We thank the reviewers for their helpful comments and recommendations, which could help us improve the paper. In the following, we address the issues raised by the reviewers in detail except for simple typographical or technical corrections, which we simply applied.

We repeat and enumerate the comments of the reviewers for convenience in bold face. Excerpts from the revised paper are shown as indented blocks; if longer unchanged sections are contained in the excerpts, changes are marked in italics.

1 Reviewer Comments 1

1. **A difficulty in the study is the heterogeneity of the data used in combination for the analysis (ECMWF, NCEP and aircraft measurement), even though the main results do not depend on the perfect match between these data. But the conclusion that filaments do not follow isentropes could be at least partly due to such a mismatch (see p.5048 l.25-28 and p.5060 l.4-5). In Fig. 3, the isentropes look as expected with respect to the PV contours: The vertical spacing between isentropes is small within the positive PV anomaly (in the northern part of the graphs) as vertical gradient of the potential temperature is enhanced; the vertical spacing then increases passed 2PVU towards the tropical troposphere; in the region of the jet, isentropes tend to be aligned with the PV gradients. But in Fig. 5, they look less natural with respect to the PV contours and they don't have the small scales that would be expected in such a region. The interpolation could well be responsible for errors on the location and tilt of isentropes in this region. Since this region is very dynamical and has lots of small scales, interpolation between two 6hourly data can be inaccurate. I recommend to add a sentence or two on this aspect in the article on p. 5048 l.25-28, and to recall it in the conclusion on p. 5060 l.4-5.**

   The employed interpolation is certainly prone to artefacts and we are actively working on a better trajectory based one. We thus added the sentences:

   > This filament seems to be not isentropic but covers instead a range of potential temperature from 365 K for the highest altitude with a VMR of 300 pptv down to 335 K. Several factors may contribute to this discrepancy. First, both the intrusion of PAN into the stratosphere as also the further development of the filamentary structure along the baroclinic jet-stream may have been non-adiabatic. In addition, the ECMWF temperature model data may be mis-aligned in time and space with the actual physical structure; further artefacts might be introduced by the linear interpolation in time of the available model data. Last, the derived trace gas VMRs represent a complicated weighted mean along the line-of-sight of the instrument, being slightly different for each derived quantity, causing further misalignment with the ECMWF temperature data sampled at tangent point locations.

In addition the conclusion was modified to

> This may indicate that the processes leading up to the imaged situation involve non-adiabatic processes such as uplift by a warm conveyor belt or gravity waves instigated by the baroclinic jet stream. Partly, this may also be an artefact of an imperfect alignment between ECMWF temperature data and the position of retrieved trace gas parcels.

2. **I was sad not to see the water vapour in 2d too! I understand that it is argued in the manuscript that this chemical does not bring enough to the main results and therefore is not included. But it's such a great tracer and it is so important radiatively. I would welcome it if you decided to include it.**

It is indeed sad, but the employed frequency range lacks the required precision for the reliable retrieval of stratospheric water vapour concentrations. However, in future work based on data acquired by the newly developed GLORIA sounder, we plan to use water vapour as tropospheric tracer as more suitable frequencies are readily available.

3. **In the explanation on the instrument and Fig 1 in Section 2, could you add a sentence or two on the resolution of the measurements in the line-of-sight direction?**

The following sentence was added at the specified place:

> The resolution along the line-of-sight is between ≈200 and 300 km, depending on the observed altitude.

4. **P. 5046 l. 5-8: Could you clarify please?**

The sentence was revised to

> The region of sampling is under double tropopause conditions during the flight period, as indicated in the figure. The primary and secondary tropopause levels shown in the figure are derived using the ECMWF temperature profiles.

5. **p. 5046, l. 23-24: This description is not accurate and its purpose is not clear at this stage. Rephrase please.**

The textual passage was changed to:

> The potential temperature structure of the cross section shows typical characteristics of the subtropical transition. The 330–360 K isentropes show upward/downward inclinations above/below the jet core level, marked by the 355 K isentrope, as the flight moving equatorward across the jet. Although these are well known structures, we point it out to connect to the tracer structure shown later.

6. **p. 5046 end of page: Can you say if there were any clouds in the region?**

There were few clouds seen during the flight and all parcels, for which we derived trace gas VMRs are cloud free. Clouds form basically the lower boundary for the retrieval before 06:50 UTC and are sometimes below 8 km altitude after 08:45 UTC.

We added

> There were few clouds seen during the flight above 8 km and all parcels, for which trace gas VMRs are derived, are cloud free.

to the section describing the synoptic situation.

7. **p. 5051, discussion on Fig. 9: Did you look at the PV and potential temperature values along the trajectories? Could that bring some additional information on the dynamics of mixing?**

Yes, but we do not see clear indications. Since the PV and potential temperature histories of the parcel are interpolated from the large-scale field, we do not expect the stretching and mixing to be explicitly represented. More discussion along this line is given in Konopka and Pan (2012).

8. **Section 3.5: You do not mention that HNO_3 is also a stratospheric compound. Is the stratospheric source of nitric acid really negligible for your purpose? I recommend to add a comment on this aspect in this section.**

We reworked the first two paragraphs to include this point:

> This section introduces a proxy for the total reactive nitrogen NO_y from retrieved trace gases and uses this to estimate the influence of tropospheric pollution on air masses in the UTLS. *The* NO_y *proxy proves especially useful when combined with the available* O_3 *measurements.* NO_y *and* O_3 *are well correlated in the lower stratosphere. This is not a consequence of a direct chemical connection, but because their source regions, sink regions, and lifetimes are similar, so that their distribution is jointly determined mostly by transport and mixing processes (Murphy et al., 1993). As the correlation is especially strong in the lowermost stratosphere, deviations thereof may be used to determine the origin of measured air masses.*
>
> The total reactive nitrogen plays an important role in the polluted and unpolluted atmosphere. It mainly consists of NO, NO_2, PAN, HNO_3, HO_2NO_2, and alkyl and multifunctional nitrates. According to Singh et al. (2007), PAN, HNO_3, and NO_x ($= NO + NO_2$) are the major contributors to NO_y in the extratropical UTLS with a combined fractional percentage of about 95 % on average. NO_2 *reacts with* OH *and a third-body to* HNO_3*, which is in turn converted back to* NO_2 *(and* OH*) by photolysis. In the altitude range of the UTLS, the conversion to* HNO_3 *is much faster, so any stable equilibrium between* NO_2 *and* HNO_3 *will heavily favour* HNO_3 *(Austin et al., 1986).* As there are no NO_x estimates available from CRISTA-NF measurements, only a proxy for NO_y can be formed by the sum of the available dominant contributors PAN, HNO_3, and $ClONO_2$. The latter trace gas could be neglected for the current atmospheric situation but would be important for the analysis of polar measurements. In the measurements of Singh et al. (2007), NO_x was the major constituent of NO_y close to the troposphere, so the given NO_y proxy might underestimate the true NO_y

by a factor of 2 to 3, depending on the altitude. But in contrast to the measurements of Singh et al. (2007), most of the air measured by CRISTA-NF should be free of recent influx caused by convection, so the NO_x content should be much lower and the HNO_3 content much higher due to prolonged ageing. Using the satellite instrument UARS, Morris et al. (1997) found a ratio of just 0.1 between NO_x and NO_y at 550 K (≈22 km), implying that the proxy should become more reliable towards the flight level.

2 Reviewer Comments 2

2.1 Major concerns

In my opinion, besides the nice 2-D visualization of the structures, the scientifically most exciting observation is the different chemical composition of the filaments/structures, e.g. the dislocation of the features in O3, PAN, and HNO3 in the eastern cross-section. This should be better carved out. I suggest the following:

1. **For better discern the features for the reader, combine the 3 figures 5-7 in one (5a-c) and locate them on top of another on one page (in which you need the caption for the x-axis only in the lowest graph). Can you somehow lay all three trace gas distributions one over the other, e.g. by using contour plots with a different colour for each gas and only 3-4 contour lines per gas (as graph 5d)?**

The suggested figure placement was not sensible in the physical format of the discussion paper, but will be used in the final paper.

We played around with a couple of visualisation options (e.g. an RGB model for the three trace gasses) and also contour plots. The visibility of even a reduced set of contour lines per gas is poor, though. In the end, the geo-spatial visualisation of tracer-tracer space as shown in the end seems to give the best overview.

2. **From the tracer-tracer-correlation plots one should learn more. First, flip x- and y-axis. O_3 is usually on the y-axis to visualize the transition into the stratosphere. The colours in the Figs. 11, 12, 13 are not well chosen, especially the light and dark purple are badly distinguishable, at least on printed hardcopies. I haven't understood from where exactly the air masses in Fig. 11 come from. Did you just show a data subset with the limits given in the box? With these limits you must get a L-shape tracer relationship, independent on the data. Then I haven't understood the sense of this figure. Is the source region of the light purple data points really so small so that the interpretation of the PAN/O3 slope as (chemically generated) enhancement ratio is valid? May**

it also be possible that the slope simply reflects spatial gradients in the trace gases? ... which would make the interpretation of the slope senseless.

With respect to choice of axis in tracer-tracer plots: we decided to follow Murphy et al. (1993) in plotting the ozone-PAN and ozone- NO_y relationship.

With respect to choice of colour scheme: The similarity of purple colours is somewhat intentional. The most important distinction we want to make is between the three groups of colours. The shading change within the group is less significant. We will nevertheless make some changes of the purple to make it easier to identify the differences especially in printed copies.

Figure 11 is indeed a subset of data points. We will make this more clear in the text. The intention of the "pink" group is to identify the differences between the tropical air and mid-latitude air in tracer-tracer space. The actual slope is not part of the discussion.

3. **The most exciting air mass is the green one in Fig.12. O3 is 200-300 ppbv and thus stratospheric, but PAN as tropospheric tracer is likewise enhanced and reaches up to 500 pptv. The green air masses thus characterize exTL air, right?! I am not an expert in PAN chemistry, but how the PAN is formed there. React e.g. certain aldehydes or oxygenated VOCs with O3, so that PAN was in-situ formed? Simple mixing of air masses in the exTL do not form such strange tracer inter-relationships. Or is O3 wrong, i.e. too high?**

Indeed, the green shaded areas mark the ExTL identified by the specified criteria. PAN is formed from many hydrocarbons as secondary pollutant. It is rather quickly formed and may stay at the depicted altitudes and temperatures for up to several weeks before being photolytically destroyed. PAN actually forms ozone when reacting with NO2, but not to an extent which may explain the ozone anomaly.

When interpreting the results, one certainly has to keep in mind, that to some extent, the relationships are also influenced by smoothing introduced by both the measurement principle and also the regularisation. Also the precision of the measurements is a notable effect as depicted in error bars in the plots. Thirdly, PAN is certainly more heterogeneously distributed in the troposphere and offers thus a different picture from, e.g. CO, which we sadly cannot retrieve from our measurements.

2.2 Minor concerns

1. **p.5040, l.22. "chemically inhomogeneous UTLS region". Change it (although I assume what you want to say)**

We revised it to

... picture of the inhomogeneous distribution of chemical species within the UTLS region.

2. **p.5048, l.23ff. "This filament ...". Try graph d (see above) to visualize possible dis- placements**

A contour plot, especially one that may be printed, sadly does not help here. The textual description details the important points.

3. **p.5050ff. It is hard to reconstruct what air mass is just meant. Thus, define in a list the different air masses with the relevant colour and ever use this definition in the text, e.g. - "tropospheric (blue)" - "high-stratospheric (red)" ...**

The suggestion is followed and the filaments are introduced as

> Four different air streams are identified:
>
> - the "high-stratospheric (red)" filament,
> - the "middle-stratospheric (green)" filament,
> - the "low-stratospheric (blue)" filament, and the
> - the "tropospheric (black)" filament.
>
> The "high-stratospheric (red)" and "high-stratospheric (green)" filament are selected to represent the filament with HNO_3 enhancement and lie mostly within the lower stratosphere poleward of the jet. The "low-stratospheric (blue)" filament is selected to represent the HNO_3 enhanced air mass below the tropopause and is associated with the stratospheric intrusion. Last, the "tropospheric (black)" filament is selected because this airmass of elevated PAN VMRs may give insight into the tropospheric sources of the PAN content of the more stratospheric filaments.

4. **p.5053, l.15. This ozone threshold of 175 ppbv is far too high for an ozone tropopause threshold value which is (seasonally dependent, typically between 70 and 130 ppbv). That is, the given categorization of air mass types and their names in Table 2 are definitely not correct.**

The 175 ppbv ozone criterion is high but the empirically sensible choice based on our ozone values from the retrieval. We will add some discussions in the text:

> The high tropospheric ozone threshold of 175 ppbv is based on the ozone distribution in the sampling region from the retrieval. This high value may be a combination of UT ozone retrieval uncertainty (see Tab 1) and pollution in the sampled airmass.

5. **p.5053.l20 – p.5054.l3. This review on the sources of PAN is something for an introduction and not for the discussion.**

The section was streamlined to the part necessary to demonstrate PAN to be a suitable tropospheric tracer:

In contrast, the major sources of PAN as a secondary pollutant are biomass burning and anthropogenic pollution in the troposphere (Stephens, 1969). Its lifetime is comparatively short, ranging from seconds in the lower troposphere to days and months in the uppermost troposphere (e.g. Roberts, 1990). Thus, it can serve as tropospheric tracer. The distribution of PAN VMRs in measured air parcels above the uppermost detected thermal tropopause shows that only few air parcels surpass 80 pptv of PAN. Air parcels with less than 80 pptv are therefore considered to be chemically stratospheric.

6. **p.5054.l4-11. A L-shape relationship especially of O3 with CO and H2O is only seen in the subtropics at sufficiently high altitudes, but e.g. never in air masses originating polewards of 40°, simply as basically everywhere there is an exTL (characterized by a non-L-shape transition from trop to strat air). Rephrase it.**

The discussion was reorganised:

Trajectory analyses presented in the previous section indicate that the filamentary structure is created by large-scale dynamical processes including Rossby wave breaking. The trajectory calculations, however, are only representing advection by the wind field. The formation of the observed structure involves both advection and mixing (Konopka and Pan, 2012). In this case, small scale processes such as turbulent mixing induced by the shear and strain in the flow, are also expected to contribute to the observed structure, which can be diagnosed using tracer-tracer relationships. Thus, this section explores *the chemical characteristics of the observed trace gas structure using tracer-tracer relationships. This analysis aims to project the information gained in chemical tracer-tracer space to geo-spatial space. As a result, a highly-resolved two-dimensional geo-spatial picture of the UTLS composition near the tropopause break reveals the accumulative effect of advection and mixing of stratospheric and tropospheric air.*

Our analyses focus on the tracer-tracer relationship between O_3 *and PAN, i.e., using* O_3 *as the stratospheric tracer and PAN as the tropospheric tracer.* O_3 *is the most frequently used stratospheric tracer. (e.g. Hoor et al., 2002; Pan et al., 2004).* Using the flight data, 175 ppbv is used as a critical value to separate the stratospheric and tropospheric air mass. *This threshold is higher than the typical 60 to 100* ppbv *used in previous studies (compare Singh et al., 2007; Pan et al., 2007). This empirical threshold is based on the* O_3 *distribution in the sampling region from the retrieval. This higher value may be a combination of UT ozone retrieval uncertainty (see Tab 1) and pollution in the sampled airmass.*

As a tropospheric tracer, PAN has some different characteristics from the frequently used compounds such as CO *and* H_2O. As a secondary pollutant are biomass burning and anthropogenic pollution in the troposphere (Stephens, 1969). Its lifetime is comparatively short, ranging from seconds in the lower troposphere to days and months in the uppermost troposphere (e.g. Roberts, 1990). *80* pptv *of PAN were chosen as the threshold for stratospheric air, i.e., air parcels with less than 80* pptv *are considered to be chemically stratospheric.*

As discussed in previous work, the tracer-tracer relationship in the UTLS for a stratospheric and tropospheric tracer pair is approximately an "L" shape (e.g. Hintsa et al., 1998; Hoor et al., 2002; Pan et al., 2007). The generally compact relationship is from the significantly different ranges of variability between the stratospheric tracers and tropospheric tracers. In this case, a stratospheric branch is formed by the air mass with widely varying O_3 *VMRs and low PAN VMRs, and a tropospheric branch is formed by*

air mass with low O_3 VMRs and high, widely varying PAN VMRs. We proceed to examine the O_3-PAN relationship to identify mixing between stratospheric and tropospheric air.

7. **p.5054ff. (Also in this respect,) I do not see the sense of the given discussion of the PAN-O3 slopes. What can we learn from them? Something on chemical processing, see also my comment above? PAN is a medium-lived compound; thus, the slopes in the exTL and the background above the exTL should tell us something about trans- port/mixing times. Is the stratospheric background of 50 pptv real? PAN is quite different to CO, as LT air is poor and UT air is rich in PAN. Thus, the information we gain from PAN should be different to the one from CO.**

The stratospheric PAN background of 50 to 70 ppt was also retrieved from MIPAS satellite measurements and thus seems rather reliable. The main task of the correlation coefficients is to allow the categorisation of air masses. That aside, the coefficients are also be compared to similar analyses carried through with polluted boundary layer air; as the coefficients for the high ozone VMR anomaly in the troposphere are similar to that of aged polluted boundary layer air is also one puzzle stone validating the high observed O_3 values.

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