

## ***Interactive comment on “Quantifying transport into the lowermost stratosphere using simultaneous in-situ measurements of SF<sub>6</sub> and CO<sub>2</sub>” by H. Bönisch et al.***

**H. Bönisch et al.**

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We thank for the helpful and constructive reviews. In general, we followed most of your suggestions. In the following you will find detailed answers to your questions and suggestions.

Abstract: Line 13: The sentence as it stands is not correct. 20% was calculated to be the extreme value of the tropospheric fraction  $\alpha_1$ . For me, it sounds like the entire LMS composition is characterized by these low values, which is not the finding of the paper. Further, any statement based on this study needs to be considered with caution, because the coverage of observations per season might not be characteristic for the entire LMS (see for example Krebsbach (2006), Conclusions)

It is correct,  $\alpha_1 < 20\%$  are extreme values during spring. Therefore we rephrase: 'Based on this concept we conclude that the stratospheric influence on LMS composition is strongest in April with extreme values of the tropospheric fractions ( $\alpha_1$ ) below 20% and that the strongest tropospheric signatures are found in October with  $\alpha_1$  greater than 80%.' It is true, the results might not be characteristic for the entire LMS (see also our answer to comment of Reviewer 1). Therefore we add the following statement: 'The high resolution SF6 and CO2 measurements obtained during the SPURT campaigns allow for a comprehensive view of the LMS region above Europe. Despite the large seasonal and spatial data coverage, SPURT alone can obviously not provide a climatology and the results presented here only apply for the region and time of the SPURT observations and have to be confirmed by further investigations and measurements.'

Introduction: Line10: Here, you can mention the study by Olsen (2008) who discussed TST in spring. Olsen M. A., A. R. Douglass, P. A. Newman, J. C. Gille, B. Nardi, V. A. Yudin, D. E. Kinnison, R. Khosravi (2008), HIRDLS observations and simulation of a lower stratospheric intrusion of tropical air to high latitudes, *Geophys. Res. Lett.*, 35, L21813, doi:10.1029/2008GL035514.

We followed your suggestion.

Section 2: Line 15ff: Please mention, how many or for which flights SF6 is not available. What observations did you use to derive a SF6/N2O relation in real-time?

SF6 are not available for 7 out of 36 flights that have been performed in total during SPURT. Further details on data availability and technical issues see Engel et al. (2006) - see our reference in section 2 (p-21233, l-10). We used measurements obtained during SPURT from the same season or ideally from the previous or on next day to derive a SF6/N2O relation (see p-21233, l-20).

3.2 Line7: Page 21237, first paragraph and Figure 2, 3, 6, and 7: The use of the  $eqlat/\theta$  coordinate system can be problematic, because PV is not conserved ev-

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erywhere in the UTLS. This is for example the case in the vicinity of the jet stream. I wonder how an altitude versus latitude plot looks like for the quantities shown. Even though the data cover eqlat bins between 40-80 degrees, the location of those data points might be close to the subtropical jet and therefore influence by upper tropical air masses. A plot like that does not have to be shown in the paper, but it would be interesting to know the location of air masses with regard to the STJ in general.

PV is conservative under adiabatic conditions. In the stratosphere diabatic timescales are long, thus PV (eqlat) and theta can be used to account for reversible excursions of planetary waves. Therefore, this coordinate system can be regarded as tropopause-following since it follows the meridional excursions of PV-contours induced by planetary waves (e.g. Hoor et al., 2004). Also, the position of the subtropical jet in the eqlat-theta coordinate system is more of climatologic nature and showing much smaller variability than in latitude based coordinate system. An altitude(-latitude) coordinate system has the disadvantage, especially for instantaneous or non-climatological in-situ data sets, that the data would show much too much scatter (e.g. Engel et al., 2006): At the same latitude belt tropospheric and stratospheric air masses are present at the same time at different longitudes.

Further, how many data points are included in each of the data bins in Figure 2,3,4, and 7? How representative is this study? A climatology based on very few flights does not allow generalized statements about transport processes (see for example Krebsbach (2006)).

A detailed description of the SPURT data coverage is given by Hoor et al. (2004) (see Section 3 and Fig. 2). We add this reference in Section 2 (Data set) p-21233, l-8: 'For a detailed analysis of the SPURT data coverage see Hoor et al. (2004).' We don't think that 36 flights are only 'very few flights'. However, the SPURT data set alone is no climatology for the entire UT/LMS. Concerning the representativity of this study, see our comment to your first question (Abstract, Line: 13) and see also our answer to comment of Reviewer 1.

Line 20: Can you be more quantitative here? What is the Theta range of the thickness of negative mean age values (and therefore the penetration height of extra-tropical tropospheric air into the LMS)? What is the location and thickness of the ExTL derived in Hoor (2004) and Birner (2006)? How do these values agree?

This is an interesting suggestion by the reviewer but a quantitative analysis of how the mean age criterion matches the ExTL derived from CO-O3 or the TIL derived from temperature profiles is beyond the scope of this paper. However, we added a reference to recent studies on this topic 'This TIL roughly corresponds to the ExTL in terms of location and vertical extent and recent studies have suggested a radiative link between the TIL and the specific water vapour and ozone distributions within the ExTL (Randel et al., 2007; Hegglin et al., 2009).'

Page 21238 Line 13ff and Figure 3. Does Figure 3 include all profiles observed in summer and fall respectively?

Yes. It was not our intention to go into details of water vapour distribution in the LMS. The interpretation of these distributions is somewhat different and much more complicated than for other trace gases due to the fact that water vapour is influenced by microphysics along the transport pathways into the LMS.

If so, how does the age of air plot look like for the combined profiles in summer and fall? Figure 2 shows two plots per season.

Combined mean age for summer or autumn looks more or less like the average of the particular two plots per season (see Engel et al., 2006). For water vapour, we only show the combined plots for summer and autumn, because the nicely illustrate the main message: 'LMS is drier but younger in autumn' (same if you look at combined mean age plots for both seasons also).

The authors suggest that LMS is flushed by very dry air from the TTL in autumn?

No, we suggest - deduced from mean age and water vapour only - that the LMS is

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flushed with dry tropospheric air from the TTL between summer (when we observed higher H<sub>2</sub>O mixing ratios and older air masses) and autumn. For clarification we rephrase p-21238, l-14ff: 'This indicates that the LMS must be flushed with very dry tropospheric air (< 5 ppm) over the course of summer till autumn.'

Stratospheric H<sub>2</sub>O is below 10ppm. From the color scale shown in Figure 3, dry stratospheric air can be found in 360 K and around 330-340 K. On the other hand, below 330 K, H<sub>2</sub>O mixing ratios are between 10-20 ppbm. If the dry air in the stratosphere would be caused by an intrusion of air from the TTL, why doesn't CO show higher tropospheric mixing ratios (as shown in Figure 8, Hoor (2004))?

We assume you mean Fig 7 and not Fig 8 because the latter displays the SPURT CO measurements in a different coordinate system (relative to the distance to the local tropopause). You do not expect 'high tropospheric mixing ratios' for CO in the LMS for air masses which were originated in the upper TTL. Measurements show that CO mixing ratios are already below 60 ppb in the upper TTL (Marcy et al. 2007). Furthermore, Hoor et al. (2005) yields similar results for the tropospheric fractions in the LMS with a CO-based mass balance approach using the SPURT measurements

Marcy, T.P., Popp, P.J., Gao, R.S., Fahey, D.W., Ray, E.A., Richard, E.C., Thompson, T.L., Atlas, E.L., Loewenstein, M., Wofsy, S.C., Park, S., Weinstock, E.M., Swartz, W.H., Mahoney, M.J.: Measurements of trace gases in the tropical tropopause layer, *Atmospheric Environment* 41:34 7253-726, 2007.

Also Kebsbach (2006) reports strongest influence of stratospheric air during winter and spring and the strongest transport of tropospheric air into the stratosphere in summer. Water vapor mixing ratios are much larger in the summer around 350 K in the stratosphere than in fall (up to 30-40 ppmv), even though the age of air is older than in fall. What is the source of these high mixing ratios? Can the influence of convection be excluded in summer?

The influence of convection can not be excluded. The seasonality of H<sub>2</sub>O in the LMS

differs clearly from other tracers observed during SPURT (see comment above also). The difference must be explained by the different mechanisms controlling water vapour in the LMS and its large gradients at the tropopause (e.g. Engel et al., 2006).

Section 4.1 The calculation of the second fraction and therefore the transport of the second pathway is defined reasonably. Considering the Earth's surface as the stratospheric entry is certainly OK, if we can exclude the stratospheric intrusions into the region of the tropical TP. However the definition to calculate the fraction of the transport of the second transport pathway can be improved: It is not clear how the control surface is defined and calculated: The text states: 'The control surface is the tropical and sub-tropical TP'. However, in the following, it is described that 'time series between 0-20 degN are used to represent the temporal behaviour of both tracers in the tropical and subtropical region.' Using surface values to calculate the entry function for the first transport pathway between 0-20 degN might be problematic: Depending on season, the STJ can be located between about 10-40 deg N the PJ up to 70 degN. Therefore many exchange processes between Troposphere and Stratosphere at the STJ and PJ are not included in this approximation.

Indeed, the formulation for the boundary condition Chi1 is misleading. The control surface for both modes is the earth surface. For clarification: All transit times are relative to the surface and not to the tropopause. Therefore we rephrase p-21242 l-19 and p-21242 l-23 (see our answer on comment of Reviewer 1). In general, the choice of the threshold 0°S to 20°N for control surface of chi1 is motivated by the work of Berthet et al. (2007) who demonstrated that nearly all 30-day backward trajectories started in the LMS above the ExTL (at the 365 K level) are leaving the boundary layer ( $z < 1$  km) between 0°N and 20°N. Therefore we add a comment at p-21242, l-23 (see our answer on comment of Reviewer 1 and 2).

Page 21244, Line 6: How different are the results derived from CO2 and SF6?

The values of DeltaAlpha1 used for the sensitivity study directly depict the difference

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between the tropospheric fractions derived from SF<sub>6</sub> and CO<sub>2</sub>. For the example of the SPURT measurements in January, the deviation  $\Delta\alpha_1$  is below 20% in all cases and about 5% on average for the best choice of parameters (see right upper panel Fig.5). For the calculation of the mass balance we use an iterative non-least square algorithm to find the best parameter, i.e. the mean transit time, to match the results for SF<sub>6</sub> and CO<sub>2</sub>.

General remarks to 21246 and 21247: A caveat needs to be added that all results described are based on a few observations per season and region.

See above our comments on the representativity of this study.

Page 21250: 'the LMS is flushed with tropospheric air during summer'. How does this statement agrees with the text on page 21238 and Figure 3 'the LMS must be flushed with dry tropospheric air between summer and autumn', so not during summer where water vapor is higher in the LMS?

The statement on page 21238 does not exclude that water vapour has been transported during summer into the LMS. The statement here is focused on the changing of water vapour and mean age between the summer and the autumn campaigns. The statement on page 21250 is the conclusion based on all results in this study. In contrast to the statement based on mean age and water vapour only, the final statement includes furthermore the conclusions derived from mean transit time from troposphere into the LMS calculated in the mass balance approach. So, we can not see a contradiction here.

TST is often connected with the existence of a double tropopause, see Olsen (2008). Randel (2007) have shown that a double tropopause occurs most frequently in winter (50-70%) and much less frequently in summer (10%). How do these findings agree with the statements found in this paper? Randel, W. J., D. J. Seidel, and L. L. Pan, 2007: Observational characteristics of double tropopauses, *J. Geophys. Res.*, 112, D07309, doi:10.1029/2006JD007904

It is not our goal and it is simply not possible with this study here to infer the underlying transport mechanisms which cause TST from the tropics into the LMS. However, TST or enhanced transport from the tropics to higher latitudes above the subtropical jet cores might be connected with the existence of a double tropopause, but it is clear that most quasi-horizontal transport relative to the downward transport (this relation determines the chemical composition of the LMS above the ExTL) from the TTL into the LMS does not occur during winter. All trace gas measurements in the LMS show that the LMS is dominated by stratospheric subsidence during winter (e.g. Sawa et al., 2008; and various other references in our paper).

Figure 4: The transport pathway across the PJ is missing. The PJ, indicated here north of 60 degrees N can occur in much lower latitudes and might contribute to the calculation of the age spectrum.

We agree that the PJ plays an important role in the ExTL region. Therefore we add the following statement at p-21238, l-10: 'Different to the region above, the formation of the ExTL is dominated by rather localised TST-processes (e.g. Dessler et al. 1995; Hoor et al. 2002) whereby irreversible cross-tropopause transport near the polar jet plays an important role (Fischer et al., 2000).' Above the ExTL the influence of the PJ seems to be non-significant (e.g. Hoor et al., 2004; Sawa et al., 2008) that's the reason why we used a lower boundary for our mass balance study.

Figure 6: The very high tropospheric fraction (100%) in October does not seem to be reasonable. Much higher H<sub>2</sub>O and CO mixing ratios should be expected as stated above.

The high tropospheric fractions (90-100 %) in the LMS in October are reasonable in case that the air is coming from the upper TTL (above 360 K). There, the water vapour mixing ratios are low, i.e. < 5 ppm (e.g. Marcy et al., 2007) and also CO is not very high in this region (see our comment above). Furthermore, CO is chemically degraded during the transport from the troposphere into the LMS having a lifetime of about 3-

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6 months. We calculated a transit time of 0.1 to 0.3 years (about 1 to 3.5 month) from earth surface into the LMS (see Fig 7). This is consistent with the results of Ray et al. (1999) who calculated a transit time from the tropopause into the LMS of about 1.5 month in September. The consequence is that there is a significant chemical degradation during the transport into the LMS. Note that, Hoor et al. (2005) accounted for the CO loss during transport to derive a CO-based mass balance. Therefore, young and dry air masses with high tropospheric fractions and CO mixing ratios below 50 ppb are no contradiction. One should keep in mind that the fraction of 90-100% means that the respective fraction has been in the troposphere within the last 3 months.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 21229, 2008.

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