



Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

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Received: 1 April 2015 – Accepted: 28 April 2015 – Published: 26 May 2015

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

A new and improved setup of the SF₆ retrieval together with a newly calibrated version of MIPAS-ENVISAT level 1b spectra (version 5, ESA data version 5.02/5.06) was used to obtain a new global SF₆-data set, covering the total observational period of MIPAS from July 2002 to April 2012 for the first time. Monthly and zonally averaged SF₆-profiles were converted into mean age of air using a tropospheric SF₆-reference curve. The obtained data set of age of air was compared to airborne and balloon-borne age of air measurements. The temporal evolution of mean age of air was then investigated in 10° latitude and 1–2 km altitude bins. A regression model consisting of a constant and a linear trend term, 2 proxies for the quasi-biennial oscillation variation, sinusoidal terms for the seasonal and semi-annual variation and overtones was fitted to the age of air time series. The annual cycle for particular regions in the stratosphere was investigated and compared to other studies. The age of air trend over the total MIPAS-period consisting of the linear term was assessed and compared to previous findings of Stiller et al. (2012). While the linear increase of mean age is confirmed to be positive for the Northern mid-latitudes and Southern polar middle stratosphere, differences are found in the Northern polar upper stratosphere, where the mean age is now found to increase as well. The magnitude of trends in the Northern mid-latitude middle stratosphere is slightly lower compared to the previous version and the trends fit remarkably well to the trend derived by Engel et al. (2009). Negative age of air trends found by Stiller et al. (2012) are confirmed for the lowermost tropical stratosphere and lowermost Southern mid-latitudinal stratosphere. Differences to the previous data versions occur in the middle tropical stratosphere around 25 km, where the trends are now negative. Overall, the new latitude–altitude distribution of trends appears to be less patchy and more coherent than the previous one. The new data provide evidence of an accelerating shallow branch of the Brewer–Dobson circulation, at least in the Southern Hemisphere. Finally the age of air decadal trends are compared to trends calculated with simulated SF₆ values by the Karlsruhe Simulation Model of the Middle

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Atmosphere (KASIMA) and good agreement is found. The hemispheric asymmetry in the trends found in the MIPAS data are also indicated in the trends calculated with simulated SF₆ values by the KASIMA model.

1 Introduction

5 While it is widely accepted that climate change with enhanced greenhouse-gas abundances leads to a warming of the troposphere and a cooling of the stratosphere, the secondary effects, however, in particular on the global residual circulation in the stratosphere, the Brewer–Dobson-Circulation (BDC), are still an issue of current research (Butchart, 2014). A changing BDC will have large impact on the overall composition
10 of the stratosphere, on the ozone budget and distribution (Shepherd, 2008; Li et al., 2009) and on the lifetimes of ozone-depleting substances such as CFCs (Butchart and Scaife, 2001; Douglass et al., 2008) and greenhouse gases. The mean age of air, which is the average transit time of an air parcel from the entry point of the stratosphere, the tropical tropopause, has become a measure for the strength of the BDC
15 in particular for observational analysis (Hall and Plumb, 1994; Waugh and Hall, 2002). The mean age of air comprises both information on the speed of the advection and the amount of mixing and stirring exerted on the air parcel. Modern general circulation models (GCMs) and chemistry-climate models (CCMs) consistently simulate an acceleration of the BDC in a greenhouse gas-induced changing climate (Rind et al., 1990; Butchart and Scaife, 2001; Butchart et al., 2006; Austin and Li, 2006; Garcia and Randel, 2008; Li et al., 2008; Calvo and Garcia, 2009; McLandress and Shepherd, 2009; Butchart et al., 2010; Okamoto et al., 2011; Bunzel and Schmidt, 2013; Oberländer et al., 2013). So far, however, this expected speeding up of the BDC has not been confirmed by observations. Engel et al. (2009) provided a 30 year record of mean age of air
20 derived from CO₂ and SF₆ balloon-borne measurements which showed a slight but insignificant increase of mean age over the years 1975–2005 for Northern mid-latitudes, which would indicate a decelerated BDC. Bönisch et al. (2011) reported an accelera-

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



tion of the shallow branch of the BDC for the time period 1979–2009, while they found an unchanged deep branch. Diallo et al. (2012) investigated the age of stratospheric air in the ERA-Interim reanalysis over the period 1989–2010 and stated that the shallow and the deep branch of the BDC may evolve differently. They found a negative and significant age of air trend in the lower stratosphere and a positive but insignificant trend in the middle stratosphere. Stiller et al. (2008, 2012) provided the first global data set on age of air derived from satellite SF₆ measurements. In their paper MIPAS-ENVISAT level 1b spectra of versions 3 and 4 were used to retrieve vertical profiles of SF₆ distributed over the whole globe for the time-period September 2002 to January 2010. Monthly zonal means were converted into mean age of air, from which decadal trends were inferred for latitude and altitude bins.

The derived age of air trends were found to be spatially inhomogeneous with regions of increasing mean age of air and regions of decreasing age of air. A hemispheric asymmetry was also found with SLIMCAT model calculations (Mahieu et al., 2014).

The work presented here is a continuation of the work of Stiller et al. (2012). An extended and improved SF₆ data set is provided on the basis of a newly calibrated version of MIPAS-ENVISAT level 1b spectra (version 5, ESA data version 5.02/5.06). This new global SF₆-data set for the first time covers the total MIPAS-period from July 2002 to April 2012.

The characteristics of the MIPAS instrument are presented in Sect. 2. The improvements on the retrieval setup are discussed in Sect. 3. The characteristics and morphology of the new global SF₆-data set and the resulting age of air data set are assessed in Sect. 4. Then the temporal development is investigated and compared with the previous findings (Sect. 5). In Sect. 6 MIPAS derived AoA trends are compared with trends calculated with simulated SF₆ values from the Karlsruhe Simulation Model of the Middle Atmosphere (KASIMA). Finally, in Sect. 7, we summarize the lessons learned about possible changes of the BDC.

2 MIPAS

MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) is a Fourier transform infrared (FTIR) spectrometer aboard ENVISAT (Environmental Satellite, Fischer et al., 2000), which was launched in March 2002 by ESA (European Space Agency).

ENVISAT's task was the permanent survey of the Earth's climate, the ocean, the land surfaces and the Earth's ecosystem in general. ENVISAT was a polar orbiting sun-synchronous satellite in an orbit of approx. 800 km with an inclination of the orbit of 98°. The mission of ENVISAT was planned with a lifetime of 5 years. In 2004 the operation of the MIPAS instrument was interrupted due to a problem with the interferometer slide. The optical path difference was then reduced, implying a deterioration of the spectral resolution from 0.025 to 0.0625 cm⁻¹, and since 2005 MIPAS was operational again in this mode. The first phase of the mission (2002–2004) is usually referred to as the MIPAS full resolution (FR) period, while the second phase is called the reduced resolution (RR) period. ENVISAT kept on sending data until April 2012, when ESA lost contact to ENVISAT.

MIPAS was designed for the detection of mid-infrared limb emission spectra in the middle and upper atmosphere. It observed a wide spectral interval ranging from 4.15 to 14.6 μm with high spectral resolution (Fischer et al., 2008). The atmospheric spectra were inverted into vertical profiles of atmospheric pressure, temperature and volume mixing ratios (vmrs) of at least 30 trace constituents. This allows studies of stratospheric chemistry and dynamics, stratosphere-troposphere exchange, chemistry and physics of the upper troposphere, chemistry and physics of the upper atmosphere, as well as climatologies and weather forecasting. One advantage of MIPAS was that as a limb emission instrument as opposed to a solar occultation instrument, it could measure globally and during day and night.

Because of the long optical path through the atmospheric layers, MIPAS could also detect trace gases with very low mixing ratios. Vertical information was gained by scanning the atmosphere at different elevation angles with different tangent altitudes. MIPAS

ACPD

15, 14685–14732, 2015

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



could observe atmospheric parameters in the altitude range from 5 to 160 km with minimum and maximum steps of 1 and 8 km, respectively (Fischer et al., 2008).

3 Improvement of the retrieval of SF₆ mixing ratios

Data processing relies on constrained least squares fitting using the Tikhonov (Tikhonov, 1963) regularization approach. Further details of the MIPAS data processor used are described in von Clarmann et al. (2003, 2009). Information on temperature and line of sight, as well as the spectral shift was taken from preceding MIPAS retrievals performed prior to SF₆ in the sequential retrieval chain.

While the retrieval of SF₆ by Stiller et al. (2012) relied on ESA version 4.61/4.62 and 4.67 calibrated radiance spectra, we have used version 5.02/5.06 spectra provided by ESA in the course of reprocessing of the data. These data are considered superior with respect to 4.61/4.62/4.67. Beyond this, the SF₆ retrieval setup has been improved over that used by Stiller et al. (2012). Improvements are related to the consideration of non-local thermodynamic equilibrium emission of interfering CO₂ lines, the treatment of interfering species in general, and the details of the joint-retrieval of the background continuum. The implementation of the altitude-dependence of the regularization strength has been slightly changed. The definition of the analysis window (941–952 cm⁻¹, see Fig. 1), the regularization strength of the inverse problem, and the spectroscopic database chosen remained unchanged since no improvements over the approach by Stiller et al. (2012) could be achieved with respect to these. Spectroscopic data were used from a dedicated MIPAS database for gases like H₂O, CO₂, O₃ and COF₂ (Flaud et al., 2003). For N₂O, NH₃, CFC-12 and SF₆ the spectroscopic database HITRAN2000 (Rothman et al., 2003) was used. Tests with varying regularization parameters did not lead to any retrieval improvements, i.e. the regularization strength chosen by Stiller et al. (2012) has been confirmed to be adequate. The new SF₆ data described and used here are version V5h_SF6_20 for the FR data product and V5r_SF6_222 and V5r_SF6_223 for the RR period. The latter two data

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



versions have no discernible differences; their different version numbers just reflect different sources of ECMWF meteorological analysis data used in the retrieval. In the FR V5h_SF6_20 data version the artefact of the previous version caused by radiance baseline oscillations in the level 1-data described in Stiller et al. (2008) is no longer an issue and has been totally overcome.

3.1 Non-local thermodynamic equilibrium

The Q branch of the ν_3 band of SF₆ at 947.9 cm⁻¹ analysed here is strongly superimposed by the CO₂ laser band (00011 → 10001) at 947.74 cm⁻¹ and lies just above the first hot band (01111 → 11101) line at 947.94 cm⁻¹ (see Fig. 1). These CO₂ emission bands deviate from local thermodynamic equilibrium (LTE) in the middle atmosphere, particularly during daytime. Stiller et al. (2012) approximated the non-LTE effect by treating the CO₂ laser band and hot band emissions as emissions from different (non-CO₂) species and by fitting their “virtual abundances” along with the SF₆-retrieval. While these virtual abundances have no physical meaning, they helped to fairly well model the CO₂ laser band emission and to avoid related spectral residuals and error propagation. Contrary to that, our refined analysis relies on explicit modelling of the non-LTE emissions of the CO₂ laser and hot bands. In the course of a preceding CO retrieval (Funke et al., 2007), the vibrational temperatures of the CO₂ laser band and the hot band were calculated for the actual atmospheric conditions. Since the radiative transfer code used in our retrieval, the Karlsruhe Optimized and Precise Radiative Transfer Algorithm (KOPRA, Stiller, 2000; Stiller et al., 2002; Funke and Höpfner, 2000) supports calculation of non-LTE emissions, these could directly be used for the calculation of the laser band signal and hot band emissions. This improves considerably the description of the CO₂ emissions and reduces the residuals between the observed and modelled spectra, leading eventually to improved SF₆ results (compare residuals at the position of CO₂ lines in Fig. 2).

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2 Interfering gases

Figure 1 shows the spectral window used for the SF₆ retrieval and the expected spectral contributions of contributing species for MIPAS reduced resolution at 20 km in mid-latitudes for July. The signature of the target species SF₆ (red solid line) is quite weak compared to some of the interfering species. Thus a careful treatment of the interfering species is essential to minimize related error propagation. Since for some of the interfering species no reliable a priori information on their abundances is available, these gases are jointly fitted along with SF₆. For other interferents, abundance information is available from preceding MIPAS retrievals; however inconsistent spectroscopic data or calibration inconsistencies in the SF₆ analysis window and the interferents' dedicated analysis windows can cause artefacts when the known abundances are used to model the contribution of these gases in the SF₆ analysis window. Thus, it is occasionally adequate to jointly fit these gases along with SF₆, too. Stiller et al. (2012) have used abundance information from climatologies or preceding MIPAS retrievals for all interfering species except for CO₂ and H₂O, which were fitted jointly along with SF₆. In the new retrieval scheme the trace gases COF₂ and ozone were additionally joint-fitted. This helped to minimize the residual of the fit and also removed a slight tilt of the residual in spectral space. For all gases a first order Tikhonov-type regularisation was chosen, which means that the slope of the profile was forced to some constraint, rather than forcing the profile towards an a priori profile as done in the Optimal Estimation approach. Usually the constant zero profile served as a priori profile and by this way oscillations in the profile are damped and the profile becomes smoother. In the following we present a gas-by-gas discussion of our treatment of all interfering species.

3.2.1 CO₂

CO₂ is the main contributing gas of all emitters in the SF₆ analysis window (microwindow) used (blue solid lines in Fig. 1). As mentioned above the maximum of the SF₆ spectral signature is just underneath of the wing of a CO₂ laser line. In order to get an

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

accurate value of the SF₆ mixing ratio from radiances emitted in the SF₆ microwindow, a very precise modelling of the CO₂ is crucial. We have used a non-LTE model to account for the CO₂ emissions in the middle atmosphere and fitted it jointly with SF₆. The first guess profile in the iterative procedure was taken from climatologies (Remedios et al., 2007).

3.2.2 H₂O

A water vapour signature is located near the SF₆ Q branch at 948.26 cm⁻¹, so a considerable information crosstalk is expected between the H₂O and the SF₆ signals. The water vapour profile resulting from the preceding retrieval in dedicated microwindows (prefit) was used as a priori and first guess profile for every geolocation. The regularization strength associated with H₂O was adjusted such that the correction with respect to the initial H₂O profile had about 1 to 1.5° of freedom. Basically the profiles have the shape of the prefit profile and only a shift of the prefit profile is allowed. The residual near the water vapour line was not reduced when the regularization for water vapour was relaxed. Variations of the regularization for water vapour or the choice of related a priori (constant zero or water prefit), did not have any discernible effect on the SF₆ retrieval.

3.2.3 COF₂

There is not much vertically resolved information contained on COF₂ in the microwindow, but fitting this trace gas jointly with the target species helped to minimize the residuals. A climatological profile served as a priori and first guess profile since there was no prefit of COF₂ available. The regularization was chosen relatively strong such that the resulting COF₂ profiles have about 1.5 to 3.5° of freedom.

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2.4 O₃

Like COF₂, ozone is not much contributing to the signal in the used microwindow, but profits from the ozone retrieval existed. Thus, the ozone profits served as a priori and first guess profiles. Together with the joint fit of COF₂, the joint fit of ozone helped to remove a tilt in the residual. The regularization applied allowed the ozone profiles to have about 1.5 to 3.5° of freedom.

3.2.5 Further species

Profiles of N₂O, NH₃ and CFC-12 were imported from a climatological database (Remedios et al., 2007). Since the signals of these gases are small, related uncertainties are tolerable.

3.3 Background continuum and radiance offset

In the previous SF₆ retrieval by Stiller et al. (2012) background continuum radiation was considered up to an altitude of 33 km. In the atmosphere continuum radiation, i.e. radiation which is only varying very slightly in spectral space (in contrast to spectral lines), is emitted by clouds, dust or other aerosol particles. Also the sum of very far wings of spectral lines, no more accounted for by the line by line calculation, can contribute to the continuum radiation. In general it was assumed that a consideration of continuum radiation above 33 km was not necessary, particularly because there are no aerosol contributions expected above the Junge layer. However, it turned out that fitting continuum radiation up to higher altitudes (50 km) could eliminate an artefact in the retrieved SF₆ profile: while in the retrieval of Stiller et al. (2012) an unexplained local maximum in SF₆ occurred around 36 km in the tropics, this supposedly unphysical feature vanishes completely with the new continuum treatment. This provides evidence that there is additional continuum radiation in the atmosphere which if not accounted for leads to elevated SF₆ mixing ratios, since the SF₆ signature is also of broad band nature. The

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



approach of a joint fit of the continuum radiation up to higher altitudes also helped to improve other retrievals of other species. In addition, a recent paper pointed out that there is evidence of aerosol particles even above the Junge layer due to meteoric dust (Neely III et al., 2011). In our retrievals we also fit a constant radiance offset jointly, in order to account for a possible residual shift in radiance due to imperfect radiance calibration. This offset had to be strongly regularized in order to cope with the pronounced linear interdependence of the continuum and offset Jacobians.

3.4 Miscellaneous

The consideration of continuum up to higher altitudes allowed the usage of more upper tangent heights. While the previous retrieval setup only used the first 19 out of 27 tangent heights in MIPAS reduced resolution mode, the new setup incorporated information from measurements of 22 tangent heights. By this way more information could be gained at higher altitudes, i.e. the averaging kernel diagonals increased slightly at higher altitudes. In addition the root mean square (RMS) of the residuals in upper tangent heights decreased. Hence, with the new retrieval setup for the first time it made sense to include 22 tangents height instead of 19. The new mean SF₆ profiles contain more information in the altitude range 40–50 km, show more structure and depend less on the prior information there.

3.5 Discussion of the retrieval refinement

In Fig. 2 the residual between measured and simulated spectrum at tangent height 12 (approx. 23 km) of the final retrieval setup (upper panels) is shown while Fig. 2 (lower panels) show the respective residual of the previous setup. To reduce the noise measured and modelled spectra have been coadded over the period of 1 day. One can see that the residuals improved substantially. Especially the CO₂ lines and the water vapour line (compare with Fig. 1) are fitted much better and overall the RMS of the

residual has been reduced from about 1.8 to 1.0 nW (cm² sr cm⁻¹)⁻¹. Also a slight tilt of the residual in spectral space could be removed.

With the new retrieval setup the unphysical “nose”, a local maximum, in the tropical SF₆ profiles at 36 km happened to disappear by considering the continuum above the standard altitude of 33 km up to an altitude of 50 km.

4 The new SF₆ database and age of air distributions

With the new retrieval setup, the complete set of nominal mode MIPAS data was processed and approx. 2.3 million SF₆ profiles have been retrieved. The profiles belong to geolocations that cover the whole globe and the full MIPAS period from July 2002 to April 2012 with several data gaps in between. The single profiles scatter a lot and the noise error is too large (in the order of 20 %) to provide useful age of air information from single profiles. But averaged profiles lead to meaningful SF₆-profiles. In Fig. 3 the time series of MIPAS SF₆ monthly zonal means at 25 km altitude is shown. SF₆ is increasing with time at almost all latitudes. Towards higher latitudes this increase is shifted in time, meaning that at higher latitudes the respective mixing ratios are reached at later times than in the tropics, as one would expect from the global circulation scheme. At high latitudes, especially in the Southern polar stratosphere seasonal influences can be identified. Every year in late austral winter to austral spring tongues with very low SF₆ mixing ratios appear in the Southern polar stratosphere. This can be explained by subsidence of very old air into the polar vortex, or even subsidence of SF₆ depleted air from the mesosphere. This effect is also indicated in the Northern polar stratosphere, but much less pronounced. This is explained by the fact that the polar vortex is less stable and pronounced in the Northern polar stratosphere and isolated subsidence inside the vortex does not occur that much.

The vertical resolution of the new SF₆ data is slightly degraded compared to the previous version to 4–6 at 20 km, 7–10 at 30 km and 12–18 at 40 km altitude due to the

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



inclusion of more gases in the fit, which was done to achieve a higher accuracy and less systematic errors.

4.1 Conversion of SF₆ into age of air

For the calculation of age of air (AoA) from SF₆ abundances a SF₆ reference curve is necessary. The theoretical concept of age of air as derived by Hall and Plumb (1994) requires the knowledge of SF₆ mixing ratios at the entry point into the stratosphere, i.e. the tropical tropopause region over a long period of time. As pointed out by Stiller et al. (2012) such a long term observational data set is not available. Only ground-based observations can provide the necessary reference data. However, transport times from the surface to the tropical tropopause are somewhat uncertain and can amount from days or even hours (to the top of convection) to weeks or months (to the top of the Tropical Tropopause Layer (TTL)). Using surface data as a reference can imply a high bias in this order of magnitude on the AoA data.

This has to be kept in mind when comparing MIPAS AoA distributions to model data, for which time zero is set by tropopause crossing of the air parcel.

We have constructed the SF₆ reference curve as described in Stiller et al. (2012) using NOAA/ESRL SF₆ data. For the period 1995 to November 2013 smoothed ground based global mean combined flask and in-situ data (Hall et al., 2011) is used while for times before 1995 a linear approximation from Hall et al. (2011) ($y = 0.125 + 0.215 \times (t - 1985)$) is applied. The reference curve is extended with a linear extrapolation until June 2014 to deal with MIPAS SF₆ values slightly higher than the reference values at that certain time that can occur sporadically due to their random errors.

The AoA is then calculated by simply mapping the measured SF₆ value on the reference curve and reading of the reference time. The time difference, the so called lag time approximates the AoA. According to Hall and Plumb (1994) this lag time is only equivalent to the mean age of air, if the used tracer is growing strictly linear, i.e. the reference curve has to be linear. Because our constructed reference curve appears to be slightly non-linear, a correction is applied. Within an iterative procedure the refer-

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

more present in the new data set. This feature of the old data set has been proven to be a retrieval artefact, which was eliminated by a refined treatment of continuum radiation (see Sect. 3). This artefact triggered an oscillation in lower layers which are no longer present in the new data set. Above 40 km, the air is now found to be younger at almost all latitudes, which appears to be more realistic. The old data version was reported to have a possible high bias of up to 2 years above 35 km, most pronounced at the summer pole due to the simplified approach concerning the non-LTE treatment of interfering CO₂ lines (Stiller et al., 2008). The full non-LTE treatment used for the new data set has removed this systematic uncertainty. In addition, part of the lower

AoA in the upper stratosphere is attributed to the revised regularization of the retrieval. Among studies of AoA (e.g. Stiller et al., 2012; Diallo et al., 2012; SPARC CCMVal, 2010) it became a standard for validation of measured or modelled AoA to compare with earlier airborne measurements from the 1990s as published by Waugh and Hall (2002) and Hall et al. (1999). In Fig. 6 we compare the new MIPAS monthly zonal means of AoA with these airborne measurements. Figure 6 shows the latitudinal cross-section of the new MIPAS AoA at 20 km for selected months together with the total AoA range covered by all monthly mean data from MIPAS as derived from the minimum and maximum value for each latitude bin (shaded in grey), and the AoA derived from airborne measurements of SF₆ and CO₂. The AoA from CO₂ refers to CO₂ observations at the tropical tropopause, so it might exhibit a slight low bias compared to the SF₆ derived AoA measurements.

As with the previous version of MIPAS AoA discussed in Stiller et al. (2012) (Fig. 4), the agreement of MIPAS AoA with the earlier airborne measurements is excellent in the Northern and Southern mid-latitudes. Overall the comparison turns out to be quite similar to the one in Fig. 4 in Stiller et al. (2012): MIPAS AoA is higher in the tropics with an AoA of about 2 years at 20 km, maybe slightly lower than in the previous comparison. The negative peak of low MIPAS AoA at about 30° N is no more present in the new version of the figure and the spread of MIPAS AoA in the tropics is lower. At high latitudes MIPAS AoA is higher than the airborne observations, however, the error bars of

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the airborne measurements still overlap with the range of the MIPAS observations. The spread of MIPAS data at high latitudes is very large, especially in the Southern polar stratosphere, and is in agreement with high amplitudes found in the seasonal cycle (see Sect. 5.2). By comparing the latitudinal cross-sections of MIPAS AoA with the aircraft data one has to keep in mind that these data were observed in the 1990s, whereas MIPAS AoA represent the decade from 2002 to 2012. We cannot expect that AoA estimates from different decades fit perfectly together, especially in the tropics where temperature changes have been reported (e.g. Randel et al., 2006). The AoA gradients at the subtropical mixing barriers are also smaller in the MIPAS data, which could be a hint to a weakening of the mixing barriers, as proposed by Stiller et al. (2012), which would also explain the higher ages in the tropics compared to the airborne data. In addition it should be noted that the airborne measurements do not represent a latitudinal cross-section at a given time, but a compilation of measurements from different years and seasons.

In Fig. 7 MIPAS AoA profiles are compared to airborne AoA profiles (in-situ CO₂ measurements by Boering et al., 1996; Andrews et al., 2001, in-situ SF₆ measurements by Ray et al., 1999 and air sample measurements by Harnisch et al., 1996) for the tropics (5° S), the Northern mid-latitudes (40° N) and the Northern high latitudes (65° N). In the tropics MIPAS AoA is older than in-situ CO₂ and SF₆ measurements at all altitudes as already observed at 20 km. In the Northern mid-latitudes the MIPAS profile fits excellently to the SF₆ in-situ data up to an altitude of 27 km and is older higher up. As expected, in-situ CO₂ measurements provide lower ages, and the AoA from SF₆ air samples by Harnisch et al. (1996) is younger, too. At Northern high latitudes, MIPAS age profiles only fit well to the SF₆ air samples taken from polar vortex air. To illustrate the high seasonality, monthly averaged MIPAS profiles are additionally shown with oldest ages found for January.

5 Observed temporal variability for the period July 2002 to April 2012

For the analysis of the temporal variability of the new AoA data set the same methods were applied as in Stiller et al. (2012), i.e. the following regression function was fitted to the data:

$$age(t) = a + bt + c_1 qbo_1(t) + d_1 qbo_2(t) + \sum_{n=2}^9 \left(c_n \sin \frac{2\pi t}{l_n} + d_n \cos \frac{2\pi t}{l_n} \right) \quad (1)$$

where t is time, qbo_1 and qbo_2 are the quasi-biennial oscillation (QBO) indices, and the sum represents 8 sine and 8 cosine functions of the period length l_n . The period of the first two sine and cosine functions is 12 and 6 month, respectively, representing the seasonal and the semi-annual cycle. The other 6 terms have period lengths of 3, 4, 8, 9, 18 and 24 months and describe deviations of the temporal variation from a pure sine or cosine wave. Fitting sine and cosine of the same period length accounts for a possible phase shift of the oscillation. The terms qbo_1 and qbo_2 are the normalized Singapore winds at 30 and 50 hPa as provided by the Free University of Berlin via <http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/index.html>. These QBO-proxies are approximately orthogonal such that their combination can emulate any QBO phase shift (Kyrölä et al., 2010). For the fit of the coefficients $a, b, c_1, \dots, c_9, d_1, \dots, d_9$ to the data, the method of von Clarmann et al. (2010) is used, which considers the full error covariance matrix of mean age data \mathbf{S}_m with the squared standard errors of the means (SEM) of the monthly zonal means as the diagonal terms (Stiller et al., 2012).

5.1 Age of air trends without consideration of autocorrelation

First we discuss the timeseries analysis within the framework of descriptive statistics, i.e. without consideration of the autocorrelations in the residuals of the trend analysis. As a second step, the analysis is repeated within the framework of inductive statistics, where autocorrelated model errors have to be considered (Sect. 5.4).

**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Figure 8 shows an example of the fit of our regression model (in orange) to MIPAS monthly zonal mean data (in blue). The derived linear trend is illustrated in orange. The fit considers a potential bias of the two MIPAS measurement periods (dashed orange line) as described in von Clarmann et al. (2010). Coloured squares indicate the measurements of Engel et al. (2009) and the green dashed line represents their estimated trend. The diagram underneath shows the residual of the fit. Such a fit is done for every $10^\circ/1\text{--}2\text{ km}$ latitude–altitude bin.

The linear increase of AoA over the whole MIPAS-period derived from our regression analysis varies with altitude and latitude. The global view can be seen in Fig. 9 top panel. Red areas indicate increasing AoA, while blue regions indicate decreasing AoA. The distribution of trends in the latitude–altitude plane roughly confirms the mean trends of those obtained by Stiller et al. (2012) and their general morphology but looks more coherent and less patchy, meaning that regions of linear increase and decrease, respectively, are more contiguous. There are basically two regions of linear decrease: A large one consisting of the tropics between about 19 to 33 km and extending to the lowermost mid-latitude Southern stratosphere, and a smaller one consisting of the upper tropical troposphere extending to the lowermost stratosphere of mid-latitudes. These regions are surrounded by regions of AoA linear increase. Largest positive linear trends were observed in the polar regions. Compared to findings of Stiller et al. (2012) a positive linear increase of mean age is confirmed for the Northern mid-latitudes and Southern upper polar stratosphere, as well as for the Northern polar lowermost stratosphere. Negative age of air trends of Stiller et al. (2012) in the lowermost tropical stratosphere and lower Southern mid-latitude stratosphere are also confirmed. Differences are found in the Northern polar stratosphere, where the mean age is now increasing as well. In the tropical stratosphere the picture is now almost opposite to the previous data of Stiller et al. (2012) meaning that AoA is increasing where it used to be decreasing and vice versa. These changes are attributed to the more adequate treatment of the background continuum emission in the retrieval and the associated removal of the spurious SF_6 maximum and subsequent errors.

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The uncertainties are rather small, even smaller than the ones derived by Stiller et al. (2012) (see Fig. 9, bottom panel), which could be a result of the longer time series, covering now the full MIPAS period, and the fact that the new AoA data set is less noisy than the previous one. The results are significant on the 2σ level for most of the altitude/latitude bins (see Fig. 9, upper panel). Hatched areas indicate where the trend is not significant.

The vertical profiles of AoA linear trends for every other latitude bin are shown in Fig. 10, top panel. Engel et al. (2009) derived a trend of AoA for 30 to 50° N of $+0.24 \pm 0.22$ yr per decade (1σ uncertainty level) for the 24 to 35 km altitude range. This trend together with its valid altitude range and its 2σ uncertainty is marked as big black cross in Fig. 10. For better illustration the same picture with the MIPAS linear trend profiles for the two relevant latitude bins is shown in Fig. 10, bottom panel. The MIPAS AoA trends of 30 to 40 and 40 to 50° N are slightly lower than in the previous version and match now impressively well with the trend estimated by Engel et al. (2009) in the 24 to 35 km altitude region. One has to keep in mind, that the trend derived by Engel et al. (2009) represents the time period 1975–2005, while MIPAS measured from 2002 to 2012. So there is only a small time overlap between the two trends. Still the agreement of both is remarkable. The MIPAS AoA trends for the latitude bins 30 to 40 and 40 to 50° N are significantly distinct from zero for almost all altitudes above 17 km even on the 2σ uncertainty level.

5.2 Annual cycle

Figure 11 shows the amplitudes and phases of the seasonal cycle, i.e. the amplitudes and phases of terms with period length 1 year determined with the regression model described above. As observed by Stiller et al. (2012), the amplitudes of the seasonal variation are strongest in the Southern polar stratosphere, which can be explained by regular intrusion of SF₆-depleted mesospheric air into the polar winter vortex, which leads to very old apparent ages. Over the year, the oldest AoA occurs there at the end of Southern hemispheric winter to spring, as expected, while the youngest air is

observed at the end of Southern summer to autumn. This process is also observed in the Northern Hemisphere, but with smaller amplitude (see also Funke et al., 2005).

The phase shift of half a year between below and above 25 km in the polar stratosphere reported by Stiller et al. (2012) is only visible between the upper stratosphere and the lowest latitude–altitude bin in both hemispheres in the new MIPAS data. Diallo et al. (2012) found polar stratospheric AoA above 25 km, with youngest air at the end of local winter to spring, to be in the opposite phase than in the lowermost extra-tropical stratosphere in their analysis of ERA Interim data. In the model analysis of Li et al. (2012) the maximum of AoA in the polar region in spring is also bounded to the lower stratosphere whereas the upper polar stratosphere exhibits younger age. This difference to MIPAS AoA can be explained by the different derivations of AoA in the respective studies: while in Diallo et al. (2012) AoA is explicitly calculated by backward trajectories of the air parcel, and in Li et al. (2012) the AoA is determined by the pulse tracer method, the MIPAS AoA is derived by SF₆ observations which exhibit an over-aging when SF₆ depleted mesospheric air subsides into the polar stratosphere during winter. This overaging in the polar stratosphere during winter shifts the phase in the MIPAS data towards oldest air in polar midwinter, when subsidence of mesospheric air is strongest.

In the tropics and most parts of the Northern mid-latitudes the amplitude of the seasonal cycle is rather small, except for the mid-latitudes above 25 km in both hemispheres, where also higher amplitudes can be found. The overall distribution of amplitudes is consistent to the one in Stiller et al. (2012), but there are also some discernible differences: the band of high seasonal amplitudes in the Northern mid-latitudes is not visible anymore in the new data.

The extra-tropical Southern lowermost stratosphere (LMS) exhibits now a higher amplitude of the seasonal cycle. Hence, now both hemispheres show enhanced seasonal amplitudes in the extra-tropical LMS, which are tentatively attributed to the seasonality of the permeability of the subtropical mixing barriers (the subtropical jet) (Stiller et al., 2012) and flooding of this region with old vortex air after the vortex breakdown at the

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



end of winter and spring. Consistently Diallo et al. (2012) found high amplitudes of the seasonal cycle in the Southern and Northern extra-tropical LMS. Most parts of both the Southern and Northern extra-tropical LMS reach their maximum in AoA in local spring in the MIPAS data set as well as in the analysis by Diallo et al. (2012). This hemispheric symmetry is a feature of the new MIPAS data set. Bönisch et al. (2009) found oldest AoA in the Northern LMS in April and youngest in October with in-situ measurements of SF₆ and CO₂ during the SPURT aircraft campaigns. MIPAS observed youngest air in hemispheric late summer to autumn when the mixing barrier in the subtropics is weakest and young air from the tropics is injected in this region, also referred to as “flushing” of the LMS (Hegglin and Shepherd, 2007). Also cross-tropopause isentropic mixing from the tropical troposphere in the extra-tropical LMS is enhanced during summer-early autumn when the subtropical jet is weak (Chen, 1995).

Model results of Li et al. (2012) of the seasonal variation of AoA also agree with MIPAS in the extra-tropical LMS.

In the Northern subtropical lower stratosphere an abrupt meridional phase shift of almost half a year occurs, which means that these air masses are well isolated by the subtropical jet. Equatorwards the air is oldest in summer, when the subtropical mixing barrier and the BDC are weakest and older air from the extra-tropics is mixed in. This process is also indicated in the Southern Hemisphere and these opposite phases between the subtropics and the extra-tropics are also observed in the model results of Li et al. (2012).

In the model simulation of Li et al. (2012) a vertical phase shift in the subtropics above 450 K (approx. 20 km) occurs. An indication of this phase shift is also visible in the MIPAS data, when the month of minimum age observed changes vertically from late winter to autumn in the Northern subtropics at 20 km.

In the mid-latitudinal middle and upper stratosphere the air is youngest in local winter, when, according to the known seasonality of the Brewer–Dobson-Circulation, younger air is brought to higher latitudes more efficiently. The mixing barriers are partially visible by abrupt phase shifts in the month of minimum and maximum age, respectively:

air masses in the polar vortex are well isolated from the rest of the hemisphere. The subtropical mixing barrier is visible in the Northern lower stratosphere at 30° N and is indicated in the upper stratosphere only in the plot of maximum age of Fig. 11. In the Southern Hemisphere the abrupt phase shifts seem to occur at 50–60° S and at 10–20° S.

In the tropics below approx. 28 km air is youngest in boreal winter, even in the Southern Hemisphere (except for altitude–latitude-bins below 20 km). The hemispheric difference is lower than expected, which was also noticed by Stiller et al. (2012). However, this minimum in AoA in the Southern tropics occurs approx. 2 month later in the new MIPAS data. In the Northern Hemisphere air is oldest in late summer, while it is oldest in austral spring to early summer in the Southern Hemisphere. Furthermore this maximum in AoA in the Southern tropics occurs 2 month earlier in the new MIPAS data set compared to the previous one.

In summary, apart from the mentioned differences, no substantial differences in the patterns of the months of maximum or minimum age, respectively, representing the phase of the seasonal variation, were found in the new data compared to the previous by Stiller et al. (2012). Thus, their respective conclusions remain valid in the light of the new data.

5.3 QBO influence

The amplitude of the QBO signal in AoA is shown in Fig. 12 for all latitudes and altitudes under assessment. High amplitudes are found not only in the tropics but also in the upper stratosphere of mid-latitudes, whereas highest amplitudes were found in the upper polar stratosphere. We also find high amplitudes in the Northern lowermost stratosphere. Overall the QBO influence seems to be more pronounced in the Southern Hemisphere above 25 km.

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



5.4 Impact of empirical errors and autocorrelation

As described in Stiller et al. (2012) our regression model only accounts for the linear trend, several periodics and the QBO-terms. Other atmospheric variability especially from non-periodic events is not included in this model. This results in fit residuals which are considerably larger than the data errors represented by the covariance matrix \mathbf{S}_m , which includes only the standard errors of the monthly means and the correlated terms to account for the possibly bias between the MIPAS data subsets (von Clarmann et al., 2010). Therefore the χ_{reduced}^2 of the fit with

$$\chi_{\text{reduced}}^2 = \frac{(\mathbf{age}_{\text{MIPAS}} - \mathbf{age}_{\text{modelled}})^T \mathbf{S}_m^{-1} (\mathbf{age}_{\text{MIPAS}} - \mathbf{age}_{\text{modelled}})}{m - n} \quad (2)$$

exceeds the value of unity in most cases, where $\mathbf{age}_{\text{MIPAS}}$ and $\mathbf{age}_{\text{modelled}}$ are the data vectors containing the measured and modelled age values, respectively, and where m and n are the number of data pairs and the number of fitted coefficients, respectively (Stiller et al., 2012). In order to consider these model errors of the regression model, the autocorrelation of two adjacent data-points was estimated in a first step. In a second step the fit was rerun with the autocorrelation and a constant error term added to the covariance matrices. These additional terms in the covariance matrices were scaled within an iterative procedure, such that the resulting χ_{reduced}^2 of the trend fit was close to unity. This iterative procedure is necessary because the additional autocorrelated error term changes the weight between the data points in the fit.

The result of the linear increase/decrease, which is referred to as “model-error corrected linear increase” in Stiller et al. (2012) is shown in Fig. 13. Compared with the respective Figure in Stiller et al. (2012) (Fig. 12) linear increase is again confirmed for the Northern mid-latitudes and the Southern polar middle stratosphere, whereas linear decrease is confirmed for the lowermost tropical stratosphere and lower and lowermost Southern mid-latitudinal stratosphere. Differences compared to Fig. 12 in Stiller et al. (2012) occur again in the tropics and in the Northern polar stratosphere, as well as in the upper Southern polar stratosphere.

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The overall pattern of linear increase/decrease resembles a lot the pattern in the trend fit without consideration of autocorrelation and empirical errors (Fig. 9). The significance of most data bins is lower, as expected, due to the additional error. Significant features which appear in both approaches, with and without consideration of autocorrelation and empirical errors, are considered to be most reliable. By comparing Fig. 13 with Fig. 9 these features are the linear increase in the Southern and Northern upper polar stratosphere and in the Northern mid-latitudes as well as the linear decrease in the tropics and Southern subtropics and the Southern lower mid-latitudinal stratosphere. A clear asymmetry between both hemispheres is confirmed.

6 Comparison with model simulation

The MIPAS SF₆-based AoA trends are compared with trends derived from SF₆ distributions calculated with the Karlsruhe Simulation Model of the Middle Atmosphere (KASIMA), see Kouker et al. (1999); Ruhnke et al. (1999); Reddmann et al. (2001, 2010) for a description of the model and some applications. Here we used the model in the configuration as described in Reddmann et al. (2001), but with a T42/L63 configuration corresponding to about 2.84° × 2.84° horizontal resolution and 63 vertical levels between 7 and 120 km. In addition, the model is nudged to ERA-Interim analyses below 1 hPa. SF₆ mixing ratio values were set at the lower boundary of the model in the troposphere using NOAA/ESRL data. Note, that the model includes the mesospheric loss of SF₆, which is implemented in the model according to Reddmann et al. (2001). Previously, Stiller et al. (2008) showed that only including mesospheric loss the apparent high mean age values in late polar stratospheric winter observed in MIPAS observations can be reproduced by the model simulations.

For the determination of the trend of SF₆ derived mean age of air in the model calculation, the SF₆ distributions were calculated on a pressure-latitude grid using 64 latitude bins. In each pressure-latitude bin monthly zonal averages of SF₆ were calculated together with their standard error of the mean. The vertical pressure coordinates

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



have been converted to geometrical altitudes assuming an isothermal atmosphere with a scale height H of 7 km ($z = -H \ln(\rho/\rho_0)$). Afterwards the monthly zonal means have been interpolated on the MIPAS altitude grid and binned in the MIPAS latitude bins. These regridded zonal SF₆ monthly means were sampled and converted to AoA in the same manner as it was done for the measured SF₆ values (see Sect. 4.1).

Figure 14 shows the distribution of age trends calculated with simulated SF₆ values from the KASIMA model in a latitude–altitude-plane with consideration of empirical errors and autocorrelations.

The model results agree remarkably well with the empirical AoA trends: positive decadal trends are found in the upper polar stratosphere in both hemispheres and at Northern mid-latitudes around 25 km while negative trends are found in the tropics and Southern subtropics as well as in the Southern lower and lowermost stratosphere of mid-latitudes and Southern polar region. The most pronounced negative trend is detected in the Southern tropics and subtropics around 25 km, as in the MIPAS measurements. At Northern mid-latitudes at about 30 km altitude a tongue of negative trend is modelled. While MIPAS detected still positive trends there, there is at least a local age trend minimum in this region in the MIPAS data. What is striking in Fig. 14 is the hemispheric asymmetry between significant negative trends in the Southern Hemisphere and significant positive trends in the Northern Hemisphere, which was also found in the MIPAS data. This hemispheric asymmetry was also noticed by Mahieu et al. (2014) with the SLIMCAT model. At high latitudes lower stratospheric trends are positive in the MIPAS but negative in the KASIMA data set. Differences in this region are most likely due to the “overaging” effect, which is more pronounced in the measured data because KASIMA underestimates polar winter subsidence.

7 Summary and conclusions

In this work the SF₆ retrieval setup for MIPAS ENVISAT spectra has been improved over the one developed by Stiller et al. (2012) and a newer version of ESA spectra (level

**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

stratosphere, where trends are now positive, and in the middle tropical stratosphere, where trends are now negative. The latter might be explained by the removal of the retrieval artefact which changed the shape of the AoA profile in the tropics considerably. The linear increase in the Southern and Northern polar stratosphere and in the Northern mid-latitudes can be considered as robust results. The significant positive trend in the Northern mid-latitudes supports the findings of Engel et al. (2009) and the inferred trends match impressively well with the estimated trend by Engel et al. (2009).

The refined MIPAS observations on AoA in this study do not corroborate the results of various model studies, which consistently predict a decreasing AoA for the whole stratosphere. However, our decadal trends cannot be compared to results from long-term model studies. Our comparison with the KASIMA model for the period 2002–2012 shows, that the linear increase in the upper polar stratosphere and in the Northern mid-latitudes can be reproduced in the model at least when data is sampled and analysed in the same manner as the MIPAS data.

Nevertheless this study finds a decreasing AoA trend in the tropics and in the lower and lowermost mid-latitudinal Southern stratosphere in agreement with long-term model studies, and hence supports the idea of an increasing shallow branch of the BDC, which was also proposed by Bönisch et al. (2011) and supported by Diallo et al. (2012), at least in the Southern Hemisphere.

Acknowledgements. This work was funded by the “CAWSES” priority programme of the German Research Foundation (DFG) under project STI 210/5-3 and by the German Federal Ministry of Education and Research (BMBF) within the “ROMIC” programme under project 01LG1221B.

E. Eckert was funded by the DFG project CL 319/2-1 (COLIBRI). We would like to acknowledge provision of MIPAS level-1b data by ESA and the SF₆ data from the NOAA/ESRL halocarbons in-situ programme. The authors also acknowledge support by Deutsche Forschungsgemeinschaft and Open Access Publishing Fund of Karlsruhe Institute of Technology.

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Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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ACPD

15, 14685–14732, 2015

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

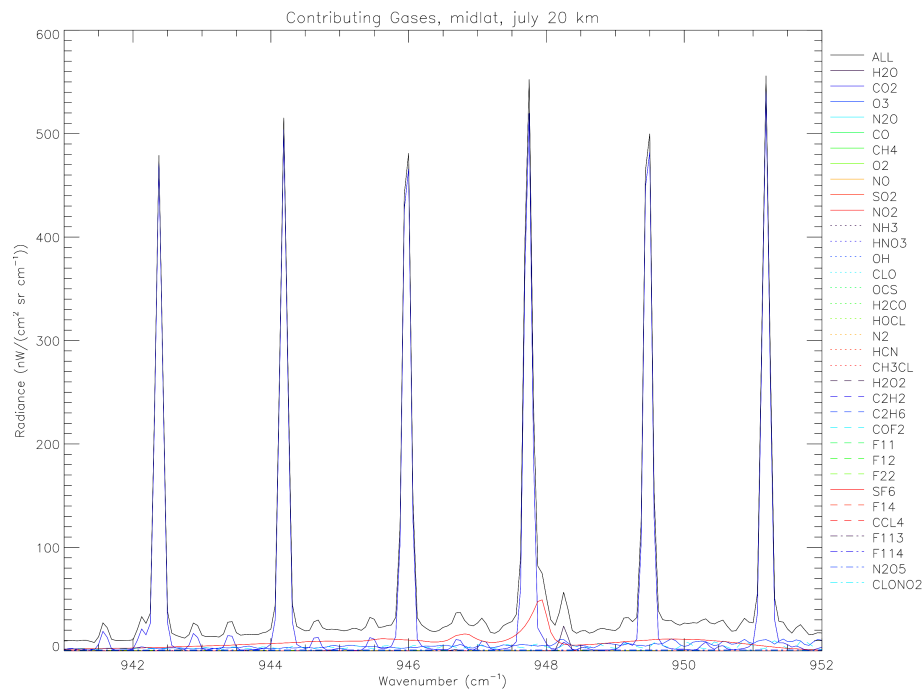


Figure 1. Contributing spectra in mid-latitudes in July at 20 km (low resolution) with the SF_6 signature in red.

Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

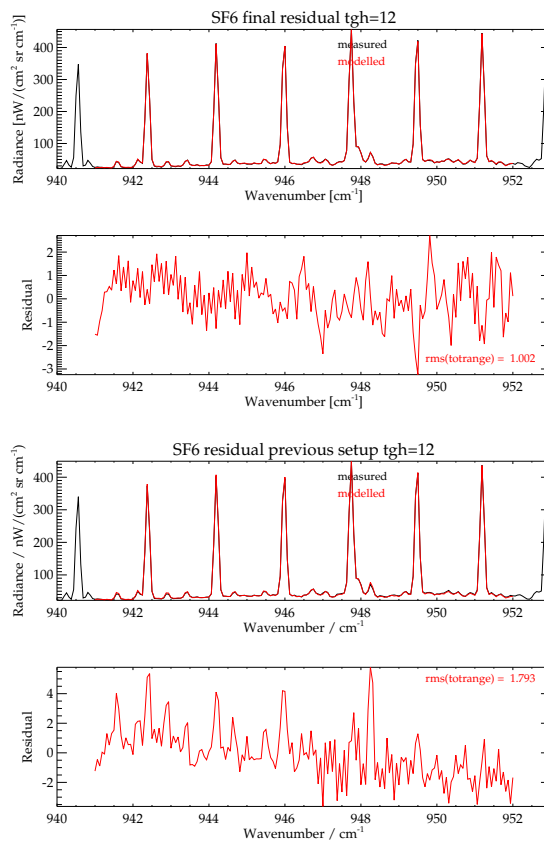


Figure 2. Coadded spectra (measured and modelled) and residuals for tangent height 12 (approx. 23 km) over one day for the final retrieval setup (upper panels) and for the the previous retrieval setup (lower panels).

**Reassessment of
MIPAS age of air
trends and variability**

F. J. Haenel et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



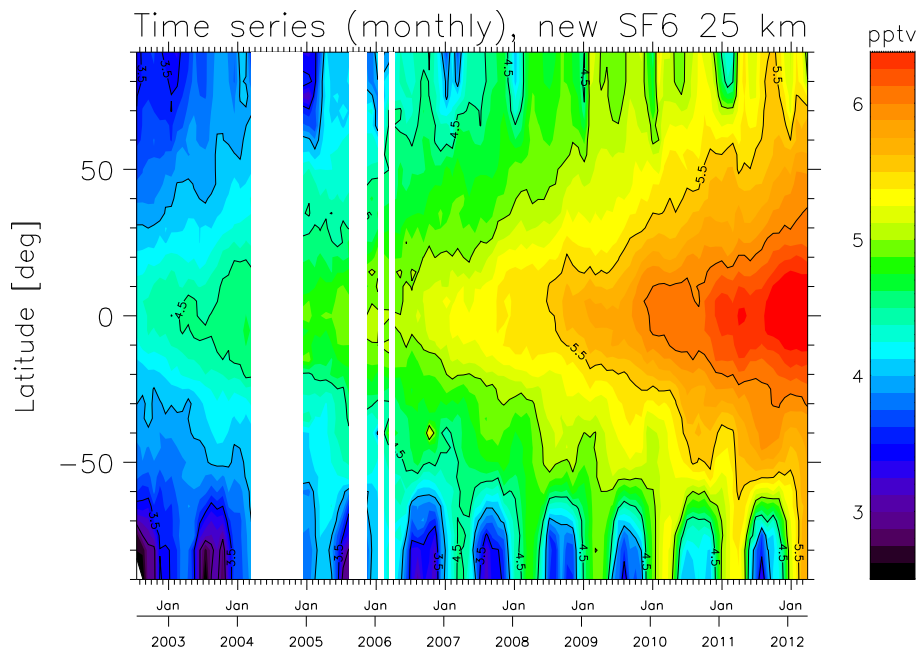
Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Figure 3.** Timeseries of SF₆ monthly zonal means at 25 km.

Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

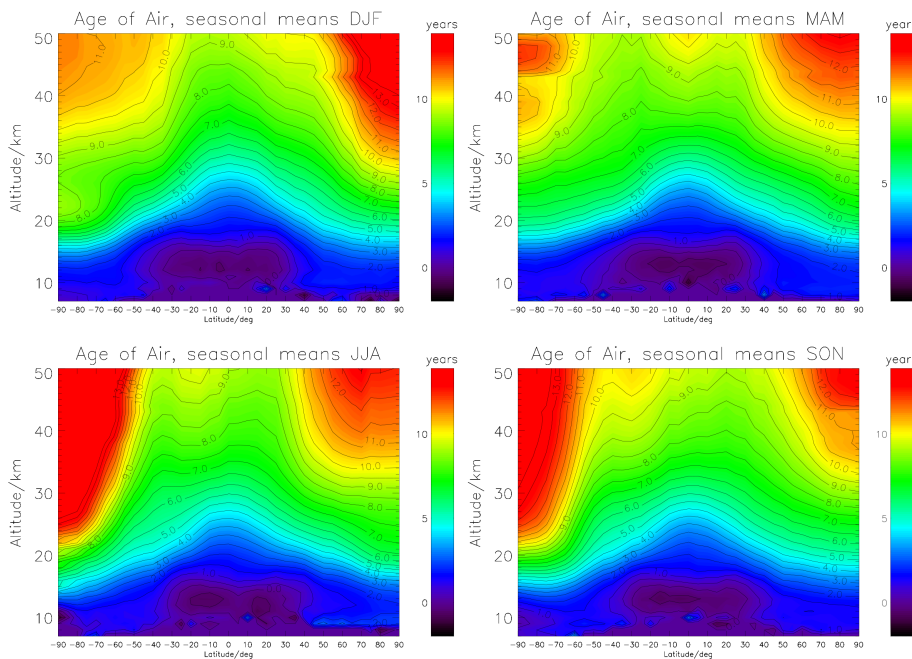


Figure 4. Zonal mean distribution of mean age of stratospheric air for the four seasons, derived by averaging MIPAS AoA data of all available years for the respective season.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

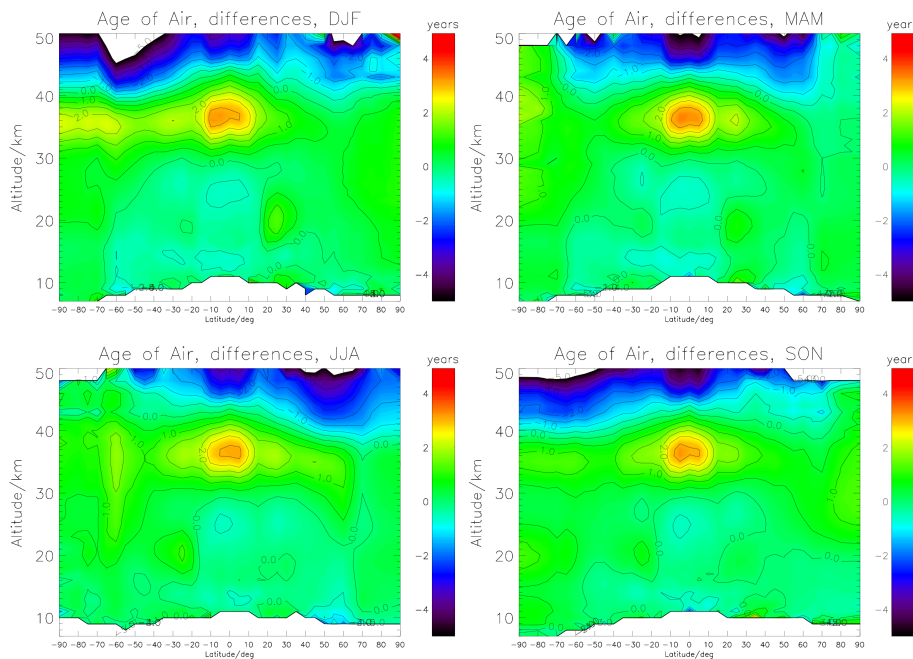


Figure 5. Differences of zonal seasonal mean distribution of mean age of stratospheric air to the previous data version averaged for the four seasons.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

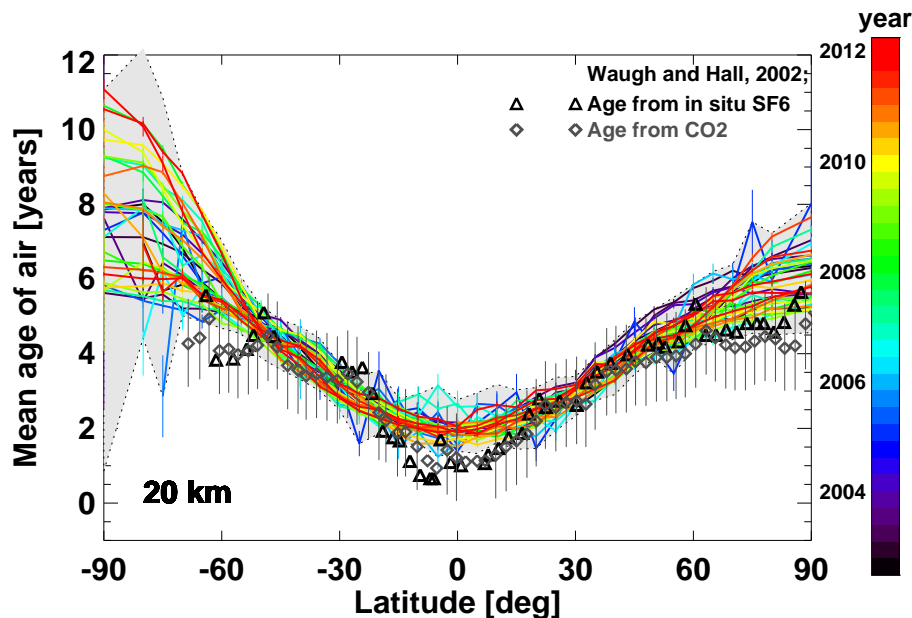


Figure 6. Comparison of MIPAS AoA latitude cross-sections at 20 km altitude (coloured curves and shaded area) with AoA derived from earlier airborne SF₆ (black triangles) and CO₂ measurements (grey diamonds with error bars) as published in Waugh and Hall (2002) and Hall et al. (1999). The shaded area represents the range of all MIPAS monthly mean AoA observations, while the coloured curves show AoA latitudinal dependence for every third month. The colour code provides the time of measurement.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

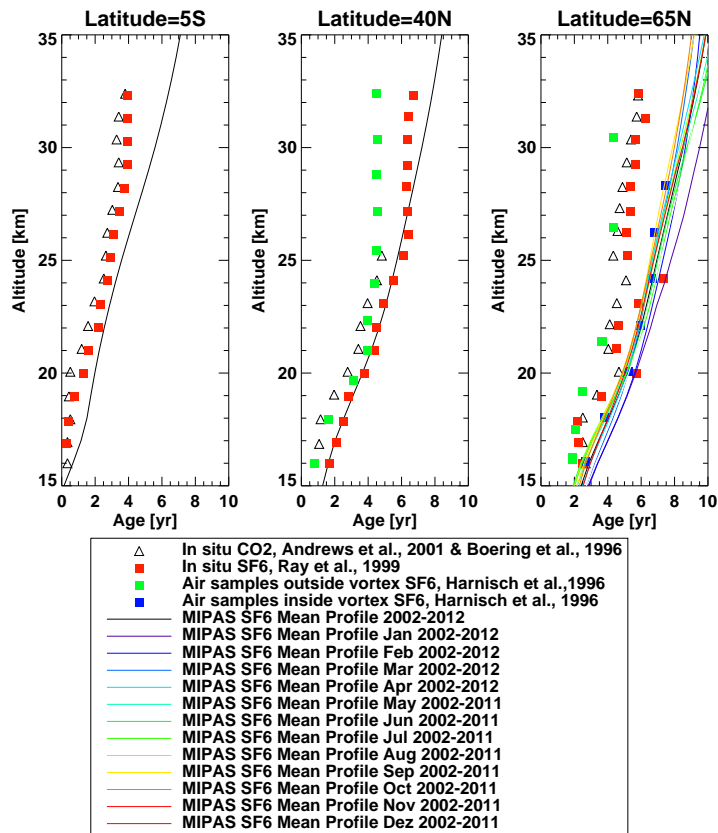


Figure 7. Comparison of MIPAS AoA profiles with airborne profiles of the 1990s for the tropics (5° S), the Northern mid-latitudes (40° N) and the Northern high latitudes (65° N).

Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

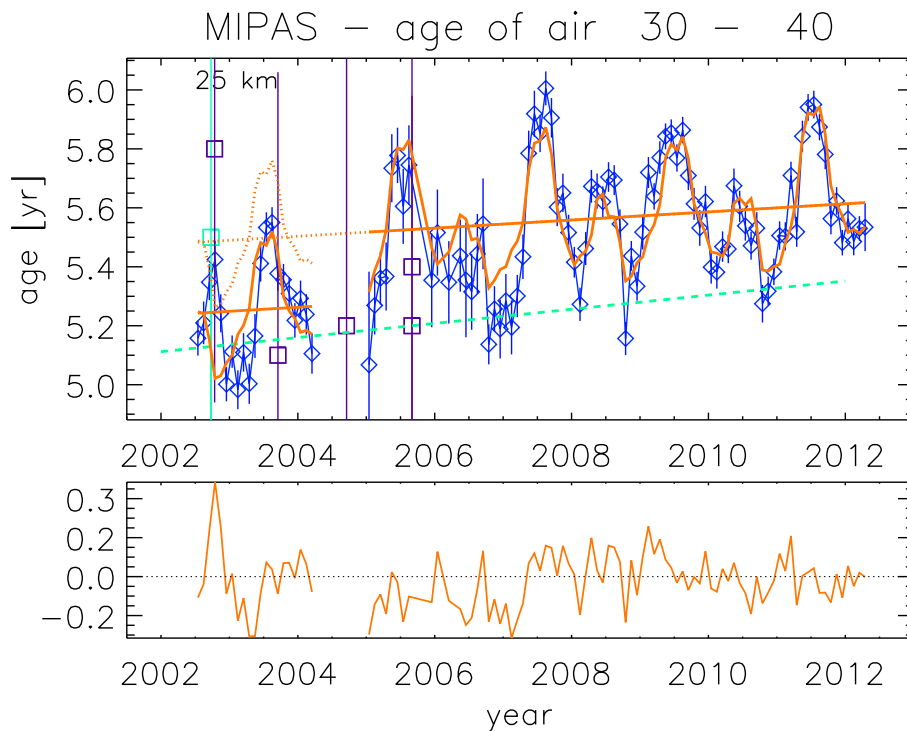


Figure 8. Example of the fit (in orange) of the regression model to MIPAS AoA monthly means (in blue) at 25 km for 30 to 40° N. The error bars represent the standard error of the mean (SEM). The orange line is the derived trend, squares represent the measurements by Engel et al. (2009) and green dashed line their estimated trend. Underneath the residual of the fit is shown.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

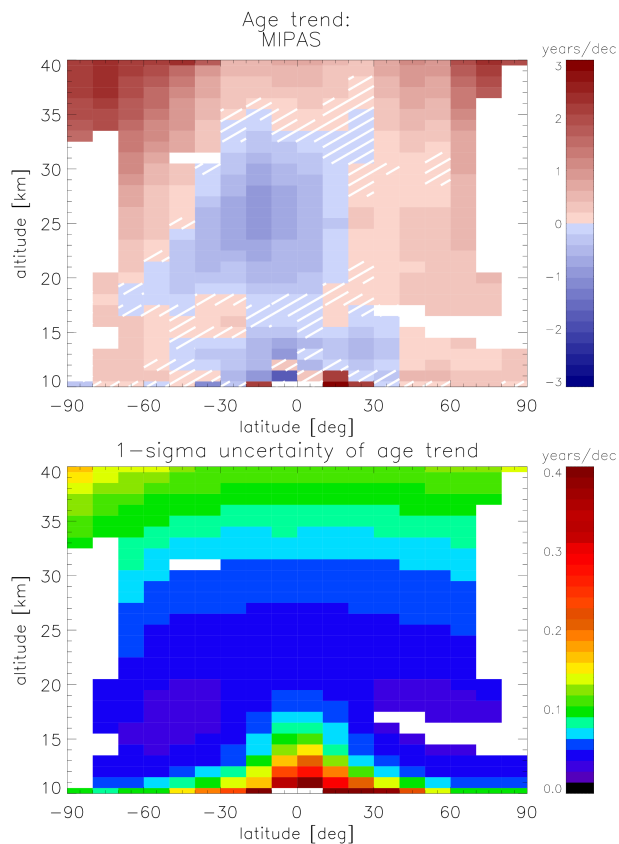


Figure 9. Altitude–latitude cross-sections of the MIPAS age of air linear increase/decrease over the years 2002 to 2012 (top), together with its 1σ uncertainties (bottom). White areas indicate where residuals between measurements and regression model get too large ($\chi^2 > 30$). Hatched areas indicate where the trend is not significant.

Reassessment of MIPAS age of air trends and variability

F. J. Haenel et al.

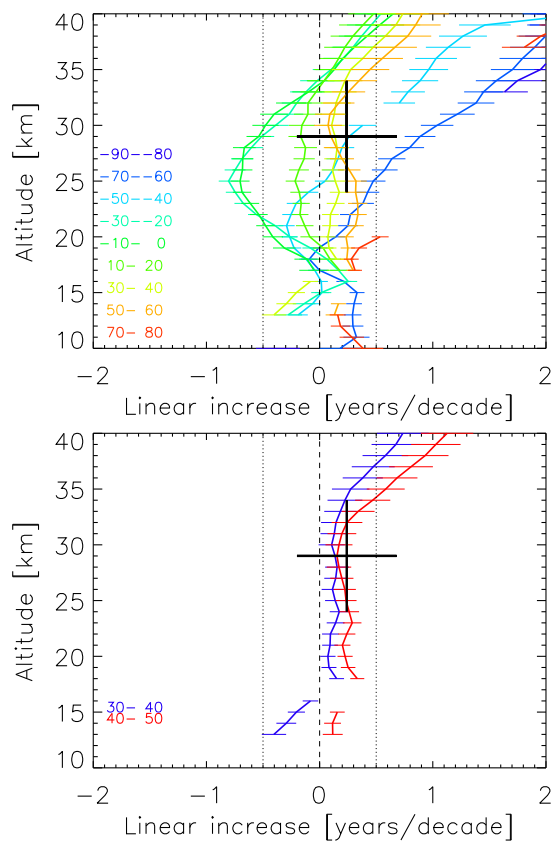


Figure 10. Vertical profiles of the age of air linear increase/decrease over the years 2002 to 2012 for example latitudes. Horizontal bars give the 2σ uncertainties of the linear variations. The 30 yr trend as derived by Engel et al. (2009) is also shown for comparison as a black cross indicating its valid altitude range and its 2σ -uncertainty.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Reassessment of
MIPAS age of air
trends and variability

F. J. Haenel et al.

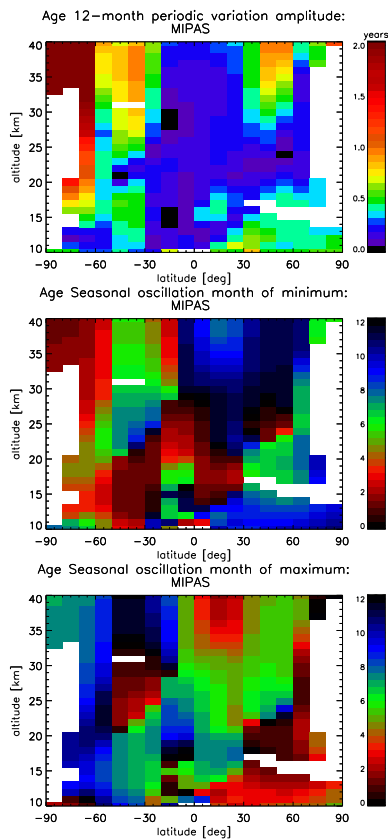


Figure 11. Altitude–latitude cross-sections of amplitudes (top) and month of the minimum (middle) and maximum (bottom) of the seasonal variation of mean age of air.

