 ppmv in the same time period neglecting water vapor contributions from air masses in the Asian monsoon region.

4 Conclusions

The mean water vapor/methane increase of 0.5 ppmv (11 %)/20 ppbv (1.2 %) from phase 1 (August) to phase 2 (September) is correlated with an increase in Asian monsoon surface tracer (MON) by 8.6 percentage points, which corresponds to a doubling of air masses in the Ex-LS influenced by the region of the Asian monsoon, southeast Asia, and tropical Pacific Ocean. Other surface regions can be neglected as a source of the moist air masses in the stratosphere. The doubling of MON is also consistent with observations of increased values of other tropospheric tracers (N_2O, SF_6, CO), which were simultaneously observed (Müller et al., 2016). Our observational results also confirm the modeling study of Ploeger et al. (2013), where a simulated increase of the water vapor from 4.5 ppmv to 5.0 ppmv in the Ex-LS corresponds to an increase of the Asian monsoon air contribution of about 10 %. Our observed increase in water vapor is also in accordance with the study by Vogel et al. (2016), who found that over the entire monsoon time period from June to October 2012, 1.5 ppmv/1.0 ppmv of the water vapor in the northern hemispheric stratosphere at 380 K/400 K originates from the AMA regions including southeast Asia and the tropical Pacific Ocean. The gradient of this increase was found to be at its strongest between the end of August and the end of September, similarly to the two phases used in this study.

Moreover, the flight path back-trajectories support the influence of the Asian monsoon on the Ex-LS. In phase 2, twice as many trajectories show a tropospheric influence compared to phase 1. The moistening in the Ex-LS is maintained by young air masses transported from the Asian monsoon with a mean mixing ratio of about 12 ppmv. The locations of the LCPs show that the imprint of in-mixed water vapor mixing ratios occurs most frequently above India and south-east Asia. The crossings of the tropopause in the case of eddy-shedding events are displaced to Rossby wave breaking regions along the subtropical jet. From these points on, air masses are transported quasi-isentropicaly into the Ex-LS as shown by Vogel et al. (2016).

All these measurements and model results contribute to a general understanding that the Asian monsoon influences the trace gas composition of the extra-tropical lower stratosphere in the northern hemisphere – at least for the year 2012. In particular, this study provides observational evidence of the water vapor transport from the Asian monsoon region to the northern Ex-LS.

Following the study by Riese et al. (2012), a 5-10% increase of water vapor in the Ex-LS produced a top of the atmosphere radiative forcing of 100-500 $\mathrm{mW\,m^{-2}}$. Depending on the residence time in the lower stratosphere, the moist and methane-rich air masses have the potential to impact surface temperatures in the northern hemisphere.

Acknowledgements. We would like to thank the coordinators of the two HALO missions TACTS and ESMVal, namely Andreas Engel, Harald Bönisch (University of Frankfurt), and Hans Schlager (DLR), for their efforts. Special thanks go to DLR-FX for the avionic data from the Bahamas instrument, Heini Wernli for providing tropopause information, and Jens-Uwe Grooß for CLaMS model forecasts and flight planning within the LASSO project (HALO-SPP,1294/GR,3786) supported by the German Research Foundation (DFG). We would also like to thank the Jülich Supercomputing Centre (JSC) for computing time on the supercomputer JUROPA within the VSR project JICG11. Our activities were part of the DFG's AMOS project (HALO-SPP 1294/VO 1276/5-1) and the Seventh Framework Program (FP7/2007–2013) of the European Union's StratoClim project (grant agreement no. 603557). Thanks also go to the Language Service of Forschungszentrum Jülich for linguistic revision. In addition, we gratefully acknowledge the ECMWF for their meteorological reanalysis data support. The observational data used in this study can be downloaded from the HALO database (DOI:10.17616/R39Q0T) at https://halo-db.pa.op.dlr.de/.

References

Baker, A. K., Schuck, T. J., Brenninkmeijer, C. A. M., Rauthe-Schoch, A., Slemr, F., van Velthoven, P. F. J., and Lelieveld, J.: Estimating the contribution of monsoon-related biogenic production to methane emissions from South Asia using CARIBIC observations, Geophysical Research Letters, 39, L10 813, https://doi.org/10.1029/2012GL051756, 2012.

Bian, J. C., Pan, L. L., Paulik, L., Vomel, H., Chen, H. B., and Lu, D. R.: In situ water vapor and ozone measurements in Lhasa and Kunming during the Asian summer monsoon, Geophysical Research Letters, 39, L19 808, https://doi.org/10.1029/2012GL052996, 2012.

Bönisch, H., Engel, A., Curtius, J., Birner, T., and Hoor, P.: Quantifying transport into the lowermost stratosphere using simultaneous in-situ measurements of SF6 and CO2, Atmospheric Chemistry and Physics, 9, 5905–5919, 2009.

Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kallberg, P., Koehler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J. . J., Park, B. . K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J. . N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, Quarterly Journal of the Royal Meteorological Society, 137, 553–597, https://doi.org/10.1002/qj.828, 2011.

Dethof, A., O'Neill, A., Slingo, J. M., and Smit, H. G. J.: A mechanism for moistening the lower stratosphere involving the Asian summer monsoon, Quarterly Journal of the Royal Meteorological Society, 125, 1079–1106, https://doi.org/10.1256/smsqj.55601, 1999.

Forster, P. M. D. and Shine, K. P.: Assessing the climate impact of trends in stratospheric water vapor, Geophysical Research Letters, 29, 1086, https://doi.org/10.1029/2001GL013909, 2002.

Gettelman, A., Hoor, P., Pan, L. L., Randel, W. J., Hegglin, M. I., and Birner, T.: The Extratropical Upper Troposphere and Lower Stratosphere, Reviews of Geophysics, 49, RG3003, https://doi.org/10.1029/2011RG000355
, 2011.

Hartmann, D. L.: Global Physical Climatology (Second Edition), vol. 103, Newnes, 2. edn., 2015.

Hegglin, M. I., Tegtmeier, S., Anderson, J., Froidevaux, L., Fuller, R., Funke, B., Jones, A., Lingenfelser, G., Lumpe, J., Pendlebury, D., Remsberg, E., Rozanov, A., Toohey, M., Urban, J., von Clarmann, T., Walker, K. A., Wang, R., and Weigel, K.: SPARC Data Initiative: Comparison of water vapor climatologies from international satellite limb sounders, Journal of Geophysical Research-atmospheres, 118, 846, https://doi.org/10.1002/jgrd.50752, 2013.

Hoor, P., Fischer, H., and Lelieveld, J.: Tropical and extratropical tropospheric air in the lowermost stratosphere over Europe: A CO-based budget, Geophysical Research Letters, 32, L07 802, https://doi.org/10.1029/2004GL022018, 2005.

Hsu, C. J. and Plumb, R. A.: Nonaxisymmetric thermally driven circulations and upper-tropospheric monsoon dynamics, Journal of the Atmospheric Sciences, 57, 1255–1276, https://doi.org/10.1175/1520-0469(2000)057<1255:NTDCAU>2.0.CO;2, 2000.

Konopka, P., Steinhorst, H. M., Grooß, J. U., Günther, G., Müller, R., Elkins, J. W., Jost, H. J., Richard, E., Schmidt, U., Toon, G., and McKenna, D. S.: Mixing and ozone loss in the 1999-2000 Arctic vortex: Simulations with the three-dimensional Chemical Lagrangian Model of the Stratosphere (CLaMS), Journal of Geophysical Research-atmospheres, 109, D02 315, https://doi.org/10.1029/2003JD003792, 2004.

Konopka, P., Grooß, J. U., Gunther, G., Ploeger, F., Pommrich, R., Muller, R., and Livesey, N.: Annual cycle of ozone at and above the tropical tropopause: observations versus simulations with the Chemical Lagrangian Model of the Stratosphere (CLaMS), Atmospheric Chemistry and Physics, 10, 121–132, https://doi.org/10.5194/acp-10-121-2010, 2010.

Krebsbach, M., Schiller, C., Brunner, D., Günther, G., Hegglin, M. I., Mottaghy, D., Riese, M., Spelten, N., and Wernli, H.: Seasonal cycles and variability of O_3 and H_2O in the UT/LMS during SPURT, Atmospheric Chemistry and Physics, 6, 109–125, 2006.

Kunz, A., Sprenger, M., and Wernli, H.: Climatology of potential vorticity streamers and associated isentropic transport pathways across PV gradient barriers, Journal of Geophysical Research-atmospheres, 120, 3802–3821, https://doi.org/10.1002/2014JD022615, 2015.

Livesey, N. J., Read, W. G., Froidevaux, L., Lambert, A., Manney, G. L., Pumphrey, H. C., Santee, M. L., Schwartz, M. J., Wang, S., Cofield, R. E., Cuddy, D. T., Fuller, R. A., Jarnot, R. F., Jiang, J. H., Knosp, B. W., Stek, P. C., Wagner, P. A., , and Wu, D. L.: EOS MLS Version 3.3 Level 2 data quality and description document, Tech. Rep. JPL D33509, Jet Propulsion Laboratory, available from http://mls.jpl.nasa.gov, 2011.

McKenna, D. S., Grooß, J. U., Günther, G., Konopka, P., Müller, R., Carver, G., and Sasano, Y.: A new Chemical Lagrangian Model of the Stratosphere (CLaMS) - 2. Formulation of chemistry scheme and initialization, Journal of Geophysical Research-atmospheres, 107, 4256, https://doi.org/10.1029/2000JD000113, 2002a.

McKenna, D. S., Konopka, P., Grooß, J. U., Günther, G., Müller, R., Spang, R., Offermann, D., and Orsolini, Y.: A new Chemical Lagrangian Model of the Stratosphere (CLaMS) - 1. Formulation of advection and mixing, Journal of Geophysical Research-atmospheres, 107, 4309, https://doi.org/10.1029/2000JD000114, 2002b.

Meyer, J., Rolf, C., Schiller, C., Rohs, S., Spelten, N., Afchine, A., Zöger, M., Sitnikov, N., Thornberry, T. D., Rollins, A. W., Bozoki, Z., Tatrai, D., Ebert, V., Kühnreich, B., Mackrodt, P., Möhler, O., Saathoff, H., Rosenlof, K. H., and Krämer, M.: Two decades of water vapor measurements with the FISH fluorescence hygrometer: a review, Atmospheric Chemistry and Physics, 15, 8521–8538, https://doi.org/10.5194/acp-15-8521-2015, 2015.

Müller, S., Hoor, P., Bozem, H., Gute, E., Vogel, B., Zahn, A., Bönisch, H., Keber, T., Krämer, M., Rolf, C., Riese, M., Schlager, H., and Engel, A.: Impact of the Asian monsoon on the extratropical lower stratosphere: trace gas observations during TACTS over Europe 2012, Atmospheric Chemistry and Physics, 16, 10 573–10 589, https://doi.org/10.5194/acp-16-10573-2016, 2016.

Pan, L. L. and Munchak, L. A.: Relationship of cloud top to the tropopause and jet structure from CALIPSO data, Journal of Geophysical Research-atmospheres, 116, D12 201, https://doi.org/10.1029/2010JD015462, 2011.

Pan, L. L., Honomichl, S. B., Kinnison, D. E., Abalos, M., Randel, W. J., Bergman, J. W., and Bian, J.: Transport of chemical tracers from the boundary layer to stratosphere associated with the dynamics of the Asian summer monsoon, Journal of Geophysical Research-atmospheres, 121, 14 159–14 174, https://doi.org/10.1002/2016JD025616, 2016.

Park, M., Randel, W. J., Kinnison, D. E., Garcia, R. R., and Choi, W.: Seasonal variation of methane, water vapor, and nitrogen oxides near the tropopause: Satellite observations and model simulations, Journal of Geophysical Research-atmospheres, 109, D03 302, https://doi.org/10.1029/2003JD003706, 2004.

Park, M., Randel, W. J., Gettelman, A., Massie, S. T., and Jiang, J. H.: Transport above the Asian summer monsoon anticyclone inferred from Aura Microwave Limb Sounder tracers, Journal of Geophysical Research-atmospheres, 112, D16 309, https://doi.org/10.1029/2006JD008294, 2007.

Patra, P. K., Niwa, Y., Schuck, T. J., Brenninkmeijer, C. A. M., Machida, T., Matsueda, H., and Sawa, Y.: Carbon balance of South Asia constrained by passenger aircraft CO2 measurements, Atmospheric Chemistry and Physics, 11, 4163–4175, https://doi.org/10.5194/acp-11-4163-2011, 2011.

Ploeger, F., Günther, G., Konopka, P., Fueglistaler, S., Müller, R., Hoppe, C., Kunz, A., Spang, R., Grooß, J. U., and Riese, M.: Horizontal water vapor transport in the lower stratosphere from subtropics to high latitudes during boreal summer, Journal of Geophysical Research-atmospheres, 118, 8111–8127, https://doi.org/10.1002/jgrd.50636, 2013.

Ploeger, F., Gottschling, C., Griessbach, S., Grooß, J. U., Günther, G., Konopka, P., Müller, R., Riese, M., Stroh, F., Tao, M., Ungermann, J., Vogel, B., and von Hobe, M.: A potential vorticity-based determination of the transport barrier in the Asian summer monsoon anticyclone, Atmospheric Chemistry and Physics, 15, 13 145–13 159, https://doi.org/10.5194/acp-15-13145-2015, 2015.

Pommrich, R., Müller, R., Grooß, J. U., Konopka, P., Ploeger, F., Vogel, B., Tao, M., Hoppe, C. M., Günther, G., Spelten, N., Hoffmann, L., Pumphrey, H. C., Viciani, S., D'Amato, F., Volk, C. M., Hoor, P., Schlager, H., and Riese, M.: Tropical troposphere to stratosphere transport of carbon monoxide and long-lived trace species in the Chemical Lagrangian Model of the Stratosphere (CLaMS), Geoscientific Model Development, 7, 2895–2916, https://doi.org/10.5194/gmd-7-2895-2014, 2014.

Popovic, J. M. and Plumb, R. A.: Eddy shedding from the upper-tropospheric Asian monsoon anticyclone, Journal of the Atmospheric Sciences, 58, 93–104, https://doi.org/10.1175/1520-0469(2001)058<0093:ESFTUT>2.0.CO;2, 2001.

Randel, W. J. and Jensen, E. J.: Physical processes in the tropical tropopause layer and their roles in a changing climate, Nature Geoscience, 6, 169–176, https://doi.org/10.1038/ngeo1733, 2013.

Randel, W. J. and Park, M.: Deep convective influence on the Asian summer monsoon anticyclone and associated tracer variability observed with Atmospheric Infrared Sounder (AIRS), Journal of Geophysical Research-atmospheres, 111, D12 314, https://doi.org/10.1029/2005JD006490, 2006.

Riese, M., Ploeger, F., Rap, A., Vogel, B., Konopka, P., Dameris, M., and Forster, P.: Impact of uncertainties in atmospheric mixing on simulated UTLS composition and related radiative effects, Journal of Geophysical Research-atmospheres, 117, D16 305, https://doi.org/10.1029/2012JD017751, 2012.

Rolf, C., Afchine, A., Bozem, H., Buchholz, B., Ebert, V., Guggenmoser, T., Hoor, P., Konopka, P., Kretschmer, E., Mueller, S., Schlager, H., Spelten, N., Suminska-Ebersoldt, O., Ungermann, J., Zahn, A., and Krämer, M.: Transport of Antarctic stratospheric strongly dehydrated air into the troposphere observed during the HALO-ESMVal campaign 2012, Atmospheric Chemistry and Physics, 15, 9143–9158, https://doi.org/10.5194/acp-15-9143-2015, 2015.

Santee, M. L., Manney, G. L., Livesey, N. J., Schwartz, M. J., Neu, J. L., and Read, W. G.: A comprehensive overview of the climatological composition of the Asian summer monsoon anticyclone based on 10 years of Aura Microwave Limb Sounder measurements, Journal of Geophysical Research: Atmospheres, 122, 5491–5514, https://doi.org/10.1002/2016JD026408, http://dx.doi.org/10.1002/2016JD026408, 2016JD026408, 2017.

Schoeberl, M. R. and Dessler, A. E.: Dehydration of the stratosphere, Atmospheric Chemistry and Physics, 11, 8433–8446, https://doi.org/10.5194/acp-11-8433-2011, 2011.

Schuck, T. J., Brenninkmeijer, C. A. M., Baker, A. K., Slemr, F., von Velthoven, P. F. J., and Zahn, A.: Greenhouse gas relationships in the Indian summer monsoon plume measured by the CARIBIC passenger aircraft, Atmospheric Chemistry and Physics, 10, 3965–3984, 2010.

Solomon, S., Rosenlof, K. H., Portmann, R. W., Daniel, J. S., Davis, S. M., Sanford, T. J., and Plattner, G. K.: Contributions of Stratospheric Water Vapor to Decadal Changes in the Rate of Global Warming, Science, 327, 1219–1223, https://doi.org/10.1126/science.1182488, 2010.

Uma, K. N., Das, S. K., and Das, S. S.: A climatological perspective of water vapor at the UTLS region over different global monsoon regions: observations inferred from the Aura-MLS and reanalysis data, Climate Dynamics, 43, 407–420, https://doi.org/10.1007/s00382-014-2085-9, 2014.

Ungermann, J., Ern, M., Kaufmann, M., Muller, R., Spang, R., Ploeger, F., Vogel, B., and Riese, M.: Observations of PAN and its confinement in the Asian summer monsoon anticyclone in high spatial resolution, Atmospheric Chemistry and Physics, 16, 8389–8403, https://doi.org/10.5194/acp-16-8389-2016, 2016.

Vogel, B., Günther, G., Müller, R., Grooß, J. U., Hoor, P., Krämer, M., Müller, S., Zahn, A., and Riese, M.: Fast transport from Southeast Asia boundary layer sources to northern Europe: rapid uplift in typhoons and eastward eddy shedding of the Asian monsoon anticyclone, Atmospheric Chemistry and Physics, 14, 12 745–12 762, https://doi.org/10.5194/acp-14-12745-2014, 2014.

Vogel, B., Günther, G., Müller, R., Grooß, J. U., and Riese, M.: Impact of different Asian source regions on the composition of the Asian monsoon anticyclone and of the extratropical lowermost stratosphere, Atmospheric Chemistry and Physics, 15, 13 699–13 716, https://doi.org/10.5194/acp-15-13699-2015, 2015.

Vogel, B., Günther, G., Müller, R., Grooß, J. U., Afchine, A., Bozem, H., Hoor, P., Krämer, M., Müller, S., Riese, M., Rolf, C., Spelten, N., Stiller, G. P., Ungermann, J., and Zahn, A.: Long-range transport pathways of tropospheric source gases originating in Asia into the northern lower stratosphere during the Asian monsoon season 2012, Atmospheric Chemistry and Physics, 16, 15 301–15 325, https://doi.org/10.5194/acp-16-15301-2016, 2016.

Wuebbles, D. J. and Hayhoe, K.: Atmospheric methane and global change, Earth-science Reviews, 57, PII S0012–8252(01)00 062–9, https://doi.org/10.1016/S0012-8252(01)00062-9, 2002.

Xiong, X., Houweling, S., Wei, J., Maddy, E., Sun, F., and Barnet, C.: Methane plume over south Asia during the monsoon season: satellite observation and model simulation, Atmospheric Chemistry and Physics, 9, 783–794, 2009.

Zahn, A., Christner, E., van Velthoven, P. F. J., Rauthe-Schoch, A., and Brenninkmeijer, C. A. M.: Processes controlling water vapor in the upper troposphere/lowermost stratosphere: An analysis of 8years of monthly measurements by the IAGOS-CARIBIC observatory, Journal of Geophysical Research-atmospheres, 119, 11 505–11 525, https://doi.org/10.1002/2014JD021687, 2014.

Zöger, M., Afchine, A., Eicke, N., Gerhards, M. T., Klein, E., McKenna, D. S., Morschel, U., Schmidt, U., Tan, V., Tuitjer, F., Woyke, T., and Schiller, C.: Fast in situ stratospheric hygrometers: A new family of balloon-borne and airborne Lyman alpha photofragment fluorescence hygrometers, Journal of Geophysical Research-atmospheres, 104, 1807–1816, https://doi.org/10.1029/1998JD100025, 1999.

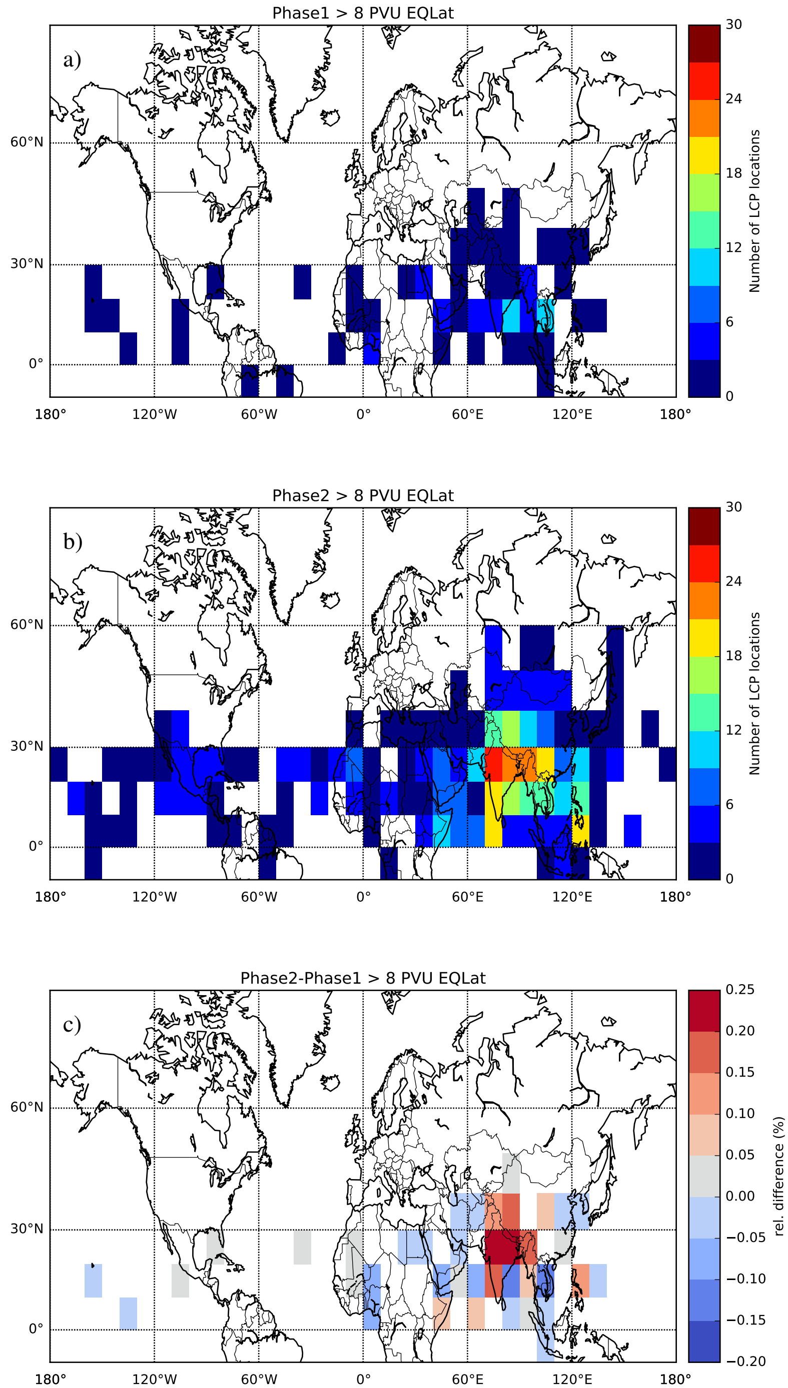

Figure 6. Location of Lagrangian Cold point (LCP) binned by longitude and latitude. LCP locations for phase 1 and phase 2 are shown in a) and b), respectively. The relative difference normalized to the total number of trajectories is shown in Fig. c).

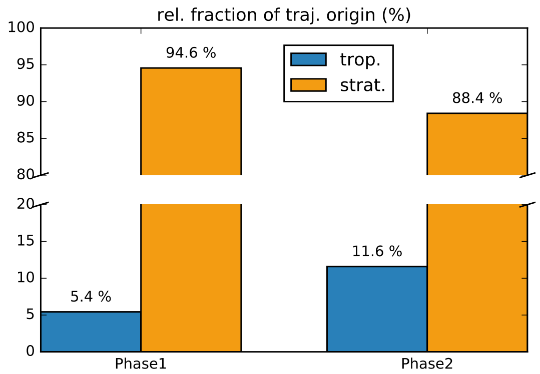

Figure 7. Fraction of trajectories with tropospheric (blue) or stratospheric (orange) origins for each phase.