

Reply to Reviewer # 1

We would like to thank the reviewer for his/her critical comments which will certainly help to significantly improve our paper. Below, the reviewer's comments are printed in **bold italic face** while our replies are printed in normal face.

This manuscript presents an analysis of the temporal variations in age calculated from MIPAS SF6 measurements. Some very interested changes in age (on seasonal and longer times) are presented, and these results have the potential to have a major impact on our understanding of stratospheric transport. However, more evidence is required to show that the age estimates are realistic, and that the temporal variability shown is real and not an artifact of the uncertainties / variability in the measurements. As described below, the ages from MIPAS are much older than other estimates, and some of the spatial variations in seasonal amplitude and linear trends are not "expected" and should be detected in long-lived tracers if real. Also, there are large trends at the tropical tropopause where age should be fixed at zero.

We'll address these points in the "Specific comments" section.

Major revisions are required before this manuscript will be acceptable for publication. In particular, the revised manuscript will need to

- (i) ***include comparisons of MIPAS age with other observations,***

Some additional material will be provided (see below for more details).

- (ii) ***focus on 2005-2010 data period (and hence eliminate the need for a bias correction),***

We'll show that restricting the analysis to the 2005 to 2010 period will not change significantly the results, but will increase the uncertainties of the derived "trends". For this reason, we'll stay with the 2002 to 2010 period.

- (iii) ***reference the age to MIPAS measurements at the tropical tropopause (so age is zero and time independent there), and***

We'll explain below why this is not feasible in practice, although we agree that it would be desirable.

- (iv) ***include discussion of independent evidence (e.g., temporal variations of long-lived tracers) that support the presented variations in age.***

We'll present a related discussion in this reply, while we are not in favor of including this point into the paper; in our opinion it would distract from the logical flow of the paper.

SPECIFIC COMMENTS

There needs to more evaluation of the MIPAS SF6 and age calculations. There is limited evaluation or comparison with previous studies of the SF6 data or age calculations used in this study. Such a comparison is needed to convince the reader that the age calculations presented are reasonable. In particular, you need to explain why the values shown in fig 5 are much older than other estimates throughout the stratosphere. The very old ages in mesosphere can be explained by SF6 loss, but what explains the age of 3 yrs at 20 km in the tropics?

We have presented in our paper: 1) a comparison of tropical SF6 abundances in the free troposphere (daily averages of the 9-15km altitude range) to ground-based SF6 observations (in situ and/or flask measurements), see Fig. 2, and 2) a comparison of mid-latitude age of air data derived from MIPAS with age of air data from balloon-borne whole air samplers during the MIPAS mission period (Fig. 4 and Fig. 6). Both comparisons provided very good agreement with the respective reference data set. Further we refer to the comparison of co-incident measurements of SF6 profiles from MIPAS and balloon-borne whole air sampling data as presented in Stiller et al., 2008.

We do not agree that "the values shown in fig 5 are much older than other estimates throughout the stratosphere". The good agreement with age of air data from Engel et al. (2009) as shown in Fig. 4

and 6 demonstrate that our age of air values are well in the range of other observations. Regarding the question “but what explains the age of 3 yrs at 20 km in the tropics” we compare in Fig. 1 of this reply the MIPAS-derived latitude cross section at 20 km of age of air to data gained in the early 1990s during aircraft campaigns (Elkins et al., 1996; Ray et al., 1999; Harnisch et al., 1996; data digitized from the publication by Waugh and Hall, 2002). While the agreement is very good at middle and high latitudes, there is indeed on average a discrepancy in the age of air data at the tropics. Not only that the MIPAS-derived age of air is higher, the shape of the latitudinal dependence is different. Furthermore, the comparison from day to day shows a variation of shape of the latitude cross-section, with variations between the hemispheres representing the seasons, and also a multi-year development of increasing and decreasing age in the inner tropics and age becoming particularly younger and older again around 25°N and S.

Even with the differences described above, the aircraft measurements are still in the range of values covered by individual MIPAS observations. For the discrepancy between the aircraft measurements and the MIPAS mean, we offer two explanations:

1. The aircraft measurements might not be representative for a global state of the atmosphere. The aircraft campaign measurements of the early 1990s are just snapshots for which single data points have been combined to a latitude cross section, while MIPAS data provide true global coverage at the same observation time. Note that the aircraft data still fall within the distribution of values measured by MIPAS.
2. These measurements are 10 - 20 yrs older than MIPAS measurements. The atmosphere might have changed since then. This would not be unexpected, given the observed cooling of the tropical tropopause around the year 2000 and the recovery during the following years, and the observed variability during the measurement period of MIPAS. The aircraft measurements show larger gradients in the regions of the mixing barriers. If mixing barriers had become weaker, as outlined in our paper as a hypothesis, tropical age of air would have increased. This seems to us to give a self-consistent picture.

We know that this is an “unexpected” result but we are very confident in the MIPAS measurements and their error analysis. Although there is no indication to us that anything is suspicious with the data, we agree that the issue of a potential artifact in the tropical MIPAS measurements can never be completely ruled out, and we’ll take care to use a careful wording in the revised version of the paper. Explaining the high age in the lower tropical stratosphere might become the task of the community in the coming years. The fact that this observation is “unexpected” is not a strong argument for preventing publication to the community.

We’ll include an improved version of the figure below and the related discussion as outlined above in the revised version of the paper.

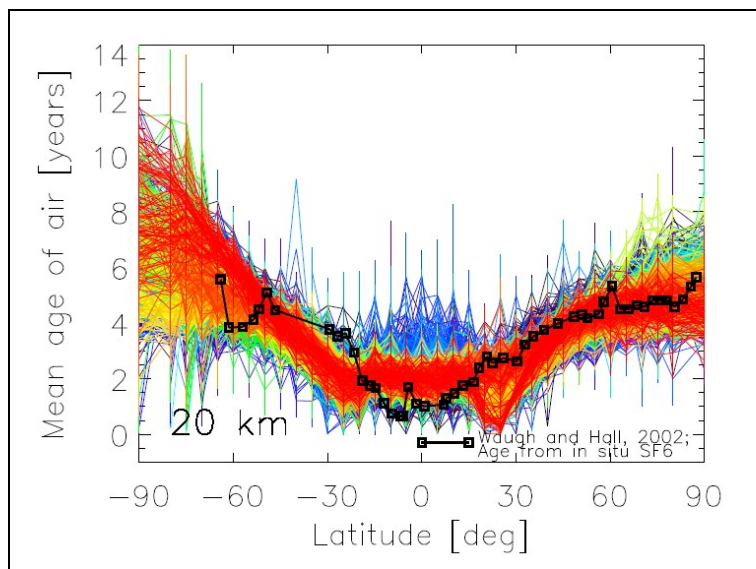


Fig.1: Latitudinal cross-sections of all daily means of age of air from MIPAS for the period 2002 to 2010 (each colored curve represents one day, color coding is from blue = early 2002 data sets over green and yellow to red = 2009/2010 data sets). The black curve with symbols is the latitude cross-section of age of air at 20 km from aircraft measurements (Elkins et al., 1996; Ray et al., 1999; Harnisch et al., 1996) published as in Waugh and Hall (2002). Figure will be improved for the inclusion in the revised version of the paper.

I suggest that before the time series plots there needs to be some plots like figure 5, showing a combination of SF6 and age, as well as different years and/or months. These plots will allow the characteristics of these distributions to be compared with other published distributions, and will also illustrate the temporal variations.

A figure with seasonal zonal means of age of air and the related description/discussion will be included, as also requested by the other reviewer. Fig. 2 of this reply gives an example of how this figure would look like.

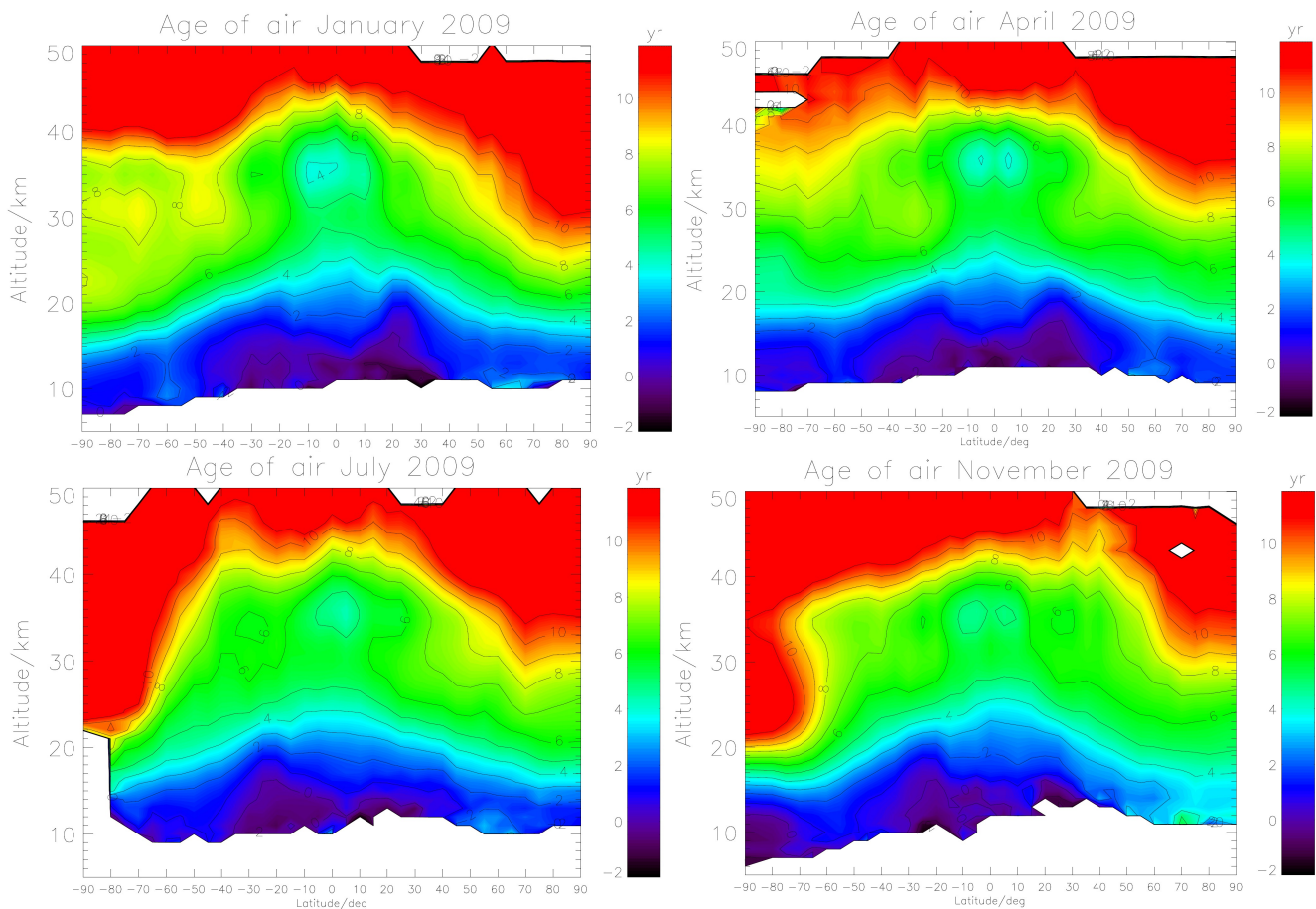


Fig. 2: Monthly zonal mean distributions of mean age of stratospheric air for the months January 2009 (top left), April 2009 (top right), July 2009 (bottom left), and November 2009 (bottom right), representing the DJF, MAM, JJA, and SON seasons.

I realize that some evaluation has been done in Stiller et al (2008) (and this paper is referenced a lot),...

Exactly, some validation was already done. The reason for its extensive referencing is that it is the precursor of this work; the retrieval approach and data characterization are discussed there in detail.

... but they focused on the 2002-2004 data and as indicated below there are clear differences between the two periods.

The comparison to co-incident balloon borne whole air sampler measurements in Stiller et al., 2008 has indeed been done for 2002-2004 data only. We also agree that there are differences between the two periods which, however, cannot be linked to the retrieval method being essentially the same for both periods. These differences can be summarized by

1. a potential bias, dependent on latitude and altitude, but not on time, which may affect the absolute age of air but not the assessments of linear increase/decrease and seasonal variation;
2. less noisy time series for the second period; the reason is the time-dependent spectral calibration artifact in the data of the first period which could be corrected for with some approximation only (see Stiller et al., 2008, Appendix); for this reason, we consider the data set of the second period as of better quality and less noisy.

Furthermore, and perhaps more importantly, the differences shown in Stiller et al (2008) are large. On page 28019 it is quoted that agreement is within 0.5 pptv, but this is not what I see in fig 4 of Stiller et al. (2008) where differences in SF6 from MIPAS and balloon of larger than 1 ppt, with similar differences between nearby MIPAS profiles. Even if agreement is within 0.5 pptv this corresponds to an error of over 2 years in age! This large difference in age needs to be discussed better, especially as much of the paper focuses on variations in MIPAS age of less than 1 yr (seasonally or trend).

The validation with co-incident balloon data needed to be done on basis of single MIPAS SF6 profiles for which the random uncertainties are high; we provide random errors in the order of 10% for single profiles which is consistent to the differences found between MIPAS and balloon data; further, we say in Stiller et al., 2008, that good agreement is found for **close** co-incidences (page 682, 1st column, line -5). We have shown in the 2008 paper in Fig. 4 all coincidences within 1 day, 1000 km and regardless which potential vorticity (PV). In each of the three panels, the coincident measurements which are **closest** in time and space and PV always agree within 0.5 pptv. The precision of a single profile with 0.5 pptv is certainly not good enough to do reasonable age of air assessments. For this reason, all further analysis has been performed on basis of zonally averaged data. Averaging provides a reduction of the random error by \sqrt{N} , with N in the order of several hundred measurements (for monthly means), while systematic errors (like a bias) cannot be reduced. The age difference between Engel et al.'s age regression line and MIPAS age regression lines is about 0.3 yrs only (see Figs. 4 and 6 in the manuscript), i.e. we can rule out that the ≤ 0.5 pptv differences found in the validation are a systematic bias. A major part of this difference is single measurement random error (noise). We'll include a more detailed discussion on the error issues in the revised version of the paper.

2. The data from 2002-2004 and 2005-2010 are combined in the analysis presented, but there are clear differences between the data from the two periods. Not only is there a large bias between the mean values but there are also significant differences in the variability within each period. This can be seen in the time series plots in fig 4 and 6. For example, the data after 2005 in fig 6 shows a clear seasonal variation but this is not the case for the initial years.

As already noted above, the differences between the two periods are

1. a potential bias, dependent on latitude and altitude, but not on time, which may affect the absolute age of air but not the assessments of linear increase/decrease and seasonal variation;
2. less noisy time series for the second period; the reason is the time-dependent spectral calibration artifact in the data of the first period which could be corrected for with some approximation only (see Stiller et al., 2008, Appendix); for this reason, we consider the data set of the second period as of better quality and less noisy in the time domain.

The bias is accounted for in the fit procedure as described by von Clarmann et al., 2010 (see section 4.1 of the manuscript). The higher noise level in the earlier period explains the apparently less pronounced seasonal variation during 2002-2004.

I don't think the inclusion of the early period adds anything, and I think the focus should just be on the latter period. This will make the paper cleaner and eliminate one uncertainty from the trend calculations.

The two periods with a potential bias among them do not introduce an uncertainty to the assessment

of the linear increase/decrease, as the method applied (von Clarmann et al., 2010) does account for the unknown bias between the two data sets in a quantitative, numerically sound and robust manner.

In order to demonstrate that the earlier noisier data set does not harm our analysis, we have repeated the regression analysis for just the second data set for several example cases and have not found any significant differences which affect our conclusions. But certainly the results are not identical. In particular, the uncertainties of the derived parameters (“trends”, amplitudes, phases) for the shorter data set are larger. Fig. 3 of this reply gives an example for the fits with and without including the first part of the time series.

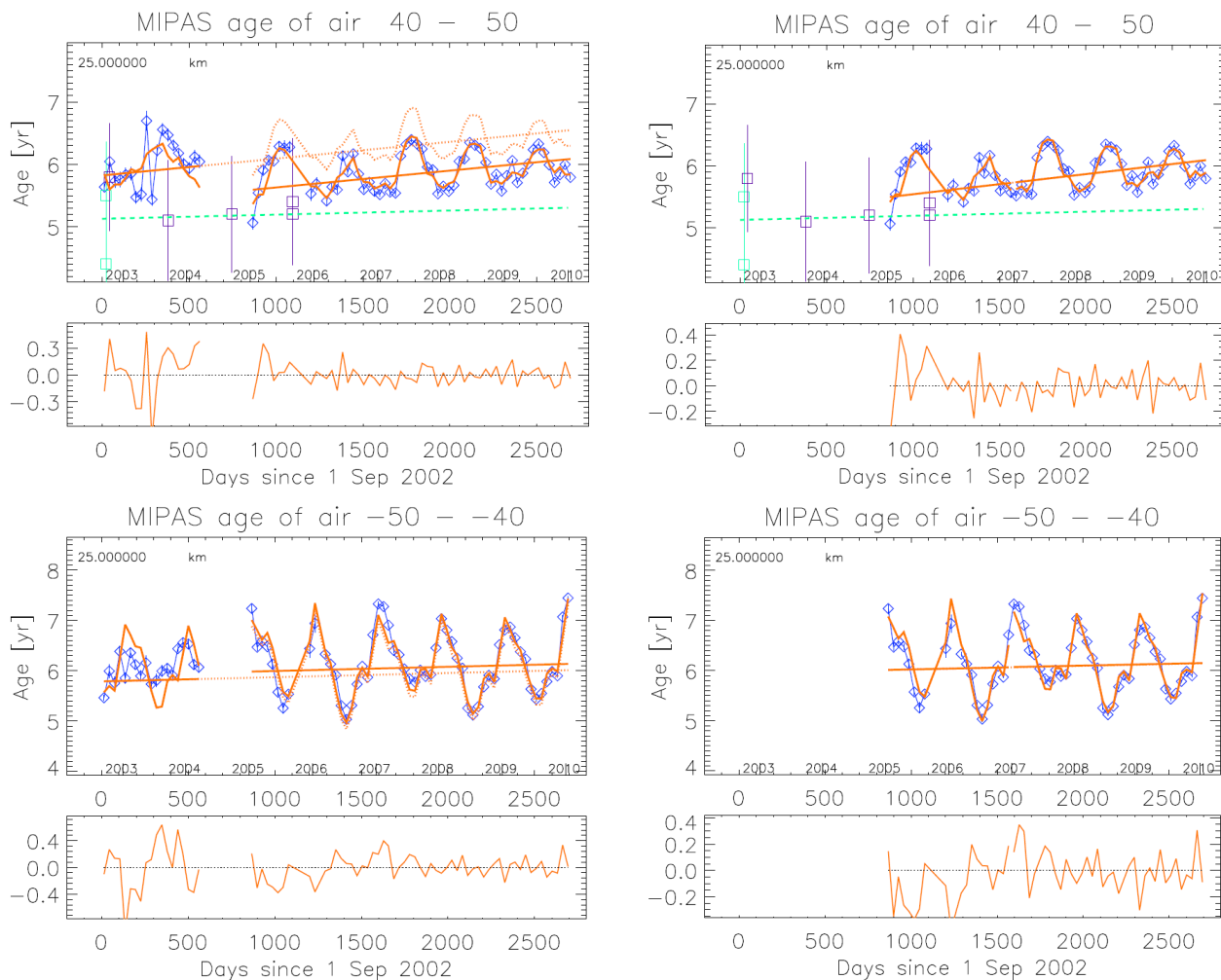


Fig. 3: Examples for regression analysis with (as in the manuscript)(left row) and without(right row) the first data set for 2002 to 2004, for 40N-50N, 25 km (top), and 50S-40S, 25 km (bottom). The fit quality and the results are similar for both tests, despite the rather large bias between the early and the later data set in the upper left case. This confirms also the validity of our method.

The data record is already quite short for a trend analysis, and since we have a method available which makes our trend estimates immune against a bias between data subsets, we are reluctant to make the data record shorter than necessary.

3. There are significant trends in the age of air entering the stratosphere, but the age should be zero and constant here (otherwise some of the trends are due to changes in tropospheric transport).

We agree that ideally, age of air should be zero at the entry point of the stratosphere by definition, i.e. due to a proper reference data set. We address the issue of variable age at 18 km below. Here, we would first like to outline that there are several difficulties with a SF6 reference data set at the tropopause when dealing with observational data:

1. Global tracer measurements at the tropopause which could serve as reference for the age of air determination are not always available; in our specific case, we would need SF6 observations at the tropopause for a significant period prior to the MIPAS observations in order to determine stratospheric age of air up to 7 years or more. Or, in other words: for the year 2010, ages up to 8 years could be determined from MIPAS observations alone, while for the year 2002, no age at all could be determined.
2. The tropopause is not constant and invariable in time and space; how can we know where exactly (in particular, at which altitude) the observed air parcel crossed the tropopause some years ago?

For these reasons, we do not see a practicable way to determine age of air without referring to the ground-based data set. We will, however, add a caveat what the term “age of stratospheric air” exactly represents in our case (and all other observational studies on age of air). Further, any other observational data set of stratospheric air refers to the ground-based observations of SF6 and/or CO2 to make the transformation from tracer vmrs to age of air. It is well documented in literature that this is problematic to some degree, but there is no other practicable way, and we do not see why it should not be allowed in our case to use the term “age of stratospheric air” in this slightly redefined sense while it is accepted in all other cases.

It is stated that SF6 needs to be referenced to abundance at the tropical tropopause (pg 28022), but a surface time series is used. (I disagree that we should "not expect any significant delay of upper tropospheric SF6 compared to surface values" (Pg 28021, line 15). I would expect the UT values to be lower, with a delay of around 6 months at the tropical tropopause.)

We agree that a delay in the order of 6 months can occur until an air parcel from the ground/boundary layer is transported through the tropical tropopause; this is a typical value provided by chemistry transport models (e.g. Felix Bunzel and Sophie Oberländer, private communication). However, we refer to the “free upper tropospheric” SF6 here; the current view is that air is transported into the tropical upper troposphere (still below the TTL) within hours or days, normally by convection, while the transport through the TTL from its lower boundary around 14 km to the tropopause around 17 km is very slow and can take months. Since we compare daily mean SF6 data from MIPAS for the 9-15 km altitude range (i.e. below and at the lower boundary of the TTL) to the ground-based data, we think our assumption is correct. We will clarify in the text that “upper tropospheric” values are limited to altitudes below 15 km and explain this reasoning.

Besides the reasons mentioned above, more related to practical considerations, we have not used MIPAS SF6 at the tropopause (around 17 km) for the transformation into age of air because tropopause SF6 was found to be already affected by seasonal and QBO variations typical for the stratosphere. This might be due to the limited altitude resolution of about 4 km which does not allow separation of stratospheric and tropospheric air completely for a location just at their boundary. However, using SF6 from the tropopause with its seasonal and multi-year variation would have introduced additional time-dependent biases in the age of air calculation which we had to avoid.

The issue with the reference point is basically a matter of definition, and not of wrong or right. Age of air measurements as provided by all instruments provide the time lag between the **entry** into the TTL and the measurement date, while modelers refer to age as the time lag between the time the air **leaves** the TTL into the stratosphere and the actual date. None of these is wrong, but we must be careful to exactly describe what we have. The modelers' AoA definition cannot be measured because relevant reference data do not exist. The problem is, that, when AoA was first discussed (and when this term was introduced), the slow transport through the TTL was not yet considered by anybody in

AoA discussions. The knowledge we have today on the transport through the TTL requires AoA definitions to be slightly modified to be still useful.

In summary, we agree that referring to a ground-based or free tropospheric SF6 abundance instead of the tropopause SF6 abundance might introduce a bias to the age of air data which could, in the worst case, even be time dependent, in the case the transport up to the tropical tropopause has some variation with time. We will include a discussion of this topic in the revised version of the paper, including the mention of a potential bias of the order of +0.5 years due to the reference altitude not being the tropopause.

Also, I don't understand why age is calculated relative to surface measurements without corrections for biases in the MIPAS measurements.

Our first reason not to correct the MIPAS age data is that we wanted to keep our age of air observations comparable to other observational data sets; all other observational data sets known to us which derive age of air from SF6 observations refer to the ground-based observations of NOAA/ESDL without any further correction. Second, we prefer not to transform our data from the one into the other system by just subtracting 0.5 yrs from our age, because this ad-hoc correction would add another uncertainty; we do not know the exact value for the correction for each time and latitude band, as it may vary with time and space. Further, an assumed constant time lag of about 0.5 years does not harm our analysis of linear increases/decreases and seasonal variations; however, it has to be taken into account when comparing to model results which set age of air to zero at the tropical tropopause. We will provide a caveat on this point.

The use of reference time series from surface measurements results in unrealistic age at the tropical tropopause:

First, as mentioned above, the age at and just above the tropical tropopause is very old. In fig 5 the age at EQ and 18 km is around 2 yrs old, whereas age should be around zero at tropical tropopause. Now this could be just a matter of a uniform shift in the age, but it is not (as next point explains).

As mentioned above, the tropopause is not constant in altitude over time, and is not at the same altitude over all tropical latitudes. The picture given by the reviewer is very simplified. In the example presented with Figure 5 of the manuscript, the zero isoline of AoA is in between 12 and 15 km which confirms Fig. 2 of the manuscript, where agreement between tropical MIPAS mean values between 9 and 15 km and the ground-based in-situ data has been demonstrated. Ages around 6 months (about the time needed for crossing the tropical tropopause layer) are found between 16 and 17 km, which is a reasonable altitude for the tropical tropopause. Around 18 km, the age is between 0.8 and 1.8 years (and not 2 years as stated by the reviewer). After subtraction of the 0.5 year time lag due to cross-TTL transport, this results at an age of 0.5 to 1.3 years after crossing the tropopause at 16-17 km. As mentioned in the manuscript, increased mixing due to weakening of mixing barriers would result in in-mixing of older midlatitudinal air and increase the age even in the tropical lower stratosphere, just by changing the shape of the age spectrum. The process of in-mixing itself is probably time dependent which explains variation of age just above the tropical tropopause.

Second, and more importantly, there are significant negative trends in age (1 yr per decade) at the tropical tropopause (figures 4 and 8). Not only should the age be zero at the tropical tropopause, it should be constant in time.

Again, the tropopause altitude varies with time and latitude; a constant = zero value of age of air at the tropopause should be expected, but cannot be expected for a fixed altitude level. In Fig. 4, we have shown, as one example, the time series of age of air for the 10°S-0 latitude bin at 16 km.

[We'll replace this figure (and the others related to 10°S-0) by figures for 20°S-10°S, because we just realized that the fit for 10°S-0/16 km was rejected for Fig. 8 because of too large χ^2 . The replacement figures are presented below as Fig. 4 of this reply.]

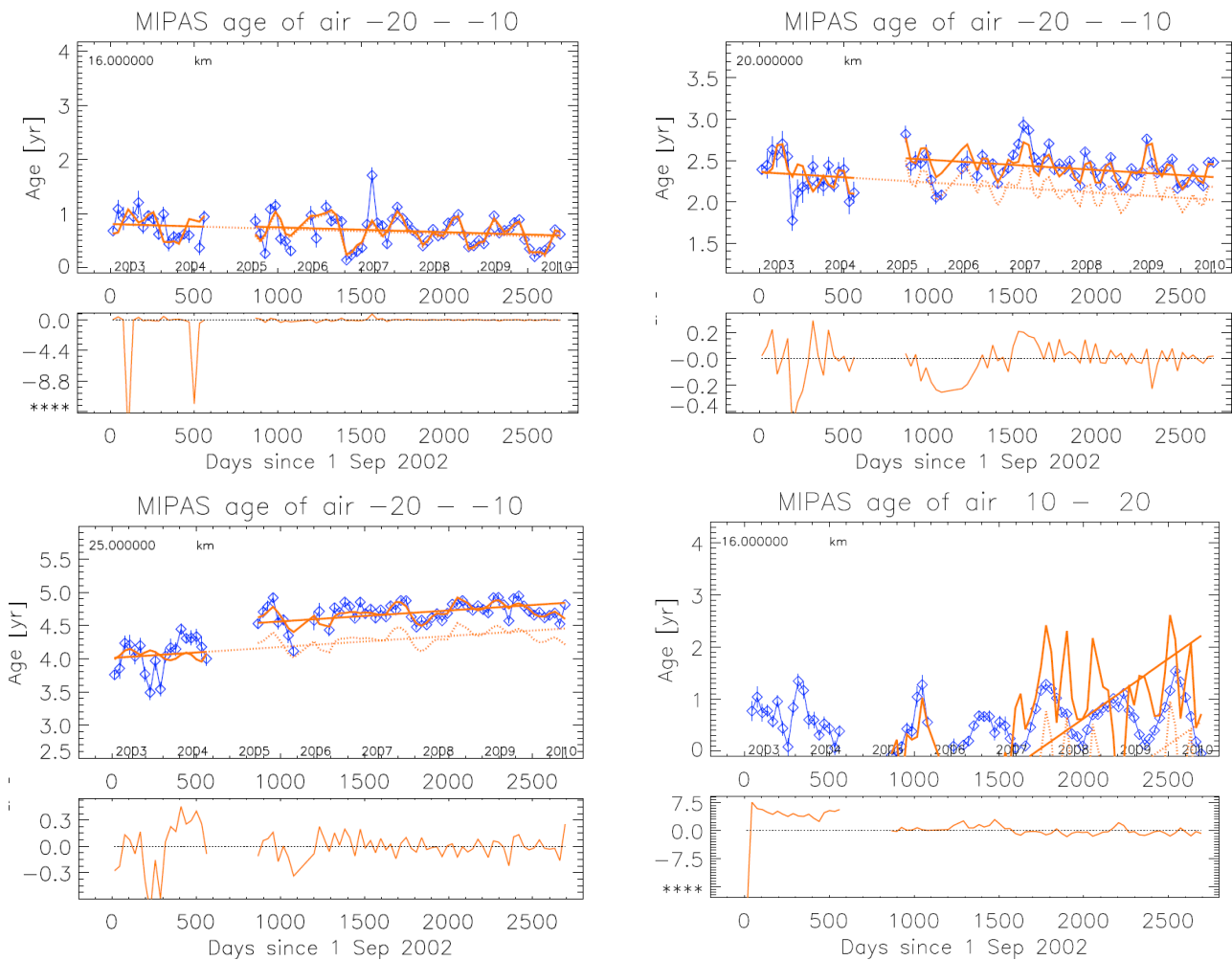


Fig. 4: Time series of age of air at 20°S-10°S for 16km, 20 km, and 25 km altitude (will replace time series for 10°S-0 of Fig. 4 of the paper), and 10°N-20°N, 16 km (bottom, right). The latter panel will not be included in the paper because the fit was not successful. However, the observational data alone confirm the statements made in the response to the reviewer.

For 20°S-10°S, 16 km, the age varies between about 0.2 and 1.2 years, with lowest age in the hemispheric winter/spring, one larger value at 1.8 years, and a slight linear decrease over the 8 years period. At the same time, we find similar ages of 0 to 1.2 years for 10°N-20°N, with lowest ages shifted by 6 months in phase, again in the hemispheric winter/spring, and a slight increase (see Fig. 4 of this reply, bottom right panel). The hemispheric differences reflect the well-known fact that upwelling in the tropical branch of the Brewer-Dobson circulation is strongest during hemispheric winter, while during hemispheric summer, the Brewer-Dobson circulation becomes rather weak. Regarding the seasonal variation and the linear increases/decreases, on average they almost cancel out, i.e. transferred to the simplified picture of constant and homogeneous tropopause, there is no variation and no “trend”. If we were asked to speculate, we would relate the observed hemispheric “trends” at or above the tropical tropopause to a change of tropopause height related to the extension of the tropical belt (e.g. Seidel et al., 2008; Seidel and Randel, 2007; Birner, 2010) with different extent in the two hemispheres; however, there is no further robust indication in the observational data for this, so we prefer not to include such a speculation in our paper.

What is the cause of the trend in your calculation of age at the tropical tropopause? Does this indicate trends in transport within the tropopause, or is this a possible artifact of the measurements near the tropopause.

See reply above. We think the “trend” at 18 km is more related to a variation of tropopause height with time and latitude, since we cannot expect the tropopause to be constant at a fixed height over all the tropics; besides, 16-17 km seems to be more reasonable for the actual tropopause height, and variation at 18 km could be also produced by time-dependent in-mixing of mid-latitude air. We are confident that our observations are correct and do reflect the actual atmosphere; nature is not as simple as a simplified scheme of the tropical tropopause might suggest.

Regardless of the cause, if age was referenced to the MIPAS SF6 tropical tropopause measurements then (by definition) the age trend there would zero and the trend at other stratospheric locations would be decreased by 1 yr / decade, i.e., locations that currently have a positive trend of 1 yr/decade would have no trend, and those with negative trend would have its magnitude increased by 1 yr/decade. The spatial structure in fig 8 would be the same but magnitude would be different.

We do not agree; we have outlined above that the temporal variation of age at fixed altitudes around the “tropopause” makes sense and the critics by the reviewer is due to a simplified picture of a homogeneous altitude-constant tropopause over all seasons and tropical latitudes, without any hemispheric differences regarding the temporal variations, and constant upwelling all over the year. There is not a constant age trend of 1 year/decade at the “tropopause”, but as shown above, variations in the various latitude bands cancel out.

Further, as outlined above, reference to MIPAS SF6 measurements at the tropopause is unfortunately not feasible. For age assessment we would need a data record extending further into the past, in order to be able to assign each observed SF6 mixing ratio in the stratosphere to the related age.

I realize there will be added uncertainties defining the reference time series, due to sensitivity to which altitude is used and extrapolating the MIPAS measurements. But these uncertainties need to be considered, and could be larger than the uncertainties you already quote for age trends.

See above. We cannot derive age of air without a data record extending much longer into the past than the MIPAS data themselves. So we have to use a slightly different definition of age of air.

4. The spatial variations in the seasonal amplitude and linear trends of age are large, and not what would be expected. For example, I would expect the sign of the trend in tropical age to be the same throughout the stratosphere, or at least not to oscillate from -1 yr/dec to +1 yr/dec. As all evidence points to a relatively isolated tropical pipe this would require a rapid speed up of vertical advection in lower stratosphere followed by an even large slow down in region above (and maybe downwelling rather than upwelling). Is there independent evidence for this?

As we suggest in Section 6 (page 28035 bottom, page 28036 top) we consider the main impact on changes of mean age of air coming from a change in mixing, i.e. changes of the shape of the age spectra, and not from changes of vertical advection. If we assume that the tropical pipe is not as isolated as the reviewer suggest, i.e. the “leaky pipe model” of Ray et al. (2010) applies, any longer-term change of age of air can be explained by changed mixing of tropical and mid-latitude air, i.e. by a change of the permeability of mixing barriers. Unfortunately, we cannot derive information on the shape of the age spectra for different spatial regions from our data; knowledge on the age spectra and if/how they have changed over recent years would elucidate the situation dramatically. We will point

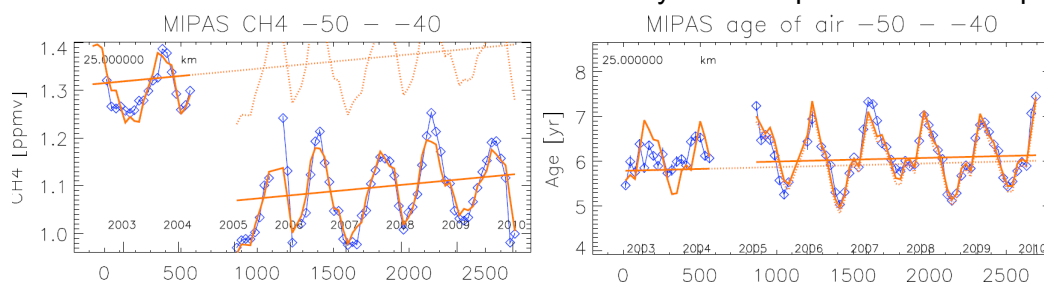
out the role of age of air spectra and the impact of mixing in more detail in the revised version of the paper.

Also, I would expect the seasonal amplitude at midlatitudes to be more similar than shown here, e.g. at 25 -30km the amplitude at 40-50S is around 1 yr whereas at 40-50N it is less than 0.5 yr. Now this could what is happening in the real atmosphere and highlight issues with my expectations. However, if the seasonal cycle amplitude in age is as shown then we should see similar variations in long-lived tracers (e.g. CH₄, N₂O). (Tropical age trends changing from -1 to +1 yr/dec would also leave a signature in these tracers.) Do data from MIPAS or other satellites provide any evidence for these changes in long-lived tracers?

The inter-hemispheric differences are caused by the different stability of the Northern and Southern polar vortices, respectively, and by the positions of the subtropical transport barriers which vary differently among the hemispheres. In another paper (Palazzi et al., 2011) we have shown that the subtropical transport barrier can be located as high as 32°N, while the Southern hemispheric subtropical transport barrier never exceeds 27°S. The difference mentioned by the reviewer with the larger seasonal cycle in the SH above 25 km is due to the stronger and more stable Southern polar vortex, filled more completely with mesospheric air, the latter being released into mid-latitudes after the vortex break-down. This is very clearly illustrated in the animation provided as supplement to the paper.

The seasonal variation is confirmed by other tracers observed by MIPAS. We show in Fig. 5 of this reply the variation of methane for tropical and middle latitudes together with the variation of age of air. We find similar seasonal variations in all long-lived tracers observed by MIPAS (we have checked CH₄, N₂O, CFC-11, CFC-12, HCFC-22 for such variations). The seasonal amplitude of methane is much larger in the Southern hemisphere, similar to the amplitude of age of air (compare Fig. 4, 1st and 2nd row). The phases of the seasonal methane variation fit to the age of air phases: lowest mixing ratios of methane are found for oldest air, reflecting the on-going destruction of methane by (photo-)chemistry. Regarding the tropical age of air “trends” and their variation with altitude, the methane observations again confirm the findings for age of air: in the lowermost stratosphere, at 18 km, where age of air has been found to decrease over several years, methane is slightly increasing (see Fig. 4, 3rd row), while in the middle stratosphere where we found the age of air to be increasing, methane has a negative “trend” (see Fig. 4, 4th row). However, we cannot explain to date why methane increases over several years while age remains almost constant in the Southern mid-latitude middle stratosphere (1st row).

We would like to point out that the methane data and their analyses are not published yet, and we consider our analysis as not yet ready for publication. We have presented here the data to support our findings regarding age of air, but we cannot explain every detail in the methane time series yet. For this reason we are not in favor to include this analysis in the present manuscript.



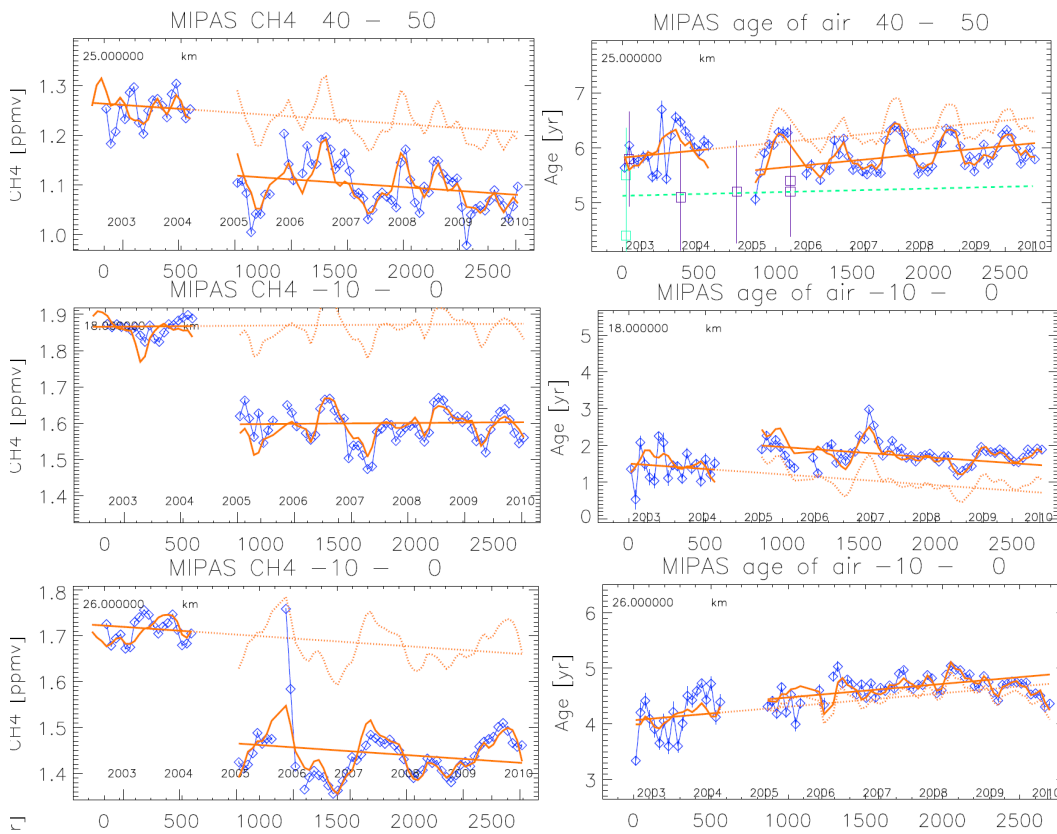


Fig. 5: Time series of methane and age of air at Southern and Northern mid-latitudes (40-50°N/S), 25 km (upper two rows) and tropics (10°S – 0), 18 km and 26 km (lower two rows). As pointed out by the reviewer, the seasonal amplitude is stronger in the Southern hemisphere than in the Northern hemisphere, both for age of air and methane. The seasonal variation of the methane abundance is such that the methane volume mixing ratio is highest for youngest air. In the tropics, methane reveals a small multi-year increase with decreasing age of air (18 km), and a multi-year decrease with increasing age of air (26 km). Around 26 km, a strong QBO signal is present in the methane time series as well.

5. The estimate of the impact of SF6 depleted air is very crude, and the values assumed used for these calculations are very uncertain. I don't see how you can give such precise estimates of the possible effect, e.g., 0.59 +/- 0.08 yr/decade. Does the uncertainty of 0.08 really account for the crudeness of the calculation? For example, in these calculations you assume an age of 5 yrs, but you own calculations indicate ages much older than this. At high latitudes you have ages between 6 and 10 years, so why not use 8 yrs in your calculation? I could make similar statements about the other assumed values in these calculations.

We admit that the estimate of the impact of SF6 depleted air in the paper is a very rough assessment only; the uncertainties given in the paper were estimated from the multi-annual variations of the numbers derived, and do not contain systematic effects like uncertainties of the oldest stratospheric age assumed. We admit that the presented uncertainties may give the impression that the numbers are more accurate than they really are; we'll therefore remove any uncertainty assessments and state explicitly that we refrain from presenting any error estimates due to the large systematic uncertainties implicated in our assessment.

MINOR COMMENTS

1. It is hard to see how good the agreement is in the upper panel of Fig 2. It would be much more useful to compare monthly mean values of the different datasets (may be with error bars to indicate variability).

For this reason we have added to the top panel the additional panels showing the residuals between each of the data sets. We want to keep the transformation of SF₆ into age of air on a daily basis as done in the paper, in order not to introduce additional uncertainty into the age data related to the averaging over months.

2. I also find figure 7 a little hard to follow. Lat-height plots showing deviation of monthly (or seasonally) mean age from the annual-mean age would be much clearer. Could show this for each month or every other month or every season. A reader could easily see the magnitude of seasonal variations and also when peak occurs from these plots.

We'll revise Figure 7 for the revised version of the paper. Another option besides that suggested by the reviewer would be to show the amplitudes as altitude-latitude cross-sections similar to the current version, but provide the months of youngest or oldest air within the seasonal variations as another altitude-latitude cross section, replacing the two lower panels of Figure 7.

References

- Birner, T., Recent widening of the tropical belt from global tropopause statistics: Sensitivities, *J. Geophys. Res.*, 115, D23109, doi:10.1029/2010JD014664, 2010.
- Elkins, J. W., et al., Airborne gas chromatograph for in situ measurements of long-lived species in the upper troposphere and lower stratosphere, *Geophys. Res. Lett.*, 23, 347–350, 1996.
- Engel, A., Möbius, T., Bönisch, H., Schmidt, U., Heinz, R., Levin, I., Atlas, E., Aoki, S., Nakazawa, T., Sugawara, S., Moore, F., Hurst, D., Elkins, J., Schauffler, S., Andrews, A., and Boering, K.: Age of stratospheric air unchanged within uncertainties over the past 30 years, *Nat. Geosci.*, 2, 28–31, doi:10.1038/ngeo388, 2009.
- Harnisch, J., R. Borchers, P. Fabian, and M. Maiss, Tropospheric trends for CF₄ and C₂F₆ since 1982 derived from SF₆ dated stratospheric air, *Geophys. Res. Lett.*, 23, 1099–1102, 1996.
- Palazzi, E., Fierli, F., Stiller, G. P., and Urban, J.: Probability density functions of long-lived tracer observations from satellite in the subtropical barrier region: data intercomparison, *Atmos. Chem. Phys.*, 11, 10579–10598, doi:10.5194/acp-11-10579-2011, 2011.
- Ray, E. A., et al., Transport into the Northern Hemisphere lowermost stratosphere revealed by in situ tracer measurements, *J. Geophys. Res.*, 104, 26,565–26,580, 1999.
- Ray, E. A., et al., Evidence for changes in stratospheric transport and mixing over the past three decades based on multiple data sets and tropical leaky pipe analysis, *J. Geophys. Res.*, 115, D21304, doi:10.1029/2010JD014206, 2010.
- Seidel, D. J., Q. Fu, W. J. Randel, and T. J. Reichler, Widening of the tropical belt in a changing climate, *Nature Geoscience* 1, 21–24 doi:10.1038/ngeo.2007.38, 2008.
- Seidel, D. J., and W. J. Randel, Recent widening of the tropical belt: Evidence from tropopause observations, *J. Geophys. Res.*, 112, D20113, doi:10.1029/2007JD008861, 2007.
- Stiller, G. P., von Clarmann, T., Höpfner, M., Glatthor, N., Grabowski, U., Kellmann, S., Kleinert, A., Linden, A., Milz, M., Reddmann, T., Steck, T., Fischer, H., Funke, B., López-Puertas, M., and Engel, A.: Global distribution of mean age of stratospheric air from MIPAS SF₆ measurements, *Atmos. Chem. Phys.*, 8, 677–695, doi:10.5194/acp-8-677-2008, 2008.
- Von Clarmann, T., Stiller, G., Grabowski, U., Eckert, E., and Orphal, J.: Technical Note: Trend estimation from irregularly sampled, correlated data, *Atmos. Chem. Phys.*, 10, 6737–6747, doi:10.5194/acp-10-6737-2010, 2010.
- Waugh, D., and T. Hall, Age of stratospheric air: Theory, observations, and models, *Rev. Geophys.*,

40(4), 1010, doi:10.1029/2000RG000101, 2002.