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Dear Editor.

Attached to this document you find a point-by-point response to the questions and comments arised from the reviewers and an updated version of our manuscript. The major changes (i.e. anything but trivial changes like typos etc.) are marked with a black box and bold font.

Thank you for your effort.

Best regards,

Jan Aschmann

We thank the referee for her/his positive and thoughtful comments to improve the manuscript. In the following, the original remarks of the referee are in *italics*.

1) Structural change of BDC:

My first major comment concerns a potential change in the structure of the BDC after the year 2000, as has been recently discussed by Boenisch et al. (2011), who argued for an increase in only the shallow circulation branch. The transition region between the shallow and deep branches of the BDC is located exactly within the 17–21 km layer considered by Aschmann et al. Hence, from the perspective of such a structural change the weakening tropical upward mass flux across 70 hPa after the year 2002 (see Fig. 4) is not necessarily in contradiction to a continued increase in tropical upwelling, if only the shallow circulation branch is strengthening. Further, if the ozone partial column between 17–21 km is dominated by the upper levels (what is very likely due to the strong increase of tropical ozone with altitude), the related ozone response could likely show the observed decrease before 2002 and a hiatus of this decrease afterwards.

Therefore, linking the trends of ozone partial columns as well as of mass flux across 70 hPa to changes in tropical upwelling in general, seems oversimplified to me. I think it could be beneficial for this study to separate the effects of changes in the shallow and deep circulation branches, to clarify whether a general weakening or a structural change in the circulation has occurred. A simple step into this direction could be to consider different levels and to infer the shallow branch from the difference, similar to the study by Lin and Fu (2013).

Thank you for this helpful suggestion. We adopted the approach of Lin and Fu (2013) and analysed the changes in the different branches of the BDC. An additional figure and discussion has been added to the revised manuscript. According to the definition by Lin and Fu (2013), we find that the acceleration of the shallow branch of the BDC (70–30 hPa) has stopped after about 2002. On the other hand, we find an increase of upwelling in the transition branch (100–70 hPa) after 2002, which is consistent with the findings of Boenisch et al. (2011).

2) Impact of mixing on tropical ozone:

My second major comment addresses the impact of horizontal mixing with mid-latitudes across the subtropical mixing barrier on tropical ozone. Such mixing transports ozone-rich mid-latitude air into the tropics and is not a secondary effect (P9953/L15), as first shown by Konopka et al. (2009). The controversial debate about the impact of such in-mixing has been reconciled in the recent paper by Abalos et al. (2013). At levels of about 1718 km about 50% of tropical ozone during summer has been in-mixed from mid-latitudes, and even at 21 km this fraction amounts to 20(Abalos et al., 2013, Fig. 4b), although the mixing itself becomes very weak above the TTL.

Consequently, a slight trend in mixing, even if occurring below the considered layer, could likely have a significant impact on the partial ozone column between 17–21 km. Hence, linking the tropical ozone trend to upwelling without taking mixing into account seems problematic to me. If the mixing effect could *be diagnosed within the model, this could proof the dominant role of upwelling in causing the ozone trend - or shed more light on a potential additional role of mixing. At least a critical discussion is needed to make the reader aware of this source of uncertainty in the analysis.*

The impact of in-mixing on LS O3 is an important point. To address this question and elucidate the role of ODS phase-out for the observed trend-change we conducted a sensitivity simulation with identical setup but with ODS emissions fixed to 1980 values. Consequently, the O3 field evolves significantly different compared to base run. However, in contrast to higher latitudes we find only minor changes of LS O3 in the tropics. More important we see a similar trend-change as in the standard simulation. This result is a strong argument that the observed trend-change is not primarily related to ODS-involved chemistry. Although we don't assess in-mixing directly, the small O3 changes in the tropical LS suggest that in-mixing is not the main driver of the observed trend-change. We added a discussion of these findings in the revised manuscript.

P9955, L11:
Is there a particular reason to use all-sky and not the total diabatic heating rate from ERA-Interim to drive the model vertical transport? The total heating rate would include all diabatic processes, although above about 100 hPa both heating rates are very similar.

No, there is no particular reason for the usage of all-sky vs. total heating rates (aside from availability issues). We made sensitivity calculations of w^* with total heating rates and found no significant differences in the tropical LS, which confirms the reviewers statement that the remaining diabatic processes (mainly latent heat release) are not relevant at the top of the tropopause and above.

P9959, L1ff:
Can effects related to chemistry totally be neglected? The authors state that neither process is sufficient to explain a short-term change. It would be good to present results supporting this statement. Further, horizontal mixing with the extratropics (see comment 2) provides a pathway for chemistry related changes in extratropical ozone to affect tropical ozone. Again, these issues have to be critically discussed.

We apologise for the unclear phrasing, we would like to point out that Meul et al. (2014) do not find indications for short-term chemical impacts on LS O3, only on longer time scales. For the role of horizontal mixing please refer to our answer above.

Fig. 4a:
Wouldn't it be better to show also the tropical heating rate as an average between turn-around latitudes, analogously to the mass flux case, and not as an 20S–20N average?

We agree that using turnaround latitudes appears to be more consistent to the rest of the figure, however, we would like to keep the 20S–20N margin for the following reasons: First we would like to retain the direct comparability to the LS O3 columns shown before, which have also been averaged between 20S–20N. Secondly, the presented heating rates are an average value over the vertical range of 17–21 km, therefore there is no clearly defined turnaround latitude, which is representative for the whole altitude range.

Technical corrections:
P9953, L5: Age of air changes indicate...
P9957, L23: ... illustrate ...
Fig. 4 caption: ... LS all-sky heating rate ...

We have fixed all of the editorial comments above and thank again the referee for the careful reading.

We thank the referee for her/his helpful comments and the invested effort. In the following, the original remarks of the referee are in *italics*.

p9953 l25 ff: Recently strong indication has been found that the large SCIAMACHY ozone trends which are in disagreement with the other satellite observations mentioned above may be due to an instrument drift (EGU 2014-5678, SI2N assessment of vertical ozone trends: Stability of limb/occultation data records over 1984-2013 against ground- based networks Daan Hubert). This drift was detected by Daan Hubert in the ESA data product, not the Bremen data product, but since drifts are often an instrumental, not a retrieval problem, one might suspect that also the Bremen data may be affected by a drift. In the discussion paper drift issues are not discussed at all. During the post- 2002 period nearly all trend information comes from SCIAMACHY data. Admittedly the SAGE data are available until 2005 but in these no inflexion is visible between 2000 and 2005 in Fig.1. Thus the observational evidence for the inflexion (at least with respect to satellite data) relies fully on SCIAMACHY. This all indicates that there is some non- negligible risk that a major part of the explained phenomenon could be an artefact. At the very least the risk of a potential drift should be critically discussed in the paper.

We agree that instrumental drifts are always a potential danger for trend calculations, in particular for relatively short timeseries as discussed in this paper. However, there are strong indications that the existence of the LS O3 trend-change is no instrumental artefact. Firstly, there are the SHADOZ data, which show essentially the same behaviour as SCIAMACHY, although admittedly the horizontal sampling provided by the sonde stations is poor compared to a satellite instrument. Perhaps more convincing is the fact that several satellite-borne instruments show positive trends in the LS, for example OSIRIS, MLS (Gebhardt et al., 2014), MIPAS (Eckert et al., 2014) and GOMOS (Kyrölä et al., 2013). Despite the considerable spread in the determined trends all mentioned instruments show no further decrease of tropical LS O3 during the last decade, as one would expect with continuous acceleration of tropical upwelling. We make this point more clear in the revised manuscript.

p9955 Eq 1: It is not clear what X_{1t} and X_{2t} are. This, however, is crucial to understand how Eq 1 can produce an inflexion point. I suspect that X_{2t} is zero before 2002 but this must be explained.

We added the definitions of the trend functions to the revised manuscript.

p9956 Eq 2-4 and p9957 l1: According to the Reinsel 2002 paper, their Eq. 1 and subsequent text, it seems to me that σ_N should be the standard deviation of the fit residuals rather than the standard error. Please check.

We apologise, you are absolutely correct. We altered the manuscript accordingly.

p9957 l8: No error bars or covariance matrices are shown or discussed but χ^2 values are presented. How are these calculated?

Again we have to apologise for unclear phrasing. We made no χ^2 test but we minimised χ^2, i.e. the sum of the squared difference between the (observational/model) data and the regression as in Jones et al. (2009). This will be fixed in the revised manuscript.

p9958 l19: The SHADOZ data should be shown in one of the figures.

Done. We've added two additional panels to Fig. 3.

p9959 1st par: However, neither process is sufficient to explain a short-term trend change. This statement needs justification. Some quantitative estimates are needed on what the competing processes can do. Have the authors ruled out that temperature trends may affect the column density but not VMR?

Please refer to our answer to referee 1 who made a similar statement. Regarding the impact of temperature trends we can confirm that the observed LS O3 trend-change is visible both in partial columns as in VMR (e.g., Gebhardt et al., 2014).

p9961 l8: The last sentence is misleading: I do not challenge that the ocean-atmosphere interaction is important to predict the BDC but I do not quite see that this emerges from the findings of this paper. I appreciate that the findings of the paper are discussed in the context of existing work, but the phrasing In conclusion, the accuracy... at a very prominent place in the paper (the last statement!) does not seem appropriate to me. Please distinguish clearly what is common knowledge and what is the immediate result of your study.

We agree that this sentence should be rephrased. We reworked the conclusions in the revised manuscript and added additional discussion about ocean-atmosphere interaction.

Minor technical and language issues:
p9954 l10 have been omitted (plural)

Fixed. Thank you.

We thank the referee for her/his interesting thoughts and suggestions. In the following, the original remarks of the referee are in *italics*.

*Aschmann et al. present an analysis of tropical lower stratospheric ozone for the period 1980 to 2013 using observations, and results from a chemical transport model driven with ERA-Interim winds and radiative heating rates. The main result shown is that the ERA-Interim radiative heating rates seem to have a trend-change around the year 2002, which then forces the tropical lower stratospheric ozone in the chemical transport model The model forced with the ERA-Interim data overestimates the observed ozone trends by a factor 2 for the pre-2002 period. The authors note that the (dis-)agreement is within the uncertainty of previous studies, but seem not concerned that the difference is a full factor 2 (i.e. -8.1% versus -3.9%). The paper provides no discussion of uncertainties in ERA-Interim heating rates - the single most critical quantity for the results presented. Given that the ERA-Interim radiative heating rates are known to give biased responses to dynamical forcing because of the use of an ozone climatology - notably the quantity central to this paper! - this is a major omission. Indeed, this problem could be the cause for the model overestimate of trends (as ozone mixing ratio in the tropical lower stratosphere decrease with an increase in upwelling, a radiative transfer calculation such as done in ERA-Interim that keeps ozone fixed and only responds to the temperature change with *overestimate* the change in radiative heating, and hence also in w^*). The topic that the paper addresses is very interesting and important, and should be addressed with rigor - such important issues concerning the methodology cannot be just ignored.*

We thank the referee for highlighting an important issue. EI heating rates play indeed a central role in our CTM simulations. However, we do not agree with the assertion that "The main result shown is that the ERA-Interim radiative heating rates seem to have a trend-change around the year 2002...". Our main focus is the fact that both observations as well as our CTM simulations show independently that there is a significant trend-change of tropical LS O3 around 2002, which ends the marked decrease in the previous decades. Given the generally good agreement between model and observations, and taking into account that EI heating rates (and the derived w^*) show a similar behaviour, we conclude that a change in tropical upwelling has occurred. We agree that the usage of a fixed O3 climatology for calculating the heating rates in Era-Interim (Seviour et al., 2012) could lead to biased responses dynamical forcing and is probably responsible for the observed overestimation of vertical transport (e.g., Ploeger et al., 2012). Nevertheless, the diabatic representation of vertical transport seems to give more realistic results in the stratosphere compared to the kinematic approach (e.g., Ploeger et al., 2011; Diallo et al., 2012). Regarding the difference between the modelled and observed pre-2002 trend, it is clear that the primary cause is the high-bias of the model relative to SAGE II in the pre-Pinatubo years (<1991) as there is remarkably good agreement between model and observations in the later years. In particular, both datasets are consistent around the critical inflexion year (2002), the central point of our study. To address the referee's concerns, we extended the discussion about the model bias and the impact of possible EI heating rate deficiencies in the revised manuscript (Sect. 3.1 and 3.2).

Further, I might challenge the authors presumption that the BD circulation was increasing before the year 2002 - the evidence for this presented in the literature is weak. I am familiar with the data and don't deny that there are indications in this direction, but what we need is firmer evidence, and this paper does not deliver this. As said above, the "observed" ozone has a much smaller trend than the model. Indeed, it could also be that the fact that the two have the same sign is coincidence (there is a fifty-fifty chance that an error in the ERA-Interim assimilation system induces a positive/negative trend). I would think that were it not for the CCM results that show an increase in the BD circulation, we might look differently at the observations (Figure 2).

We agree that it is challenging to pinpoint changes of the BDC given its complexity and inaccessibility to direct measurements. We further concur that more analysis and in particular more observational data is definitely needed to get a clearer picture. However, from the reported observations and simulation studies there are more indications for an acceleration of the (lower-branch) BDC at least in the last decades of the 20th century than against this hypothesis ("... authors presumption that the BD circulation was increasing before the year 2002"). On this basis, this study does not try to "prove" an acceleration of the BDC, as we are only show well-known data. Our focus is on the consistency of recent observations with a postulated acceleration of the BDC (e.g., Butchart et al., 2010; Randel and Thompson, 2011), which has not been discussed in detail in present literature so far.

I think the paper needs major revisions, and would encourage the authors to substantially strengthen the discussion of the following aspects: (i) What do ERA-Interim radiative heating rates represent, what are sources of biases, and how do they potentially affect the model results? (ii) Make it clear (also in abstract) that the key to the model result is the ERA-Interim heating rate - no more, no less. (iii) Fidelity of ozone observations - in particular, I would like to see a proper error calculation for the impact of merging data from different data sources. That is, metrics should be provided how well different observations agree in periods of overlap, the uncertainty in the offset, and how that affects the trend estimates. (iv) The link to the warming hiatus in the troposphere is really not established at all in this paper. While we all "hypothesize" (Line 21) that this is the case, we also expect that a paper that discusses this aspect provides the mechanism and evidence.

Regarding points (i) and (ii): As stated above, we have strengthened the discussion about the impact of Era-Interim heating rates.
Regarding point (iii): Thank you for this suggestion. We agree that in principle a complete error calculation would be beneficial for the analysis. However, in practice, this step is often omitted as the most relevant potential problems such as instrumental drifts are difficult to assess and require a dedicated analysis on its own (e.g., Jones et al., 2009; Rahpoe et al., 2013; Eckert et al., 2014). Therefore we follow a similar approach as in previous studies (e.g., Randel and Thompson, 2011) and briefly discuss offset and correlation in the overlap period (p. 9958 l. 3f), showing the data in Fig. 3. Regarding the fidelity of the O3 data in general: We present two sources for the post-2002 period (SCIAMACHY and SHADOZ), which both show no significant negative trend as one would

expect for a continued acceleration of the BDC. This feature is also present in the records of other instruments as pointed out in the paper (p. 9958, l. 21f). This agreement prompts confidence that the observed trend-change in LS O3 is indeed real and no instrumental artefact.
Regarding point (iv): We agree that the paper does not establish a link between the SST warming hiatus and tropical upwelling. A detailed analysis is way beyond the scope of this paper, however, we would like to point out that there are good arguments that the observed trend-change in O3/upwelling is possibly related to this phenomenon. Although the connection between SST and upwelling is well established in the literature, the possible impact of the recent cooling of the Eastern Pacific on LS O3 has not been discussed so far, to the best of our knowledge. In either case we agree that the conclusion section needs rephrasing, which will be done for the revised version.

P1/L41: They are consistent with, but not really proof that the BD is accelerating.

We agree.

P1/L63: Meridional mixing is probably not a secondary effect, but of similar importance. To the best of my knowledge no study has been conducted to quantify a possible trend in the mixing contribution, so I don't think that you can rule it out without any analysis of the problem.

The first referee raises a similar concern. Therefore we conducted an additional sensitivity simulation to assess the impact of in-mixing. Details are given in the corresponding reply; furthermore we added an additional paragraph to Sect. 3.1. The bottom line is that in-mixing does indeed impact the trends, but its contribution is small compared to changes in vertical transport.

P2/L35: See above - The absolute offset mentioned is less critical than the degree of agreement during the overlap period; please provide a careful error propagation calculation.

Please refer to our reply above (re. (iv)).

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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. Our model reproduces the observed tropical lower stratosphere ozone record, showing a significant decrease in the early period followed by a statistically robust trend-change after 2002. [redacted]**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 during the last decade, which appears to be the primary cause for the observed trend-change in ozone.**

1 Introduction

The issue of whether the large-scale Brewer-Dobson Circulation (BDC) has strengthened in the recent past, as a result 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 (SF_6) 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 (O_3) 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 (O_2) photo-dissociation and a steady influx of O_3-poor tropospheric air from below (Avallone 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 O_3 mixing ratios (Konopka et al., 2009; Ploeger et al., 2012; Abalos et al., 2013), with the largest impact during boreal summer directly above the tropopause ($\approx$ 380 K/17 km). Several studies have reported a negative trend of O_3 in the tropical LS in the range

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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[1] 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. × 5°lat. × 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 considerable 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.

[1] First reported at the Quadrennial Ozone Symposium 2012 Toronto, August 27–31 2012 and published in Gebhardt et al. (2014)

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. × 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:

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

[redacted]**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.,

Table 1. Geolocation, temporal coverage and average LS O_3 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
Watukosek-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

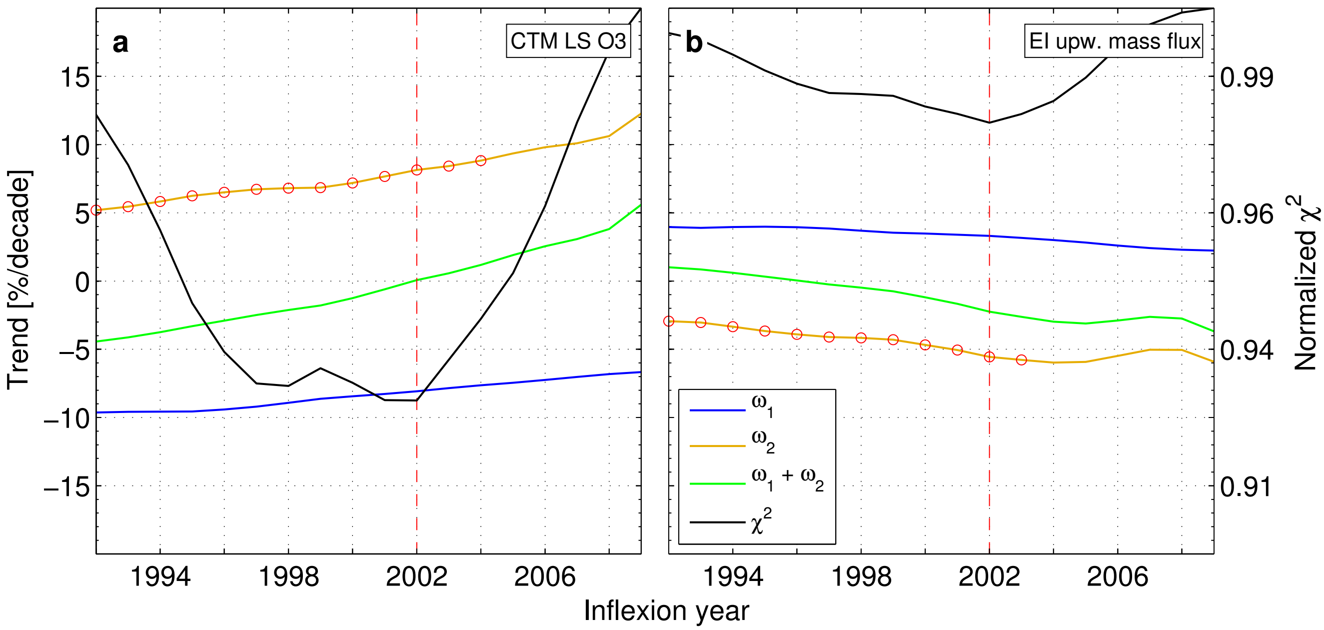

Fig. 1. The dependence of the linear fit parameters ω_1, ω_2 and ω ($\omega_1 + \omega_2$) on the inflexion year T_0 is shown for the regression of modelled tropical LS O_3 column (**a**) and EI upward mass flux at 70 hPa (**b**). Red circles denote the years where the trend-change (ω_2) exceeds the 95% confidence threshold. The black lines are the normalised χ^2 values of the fit residuals.

2014), our regression model uses two linear components to 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 El 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 Center[2], the ENSO proxy is represented by the Multivariate ENSO Index (MEI) from the NOAA Earth System Research Laboratory[3] (Wolter and Timlin, 2011) lagged by two months and the solar cycle by the Bremen composite Mg II index[4] (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 O_3 timeseries (e.g., Reinsel et al., 2002; Jones et al., 2009; Sioris et al., 2014), the corresponding standard deviations for the trend components

[2]www.cpc.ncep.noaa.gov/data/indices/

[3]www.esrl.noaa.gov/psd/enso/mei/

[4]www.iup.uni-bremen.de/gome/solar/MgII_composite.dat

are given by

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

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

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

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: 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 ± 0.9 % per decade (ω_1) in the pre-2002 period and 0.1 ± 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. $\omega_2 > 2\sigma_\omega$).

[redacted]**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 ± 0.5 % per decade (ω_1) for the pre-2002 period, consistent with the range of -(3–6) % per decade given by earlier studies (Fig. 3c, d; Randel and Thompson, 2011; Sioris et al., 2014; Bourassa et al., 2014). [redacted]**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 is possibly explained by a spin-up effect of the model stemming from the usage of replicated input data for the initialisation phase (Sect. 2.2). Another point could be the overestimation of the vertical transport velocity based on diabatic heating rates in the EI dataset (Ploeger et al., 2012; Diallo et al., 2012), which is discussed in more detail in Sect. 3.2. After 2002, the agreement between model and observations improves considerably and the trend is 0.5 ± 1.5 % per decade (ω), yielding a statistically significant trend-change of 4.4 % per decade (ω_2) for the SAGE II-SCIAMACHY dataset**. We obtain similar values (-3.6 ± 0.5, 0.4 ± 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 those studies, which focus solely on the most recent observational record of O_3, although the differences in utilised regression models and timeseries length make a direct comparison difficult. 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) find a slightly positive trend of 0–1 % per decade in the same region in MIPAS observations (2002–2012). [redacted]**In summary, despite 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 explana-

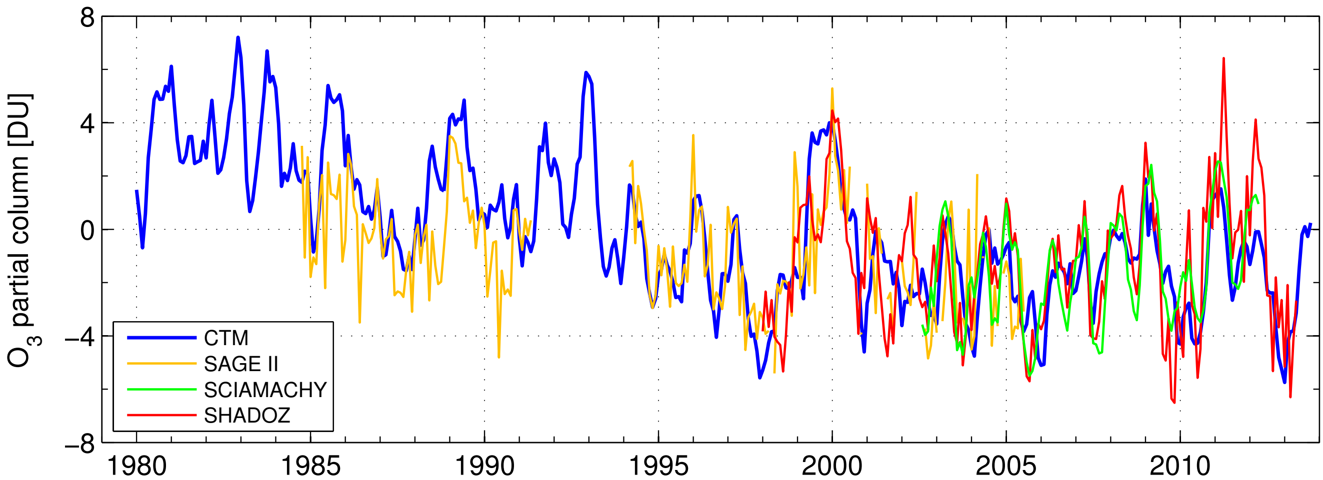

Fig. 2. Observed and simulated tropical (20°N–20°S) LS O_3 partial columns (17–21 km). Anomalies are deviations from the modelled 1980–2013 averages.

tion 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 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. [redacted]**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 %) and we calculate very similar trends (-7.8 ± 0.9, -0.6 ± 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 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).** 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

Some studies point out that the increase of tropical upwelling may be compensated by an, as yet, unexplained weakening or shifting of tropical mixing barriers (Stiller et al., 2012; Eckert et al., 2014). However, it is also possible that the increase of tropical upwelling itself has ceased. To investigate this hypothesis, we analyse tropical upwelling in the EI reanalysis that drives our model. 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. [redacted]**The diabatic representation of vertical transport yields more realistic estimates of stratospheric age of air in comparison to the kinematic approach (Diallo et al., 2012) and is also less dispersive (Ploeger et al., 2011). On the downside, there are indications that EI heating rates overestimate the ascent in the tropics (Ploeger et al., 2012; Diallo et al., 2012). One possible cause for this overestimation could be the usage of a fixed O_3 climatology for calculating the heating rates in the reanalysis (Seviour et al., 2012). Keeping O_3 constant could lead to biased responses to dynamic forcings and thus exaggerating changes in vertical transport. This probably contributes to the differences between model and SAGE II data in the pre-Pinatubo period discussed in Sect. 3.1.**

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 cor-

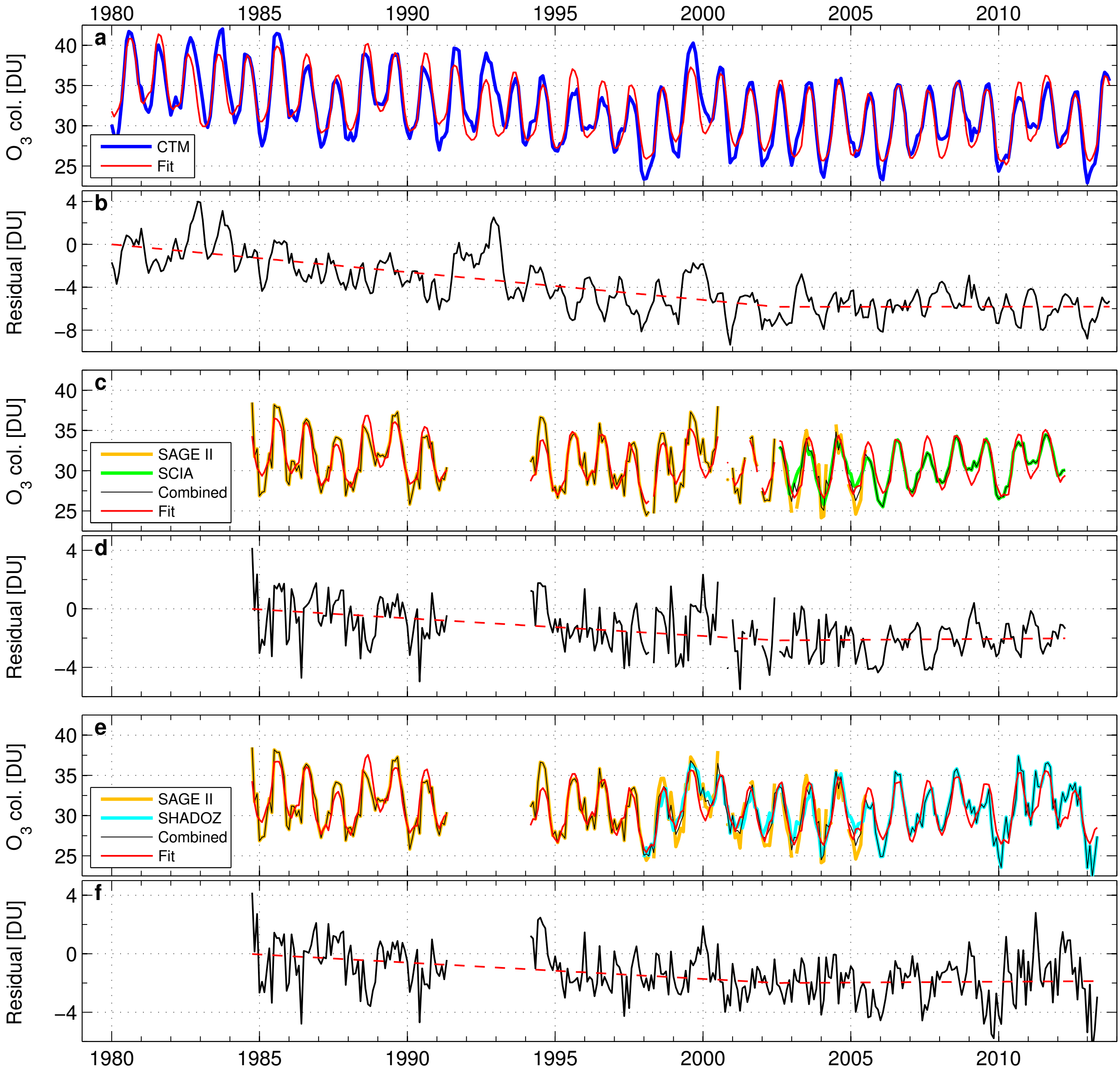

Fig. 3. Regression analysis of observed and simulated O_3 partial columns. Model, combined SAGE II/SCIAMACHY and combined SAGE II/SHADOZ LS O_3 with regression function (**a**, **c**, **e**). Corresponding fit residuals excluding the linear terms (**b**, **d**, **f**). The dashed red lines depict the resulting linear trends before and after 2002.

responding 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 as described in Seviour et al. (2012). 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 $\pm$ 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, reflecting the overestimation of vertical transport mentioned above. After 2002, however, there is a statistically significant trend-change around 2002 leading to a negative trend of -2.3 $\pm$ 2.5 % per decade (ω) mirroring the trend-change in the LS O_3 timeseries. [redacted]**Further insight into structural changes of the BDC can be gained by decomposing the circulation into different branches. Here,**

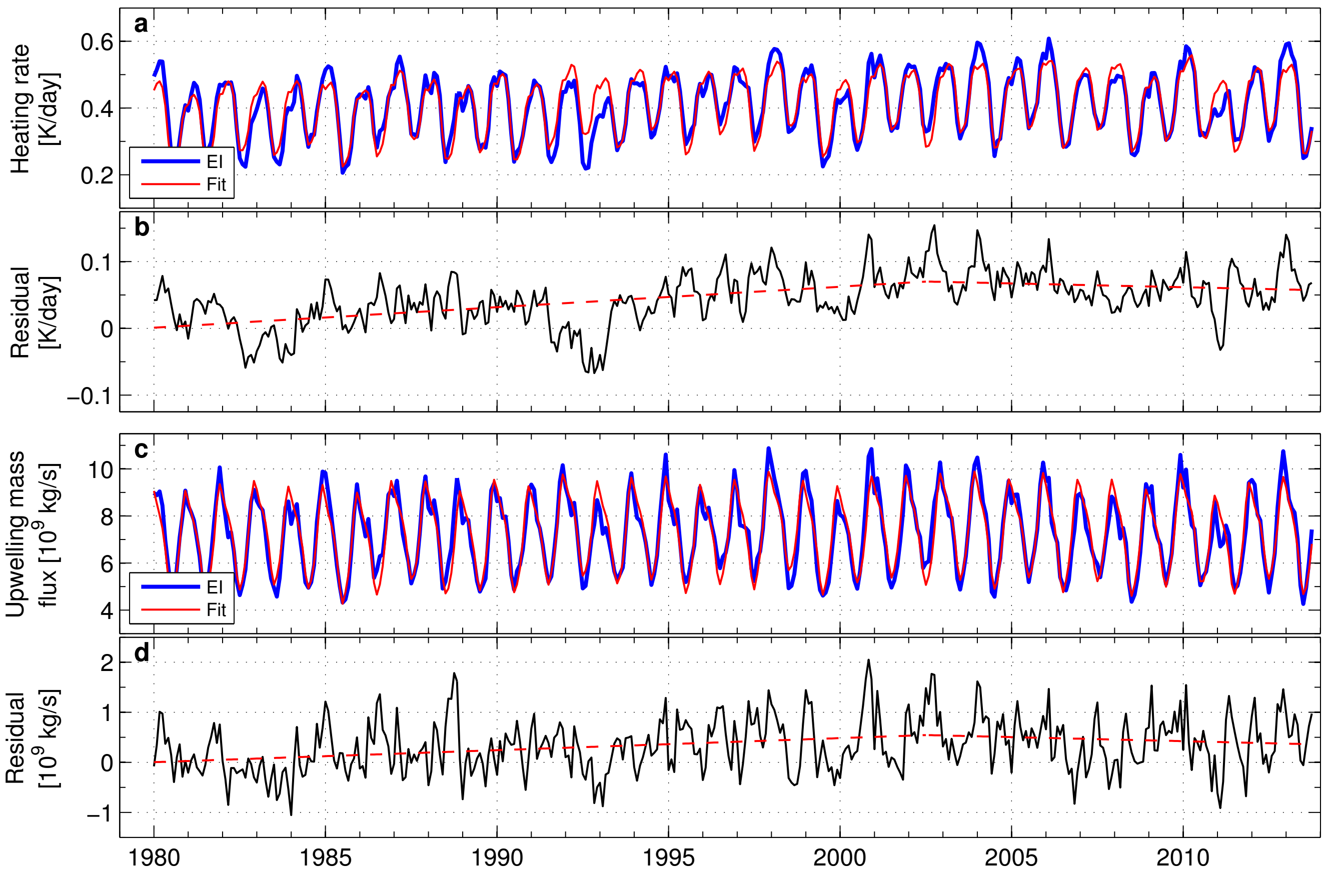

Fig. 4. Regression analysis of EI LS all-sky heating rate (17–21 km; **a, b**) and upwelling mass flux (70 hPa; **c, d**). Setup identical to Fig. 3 otherwise.

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; 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 N_2O and O_3 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.

Taking into account the sensitivity of LS O_3 to vertical transport, we conclude 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

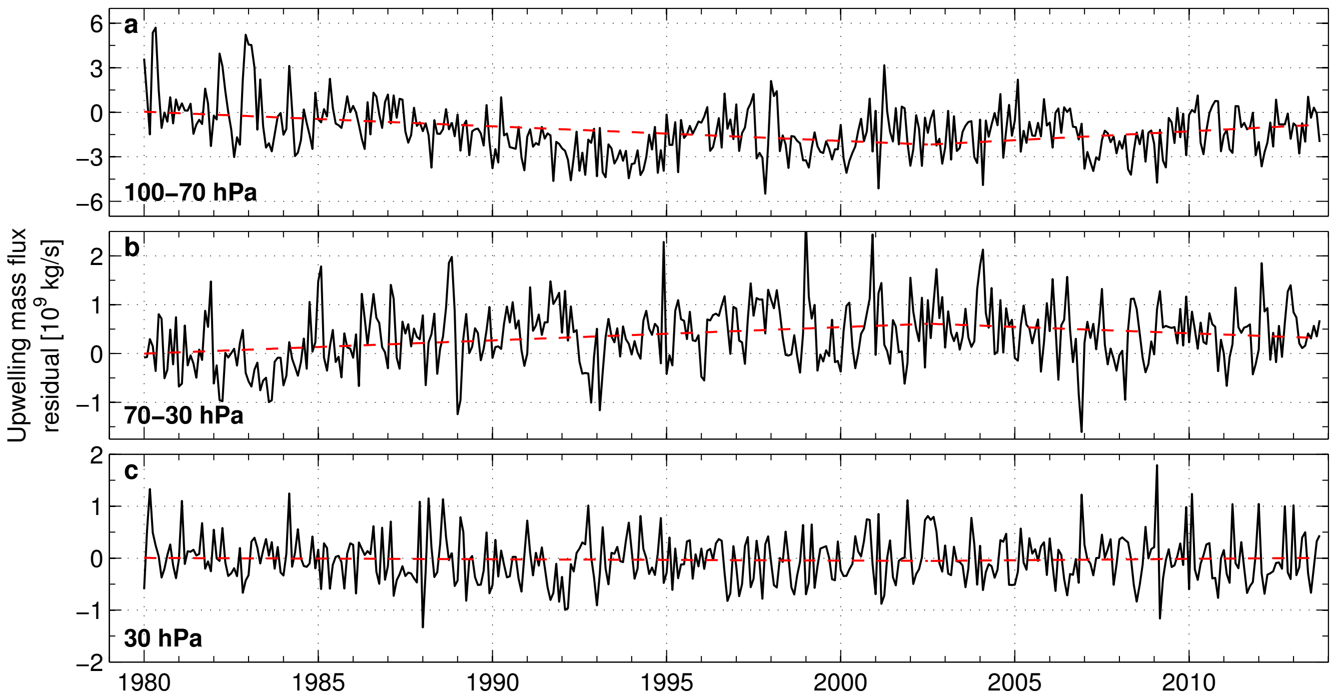

Fig. 5. Fit residuals (excluding linear terms) of EI upwelling mass fluxes in different BDC branches (**a** 100–70 hPa; **b** 70–30 hPa; **c** 30 hPa). The dashed red lines depict the resulting linear trends before and after 2002.

with either heating rates (-0.83), or 70 hPa upward mass flux anomalies (-0.55).

[redacted]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 latter 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 heat uptake by the equatorial Eastern Pacific (Kosaka and Xie, 2013; England et al., 2014), this feature can be clearly observed in the data-assimilated EI dataset (Fig. 6). 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. 7).

4 Conclusions

[redacted]In this study, we compile observations and model simulations of tropical LS O_3 from 1980–2013. We find negative trends of O_3 both in observation and model before 2002, consistent with earlier studies (e.g., Randel and Thompson, 2011). These trends in O_3 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 tropical upwelling (e.g., Butchart et al., 2010). However, we also find an unexpected hiatus of the negative trend in LS O_3 after 2002. As we find a similar feature in the behaviour of the vertical transport in the EI dataset, we conclude that the change in LS O_3 is primarily caused by changes in tropical upwelling. 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. Previous modelling studies suggest a dynamical link between tropical SST and upwelling (e.g., Oman et al., 2009). Consequently, it is possible that the detected trend-change in O_3/upwelling is associated with the recently observed hiatus in tropical SST, caused by an unexpected cooling of the Eastern Pacific (e.g., Meehl et al., 2011). 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.

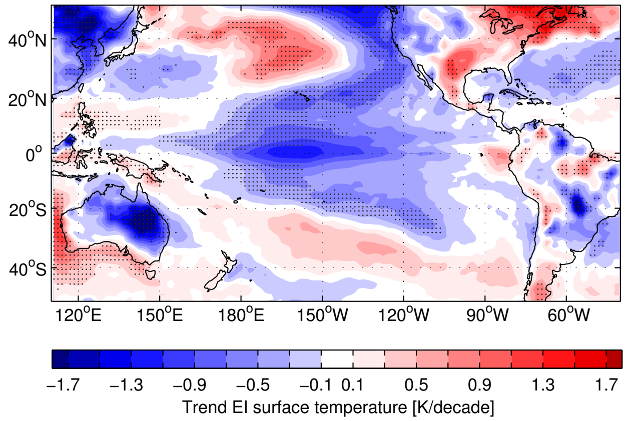

Fig. 6. Linear trends of EI surface temperature from 2002–2013. Stippling indicates where the trend exceeds the 95% confidence threshold. Setup adapted from Kosaka and Xie (2013).

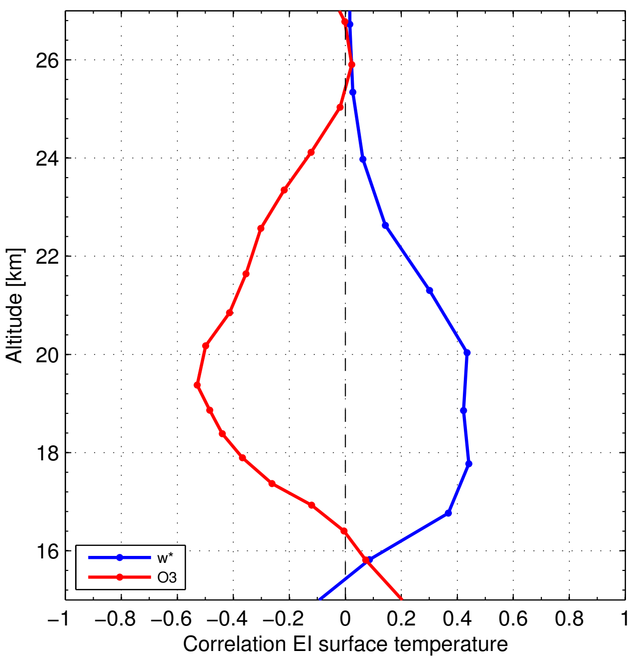

Fig. 7. Correlation of EI surface temperature anomalies with anomalies of w^* calculated from EI all-sky heating rates and modelled O_3 mixing ratios in the tropics (20° N–20° S). The corresponding timeseries range from 01-1980 to 10-2013.

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