

***Interactive comment on* “Global distribution of mean age of stratospheric air from MIPAS SF₆ measurements” by G. P. Stiller et al.**

G. P. Stiller et al.

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We would like to thank the referee for his/her suggestions which definitely help to improve the paper (referee comments are inserted *in italics* for convenience).

Summary and General Comments:

This paper reports a new, less biased retrieval of SF₆ observations from the MIPAS instrument on Envisat for the period Sept 2002 to March 2004. The new retrieval method is discussed in detail, with further elaboration in an appendix; comparisons for the sake of validation are made with ground-based and balloon observations. The KASIMA model is used to demonstrate that mesospheric loss processes are essential for correct interpretation of stratospheric air derived from the SF₆ measurements. The SF₆ observations are used to identify episodes of descent of mesospheric air in to the

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polar vortices in winter.

The paper contains valuable new measurements of SF₆ which give us 8217; observations8217; of stratospheric age. The comparison with the model is also important for demonstrating the importance of losses and thus in the calculation of the correct age of air. A considerable fraction of the paper is spent discussing the details of the retrieval. I suppose it is important to document this, but it may not be that interesting for many readers.

Since another previously published data set of SF₆ global distributions derived from MIPAS data already exists (Burgess et al., 2004; 2006) we consider it necessary to describe the retrieval procedure in detail, so that interested readers can deduce where the differences in the data sets come from.

The authors frequently interpret their 1.5 year data set in terms of trends and interannual variability. This data set is far too short for such interpretation and those aspects of the paper need to be eliminated or modified.

The referee is correct that we interpret the tropospheric variation in SF₆ over time as a trend; this is based on the a priori knowledge provided by ground-based measurements from, e.g. NOAA/ESRL/GMD; more justification for this interpretation is given below.

Besides this point, we tried to be careful in the original version not to give the impression that we deduce long-term trends or climatological variabilities from the 19-months MIPAS data set. In the revised version we will take even more care to talk about differences between years, seasons, or hemispheres, and not to indicate any conclusions on climatological variability or trends.

While I think it is very useful, especially to the chemistry-climate modeling community, to have empirical age of air for the entire stratosphere, I rather dislike that the figures are nearly always of “apparent” mean age rather than the true age calculated when the mesospheric loss is included. Someone skimming this paper may come

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away thinking that stratospheric high latitude age is much greater than it really is. This aspect may not be something you want to change, but I think this paper would serve its readers better with an emphasis on true age rather than apparent age.

It is well known in the community that SF₆ observations directly can provide apparent age only; in order to avoid any doubt about this, we have included “from MIPAS SF₆ measurements” in the title of the paper. As the referee states, the data set will possibly be useful for validation of chemistry-climate models. The only way to correct the apparent age to the true age we can see is to correct the observational data set by differences provided from model runs with/without the mesospheric loss included. I.e., the modified data set would rely on assumptions and qualities of the model runs used; in order to avoid introduction of properties of one model for validation of another, we prefer to present the unmodified observational data set.

This paper makes a valuable contribution and should be published after revisions.

Specific Comments

p. 13656, lines 24-26. Since the MIPAS retrievals do not go below 6 km, how is it possible to compare with ground-based measurements? Are you comparing the lowest MIPAS measurements (appr. 6-9 km kernel?) with ground data? If so, please state so explicitly and comment on the validity of comparing boundary layer and mid-tropospheric observations.

This statement refers to tropospheric SF₆ mean values as derived by Burgess et al., 2006, their Fig. 5. They do not describe in more detail how they derived tropospheric mean values, nor how they did the comparison to NOAA/ESRL/GMD data (called GMD/CATS in their paper), but certainly their tropospheric mean values must refer to the free-troposphere measurements of MIPAS, similar to ours. Burgess et al. refer to an “almost constant offset” between their MIPAS data and the ground-based observations and attribute it to the uncertainty of spectroscopic line data used.

In our case (see page 13664, lines 18/19) we compare tropical tropospheric mean values derived from daily averages over 9 to 15 km altitude and 17.5°S to 17.5°N latitude to global mean values of NOAA/ESRL/GMD ground-based data. This is explicitly stated on page 13664, line 19. We believe that there are good reasons to compare data from the free-troposphere to ground-based data since we expect homogeneous mixing in the troposphere for a tracer like SF₆ which has no tropospheric sinks. This assumption is confirmed by findings of Gloor et al. (2007). These authors present vertical profiles of tropospheric SF₆ vmrs measured in-situ at various sites and seasons (see their Fig. 7). In general, their profiles agree very well with the assumption of constant values in the planetary boundary layer (PBL) and the free troposphere. The only site where SF₆ vmrs in the PBL were significantly higher than in the free troposphere was Harvard, Forest, Massachusetts (called HFM) which “is located near a strong emission region” (Gloor et al., 2007). In their Table 4, they present PBL – Free troposphere differences in the order of -0.03 to +0.05 pptv, (with one exception, again belonging to HFM where the difference is +0.22 pptv). Differences in the order of 0.025 pptv (about 0.5% of the actual SF₆ vmr) are below the standard error of the MIPAS daily tropical tropospheric mean values and just in the range of the standard error of the GMD ground-based in-situ data and would, thus, not be detectable in the comparison to GMD values. We conclude from the Gloor et al. paper that the bias between the PBL and the free troposphere is so small that it is not relevant within the MIPAS-GMD comparison. We will discuss this aspect in more detail in Section 6 of the revised paper.

p. 13662, all of Section 3.2: This section talks about a CO retrieval using CO₂ non-LTE modeling and the implications for the SF₆ retrieval. This is implied but not stated and led to confusion the first few times I read this. The paragraph from line 8-14 never actually refers to SF₆, but in talking about biases of pptv you must be referring to SF₆. Please be clear here. This will make it much easier to follow your argument about low biases in SF₆ leading to high biased aged, and vice-versa.

It will be stated more clearly and explicitly that the SF₆ vmr bias introduced by

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LTE-handling of CO₂ lines is meant.

p. 13664, beginning of Section 5. In two preceding sections, 2 different biases are discussed, one of which is corrected for (the one caused by oscillations) and the other which is not (the one requiring a full non-LTE calculation). However, this section begins with the statement “Global distributions of SF₆ on basis of bias-corrected data have been derived...”. This is misleading because the data are not fully bias corrected. Please acknowledge this here and be clear about what has not been corrected at this time.

In order to make clear which bias has been corrected and which one not, we will add at the end of section 3.3 the following sentences: “For further use, daily and monthly zonally averaged SF₆ data have been corrected for the systematic contributions from the radiance baseline oscillations according to the method described in the Appendix. All daily and monthly averages of SF₆ vmrs and age of air data presented in the following sections are corrected for the bias caused by the radiance baseline oscillations (called gain-bias in the following).”

We have stated already at the end of Section 3.2, that the bias caused by the LTE handling of CO₂ lines has not been corrected and cannot be corrected at this time, since CO₂ vibrational temperatures are currently not available for most of the days included in the analysis.

p. 13664, Figure 6. I find it hard to extract useful information from this figure. The information on seasonal and latitudinal variation that you wish to convey is already contained in Figure 8. Figure 6 can be eliminated.

Figure 8 presents global distributions of the mean age of stratospheric air, while Figure 6 presents SF₆ profiles for various latitudes and seasons. Our intention with this figure was to provide information in a way that numbers can be derived from it (which is always very hard from contour maps). We appreciate the referee’s comment that this figure might not be especially useful and will move it into the electronic supplement.

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p. 13664, lines 18-20. Just to be clear, the MIPAS-derived trend is a mid or free tropospheric trend, correct?

This is correct, see above for reply to comment on p. 13656, lines 24-26.

p. 13665, lines 6-8. This statement that the MIPAS-derived trend has been used to convert SF₆ into mean age cannot be completely true. To get stratospheric ages of 10-12 years requires 10-12 years of tropospheric measurement. Since the MIPAS dataset is only 1.5 years, you must be relying on 1) the ground measurements, or 2) the assumption that MIPAS SF₆ trends are perfectly linear going back at least 10 years. I think this should be clarified and I think it would be useful to include 10-12 years of SF₆ surface data in Figure 7 so the reader to see how linear the data actually are.

We will extend the time range of Fig. 7 to the period 1996 to 2006 in order to demonstrate that the NOAA/ESRL/GMD measurements indeed provide a fairly linear trend, and add some comments on this point. Further we will include the linear regression lines for the flask and in situ measurements, and the extrapolation to 1996 - 2006 of the quadratic growth function derived by Geller et al. (1997) for the period 1987 - 1996. We will add the following sentences to Section 6:

“The MIPAS-derived linear increase has been used to convert SF₆ global distributions into mean age of stratospheric air by assigning the SF₆ vmr difference observed in the troposphere and at some location in the stratosphere, respectively, to the time lag since the troposphere showed the mixing ratio measured in the stratosphere, according to the following linear relationship:

$$\text{age} = t - t_0 - \frac{\text{SF}_6 - a}{b} \quad (1)$$

with t = time of the observation (in years), $t_0 = 2002.0$ (i.e. 1 January 2002), $a = 4.89$ pptv (the tropical tropospheric vmr on 1 January 2002 as derived from Fig. 7), $b =$

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0.230 pptv/yr (the yearly tropical tropospheric increase as derived from Fig. 7). By doing this we implicitly assume that the yearly increase of the tropical tropospheric SF₆ vmr as derived from MIPAS has remained linear and constant within the relevant period given by the actual ages observed in the atmosphere, i.e. for about 10 to 15 years. For about 8 years, this assumption is confirmed by the time series of ground-based NOAA/ESRL/GMD measurements covering the years 1996 to 2006, since the time series of the globally averaged SF₆ vmr is well consistent with a linear increase (see Fig. 7, dotted lines). For the period 1987-1996, Geller et al. (1997) found that the surface SF₆ increase was consistent with an overall quadratic growth rate, where the quadratic term, however, was rather small compared to the linear term (the coefficients are 0.0049 pptv/yr² (quadratic term) vs. 0.2376 pptv/yr (linear term)), while Maiss and Levin (1994) found Southern hemispheric SF₆ observations between 1970 and 1991 to be consistent with a purely quadratic increase described by $0.004763 \times (t - 1968.82)^2$. The extrapolation of the Geller et al. (1997) parameterization to the 1996-2006 period is shown for comparison in Fig. 7 as light-blue line. It is obvious that the yearly increase in the 1996 - 2006 period is smaller and more linear than the extrapolation from the previous 10 years. For the period 1987 - 1996, however, the extrapolation of the linear trends from MIPAS and NOAA/ESRL/GMD will overestimate the steepness of the SF₆ increase, introducing a systematic error into the age-of-air assessment from these trends. If we assume that the SF₆ increase is described well by the quadratic parameterization derived by Geller et al. (1997) for the period 1987-1996 and by the MIPAS-derived linear increase since 1996, and we use the MIPAS-derived linear increase for age-of-air assessment for ages between 6 and 15 years, we underestimate the inferred ages by at most 1.2 years, i.e. for 15 year old air the MIPAS linear increase would provide an age of 13.8 years. If we correct the Geller et al. growth parameterization by an additive term of +0.14 pptv in order to better match the most recent NOAA/ESRL/GMD global mean flask data of 1 Jan 1996 (3.58 pptv, from the regression line), we estimate that the MIPAS linear trend will underestimate the inferred ages even by up to 1.7 years (for 15 years of “real” age).

However, one should keep in mind that SF₆-derived ages higher than 6 to 8 years have been observed within polar vortices only; the respective low SF₆ vmrs are due to intrusions of mesospheric air which had undergone mesospheric SF₆ loss processes (see Section 7.3); the assessment of real age of air from these SF₆ observations suffers from further uncertainties like the details of mesospheric loss modelling in chemistry transport models.

We used the MIPAS-derived increase instead the NOAA/ESRL/GMD trend in order to account for the small additive bias between MIPAS and the ground-based measurements which is apparent by the vertical shift of the MIPAS regression line (red solid line in Fig. 7) versus the NOAA/ESRL/GMD global mean time series (middle green and violet solid lines and dotted regression lines in Fig. 7)."

p. 13665, l. 15: In contrast to the statement about tropical ascent being greatest in summer, the greatest tropical ascent by the Brewer-Dobson circulation occurs during northern winter. This figure shows relatively low SF₆ in the tropical middle stratosphere in summer; could this be an effect of the QBO circulation?

Figure 8 shows age of air, not SF₆.

The timeseries at higher potential levels (like 850 K, 1000K) which will be added to the revised version of the manuscript, reveal the following: tropical air is youngest in Northern winter 2002/2003, but even Northern winter 2003/2004 has younger air in the tropics than Northern summer 2003, except June 2003. In June 2003, very young air is found in the tropics (strong upwelling) which might be related to the very old air at the South pole (strong subsidence). We admit that it might not have been the best choice to show June 2003 in the Figure and our conclusions were somewhat misled by picking out this month; we will replace the zonal means of June 2003 by July 2003, and will reword the conclusions in the sense of this reply.

However, as discussed later in the original version of the manuscript (see p, 13670, 125ff and Fig. 12) there seems to be an increase of the mean age of air in the tropics for altitudes around 625 K over the complete period. We have speculated that this

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might be a QBO effect. A change of phase of the QBO occurred around mid 2003 for the relevant altitude range (easterly over westerly phase before mid 2003, westerly over easterly phase after mid 2003) (K. Krüger, pers. communication). However, at this time we cannot comment conclusively on this issue, and therefore we will not discuss this topic in the paper, according to your suggestion (comment on p. 13669, I.1, see below) to remove all comments on age of air trends and interannual variability.

p. 13665, l. 19: What point are you making when you say “...providing nearly mid- latitudinal conditions in the polar region8217;”? Are you referring to chemical composition? Certainly temperature and sunlight are not like the midlatitudes here.

We referred here to SF₆ mixing ratios and mean ages, respectively, which revealed values typically occurring in midlatitudes. We will rephrase this sentence to make clearer what we mean.

p. 13666, l. 23, interannual variation: This dataset has only 1 full SH winter and most of 2 NH winters. It is unreasonable to talk about interannual variability! You may point to differences between one year and the next, but you cannot draw conclusions about in- terannual variability. It is unreasonable to talk about 8217;’increasing impact of mesospheric intrusions8217;’; based on this scant data set. Do not forget that 2002 was an extremely anomalous winter/spring in the southern hemisphere, and thus to use 2002 as a base- line for comparisons would be wrong.

We will take more care in the revised version not to give the impression that we mean inter-annual variability when we talk about in-annual variations, i.e. differences between years. The sentence “suggesting increased impact of mesospheric intrusions” was related to the different altitudes (i.e. the impact is higher at 25 km than at 16 km). We will rephrase this paragraph to make clearer what we mean.

Figure 9: How would the uncorrected biases increase the uncertainties in these 3 mean age plots?

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If we could correct for the bias due to LTE treatment of the CO₂ lines, we would do so; at the current state, the seasonal variation of the bias is unclear. For the Northern winter case, we find the following: correction for the bias would increase the age of air at 16 km by about 0.5 years in the tropics and 0 to 0.5 years at other latitudes; it would not change the latitudinal distribution at 20 km; it would decrease the age of air by about 0.5 years for all latitudes at 25 km.

p. 13666, Figure 10. I like this figure even less than Figure 6: too busy, too hard to extract information. However, Figure 11 conveys much of this information in an easier- to-interpret format (the contour plots). I suggest you eliminate Fig. 10 and add 1 or 2 levels (say 900K and 1200K) to figure 11. This will better show the altitude, seasonal, and meridional variability.

We will show more time series of age of air at other potential temperature levels and move Figure 10 into the electronic supplement; however, we would like to point out that 1200 K (appr. 38 km) is beyond the altitude range the MIPAS SF₆/age of air data should be used, because of low precision and increasing systematic uncertainties (compare Figs. 2 and 3). The useful altitude range (6 to 35 km) is explicitly stated in the manuscript.

p. 13668, l. 20: With such a short data set, there is no basis for commentary about whether something is unusual in the NH. A very interesting paper on the subject of interannual variability in mesospheric descent in the NH and SH, based on UARS data and using trajectory calculations, is Rosenfield and Schoeberl ("On the origin of polar vortex air", JGR 2001). Please check this out; it may help the discussion.

The sentence will be reworded as follows: "However, despite severe subsidence events, similar high ages of air as for the Southern hemispheric winter 2003 have not been observed during the two Northern hemispheric winters covered by MIPAS (see Fig.11)." We can't see a direct link to the Rosenfield and Schoeberl paper, since we do not estimate descent in the polar vortex from comparison of tracer-tracer correlations

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in fall and spring, respectively, but continuously observe the apparent age of air during all the winter.

p. 13669, l.1: "...the mean age of air seems to increase with time". Between the uncorrected bias and the short data set, it is too speculative to make such a statement. Please remove all statements about age trends and interannual variability from the discussion and conclusions.

We will reword respective sentences in the discussion and conclusions sections.

p. 13670, l. 3: "...in the first case". It would be clearer if you said which case you meant, for example, "for the simulation with mesospheric loss,...".

Will be done.

p. 13670, l, 26: here it says "a potential bias of up to 1 year...", but earlier it was stated that above 35 km there were biases of 2 years. Please be consistent.

Earlier we stated that a bias up to 2 years was found for altitudes **above** 35 km. Here we restrict the available MIPAS data set to altitudes **below** 35 km (see p 13670, l20). Nevertheless, we will make clear here and earlier, which bias belongs to which altitude range.

p. 13671, lines 25-27. It is not necessary to state the obvious, namely, that you cannot detect change in middle atmosphere circulation with a 1.5 year data set.

Agreed, will be corrected.

Language/grammar issues

Abstract, line 26: change "considerable amount" to "considerable degree".

p. 13661, l. 4: "Further contributing model parameter...". Suggested rewording: Additional contributions to model parameter errors include uncertainties in...

p. 13661, l. 8: "Other model parameter errors were found to be negligible." Unneces-

sary statement and can be deleted.

p. 13663, l. 17: use “Kiruna, Sweden”. North Sweden is not a country.

p. 13664, l. 2: Change “on basis of” to “using”.

p. 13664, lines 3-4: Change “Figure 5 provides as an example” to “Figure 5 shows...”

p. 13664, lines 9-10: change “The SF₆ vmr is decreasing both” to “The SF₆ vmr decreases...”. Also, remove the word “traveling” from in front of time.

p. 13665, l. 17: Use “both” polar vortices, not “all”.

p. 13667, l. 11: unrealistically, not unrealistic.

p. 13667, l. 21: not “severe” transport. Perhaps try strong or far-reaching.

p. 13673, lines 2-6: “artifact” not “artefact”. There might be other occurrences of this, including Fig. 16 caption.

p. 13673, l. 7: Change to “correcting the affected...”.

p. 13673, l. 10: I don't think “equidistributed” is a word. Try equally distributed.

Figures

I know that the figures in ACPD are smaller than they would actually appear in ACP, however, there are some legends and labels in the figures that I think may be too small to be readable even in ACP. Please recheck all font sizes, especially for Figures 1, 2, 7, 4, 9.

Fig. 1: There are more than 20 species in the legend (too small to read!) but no way are 20 species visible on the plot (bottom panel). Please reduce this list to match the figure and make the font larger.

Fig. 7: Thicker lines would be better.

Fig. 8: Month labels atop each panel would make this figure easier to understand at a glance.

Fig. 11: Same comment as above, but needs theta labels.

Fig. 12: Same comment as above, but needs latitude range labels.

Fig. 13: Same comment. Please title each plot for quicker understanding (e.g., “MIPAS”, “with SF₆ Loss”, “without SF₆ loss”).

Fig. 15: The caption refers to a 2nd, 3rd, and 4th line but I only see one in the plot.

All these suggestions will be considered.

References

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Gloor, M., Dlugokencky, E., Brenninkmeijer, C., Horowitz, L., Hurst, D.F., Dutton, G., Crevoisier, C., Machida, T., and Tans, P., Three-dimensional SF₆ data and tropospheric transport simulations: Signals, modeling accuracy, and implications for inverse modeling, *J. Geophys. Res.*, 112, D15112, doi:10.1029/2006JD007973, 2007.

Maiss, M., and Levin, I., Global increase of SF₆ observed in the atmosphere, *Geophys. Res. Lett.*, 21, 569-572, 1994.

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