An influence of solar spectral variations on radiative forcing of climate

Joanna D. Haigh, Ann R. Winning, Ralf Toumi, Jerald W. Harder

Institutions: Imperial College London, University of Colorado Boulder

Published on: 07 Oct 2010 - Nature (Nature Publishing Group)

Topics: Solar irradiance, Solar cycle, Radiative forcing, Atmosphere and Ozone layer

An influence of solar spectral variations on radiative forcing of climate

Joanna D. Haigh[1], Ann R. Winning[1], Ralf Toumi[1] and Jerald W. Harder[2]

[1] *Blackett Laboratory, Imperial College London, UK.*

[2] *Laboratory for Atmospheric and Space Physics, University of Colorado, Boulder, Colorado, USA*

Correspondence to: J. D. Haigh[1] (Email: j.haigh@imperial.ac.uk)

The thermal structure and composition of the atmosphere is determined fundamentally by the incoming solar irradiance. Radiation at ultraviolet (UV) wavelengths dissociates atmospheric molecules, initiating chains of chemical reactions, specifically those producing stratospheric ozone, and providing the major source of heating for the middle atmosphere, while radiation at visible and near-infrared wavelengths mainly reaches and warms the lower atmosphere and the Earth's surface[1]. Thus the spectral composition of solar radiation is crucial in determining atmospheric structure, as well as surface temperature, and it follows that the response of the atmosphere to variations in solar irradiance depends on the spectrum[2]. Daily measurements of the solar spectrum between 0.2 and 2.4µm, made by the Spectral Irradiance Monitor (SIM) instrument on the SORCE satellite[3] since April 2004, have revealed[4] that over this declining phase of the solar cycle there was a four to six times larger decline in UV than would have been predicted based on previous understanding. This reduction was partially compensated in the total solar output by an increase in radiation at visible

wavelengths. Here we show that these spectral changes appear to have led to significant decline from 2004 to 2007 in stratospheric ozone below 45km altitude, with an increase above. Our results, simulated with a radiative-photochemical model, are consistent with contemporaneous measurements of ozone from the Aura-MLS satellite, although the short time period makes precise attribution to solar effects difficult. We also show that solar radiative forcing of surface climate - based on the SIM data - is out of phase with solar activity. Currently there is insufficient observational evidence to validate the spectral variations observed by SIM, or to fully characterise other solar cycles, but our findings raise the possibility that the effects of solar variability on temperature throughout the atmosphere may be contrary to current expectations.

The peak of the most recent '11-year' solar cycle (identified as number 23) occurred 2000-2002, and from then until about December 2009 the Sun was declining in activity. Fig. 1 shows the difference between 2004 and 2007 in solar spectral irradiance measured by SIM. This is quite unlike that predicted by multi-component empirical models, based on activity indicators such as sunspot number and area, as exemplified by that of Lean[5] (also shown in Fig. 1). The SIM data indicate a factor 4-6 larger decline in UV from 2004 to 2007 and an increase in visible radiation, compared with a small decline in the model. Other empirical models[6,7] show larger amplitude variations in the near UV than the Lean model but none reflect the behaviour apparent in the SIM data. Also shown in Fig. 1, for wavelengths 116-290 nm, are independent measurements made by the SOLSTICE instrument on SORCE. The data from SIM and SOLSTICE both indicate substantially more UV variability than the Lean model. SIM calibration, and instrument comparisons, are discussed in detail by Harder et al[8].

In order to investigate how these very different spectral changes might impact the stratosphere experiments have been carried out using a two-dimensional (latitude-height) radiative-chemical-transport model of the atmosphere[9]. This model includes detailed representations of photochemistry and radiative transfer and has been utilised in many studies involving radiation-chemistry interactions.[10,11] [See Supplementary Information for further details]. This type of model produces realistic simulations of the upper stratosphere (above about 25km) but is less reliable at lower altitudes where photochemical time constants are longer and a more accurate representation of transport processes is required. The results below come from four model runs using solar spectra derived from the SIM measurements (with SOLSTICE data for wavelengths less than 200 nm) and those produced by the Lean model, each for both 2004 and 2007.

In Fig. 2 are presented latitude-height maps of the difference between 2004 and 2007 in December ozone concentrations. The Lean spectral data produce a broad structure of greater ozone concentrations, in 2004 relative to 2007, with maximum values of around 0.8% near 40km, while the SIM data produce a peak enhancement of over 2% in low latitudes around 35km along with significant reductions above 45km. The predicted temperature differences (Supplementary Fig. 1) are also very different: with the Lean dataset showing temperatures 0.3-0.4 K greater in 2004 relative to 2007 at the top of the model domain while the SIM dataset produces a peak warming of 1.8K at the summer polar stratopause. These are qualitatively similar, but about 50% larger than, those estimated by Cahalan et al[12] with an idealised forcing in a full climate model, possibly due to the broader spectral resolution imposed and the lack of ozone-temperature feedback in that model version.

The very different scenarios produced by the two spectral datasets suggest they might be distinguishable in observational records. A multiple regression analysis has been carried out of EOS Aura-MLS deseasonalised monthly mean ozone data. Four regression indices were used: a constant, 2 orthogonal indices representing the quasi-biennial oscillation (QBO, which dominates ozone variability in the tropical stratosphere) [13] and a solar index constructed from SIM data integrated over 200-400nm. Motivated by the model results (Fig. 2) two spatial regions were chosen, both spanning the tropics, one at altitude 10-6.8 hPa, where the model predicts the largest difference 2004-2007, and one, 0.68-0.32 hPa, where the model shows largest negative values. Fig. 3 shows the raw data and the fits reconstructed from the four regression components; it also shows (in red) the derived solar component which is statistically significant at >95%/>99% at the upper/lower levels (see Supplementary Information).

Over the period from the late 1970s to the late 1990s tropical ozone at altitudes 35-50 km decreased by about 9%[13] in response to increasing concentrations of active chlorine species. Since about 2000, however, the trend in chlorine has reversed and ozone has stopped declining. Stratospheric cooling by greenhouse gases has likely also contributed to the ozone trend reversal by slowing the chemical reactions which destroy it[14]. Over the short period of the present study it is not possible to differentiate statistically these factors from each other, or from any solar influence. Nevertheless, it seems likely that the Sun is important in the apparent decrease in ozone below 45 km from 2004 to 2007. The change in sign near 45 km is also more consistent with the modelled response to the SIM spectral variations than to the Lean spectra. Previous analyses[13,15] of the solar signal in ozone, averaged over approximately 2.5 solar cycles (1979 to 2005 or 2003), have not shown this structure. This suggests that the declining phase of solar cycle 23 is behaving differently, or possibly that the solar cycle exhibits different behaviours during its ascending and descending phases.

In order to understand the different spatial structures, and magnitudes, of the modelled ozone responses we consider photochemical processes. The sharp decrease in ozone above 45km with the SIM spectra (Fig. 2b) is consistent with it being in photochemical steady state with the dominant sinks, *viz.* increased levels of HO_x and O. These losses are compensated by the greater production of O_x through photodissociation of O_2 in the Huggins band and this dominates the loss lower down. Furthermore, the ozone decreases produce a self-healing effect whereby more UV radiation is transmitted to lower levels resulting in greater O_2 photolysis and thus more O_3. [See Supplementary Information].

In order to assess the sensitivity of our results to uncertainty in the measured irradiance values at 200-240 nm (see Fig. 1) we carried out another set of experiments (not shown) in which the switchover from SOLSTICE to SIM was imposed at 240 nm (rather than 200 nm). There are differences in detail in the resulting temperature and ozone fields but the general picture is the same: reduced ozone in the upper stratosphere and mesosphere and a positive peak in the middle stratosphere. Thus there is uncertainty in the magnitude of the response but this does not impact our conclusions with respect to the impact on the middle atmosphere. It also has little bearing on the radiative forcing estimates now presented.

The response of tropospheric and surface climate to variations in solar activity is an important consideration in the attribution of surface temperature trends to human or natural factors. Radiative forcing of climate is defined by the Intergovernmental Panel on Climate Change as the change in net flux at the tropopause, taking into account the effects of any stratospheric adjustment[16]. It is known that solar radiative forcing is modulated by the ozone response to changes in solar UV[2]; the effect of an increase in ozone is twofold: firstly to reduce the flux of solar radiation reaching the tropopause and secondly to increase the flux of infrared radiation, mainly through its impact on stratospheric temperatures. The net effect has been assessed as a small increase in the net downward flux[17].

Simulations of the effect of solar variability on tropospheric climate, such as those reviewed in the IPCC assessments[16], tend to incorporate broad spectral bands which resolve neither details of the spectrum nor the effect of stratospheric ozone. The SIM measurements place an additional perspective on this. Although the change in total irradiance from 2004 to 2007 is similar in the Lean and SIM datasets their very different spectral compositions, and the resulting impacts on the stratosphere, produce quite different pictures of the transmission of radiation to the tropopause, and thus different modulations of radiative forcing. Table 1 presents the change, 2004-2007, in global average downward solar flux at the top of the atmosphere, and at the tropopause, in four spectral bands, along with the resulting change in downward thermal radiation at the tropopause.

Little of the radiation in the 200-310 nm wavelength band reaches the tropopause so that the large increase in the SIM irradiance at the top-of-atmosphere is not found here. In the 310-500 nm region the radiation reaching the tropopause is modulated by the ozone column above, so that the larger increase in ozone produced by the SIM spectra significantly diminishes radiation reaching 105hPa. In the 500-700 nm region ozone absorption again plays a part so that the decrease in the TOA value in the SIM experiment becomes an even larger decrease at the tropopause. The change in spectrally integrated solar irradiance at TOA in the two experiments is very similar: 0.11 Wm^{-2} with Lean and 0.09 Wm^{-2} with SIM, but at the tropopause, while the Lean experiment shows an increase of 0.06 Wm^{-2}, the SIM shows a decrease of 0.16 Wm^{-2}. The thermal radiation increases the Lean radiative forcing slightly, and moderates the decrease in SIM, so that the net solar radiative forcing 2004-2007 estimated using the two datasets is +0.08 Wm^{-2} with Lean (consistent with previous studies of radiative forcing over a solar cycle[17]) but -0.10 Wm^{-2} when the SIM data are used. The latter suggests that radiative forcing of surface climate by the Sun is out of phase with solar activity, at least over this declining phase of solar cycle 23. In their study Cahalan et al[12] did not find this out-of-phase relationship in near-surface air temperature, perhaps because their radiative-convective model did not incorporate the ozone response.

The SIM data provide an entirely different picture than currently accepted of how solar irradiance varies. It is pertinent to ask whether this spectral variability is typical of solar activity cycles and, if so, why it has not been observed previously. It is possible that the Sun has been behaving in an anomalous fashion recently; certainly the current solar minimum is lower and longer than any of those observed over recent decades[18] and perhaps the solar spectrum has different characteristics when the Sun is in a state of very low activity. Gaps in understanding will only be resolved by the acquisition of long-term, well-calibrated, high vertical resolution measurements of stratospheric composition and temperature acquired coincidently with the essential solar spectral data that have also been properly degradation-corrected and calibrated.

The SORCE observations are, however, consistent with a solar activity-dependent change in the temperature gradient of the solar photosphere[4] suggesting that the offsetting irradiance trends with wavelength seen in SIM should appear in each solar cycle. If this is the case then it is necessary to reconsider current understanding[19] of the mechanisms whereby solar cycle variability influences climate: the impact on the stratosphere is much larger than previously thought and the radiative forcing of surface climate is out of phase with solar activity. Currently there is no evidence to ascertain whether this behaviour has occurred before but if this were the case during previous multi-decadal periods of low solar activity it would be necessary to revisit assessments of the solar influence on climate and to revise the methods whereby these are represented in global models.

METHODS SUMMARY

Solar spectra The spectra used were 10-day averages of the Lean model and SORCE data centred on 21 April 2004 and 7 November 2007, chosen as furthest spaced dates of calibrated SORCE data. The data were interpolated onto the 171 wavebands of the 2D model in the range 116-730 nm.

2D model. The 2D (latitude-log pressure) zonal mean model incorporates interactions between radiative, chemical and dynamical processes. The same solar spectra are used for the calculation of chemical photodissocation and heating rates. The model was run to (seasonally-varying) equilibrium with each of the 4 spectral datasets. All results are presented for December.

Multiple regression analysis. The code (Myles Allen, Oxford University, personal communication) estimates the coefficients of regression indices simultaneously with the parameters of a red noise model, here taken to be of order unity. The fit is iterated until the noise model fits within a pre-defined threshold. This method minimises the possibility of noise being interpreted as a signal and can produce, using a Student's t-test, measures of the confidence intervals of the resultant regression coefficients, taking into account any covariance between the indices.

Received 16 February 2010;

1. Brasseur, G. & Solomon, S. Aeronomy of the middle atmosphere: chemistry and physics of the stratosphere, (Springer, 2005).

2. Haigh, J.D. The role of stratospheric ozone in modulating the solar radiative forcing of climate. Nature 370, 544-546 (1994).

3. Harder, J., Lawrence, G., Fontenla, J., Rottman, G. & Woods, T. The Spectral Irradiance Monitor: Scientific requirements, instrument design, and operation modes. Solar Physics 230, 141-167 (2005).

4. Harder, J.W., Fontenla, J.M., Pilewskie, P., Richard, E.C. & Woods, T.N. Trends in solar spectral irradiance variability in the visible and infrared. Geophysical Research Letters 36(2009).

5. Lean, J. Evolution of the sun's spectral irradiance since the Maunder Minimum. Geophysical Research Letters 27, 2425-2428 (2000).

6. Krivova, N.A., Solanki, S.K. & Floyd, L. Reconstruction of solar UV irradiance in cycle 23. Astronomy & Astrophysics 452, 631-639 (2006).

7. Krivova, N.A., Solanki, S.K., Fligge, A. & Unruh, Y.C. Reconstruction of solar irradiance variations in cycle 23: Is solar surface magnetism the cause? Astronomy & Astrophysics 399, L1-L4 (2003).

8. Harder, J.W., et al. The SORCE SIM Solar Spectrum: Comparison with Recent Observations Solar Physics 263, 3-24 (2010).

9. Harwood, R.S. & Pyle, J.A. A 2-dimensional mean circulation model for the atmosphere below 80km. Quarterly Journal of the Royal Meteorological Society 101, 723-747 (1975).

10. Bekki, S., et al. The role of microphysical and chemical processes in prolonging the climate forcing of the Toba eruption. Geophysical Research Letters 23, 2669-2672 (1996).

11. Warwick, N.J., Bekki, S., Nisbet, E.G. & Pyle, J.A. Impact of a hydrogen economy on the stratosphere and troposphere studied in a 2-D model. Geophysical Research Letters 31(2004).

12. Cahalan, R.F., Wen, G.Y., Harder, J.W. & Pilewskie, P. Temperature responses to spectral solar variability on decadal time scales. Geophysical Research Letters 37(2010).

13. Randel, W.J. & Wu, F. A stratospheric ozone profile data set for 1979-2005: Variability, trends, and comparisons with column ozone data. Journal of Geophysical Research-Atmospheres 112(2007).

14. Haigh, J.D. and Pyle, J.A. A two-dimensional calculation including atmospheric carbon dioxide and stratospheric ozone. Nature, 279, 222-224 (1979).

15. Soukharev, B.E. & Hood, L.L. Solar cycle variation of stratospheric ozone: Multiple regression analysis of long-term satellite data sets and comparisons with models. J. Geophys. Res. 111 (2006).

16. Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor and H.L. Miller (eds.). Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, 2007. Cambridge University Press (2007).

17. Larkin, A., Haigh, J.D. & Djavidnia, S. The effect of solar UV irradiance variations on the Earth's atmosphere. Space Sci. Rev. 94, 199-214 (2000).

18. Fröhlich, C. Evidence of a long-term trend in total solar irradiance. Astron. & Astrophys. 501, L27-U508 (2009).

19. Gray, L.J., et al. Solar influences on climate. Rev. Geophys., in press (2010).

Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

Acknowledgements This work was partially funded by the UK Natural Environment Research Council SOLCLI consortium project. We are grateful to Judith Lean for providing solar spectra, to Bill Randel for advice on data analysis and to Lucien Froidevaux for advice on the EOS Aura-MLS data. The MLS data were obtained from the instrument website http://mls.jpl.nasa.gov/index-eos-mls.php.

Author Contributions J.W.H. provided the SIM and SOLSTICE data, information on its interpretation and on solar variability, R.T. provided input on stratospheric photochemistry, A.R.W. edited the SIM data into a format suitable for the model and carried out preliminary model runs, J.D.H. performed the model experiments and diagnostics, carried out the MLS data analysis and wrote the paper.

Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of this article at www.nature.com/nature. Correspondence and requests for materials should be addressed to. J.D.H. (j.haigh@imperial.ac.uk).

Table 1. Difference in global average downward radiative flux (Wm^{-2}) between 2004 and 2007 at the top of the atmosphere (TOA) and at 105hPa (TPS) calculated for December in the model using the two spectral datasets.

Wavelength	200-310 nm		310-500 nm		500-700 nm		700-1600 nm		Total solar 200-1600 nm		Thermal (LW)		Net	
Level	TOA	TPS	TOA	TPS	TOA	TPS	TOA	TPS[1]	TOA	TPS	TOA	TPS	TOA	TPS
Lean data	0.02	0.00	0.04	0.03	0.03	0.01	0.02	0.02	0.11	0.06	0	0.02	0.11	0.08
SIM data	0.16	0.00	0.11	0.06	-0.13	-0.17	-0.05	-0.05	0.09	-0.16	0	0.06	0.09	-0.10

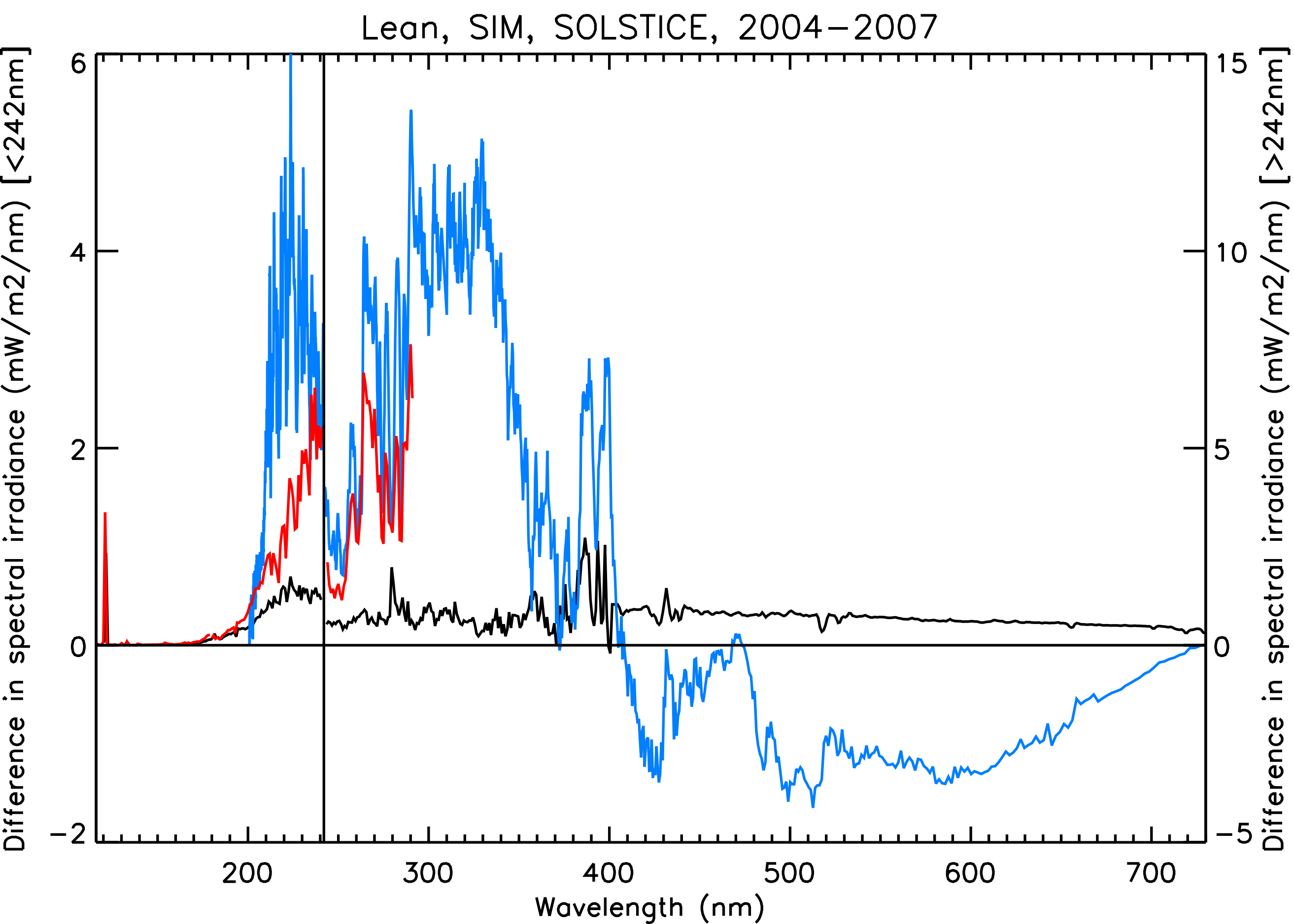
[1] The radiation scheme in the 2D model does not calculate flux propagation in this spectral region. As radiation at these wavelengths is absorbed very little by stratospheric gases the tropopause values here are assumed to be the same as the top-of-atmosphere values.

Figure Captions

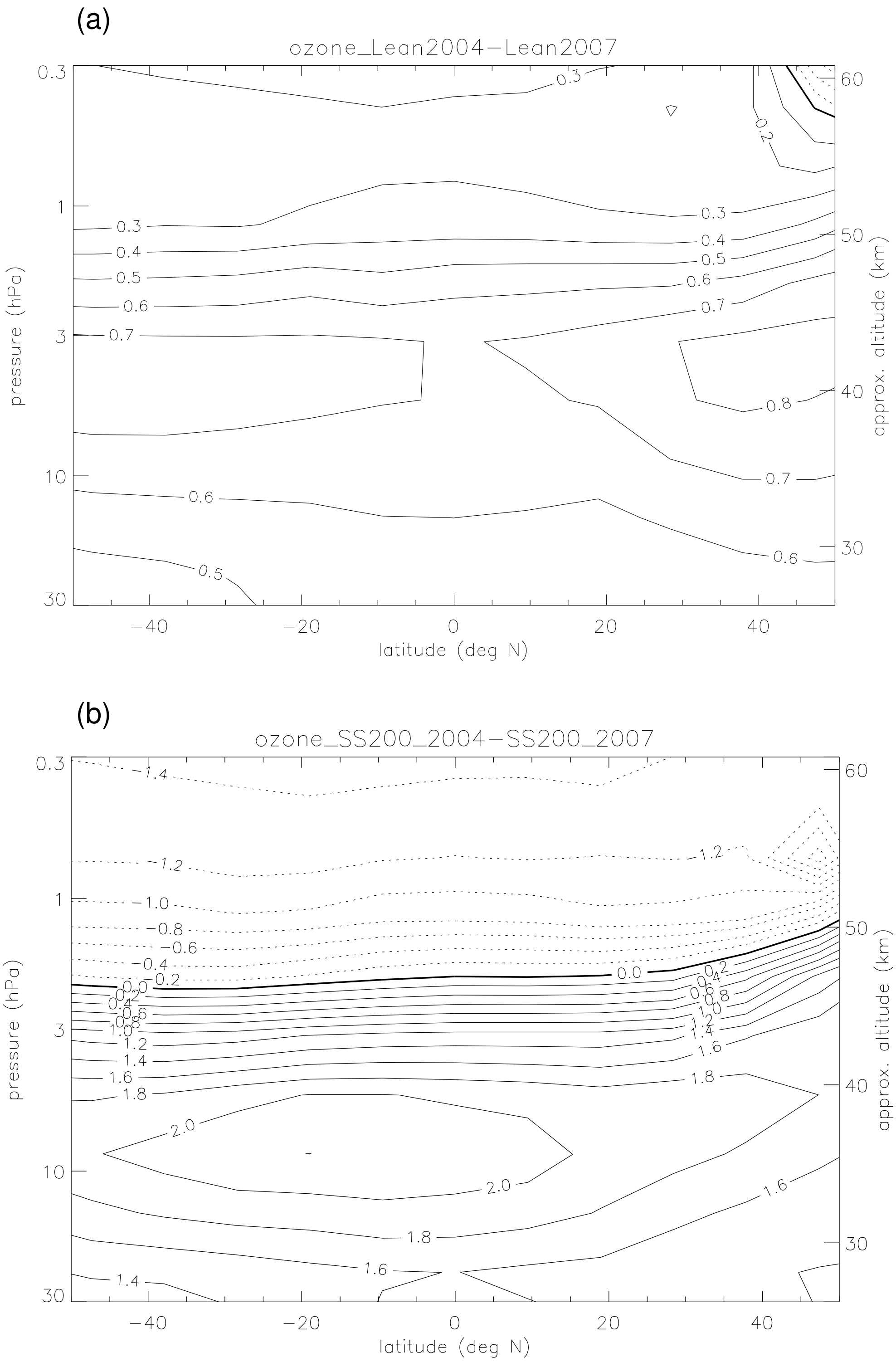
Figure 1 Difference in solar spectrum between April 2004 and November 2007. The difference (2004-2007) in solar spectral irradiance (W m^{-2} nm^{-1}) derived from SIM data[4] (in blue), SOLSTICE data[8] (in red) and from the Lean model[5] (black). Different scales are used for values at wavelengths less and more than 242 nm (see left and right hand axes respectively).

Figure 2 Modelled difference in ozone between December 2004 and 2007. Estimates of the percentage difference (2004-2007) in zonal mean ozone concentration produced by the model using solar spectra from (a) the Lean model and (b) SIM/SOLSTICE data.

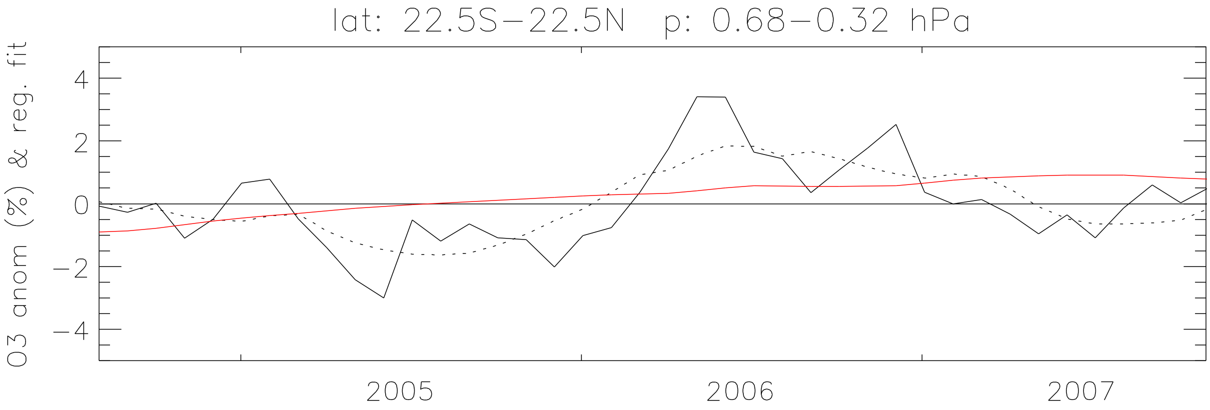
Figure 3 Time series of AURA-MLS v2.2 ozone concentrations. The data (solid black lines) are anomalies (%) of tropical (22.5 °S-22.5 °N) deseasonalised monthly means from August 2004 to November 2007. The values reconstructed from the 4-component regression model are shown as dashed lines. The solar component of the regression is shown in red. Other components are shown, along with the solar, in Supplementary Fig. 2. Data averaged between (a) 0.68 and 0.32 hPa and (b) 10 and 6.8 hPa.



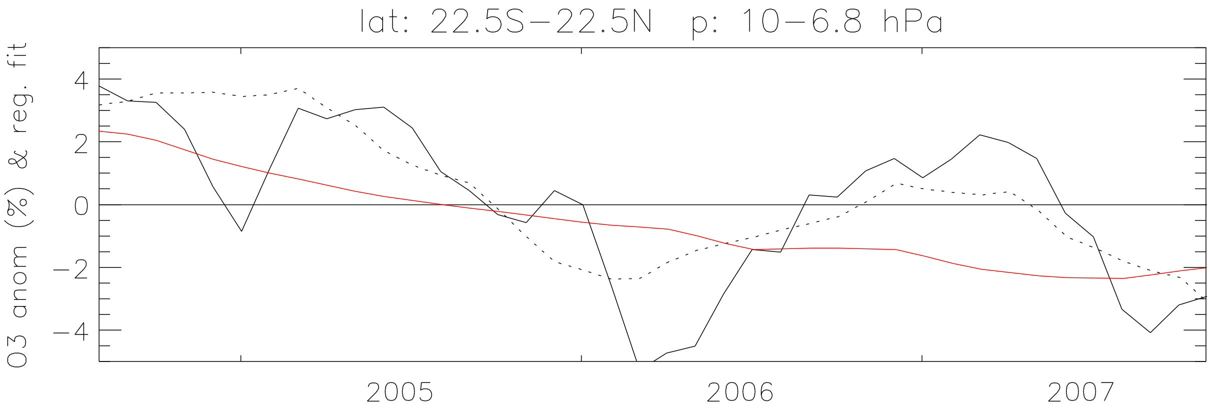
Lean, SIM, SOLSTICE, 2004−2007
Difference in spectral irradiance (mW/m2/nm) [<242nm]
Difference in spectral irradiance (mW/m2/nm) [>242nm]
Wavelength (nm)
6
4
2
0
−2
15
10
5
0
−5
200
300
400
500
600
700


(a)
ozone_Lean2004–Lean2007
pressure (hPa)
approx. altitude (km)
latitude (deg N)
0.3
1
3
10
30
60
50
40
30
−40
−20
0
20
40
0.2
0.3
0.4
0.5
0.6
0.7
0.8
(b)
ozone_SS200_2004–SS200_2007
pressure (hPa)
approx. altitude (km)
latitude (deg N)
−1.4
−1.2
−1.0
−0.8
−0.6
−0.4
−0.2
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

(a)


lat: 22.5S–22.5N p: 0.68–0.32 hPa
O3 anom (%) & reg. fit
4
2
0
−2
−4
2005
2006
2007

(b)


lat: 22.5S–22.5N p: 10–6.8 hPa
O3 anom (%) & reg. fit
4
2
0
−2
−4
2005
2006
2007

2D Model

The model extends from pole to pole and from the ground to 60 km with a horizontal resolution of 9.5° and a vertical resolution of half a pressure scale height (~3.5 km). Atmospheric circulation is calculated from forcing terms which include solar heating in the stratosphere by O_2 and O_3 and longwave heating in the stratosphere by CO_2, H_2O, O_3, CH_4 and N_2O.[1] The photochemical scheme includes reactions in the O_x, NO_x, HO_x, ClO_x, BrO_x, CHO_x, and SO_x families.[2] The top-of-atmosphere solar spectrum is specified in 171 spectral intervals between 116 and 720 nm, as are the absorption cross-sections of O_2, O_3. and other photoactive species. The model runs described in this paper were carried with fixed background concentrations of CO_2 and CFCs.

MLS ozone data analysis

The data used for the detection of observed signals in ozone were EOS Aura-MLS v2.2 monthly means. MLS has a limb-scanning approach which provides measurements with a vertical resolution of 3km between 215 and 0.2 hPa. We average data over the tropics and over two separate vertical slabs, as described in the main text. These time series are then used in the multiple regression analysis. The 2 QBO indices are the first two principle component time series derived from 55 years of monthly mean equatorial zonal wind at 14 pressure levels between 90 and 10 hPa (Barbara Naujokat, Frei Universität Berlin, personal communication). The 2 indices incorporate about 95% of the total QBO variance and thus in weighted combination can represent QBO variability at any level within the range provided. Solar variability is represented by SIM UV data but the use of a linear trend produces very similar results.

Supplementary Fig. 2 shows the time series of the contributions of each of the three varying regressions indices for the two atmospheric layers. The sum of these components (plus the offset) gives the fit shown in Fig.3 of the main document. There is a dominant contribution from the QBO in each layer, but the timing is such that it is a different QBO component to the fore. The solar signal, also reproduced in Main Fig. 3, shows a decline from 2004 to 2007 at the lower level (corresponding to the positive signal 2004-2007 shown in Main Fig. 2) and the opposite at the higher level. Supplementary Table 1 presents the regression statistics: the solar signal is strongly significant in both layers but, as discussed in the main text, not distinguishable from a monotonic change due to other factors such as climate change. Nevertheless, it is the different behaviour at the two pressure levels that suggests that the solar influence is important.

Ozone production and destruction rates

Supplementary Fig.3 shows how each of the three major catalytic ozone destruction cycles (O_x, HO_x and NO_x), as well as ozone production through photodissociation, contribute to the difference in ozone concentrations found for 2004-2007using the Lean model (shown in Fig.2a of the main text). Supplementary Fig. 4 shows the same for the runs using the SIM/SOLSTICE data. All magnitudes are larger in the latter case due to the impact of the larger differences in UV radiation In the lower mesosphere the major loss mechanism for O_x ($=O_3+O(^3P)+O(^1D)$) is by reaction with HO_x ($=OH+HO_2$) which is mainly produced through the creation of OH by the reaction of H_2O with $O(^1D)$. The change in model $O(^1D)$ (not shown) is 0.5 to 1% at 40-60km in the Lean experiment but 3 to >4% in the same region with the SIM spectral data, due to the enhanced photodissociation of O_3 by UV radiation (Fig. 1 of main text). The resulting increases in HO_x (not shown) are over 2% throughout the stratosphere in the SIM experiment but less than 0.4%, peaking at the top of the model domain, with the Lean model. In the upper stratosphere the major loss mechanism for O_3 is by recombination with O and higher concentrations of the latter (~5% for SIM and ~1% for Lean) also tend to reduce O_3 concentrations. Photochemical production peaks around 3 hPa in the Lean experiment and somewhat lower in the SIM due to the increased ratio of longer to shorter wavelength UV radiation, and also a self-healing effect.

References

1. Haigh, J.D. Radiative heating in the lower stratosphere and the distribution of ozone in a two-dimensional model. *Quarterly Journal of the Royal Meteorological Society* **110**, 167-185 (1984).

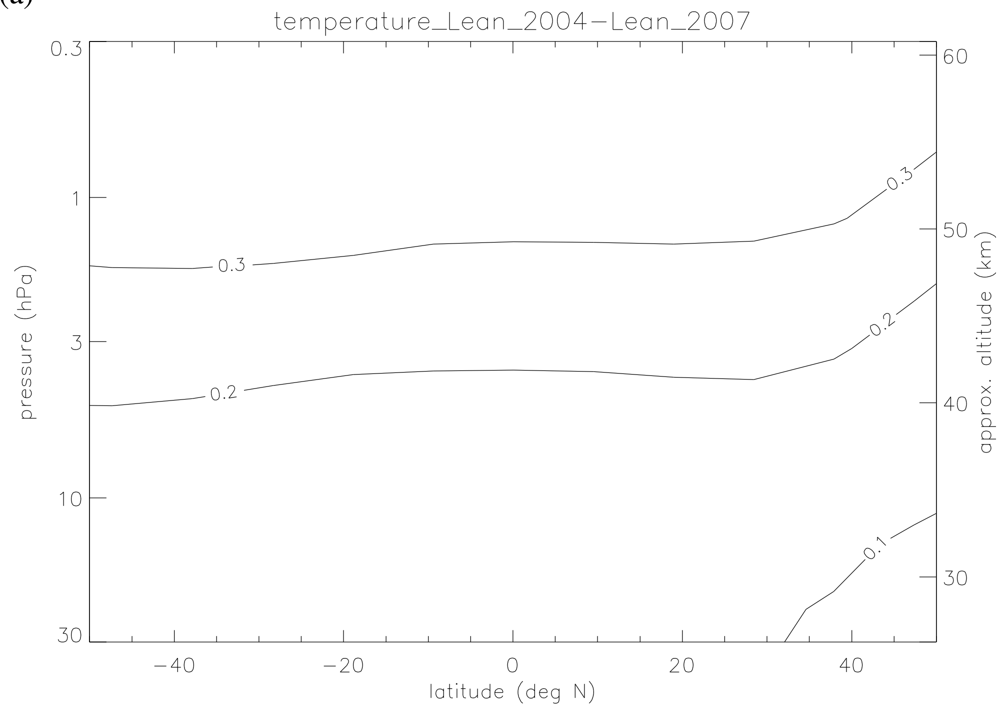
2. Law, K.S. & Pyle, J.A. Modelling trace gas budgets in the troposphere. 1. Ozone and odd nitrogen. *Journal of Geophysical Research-Atmospheres* **98**, 18377-18400 (1993).

Supplementary Table 1 Results of regression analysis of MLS ozone data. For each layer and for each regression index are given the magnitude of the signal (O_3 variation (%)), the t-value of that signal from a Student's t-test and the significance level of the result, given 35 degrees of freedom. The solar (or trend) signal is highly significant at both layers.

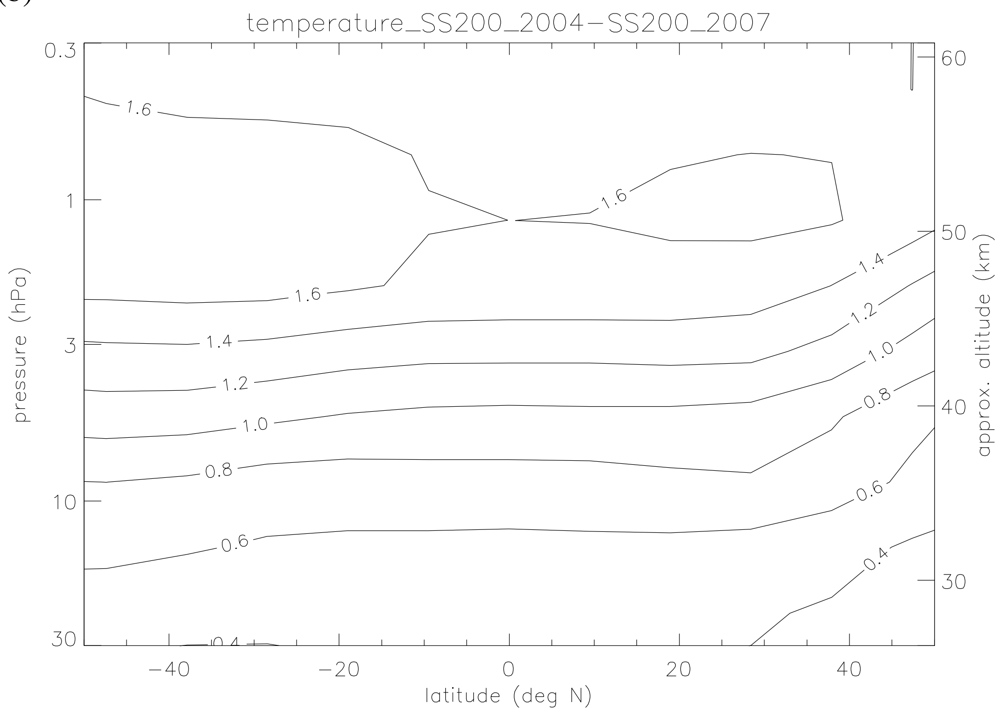
	QBO1			QBO2			solar		
	O_3 (%)	t-val	signif(%)	O_3 (%)	t-val	signif(%)	O_3 (%)	t-val	signif(%)
0.7-0.3 hPa	3.0	4.2	>99	0.1	0.1		-1.8	2.3	>95
10-6.8 hPa	-0.8	0.5		4.6	2.9	>99	4.7	2.8	>99

Supplementary Figure 1 Modelled difference in temperature between 2004 and 2007. Estimates of the difference (2004-2007) in zonal mean temperature (K) in December produced by the model using solar spectra from (a) the Lean model and (b) SIM/SOLSTICE data.

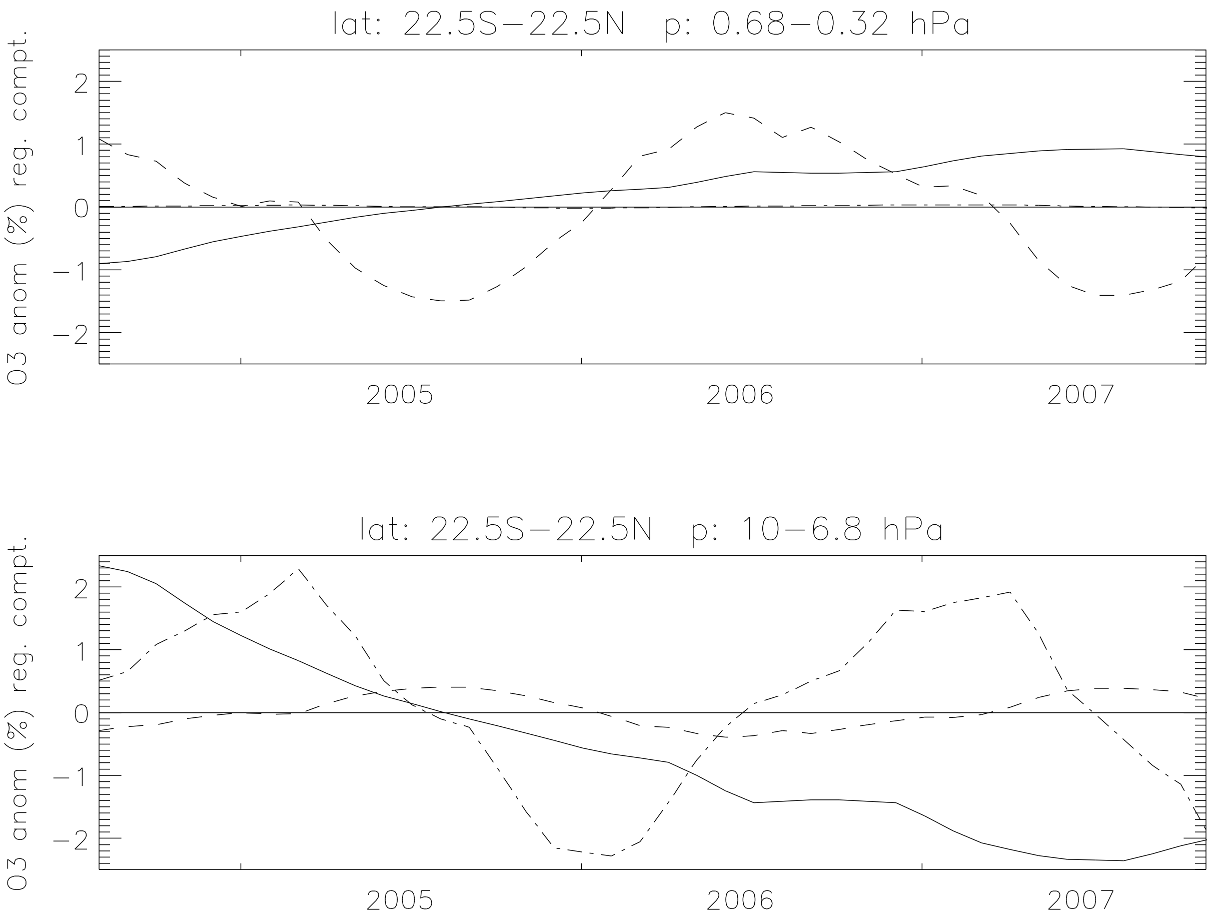
(a)



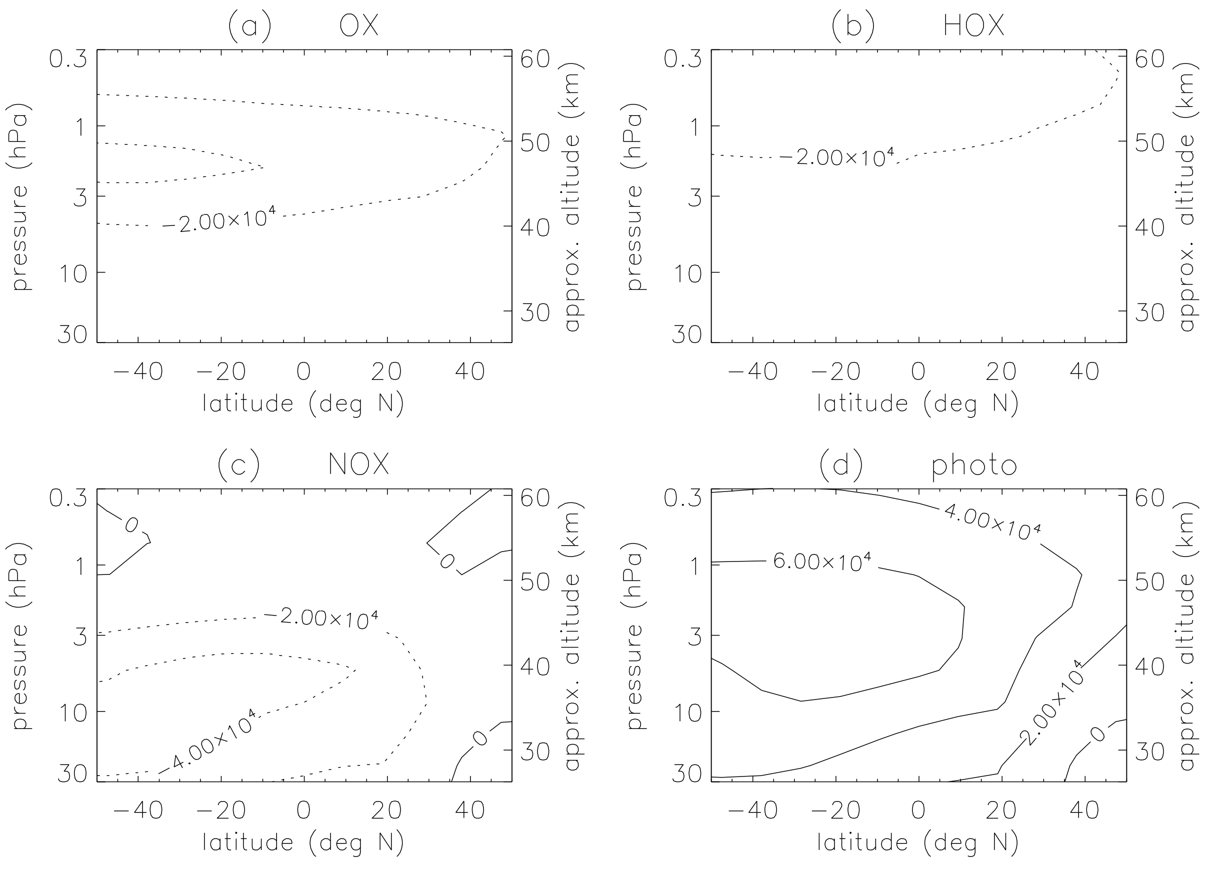
(b)



Supplementary Figure 2. Time series of MLS ozone concentrations showing the individual components from the regression analysis. The data are anomalies (%) of tropical (22.5 °S-22.5 °N) deseasonalised monthly means from August 2004 to November 2007. The regression components are a constant (offset, not shown) two orthogonal time series for the QBO (dashed and dash-dotted lines, respectively) and a solar UV time series (solid line). The upper panel used data averaged between 0.68 and 0.32 hPa, the lower data between 10 and 6.8 hPa.



Supplementary Figure 3 Modelled difference between 2004 and 2007 in rates of production and destruction of ozone by photochemical processes in December. Differences estimated using solar spectra from the Lean model. (a) Destruction by combination of $O+O_3$, (b) Catalytic destruction by HO_x compounds, (c) Catalytic destruction by NO_x compounds, (d) Production through photodissociation of O_2 (and other minor); Contour interval = $2x10^4$ molecules cm^{-3} s^{-1}. Negative [positive] values in panels (a)-(c) [(d)] indicate greater destruction [production] rates in 2004 relative to 2007.



Supplementary Figure 4 As Supplementary Figure 3 but for model experiment with SIM/SOLSTICE spectra. Contour interval = 4×10^4 molecules cm^{-3} s^{-1}.