

We thank editor Gabriele Stiller and the anonymous reviewer for their continued effort to improve the manuscript. In the attached manuscript, the major changes are marked with a black box and bold font. In the following, the original remarks are in *italics*.

Comments by Gabriele Stiller

Abstract, l14-18 (major comment): In the light of Fig. 2, and in accordance with referee #3's comments, this statement seems to be too strong. The deviations of the trends derived from models and observations, respectively, and in particular in the early phase of the analysis period, seem not to allow to state that the model "reproduces" the observed records; the model is similar in the sign of trends, at best. I realise that the deviations and their potential causes are discussed later-on in the body of the paper (although this discussion remains rather vague). Nevertheless, this discussion does not justify such a strong statement in the abstract.

We think there is possibly a misunderstanding. We agree that there are differences between the modelled and observed O3 **trends**, however, the O3 record mentioned in the abstract refers to the **absolute LS partial columns**. Here, the model actually does an excellent job in reproducing the observed timeseries, admittedly with the exception of the 5 years from 1985–1990 where there is a 1 DU (3 %) offset. Afterwards, the modelled O3 record is well within the variation range of the observations. We make this more clear in the revised abstract.

Footnote on page 2, 1st col (minor comment): I do not see the need for this footnote. This work can be referenced in the regular way.

The footnote will be removed.

Page 4, l87-91 (major comment): This is one of my most major and also general comments: The trend derived within this work from the combination of SAGE-II and SCIAMACHY after 2002 is about 0.5 % per decade, while the trend presented by Gebhardt et al. (2014) from SCIAMACHY alone after 2004 is about 4 % per decade. This is an order of magnitude difference. You earlier state that SAGE-II and SCIAMACHY fit very well, i.e. this cannot be the reason for the different trends. The only remaining reason is the slightly different period (2002 in your case, 2004 in Gebhardt et al.s case). When looking into the time series of Fig 2., one might have the impression that the increase became considerably steeper after 2004. This puts into doubt if the turnaround point derived from the model time series is appropriately to be applied to the observations, or if the assessment of the turnaround point should not be done for each individual time series as presented in Fig. 3.

Said all this, I would like to clearly state that it is not the duty of the reader or the editor to find out why the trend numbers, published within 6 months in the same journal by the same research group with overlapping author lists, differ by a factor of ten. Instead it is the duty of the authors to give a satisfying

explanation why the numbers published here are not in accordance at all (not even within 2 sigma!) with numbers published slightly earlier. Please provide a careful analysis (and not only some hand-waving arguments) on the discrepancy between your trend estimate and that of Gebhardt et al.

The calculated trends are not quantitatively comparable. Aside from the fact that Gebhardt et al. present trends of profile O₃ whereas we use partial columns, the most important difference is applied regression model. The analysis of Gebhardt et al. is necessarily restricted to 2002/2004–2012 using a single linear component. In contrast, we use a much longer timeseries (1985–2012 in case of the combined SAGEII-SCIAMACHY dataset) with two linear components. As the two linear terms are not calculated independently, a combined fit will necessarily yield a different result compared to two separate fits with only a single linear term each. As an example, Fig. 1 shows the resulting O₃ profile trends if we apply our regression model to the 2002–2012 SCIAMACHY data with only a single linear term. The resulting trend is about 5 % per decade in the LS region, i.e. reasonably close to the results of Gebhardt et al. considering the remaining differences in the regression, for example the choice of fit proxies. We rephrased the objected paragraph in the manuscript to make clear that the important point is that several instruments show positive O₃ trends since the beginning of the 21st century, not supporting the hypothesis of continued increase of upwelling. The absolute trends are significantly larger compared to our results due to the different regression approach and can only be compared qualitatively.

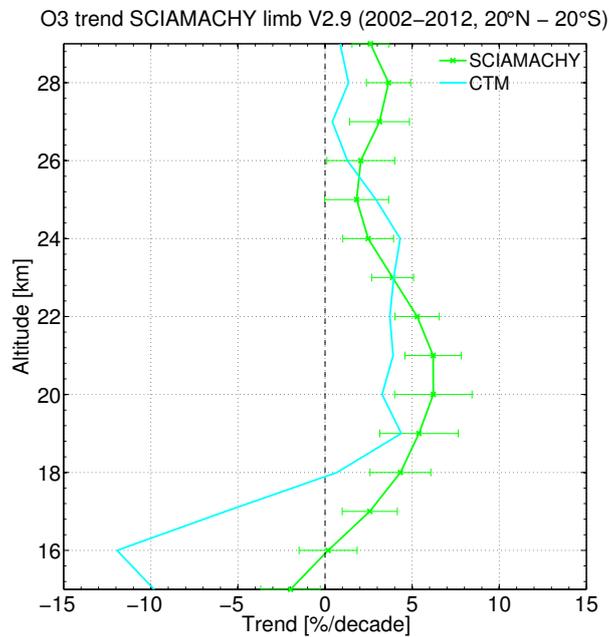


Figure 1: Linear O₃ trend in the tropics from 2002–2012 from SCIAMACHY and B3DCTM.

Page 5, l8 to end of section (major comment): Change of ODS is not the only process relevant for extra-tropical ozone variability, in particular in the UTLS; pollution transported to the UTLS and affecting the ozone budget there seems to be another source of variability with at least similar importance. For this reason I don't right see how the sensitivity studies related to ODS emissions could help to analyse the impact of in-mixing of ozone into the tropical LS.

First of all, there is strong chemical net production of O₃ in the LS, balanced by upward transport (see introduction for references). Consequently, the O₃ concentration in the LS is not very sensitive to changes in chemical loss. We tested this sensitivity by artificially increasing the abundance of ODS by fixing their emissions and found only minor changes in tropical O₃ concentration. If changes in ODS are not relevant for tropical LS O₃, we deem it reasonable to assume that this also applies for other pollutants. Of course, we cannot explicitly rule out an impact of extra-tropical pollutants since these are not included in our model. Therefore we will add a qualifying statement to the manuscript.

Page 5, l33-35 (minor comment): This is not necessarily a contradiction; shift of the mixing barriers may show up as a reduced tropical upwelling if the latitude band under analysis is not shifted accordingly.

This is true, however, when analysing the mass fluxes we always use the corresponding turnaround latitudes, that means a confusion of both processes is not possible.

Page 5, l53-63 (major comment): The discussion of the weaknesses of the EI heating rates remains rather vague, given that this was a major point in the comments of reviewer#3, and no attempt is made to provide a quantitative assessment of the errors introduced by this. Could you give an estimate, or is there any reference in the literature, what the magnitude of the overestimation of the upwelling in the tropics is, and how it changes with altitude and, most importantly, with time? Which period is represented best by the fixed ozone climatology used within EI? Why do you assume that the discrepancies between the model and the SAGE-II data are due to these shortcomings, and why should only this period be affected? Do you have any indication for this assumption?

An evaluation of upwelling is difficult, as it cannot be measured directly. Therefore, one has to rely on proxies or model calculations. Diallo et al. (2012) demonstrated that the usage of EI heating rates improved the age of air (AoA) in the LS compared to the kinematic approach. However, they also found significant underestimation of AoA in the northern tropics of up to 1 year (50 %) compared to observations. In addition, Fueglistaler et al. (2009) and Ploeger et al. (2012) state that EI heating rates are too fast by 40–50 % in the tropical UTLS, based on model calculations. Fueglistaler et al. (2009) calculated that a significant part of this overestimation is due to the usage of a fixed O₃ climatology, as pointed out by reviewer 3. The climatology used in EI was created by Fortuin and Langematz (1995) based on data from 1966-1990. As O₃ concentration in the real atmosphere decreased since then it is possible that the fixed O₃ abundance in the EI heating rate calculations produce an artificial drift, which may partly explain the differences between modelled and observed O₃ trend. How-

ever, since both observations and model show a distinctive downward trend of LS O3 which stops after 2002, it is highly probable that this phenomenon is not a simple artefact. On the other hand, this does not explain the localised difference between model and SAGE II in the pre-Pinatubo years. We rephrased the paragraph in Sect. 3.1 to make this more clear. The discussion of EI heating rates in Sect. 3.2 have been also expanded to include the points raised above.

Figs. 3 and 4 (major comment): Please provide the trends shown in panels b, d, and f (Fig. 3) and b and d (Fig. 4) additionally as numbers (i.e. values of w_1 and w_2 or w) in a legend so that the reader has a chance to compare the derived trends for the various data records.

Done.

Page 7, l14-21 (major comment): The different signs for the mass flux trends in and out of the shallow branch indicate some kind of a divergence before 2002 and convergence after 2002. These source and sink terms need to be compensated for, for example by decreasing (before 2002) and increasing (after 2002) pseudo-horizontal transport, which in turn, should affect the strength of in- and out-mixing into/from the tropical pipe. How does this, in turn, affect tropical LS ozone and its trends? This question is also related to one of the major concerns of reviewer # 1 which has, to my opinion, not been properly addressed.

This is a convincing argument and we added a new section to the manuscript to discuss the changes in the different branches in more detail, including the role of mixing and the location of the tropical transport barriers. In fact, there is not necessarily a contradiction between a major role of upwelling in controlling LS O3 abundances and in-mixing from higher latitudes, as both processes are not independent. Increased upwelling will lead to more meridional exchange due to mass conservation, as pointed out by your comment. On the other hand, even if the mixing process is not directly related to upwelling (e.g. the impact of the Asian monsoon), the vertical transport velocity will impact the effectiveness of the in-mixing (Ploeger et al., 2012). We made this more clear in Sect. 3.2 in the manuscript.

In the new Sect. 3.3 we discuss the changes of the tropical transport barriers, which show a similar behaviour as the upwelling mass fluxes. Basically, this relationship has been pointed out earlier by, e.g., Li et al. (2010). They state that the same dynamical processes, which drive the tropical upwelling also control the extent of the tropical pipe. We find a distinctive south-shift of the mixing barriers after 2002 at 100 and 30 hPa accompanying the mass flux changes, which is consistent with the findings of Eckert et al. (2014).

Page 8, l17-21 (major comment): Given what has been shown on Page 7, lines 15ff, I would expect the blue line in Fig.7 to change from correlation to anti-correlation (or vice versa) around 70 hPa (19 km) since the trends in the mass fluxes for the transition and the shallow branch have always differing signs. However, the w^ is always positively correlated with the surface temperature. Any explanation for this?*

The “turning point” is not at 70 hPa but actually around 100 hPa (≈ 16.4

km). The correlation as shown in Fig. 7 does not become negative there, this is correct, but is statistically no longer significant.

Conclusion (major comment): I miss in the conclusion completely the discussion of the role of mixing processes; in particular, since the paper gives indirect evidence (see my comment to page 7, l14-21) that mixing with the extra-tropics must have changed considerably during the period analysed, and, moreover, at the same turnaround points as the trend change in ozone. Change of ozone trends due to change in the strength of mixing cannot be excluded in the first place, and the conclusions need to discuss why this work has provided evidence that change in mixing strength is not the driver for ozone trend changes.

We agree and reworked the conclusion section completely to take into account the new findings mentioned above.

Comments by anonymous referee 1

1. *I also see that the correlation between increasing upwelling and decreasing ozone, which even holds for the trend change around 2002, provides a strong argument for the upwelling change causing the ozone change. However, a correlation is no proof and from my point of view the authors cannot ultimately rule out an impact of changes in tropics-extratropics mixing, as long as they do not explicitly calculate this term and show its effect on tropical ozone. Therefore, I think they should clearly state this, perhaps at the end of Sect. 3.1 or in Sect. 3.2 (e.g., ‘Although we cannot ultimately rule out an effect of mixing ..., the most probable explanation ...’).*

This is correct and we reworked the manuscript accordingly (see also the related comments of G. Stiller above).

2. *(P6/L3ff) Why should a high bias of vertical transport cause a high bias in the relative upwelling trend (which is here given in %)? Please clarify.*

We meant the biased response of EI upwelling due to the fixed O3 climatology. We rephrased the sentence.

3. *(P6/L6) ‘After 2002 ..., around 2002 ...’ remove the ‘around 2002’.*

Done.

On the hiatus in the acceleration of tropical upwelling since the beginning of the 21st century

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Abstract. Chemistry-climate models predict an acceleration of the upwelling branch of the Brewer-Dobson circulation as a consequence of increasing global surface temperatures, resulting from elevated levels of atmospheric greenhouse gases. The observed decrease of ozone in the tropical lower stratosphere during the last decades of the 20th century is consistent with the anticipated acceleration of upwelling. However, more recent satellite observations of ozone reveal that this decrease has unexpectedly stopped in the first decade of the 21st century, challenging the implicit assumption of a continuous acceleration of tropical upwelling. In this study we use three decades of chemistry-transport-model simulations (1980–2013) to investigate this phenomenon and resolve this apparent contradiction. **Aside from a high-bias between 1985–1990, our model is able to reproduce the observed tropical lower stratosphere ozone record. A regression analysis identifies a significant decrease in the early period followed by a statistically robust trend-change after 2002, in qualitative agreement with the observations.** We demonstrate that this trend-change is correlated with structural changes in the vertical transport, represented in the model by diabatic heating rates taken from the reanalysis product Era-Interim. **These changes lead to a hiatus in the acceleration of tropical upwelling between 70–30 hPa and a southward shift of the tropical pipe at 30 and 100 hPa during the last decade, which appear to be the primary causes for the observed trend-change in ozone.**

sult of anthropogenic activity, has been raised (Oman et al., 2009; Butchart et al., 2010; Randel and Jensen, 2013). Recent chemistry-climate model (CCM) simulations predict an increase of resolved wave activity and orographic gravity wave drag resulting from increasing sea surface temperatures (SST; Garcia and Randel, 2008; Oman et al., 2009; Waugh et al., 2009; Butchart et al., 2010; Garny et al., 2011). This strengthens the upwelling branch of the BDC, commonly referred to as the tropical upwelling. In comparison, the behaviour of the observations available since about 1980 is ambiguous. The long-term cooling of the tropical lower stratosphere (LS, about 17–21 km; Thompson and Solomon, 2005; Young et al., 2012) and the observed weakening of the stratospheric quasi-biennial oscillation (QBO; Kawatani and Hamilton, 2013) are consistent with the predicted increase of upwelling. On the other hand, the mean residence time of air parcels in the stratosphere (age of air) inferred from sulfur hexafluoride (SF₆) measurements is inconsistent with an overall acceleration of the BDC (Engel et al., 2009; Stiller et al., 2012). Age of air changes indicate no significant changes or even deceleration of the vertical transport in the middle stratosphere. To reconcile the observed discrepancies it has been argued that the individual branches of the BDC are evolving differently, i.e. an increase of tropical upwelling does not necessarily imply an acceleration of the overall circulation (Bönisch et al., 2011; Diallo et al., 2012; Lin and Fu, 2013).

Ozone (O₃) is a sensitive proxy for vertical transport in the tropical LS (Randel et al., 2006; Waugh et al., 2009; Randel and Thompson, 2011; Polvani and Solomon, 2012). Its local mixing ratio is considered to result from a stationary state involving production by oxygen (O₂) photo-dissociation and a steady influx of O₃-poor tropospheric air from below (Avalone and Prather, 1996; Waugh et al., 2009; Meul et al., 2014). Meridional mixing from higher latitudes is another factor that contributes to the seasonality in the O₃ mixing ratios (Konopka et al., 2009; Ploeger et al., 2012; Abalos et al.,

1 Introduction

The issue of whether the large-scale Brewer-Dobson Circulation (BDC) has strengthened in the recent past, as a re-

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2013), with the largest impact during boreal summer directly above the tropopause (≈ 380 K/17 km). Several studies have reported a negative trend of O_3 in the tropical LS in the range of $-(3-6)$ % per decade from about 1985 onwards, consistent with the CCM predicted increase of tropical upwelling (e.g., Randel and Thompson, 2011; Sioris et al., 2014; Bourassa et al., 2014). In contrast, more recent O_3 observations from various satellite instruments indicate no statistically significant decrease of LS O_3 since the beginning of the 21st century (Kyrölä et al., 2013; Eckert et al., 2014; Gebhardt et al., 2014).

Stimulated by the need to explain the unusual linear trends revealed from the vertical profile of O_3 retrieved from SCIAMACHY we use three decades of O_3 observations and simulations to investigate this phenomenon. Section 2 describes the observations, model and regression analysis used in this study. The results are discussed in Sect. 3.

2 Data and analysis

2.1 Observations

For a quantitative analysis of tropical upwelling, we use combined O_3 observations from satellite instruments and sondes. The earlier decades (1985–2005) are covered by the ERBS/SAGE II instrument (McCormick et al., 1989), providing O_3 profiles based on solar occultation measurements. Due to its viewing geometry, the vertical resolution of the profiles is high (1 km, range 15–50 km), although the horizontal sampling is relatively sparse (global coverage in 1 month). Here we use version 7.0 of the data (Damadeo et al., 2013), screened for cloud and aerosol contaminated profiles as suggested by Wang et al. (2002). Two years of data after June 1991 have been omitted due to contamination by the eruption of Mt. Pinatubo. For the last decade (2002–2012), we use O_3 observations from ENVISAT/SCIAMACHY (Burrows et al., 1995) based on limb geometry (retrieval version 2.9; Sonkaew et al., 2009). The vertical resolution is about 3–4 km over an altitude range of 10–75 km; global coverage is achieved every 6 days. Data from both instruments has been binned into monthly samples on a uniform horizontal and vertical grid (15° lon. \times 5° lat. \times 1 km). To minimise sampling issues and taking into account the differences in horizontal and vertical resolution of the instruments, any further analysis is based on partial columns of O_3 between 17–21 km and 20° N– 20° S, similar to the approach of Randel and Thompson (2011).

The satellite data is augmented by an ensemble of tropical sonde measurements from the Southern Hemisphere Additional Ozonesondes network (SHADOZ; 1998–2013; Thompson et al., 2003, 2012). We use 10 sites located in the tropics with long and continuous records. The selected stations along with their temporal coverage and mean value are listed in Table 1. Typically there are 2–4 observations per

month for each SHADOZ station, which provide O_3 profiles in a considerably higher vertical resolution (50–100 m) compared to the satellite instruments. As there is a high degree of longitudinal symmetry in the stratospheric ozone profiles (Thompson et al., 2003), we average the individual records to obtain a representative mean for the tropics.

2.2 Model

To obtain a consistent timeseries of LS O_3 of the last decades for direct comparison with observations, we conducted a 33-year simulation with the Bremen three-dimensional chemistry-transport-model (B3DCTM; Sinnhuber et al., 2003; Aschmann et al., 2009; Aschmann and Sinnhuber, 2013). The current version of the model has a horizontal resolution of 3.75° lon. \times 2.5° lat. and covers the vertical domain from the surface up to approximately 55 km using a hybrid $\sigma - \theta$ coordinate system (e.g., Chipperfield, 2006). The vertical resolution in the tropical LS is about 600 m. The model is driven by 6-hourly input of European Centre for Medium-range Weather Forecast (ECMWF) Era-Interim (EI; Dee et al., 2011) reanalysis data. Vertical transport in the purely isentropic domain (above ≈ 16 km in the tropics) is prescribed by EI all-sky heating rates. The B3DCTM incorporates a comprehensive chemistry scheme originally based on the chemistry part of the SLIMCAT model (Chipperfield, 1999), covering all relevant photochemical reactions for stratospheric O_3 chemistry. Reaction rates and absorption cross sections are taken from the Jet Propulsion Laboratory recommendations (Sander et al., 2011). Injection of ozone-depleting substances (ODS) is prescribed according to WMO scenario A1 (World Meteorological Organization, 2011). To avoid initialisation artefacts, the model has been run with replicated input data to reach steady state before starting the actual integration from January 1979 to October 2013.

2.3 Regression

The multivariate regression analysis used throughout this study is based on Reinsel et al. (2002) with Y_t as the monthly mean variable to be fitted:

$$Y_t = \mu + S_t + \omega_1 X_{1t} + \omega_2 X_{2t} + QBO_t + ENSO_t + SC_t + N_t \quad (1)$$

$$t = 1, \dots, T$$

where μ is the baseline constant, S_t a seasonal component, $\omega_{1,2}$ are the trend coefficients with $X_{1,2t}$ as trend functions:

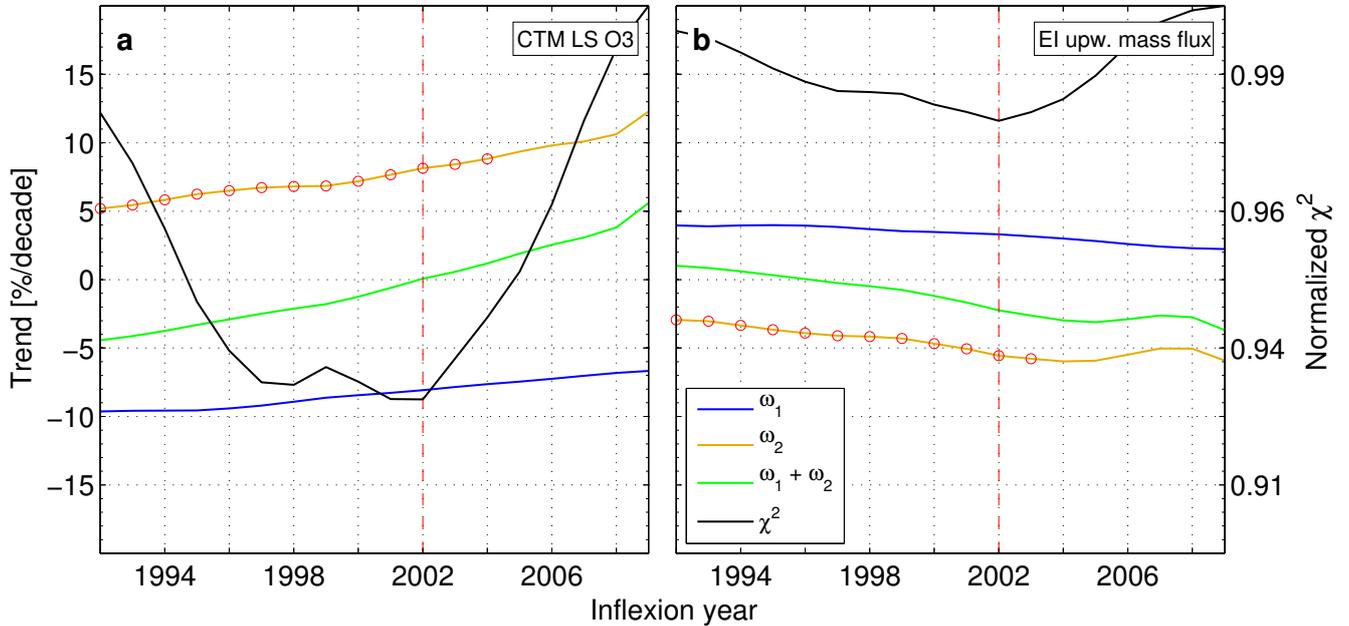
$$X_{1t} = t/12 \quad (2)$$

$$X_{2t} = \begin{cases} 0 & 0 < t \leq T_0 \\ (t - T_0)/12 & T_0 < t \leq T \end{cases} \quad (3)$$

Note that in contrast to most previous studies, which examined LS O_3 (e.g., Randel and Thompson, 2011; Sioris et al., 2014), our regression model uses two linear components to

Table 1. Geolocation, temporal coverage and average LS O₃ column of utilised SHADOZ sites.

Name	Location		Coverage	Average [DU]
Ascension Is.	14.4°W	8.0°S	01/1998 – 08/2010	28.76
Costa Rica	84.0°W	9.9°N	07/2005 – 12/2012	30.66
Hilo	155.0°W	19.4°N	01/1998 – 02/2013	36.97
Watakosek-Java	112.6°E	7.5°S	01/1998 – 06/2013	27.06
Kuala Lumpur	101.7°E	2.7°N	01/1998 – 12/2011	30.26
Nairobi	36.8°E	1.3°S	01/1998 – 06/2013	30.66
Natal	35.3°W	5.5°S	01/1998 – 05/2011	29.74
Paramaribo	55.2°W	5.8°N	09/1999 – 12/2011	31.53
Samoa	170.6°W	14.2°S	01/1998 – 12/2012	30.91
San Cristobal	89.6°W	0.9°S	03/1998 – 10/2008	29.26

**Fig. 1.** The dependence of the linear fit parameters ω_1 , ω_2 and ω ($\omega_1 + \omega_2$) on the inflexion year T_0 is shown for the regression of modelled tropical LS O₃ column (a) and EI upward mass flux at 70 hPa (b). Red circles denote the years where the trend-change (ω_2) exceeds the 95% confidence threshold. The black lines are the normalised χ^2 values of the fit residuals.

take into account a possible change of trend at a given point in time. ω_1 is the linear trend up to a specified inflexion date T_0 . After T_0 , the new linear trend ω comprises the sum of the earlier trend ω_1 and the trend-change component ω_2 . The additional regression terms are QBO_t for QBO, $ENSO_t$ for the EI Niño Southern Oscillation (ENSO) and SC_t for solar cycle. The QBO proxy consists of the QBO.U30 and QBO.U50 (zonal wind 30/50 hPa) from the NOAA Climate Prediction Center¹, the ENSO proxy is represented by the Multivariate ENSO Index (MEI) from the NOAA Earth System Research Laboratory² (Wolter and Timlin, 2011) lagged by two

months and the solar cycle by the Bremen composite Mg II index³ (Snow et al., 2014). Finally, N_t represents the unexplained noise.

Assuming first order autocorrelation noise (AR(1) model), as commonly used in the regression of O₃ timeseries (e.g., Reinsel et al., 2002; Jones et al., 2009; Sioris et al., 2014), the corresponding standard deviations for the trend components

¹ www.cpc.ncep.noaa.gov/data/indices/² www.esrl.noaa.gov/psd/enso/mei/³ www.iup.uni-bremen.de/gome/solar/MgII.composite.dat

are given by

$$\sigma_{\omega_1} \approx \frac{\sigma_N}{n^{3/2}} \sqrt{\frac{1+\phi}{1-\phi}} \quad (4)$$

$$\sigma_{\omega_2} \approx \frac{\sigma_N}{2} \sqrt{\frac{1+\phi}{1-\phi}} \left(\frac{n}{n_0 n_1} \right)^{3/2} \quad (5)$$

$$\sigma_{\omega} \approx \frac{\sigma_N}{n_1^{3/2}} \sqrt{\frac{1+\phi}{1-\phi}} \sqrt{\frac{n_0+4n_1}{4n}} \quad (6)$$

where σ_N is the standard deviation of the fit residuals, n_0 , n_1 are the numbers of years of data before and after the trend-change, respectively, with $n = n_0 + n_1$. ϕ represents the autocorrelation of the residuals with a time lag of 1 month.

The choice of the inflexion year T_0 is a free parameter found in the regression analysis. Figure 1 illustrates the impact of the choice of T_0 on the regression of modelled LS O_3 columns and EI upward mass flux (as discussed below in Sect. 3). A 2σ -significant trend-change (ω_2) is obtained for a range of possible inflexion years (marked by red circles). We therefore use a χ^2 fit based on the regression residuals, similar to the approach described by Jones et al. (2009), to identify the most probable inflexion year. We find a clear minimum in the χ^2 values close to 2002 and consequently select this year as the turning point in the trend analysis.

3 Results and Discussion

3.1 Lower stratosphere ozone column

Figure 2 presents tropical LS O_3 column anomalies (20°N – 20°S , 17–21 km) from measurements and the simulation. The agreement between model and observations is good, except for a small high-bias relative to the earlier SAGE II data (1985–1990) of approximately 1 DU ($\approx 3\%$): correlation coefficients are 0.65 between modelled and observed datasets.

A decline of O_3 is evident in the tropical LS during the first two decades (1980–2002), both in the observed and modelled timeseries. This is consistent with an increase of tropical upwelling during this period. However, this trend vanishes in the third decade (2002–2013). Figures 3a and b illustrate the results from the regression analysis of the modelled timeseries showing the fit function and the corresponding residuals, respectively. The linear trend amounts to $-8.1 \pm 0.9\%$ per decade (ω_1) in the pre-2002 period and $0.1 \pm 3.3\%$ per decade (ω) for the remaining years. The resulting trend-change of 8.2% per decade (ω_2) is statistically significant within the 95% confidence interval (i.e. $\omega_2 > 2\sigma_{\omega}$).

To apply our analysis to the observational data we merge the available datasets (SAGE II–SCIAMACHY; SAGE II–SHADOZ) by joining the two individual timeseries and average the overlap period. In case of SAGE II–SHADOZ, this method has been applied before by Randel and Thompson (2011) who found excellent agreement between SHADOZ

and SAGE II in tropical LS O_3 , despite the sparse horizontal sampling provided by the sondes. SAGE II and SCIAMACHY show similarly good agreement in this area, with a correlation of 0.82 and an average bias of 0.5 DU ($< 2\%$) during the overlap period. Considering the good agreement between the observations (Fig. 3), it is reasonable to combine them into a continuous timeseries. When we apply the regression to the combined SAGE II–SCIAMACHY timeseries, we calculate a trend of $-3.9 \pm 0.5\%$ per decade (ω_1) for the pre-2002 period, consistent with the range of $-(3\text{--}6)\%$ per decade given by earlier studies (Fig. 3c, d; Randel and Thompson, 2011; Sioris et al., 2014; Bourassa et al., 2014). However, this value is smaller than the pre-2002 trend determined from our CTM simulations. Considering the good agreement between observations and model after the Mt. Pinatubo data gap (1991–1994), the discrepancy must be mainly caused by the model high-bias compared to the early SAGE-II measurements. The origin of this bias is not entirely clear. Assuming that the SAGE II record is consistent before and after the Mt. Pinatubo eruption, the bias possibly results from a spin-up effect of the model stemming from the usage of replicated input data for the initialisation phase (Sect. 2.2). This would explain why the bias is only present in the first third of the timeseries. Another point could be the systematic overestimation of LS vertical transport based on diabatic heating rates in the EI dataset, which is discussed in more detail in Sect. 3.2. After 2002, the agreement between modelled and observed trends improves considerably. For the SAGE II–SCIAMACHY dataset, the trend amounts to $0.5 \pm 1.5\%$ per decade (ω), yielding a statistically significant trend-change of 4.4% per decade (ω_2). We obtain similar values (-3.6 ± 0.5 , $0.4 \pm 1.4\%$ per decade for ω_1 , ω) if we use the SHADOZ data instead of SCIAMACHY in the combined dataset (Fig. 3e, f). Consequently, both observational and model data show that the decrease of LS O_3 effectively stopped around 2002 and there has been no significant change afterwards. **This is in qualitative agreement with previous studies, which focus solely on the most recent observational record of O_3 . Gebhardt et al. (2014) compared several satellite instruments and report consistently positive trends of tropical O_3 between 17–21 km, ranging from about 2 (OSIRIS), 4 (SCIAMACHY) up to 14 % per decade (MLS), covering the years 2004–2012. Eckert et al. (2014) found a slightly positive trend of 0–1 % per decade in the same region in MIPAS observations (2002–2012). These results cannot be quantitatively compared to our results, as the trends have been calculated for profile ozone and with a significantly different regression approach, which uses only a single linear component and is obviously restricted to 2002/2004–2012. If we apply these restrictions to our own regression analysis (i.e., only ω_1 , range 2002–2012), the resulting trends are consistent with the earlier studies, i.e. 3–5 % per decade for model and SCIAMACHY profile ozone between 17–21 km in the tropics (not shown here). The important point is, that de-**

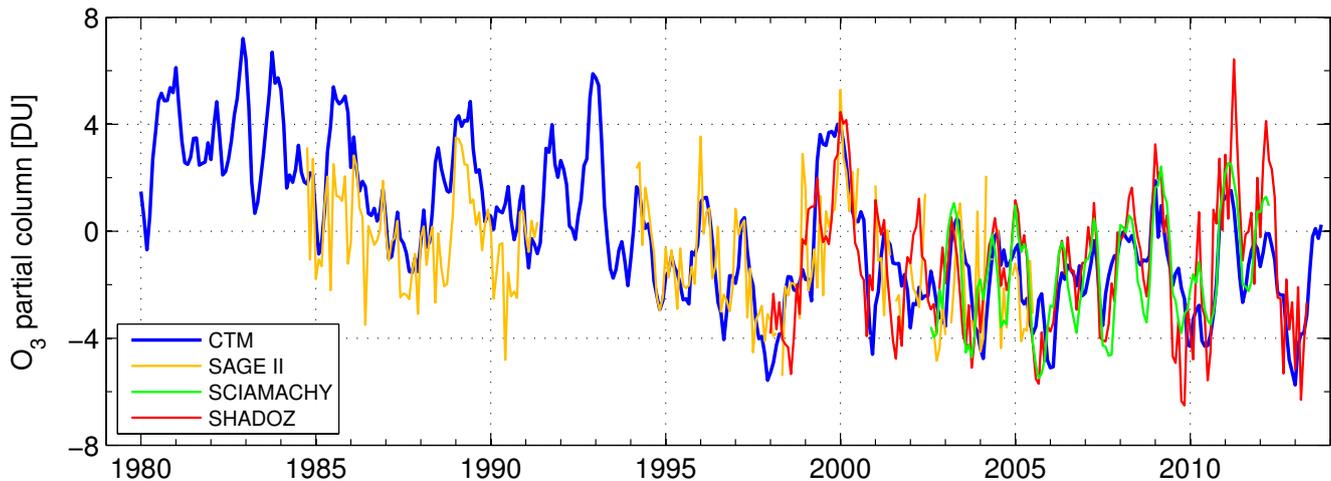


Fig. 2. Observed and simulated tropical (20°N – 20°S) LS O_3 partial columns (17–21 km). Anomalies are deviations from the modelled 1980–2013 averages.

spite the considerable spread in the determined trends all instruments described above show no further decrease of tropical LS O_3 during the last decade. This agreement justifies our confidence that this phenomenon is not an instrumental or retrieval artefact.

Local chemical effects can be largely ruled out as explanation for the detected trend-change of LS O_3 . As stated above, O_3 abundance in the tropical LS is mainly determined by vertical transport and chemical net production (by O_2 photolysis; Avallone and Prather, 1996; Waugh et al., 2009; Meul et al., 2014). O_3 -destroying catalytic species are scarce in the tropical LS, therefore the phase-out of ODS, and the associated recovery (e.g., World Meteorological Organization, 2011), has no direct impact on O_3 concentrations in this region. To verify this assumption, we have conducted a sensitivity simulation with identical setup but with ODS emissions fixed to the values of 1980 (not shown here). In contrast to mid and high latitudes, the O_3 mixing ratios in the tropical LS show little difference to the standard simulation ($<2\%$ compared to 15 – 20% at higher latitudes) and we calculate very similar trends (-7.8 ± 0.9 , $-0.6 \pm 3.4\%$ per decade for ω_1 , ω). The relatively small impact on the post-2002 trend ω , compared to the overall trend-change, is likely related to O_3 in-mixing from mid-latitudes. Not explicitly accounted for is a possible indirect relationship between ODS-related polar O_3 depletion and tropical LS O_3 by dynamical coupling, as pointed out by several studies (Waugh et al., 2009; Oman et al., 2009). **Furthermore, the model does not consider in-mixing of extra-tropical pollutants aside from ODS.** Meul et al. (2014) predict an increase of photolytic O_3 production as a result from long-term changes in the overhead O_3 column. Furthermore, an increase of odd nitrogen (NO_x) might lead to additional O_3 production. However,

they found no indication that either process is sufficient to explain a short-term trend-change. Overall the most probable explanation of the observed behaviour is that changes in dynamics must be involved.

3.2 Tropical upwelling

On the dynamical side, the obvious impact factors are tropical upwelling and in-mixing from higher latitudes. An accurate attribution of the relative impact of each process is difficult, as they are not completely independent from each other. From an Eulerian point of view, changes in the vertical flux must be balanced by horizontal exchange due to mass conservation. From a Lagrangian perspective, the vertical velocity of an air parcel determines the effectiveness of a local in-mixing region, for example by the Asian monsoon anticyclone as described by Ploeger et al. (2012); Abalos et al. (2013). In the following, we concentrate on tropical upwelling as it is easier to quantify than in-mixing. A typical representative quantity for the tropical upwelling is the upward mass flux at 70 hPa (≈ 18.5 km in the tropics; Butchart et al., 2010; Seviour et al., 2012). A recent study assessing the upward mass flux in EI found a negative trend of -5% per decade for the years 1989–2009, based on EI kinematic vertical winds (Seviour et al., 2012). This is in contradiction with the results of current CCMs, which predict an increase of upwelling of about 2.0% per decade (ensemble mean; Butchart et al., 2010). The quality of stratospheric vertical transport in EI improves considerably, when diabatic heating rates are used instead of the kinematic wind. The diabatic representation of vertical transport generally yields more realistic estimates of stratospheric age of air in comparison to the kinematic approach (Diallo et al., 2012) and is also less

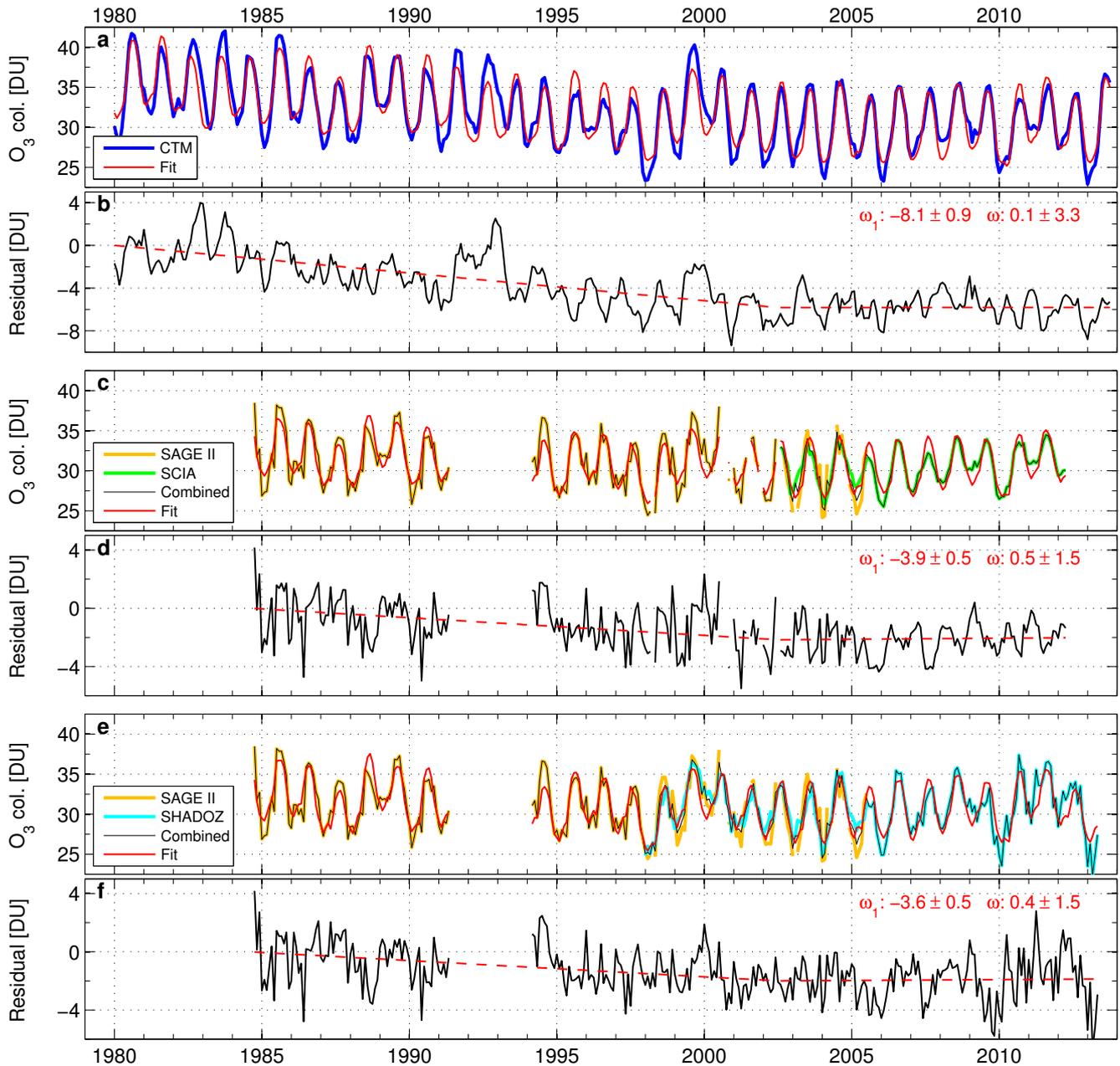


Fig. 3. Regression analysis of observed and simulated O₃ partial columns. Model, combined SAGE II/SCIAMACHY and combined SAGE II/SHADOZ LS O₃ with regression function (a, c, e). Corresponding fit residuals excluding the linear terms (b, d, f). The dashed red lines depict the resulting linear trends before and after 2002 with the fit coefficients in red font (unit: % per decade).

dispersive (Ploeger et al., 2011). **On the downside,** there are indications that EI heating rates overestimate the ascent in the tropics. Diallo et al. (2012) report an underestimation of age of air compared to observations in the tropical northern hemisphere reaching up to 50 % when using EI heating rates. Similarly, Fueglistaler et al. (2009) and Ploeger et al. (2012) state that EI heating rates are too fast by 40–50 % in the tropical up-

per troposphere/lower stratosphere (UTLS). According to Fueglistaler et al. (2009), a significant part of this overestimation can be attributed to the fixed O₃ climatology for calculating the heating rates in the reanalysis. As the utilised climatology by Fortuin and Langematz (1995) is based on data from 1966–1990 and the real O₃ mixing ratio decreases until 2002 (Sect. 3.1), this will lead to increasingly biased responses of the EI radiative transfer

scheme during this period. This would partly explain the overestimation of the pre-2002 trend of LS O₃ in the model (Sect. 3.1) and is consistent with the similar overestimation of the upwelling trend in EI discussed below.

Figure 4 shows the tropical LS EI all-sky heating rates (20°N–20°S, 17–21 km; panel a), which are used to drive the vertical transport in our isentropic model, and the corresponding EI upward mass flux at 70 hPa (panel c). The upward mass flux is the integral of the residual vertical velocity w^* between turnaround latitudes ($\phi^{\text{SH}}, \phi^{\text{NH}}$) as described in Seviour et al. (2012)

$$\text{upward mass flux} = 2\pi \int_{\phi^{\text{SH}}}^{\phi^{\text{NH}}} w^* \rho_0 a^2 \cos\phi \, d\phi \quad (7)$$

with basic state density ρ_0 and Earth’s radius a . In turn, w^* is calculated from the EI heating rates using the iterative algorithm described by Solomon et al. (1986). Applying the regression analysis to the upward mass flux yields a positive trend of 3.3 ± 0.7 % per decade for the pre-2002 period (ω_1 ; Fig. 4d). This value is consistent with the CCM results (2.0 % per decade) although somewhat high-biased, likely reflecting the impact of the fixed O₃ climatology for calculating the EI heating rates as discussed above. After 2002, however, there is a statistically significant trend-change leading to a negative trend of -2.3 ± 2.5 % per decade (ω) mirroring the trend-change in the LS O₃ timeseries.

3.3 Structural BDC changes

Further insight into structural changes of the BDC can be gained by decomposing the circulation into different branches. Here, we adopt the method of Lin and Fu (2013) and define the tropically controlled transition branch, ranging from 100 to 70 hPa and the stratospheric shallow and deep branch (70–30 hPa and <30 hPa, respectively). The strength of the individual branches is estimated by the differences of upward mass fluxes across the corresponding boundaries (determined between turnaround latitudes); in case of the deep branch it is simply the flux across the 30 hPa boundary. Figure 5 presents the results of the regression analysis of the mass fluxes in the individual branches, calculated from EI all-sky heating rates as above. In the stratospheric deep branch, there is no significant non-zero trend during the last decades, which is consistent with earlier studies (e.g., Engel et al., 2009; Bönisch et al., 2011; Lin and Fu, 2013). Consequently, the evolution of the shallow branch is dominated by the changes in the 70 hPa flux as discussed above (Fig. 4d), displaying the characteristic trend change around 2002 (5.8 ± 0.9 , -5.6 ± 3.3 % per decade for ω_1, ω). Interestingly, the transition branch shows the inverted behaviour, a decrease prior to 2002 and an increase afterwards (-9.0 ± 1.1 , 11.1 ± 4.0 % per decade for ω_1, ω). Apparently there is a shift in mass flux balance from the transition branch towards the

shallow branch in earlier decades, which begins to reverse at the beginning of the 21st century. This result does not agree with the findings of Lin and Fu (2013), who calculate positive trends both in the transition and shallow branches based on current CCM simulations. However, Bönisch et al. (2011) detect a significant increase of the residual circulation around 2000, based on N₂O and O₃ observations. They state that this increase is mainly confined to the lower stratosphere between 100–63 hPa, which is consistent with our definition of the transition branch.

Another important aspect is the evolution of the tropical mixing barriers, i.e. the extent of the “tropical pipe” (Plumb, 1996). Stiller et al. (2012) and Eckert et al. (2014) state that the observed trend patterns of SF₆ and O₃ could be possibly explained by a weakening or a shift of transport barriers. In turn, the extent of the tropical pipe is affected by the same dynamical processes which cause the increase in tropical upwelling (Li et al., 2010). Therefore it is likely that both phenomena are related. Figure 6 shows the development of the turnaround latitudes determined from upward mass fluxes at 100, 70 and 30 hPa, separated for the northern and southern hemisphere (NH, SH). The resulting linear trends are listed in Table 2. The tropical pipe is expanding at all three levels in the pre-2002 period. In the 100 and 30 hPa level the expansion is about 2.7° per decade whereas the trend in the NH is roughly twice as large as in the SH. At 70 hPa, the expansion is only 0.6° per decade, mainly restricted to the SH as there is no significant trend in the NH. After 2002, the tropical pipe shrinks at all levels. Again, the effect is most pronounced at 100 and 30 hPa with -6.2° per decade and -2.8° per decade, respectively and there is the same asymmetry that the change is much stronger in the NH. At 70 hPa, the effect is smaller (-0.8° per decade) and restricted to the SH. The development at the lowest level (100 hPa) is consistent with the observed widening of the tropical belt in the troposphere (Seidel et al., 2008). Relying on independent observational parameters, they state that the tropical belt expanded by 2–5° per decade between 1980–2005. The expansion accompanying the mass flux decrease in the pre-2002 period and the inverted picture afterwards is also in agreement with the findings of (Li et al., 2010), who predict an anti-correlation between upwelling strength and expansion of tropical mixing barriers. However, we cannot reproduce this anti-correlation at 70 and 30 hPa. At 70 hPa, there is hardly any statistically significant change in the extent of the tropical pipe although the mass flux is changing whereas Li et al. (2010) predict a narrowing at the same pressure level of -0.5° per decade. Just the opposite situation occurs at 30 hPa. Here, the extent of the tropical pipe is changing despite the absence of significant vertical mass flux changes. An interesting detail is the south-shift of the tropical pipe after 2002. Both at 100 hPa and 30 hPa there is a net south-shift of tropical upwelling of

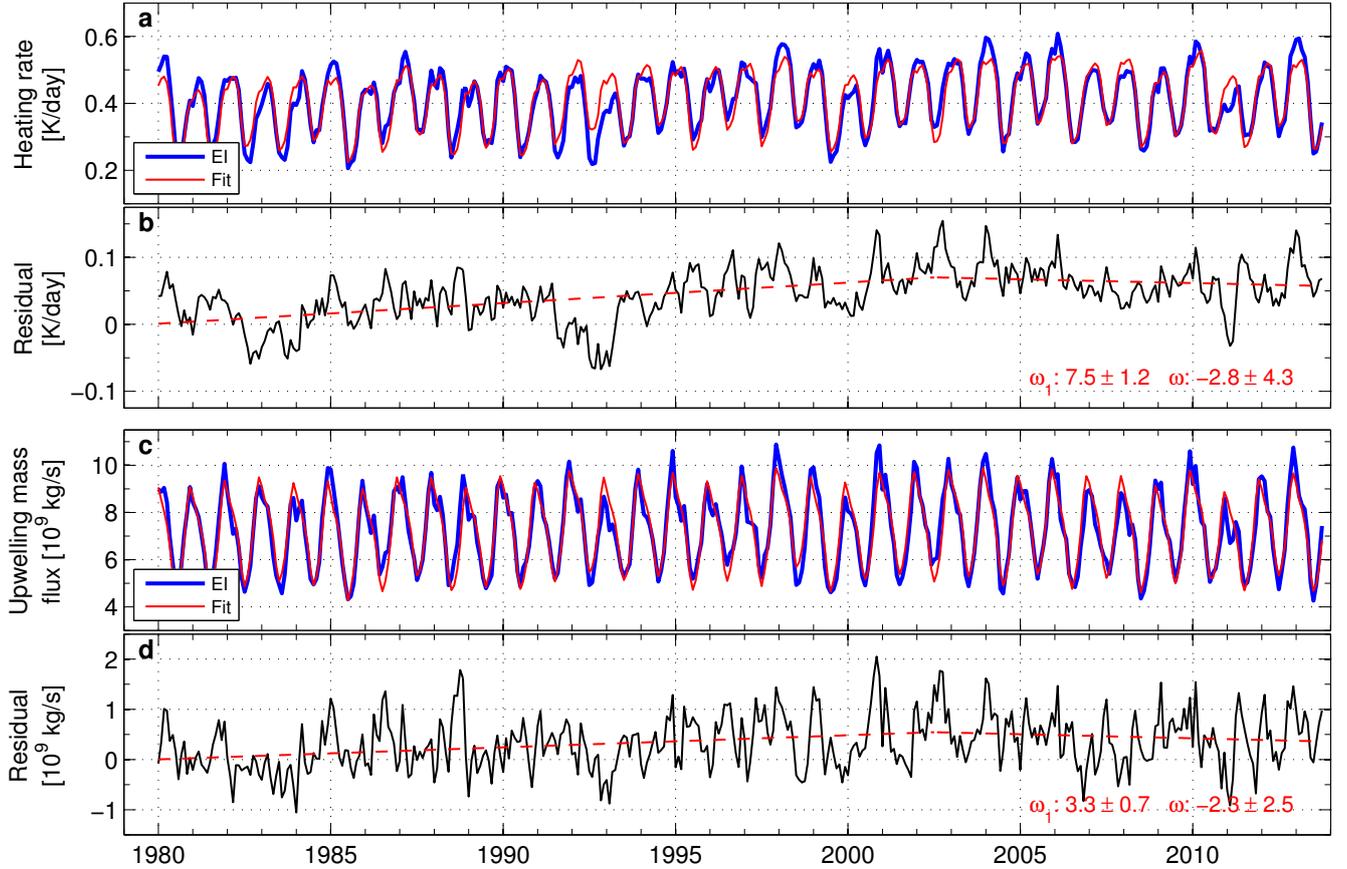


Fig. 4. Regression analysis of EI LS all-sky heating rate (17–21 km; **a, b**) and upwelling mass flux (70 hPa; **c, d**). Setup identical to Fig. 3 otherwise.

3.4–3.7° per decade, which is in good agreement with the estimated south-shift of 5° on the basis of MIPAS O₃ observations (2002–2012; Eckert et al., 2014).

Table 2. Average turn-around latitude per level and hemisphere and corresponding linear trend coefficients obtained from the regression analysis (Fig. 6). The unit of the trends is degree per decade.

p [hPa]	ϕ [°]	ω ₁	ω ₂	ω
NH 100	21.9	1.81 ± 0.15	-6.58 ± 0.73	-4.77 ± 0.56
SH 100	18.6	0.90 ± 0.13	-2.31 ± 0.62	-1.41 ± 0.48
NH 70	31.4	0.16 ± 0.10	0.58 ± 0.47	0.74 ± 0.37
SH 70	29.8	0.44 ± 0.13	-2.01 ± 0.63	-1.56 ± 0.49
NH 30	37.5	1.89 ± 0.34	-5.14 ± 1.60	-3.25 ± 1.24
SH 30	32.1	0.69 ± 0.23	-0.29 ± 1.11	0.41 ± 0.86

4 Discussion and conclusions

In this study, we compile observations and model simulations of tropical LS O₃ from 1980–2013. We find

negative trends of O₃ both in observation and model before 2002, consistent with earlier studies (e.g., Randel and Thompson, 2011). These trends in O₃ are accompanied by an increase of tropical upwelling found in the EI dataset based upon diabatic heating calculation, confined to the shallow branch of the BDC (70–30 hPa). This is also in agreement with previous modelling studies, which predict an increase of upwelling (e.g., Butchart et al., 2010). However, we also find an unexpected hiatus of the negative trend in LS O₃ after 2002, mirrored by a similar feature in the behaviour of the vertical transport in the EI dataset. Our analysis shows that the acceleration of the shallow branch has ceased; at the same time the strength of the transition branch (100–70 hPa) increases after about 2002, in agreement with the findings of Bönisch et al. (2011). The deep branch (<30 hPa) does not show any significant changes. We further see an expansion of the tropical pipe before 2002, followed by a narrowing afterwards. This effect is most pronounced at 100 and 30 hPa, where the upwelling is shifted also northwards before and southwards after 2002.

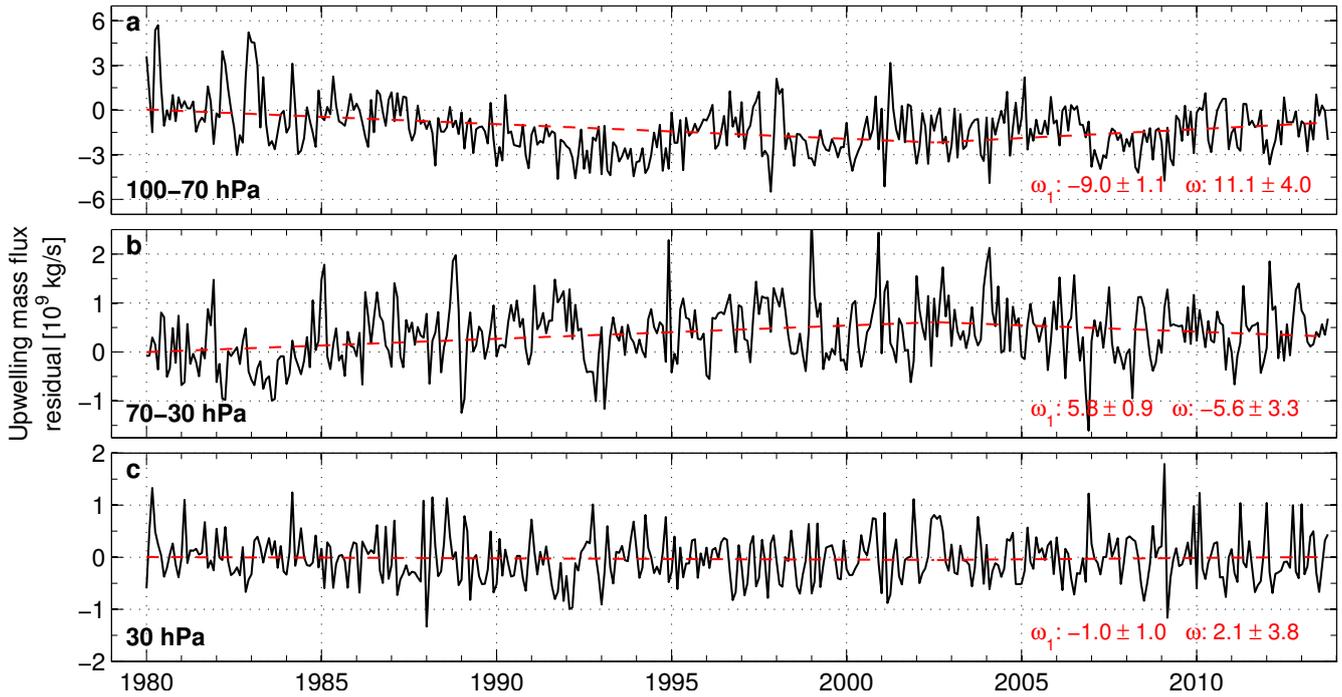


Fig. 5. Fit residuals (excluding linear terms) of EI upwelling mass fluxes in different BDC branches (**a** 100–70 hPa; **b** 70–30 hPa; **c** 30 hPa). The dashed red lines depict the resulting linear trends before and after 2002 with the fit coefficients in red font (unit: % per decade).

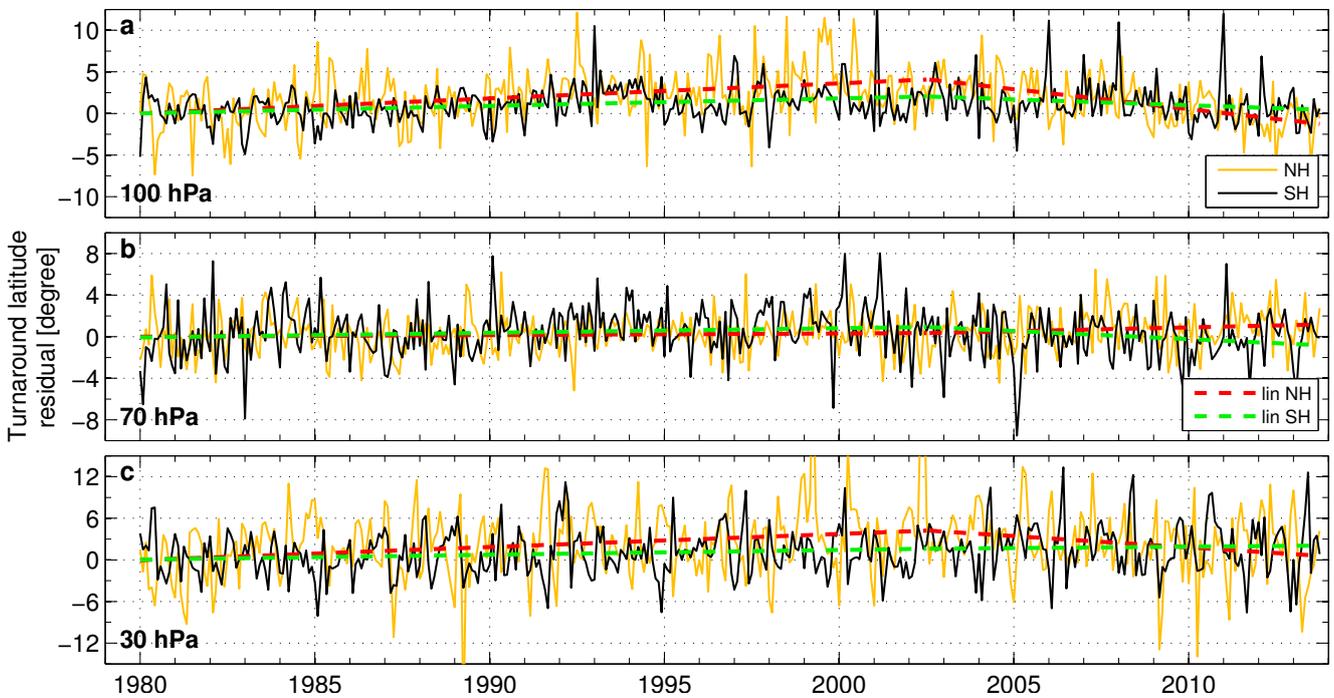


Fig. 6. Fit residuals (excluding linear terms) of turnaround latitudes determined from EI upwelling mass fluxes at different pressure levels (**a** 100 hPa; **b** 70 hPa; **c** 30 hPa). The black and orange lines denote the fit residuals whereas the red and green dashed lines depict the resulting linear trends in the northern and southern hemisphere (NH/SH), respectively.

Taking into account the sensitivity of LS O_3 to vertical transport, there are strong indications that the observed trend-change in O_3 is primarily a consequence of the simultaneous trend-change in tropical upwelling. As the analysed partial columns are dominated by the upper altitudes due to the steep vertical gradient in tropical O_3 , they are particularly sensitive to changes in the shallow branch of the BDC between 70–30 hPa. This hypothesis is corroborated by significant anti-correlation between LS O_3 anomalies with either heating rates (-0.83), or 70 hPa upward mass flux anomalies (-0.55). However, we cannot rule out a possible impact of in-mixing, which is not covered by our analysis. Further, the detected southward shift of upwelling after 2002 in the EI dataset is consistent with the hypothesis of Eckert et al. (2014), who demonstrated that the recently observed O_3 trends could be partly explained by a south-shift of the tropical mixing barriers. This is not necessarily a contradiction to our conclusion, as both upwelling and location of mixing barriers are impacted by the same dynamical processes (Li et al., 2010).

The cause of these changes is currently unknown. One plausible explanation could be the unexpected La-Niña-like cooling of the equatorial Eastern Pacific since the beginning of the 21st century (Meehl et al., 2011). The cause of this cooling is not yet fully understood and currently debated (Chen and Tung, 2014). However, it has a significant impact on global surface temperatures (Kosaka and Xie, 2013) and ultimately, by dynamical coupling, on tropical upwelling (Oman et al., 2009; Butchart et al., 2010; Garny et al., 2011). Recent studies describe the associated circulation changes (England et al., 2014) and their impact on tropospheric O_3 (Lin et al., 2014). In contrast to current unconstrained CCM, which generally do not predict this exceptional cooling of the equatorial Eastern Pacific surface (Kosaka and Xie, 2013; England et al., 2014), this feature can be clearly observed in the data-assimilated EI dataset (Fig. 7). This hypothesis is further corroborated by significant (anti-) correlation between tropical surface temperatures and LS upwelling/ O_3 mixing ratios, which is most prominent in the LS (Fig. 8; Hardiman et al., 2007). This particular relationship between ocean and atmosphere must be investigated in more detail, as it is likely that the accuracy of our predictions of future BDC development and its consequences for stratospheric O_3 critically depends on our understanding of this interaction.

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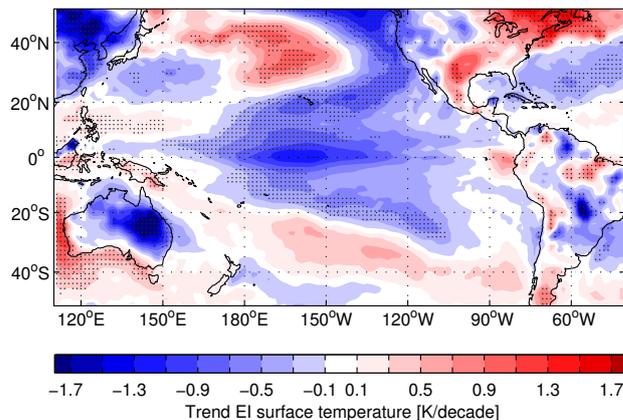


Fig. 7. Linear trends of EI surface temperature from 2002–2013. Stippling indicates where the trend exceeds the 95% confidence threshold. Setup adapted from Kosaka and Xie (2013).

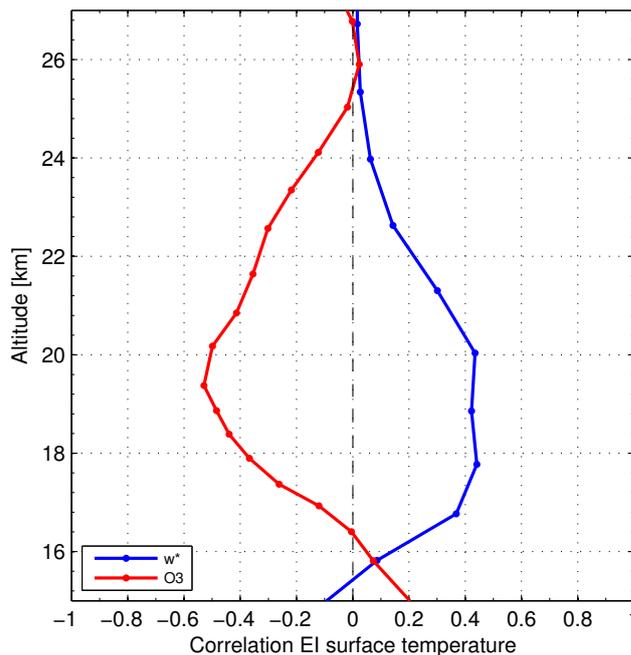


Fig. 8. Correlation of EI surface temperature anomalies with anomalies of w^* calculated from EI all-sky heating rates and modelled O_3 mixing ratios in the tropics (20°N–20°S). The corresponding timeseries range from 01-1980 to 10-2013.

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