**Atmospheric
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Radiative forcing of
ozone during a
solar-cycle

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Radiative forcing from modelled and observed stratospheric ozone changes due to the 11-year solar cycle

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Abstract

Three analyses of satellite observations and two sets of model studies are used to estimate changes in the stratospheric ozone distribution from solar minimum to solar maximum and are presented for three different latitudinal bands: Poleward of 30° north, between 30° north and 30° south and poleward of 30° south. In the model studies the solar cycle impact is limited to changes in UV fluxes. There is a general agreement between satellite observation and model studies, particular at middle and high northern latitudes. Ozone increases at solar maximum with peak values around 40 km. The profiles are used to calculate the radiative forcing (RF) from solar minimum to solar maximum. The ozone RF, calculated with two different radiative transfer schemes is found to be negligible (a magnitude of $0.01\,\mathrm{Wm^{-2}}$ or less), compared to the direct RF due to changes in solar irradiance, since contributions from the longwave and shortwave nearly cancel each other. The largest uncertainties in the estimates come from the lower stratosphere, where there is significant disagreement between the different ozone profiles.

1 Introduction

Variations in total solar irradiance (TSI) and its spectral distribution are expected to influence climate in different ways. Sun-climate connections have an impact on the chemical distribution in the atmosphere, including effects on ozone and other chemically active greenhouse gases (Lean, 1997); the changes in TSI influence atmospheric ozone directly through absorption of shortwave solar radiation and thereby atmospheric temperatures (Haigh, 1996; Shindell, et al., 1999). The absorption of solar radiation by ozone is vital for shielding of UV radiation and the temperature profile in the middle and upper atmosphere. Although it is well established that variations in TSI, and in particular the spectral differences in this variation, has an impact on the ozone, the climate impact of this change in ozone, however, is uncertain. There are significant differences

in earlier estimates of the radiative forcing due to ozone changes caused by variations in TSI where even the sign of the radiative forcing has differed (Houghton et al., 2001). Finally it has also been suggested that changes in TSI can alter clouds (Svensmark, 1998). The latter effect is controversial and it is highly uncertain if such a mechanism exists at all, or whether it is significant in magnitude (Kristjansson, et al., 2004; Kuang, et al., 1998).

Stratospheric ozone is affected by solar cycle variations through changes in the UV fluxes which affect the photo-dissociation of chemical species. Changes in stratospheric dynamics resulting from solar cycle variations, are another possible cause and consequence of ozone variation. The impact on the total ozone column through such variations has been demonstrated through both observations and model studies (Brasseur, 1993; Jackman et al., 1996; Zerefos and Crutzen, 1975; van Loon and Labitzke, 1994; Zerefos et al., 1997; Hood, 1997; Haigh, 1994; Shindell et al., 1999). Recently, the ozone response to the solar cycle has been studied also by fully interactive 3-D chemistry-climate models (e.g. Tourpali et al., 2003; Egorova et al., 2004). The total ozone column increase from solar minimum to solar maximum is in the range 1 to 2% (Zerefos et al., 1997).

Several studies have shown ozone changes near the tropopause level has the largest climate impact (Forster and Shine, 1997; Hansen et al., 1997; Lacis et al., 1990; Wang and Sze, 1980). An ozone increase leads to an enhanced atmospheric trapping of longwave radiation (positive radiative forcing) as well as absorbing more solar radiation. An ozone increase in the stratosphere leads to less solar radiation reaching the surface-troposphere system and a negative solar radiative forcing which opposes the positive longwave radiative forcing. The net radiative forcing (sum of solar and longwave radiative forcing) is dependent on the altitude of the stratospheric ozone change (Houghton et al., 2001).

2 Atmospheric processes

2.1 Changes in solar UV radiation over the solar cycle

The 11-year solar cycle is connected with large variability of the solar radiation in the UV part of the spectrum. Accurate assessments of the solar irradiance variation are now available through satellite measurements. Two solar cycles are well document through the work of Lean et al. (1997). Based on observations for September 1986 for solar minimum and November 1989 for solar maximum, the variation in the solar cycle was estimated in the wavelength interval 119.5 to 419.5 nm. The data for the 11-year cycle variations used in the model studies are based on this work and are shown in Fig. 1. Note that since the change in the F10.7 cm flux during this period is 160 units, compared to the 120 units for a typical solar cycle, the radiative forcing results obtained using the model-derived ozone changes have been scaled by a factor of 120/160, for comparison with the forcings derived from the satellite regressions. The variation increases strongly towards shorter wavelengths where compounds like O_2 and N_2O are dissociated in the stratosphere (up to 10%).

2.2 Impact on stratospheric chemistry

Since changes in the solar flux from solar minimum to solar maximum are most pronounced toward the shorter wavelength range $\lambda \sim 200$ nm (Fig. 1), which is absorbed mainly in the middle to upper stratosphere, this is the height range where the largest direct impact on the chemistry is expected to occur.

Stratospheric ozone production takes place through O_2 photo-dissociation:

$$O_2 + h\upsilon \rightarrow O + O \quad \lambda \leq 242 \, \text{nm} \tag{R1}$$

$$O + O_2 + M \rightarrow O_3 + M. \tag{R2}$$

Enhanced UV fluxes going from solar minimum to solar maximum lead to enhanced

ozone production in the stratosphere above approximately 30 km, limited by the 242 nm cut-off in photodissociation.

The main loss of ozone in the middle and upper stratosphere is through catalytic processes involving nitrogen oxides. The main source of nitrogen oxides in the stratosphere is through the reaction of excited state atomic oxygen O (^{1}D) with N_2O (Crutzen et al., 1975):

$$O(^1D) + N_2O \rightarrow NO + NO. \tag{R3}$$

However, only approximately 5% of the N_2O loss occurs via this reaction; the main loss which determines the profile in the stratosphere is through short wave photodissociation giving molecular nitrogen:

$$N_2O + h\upsilon \quad \rightarrow N_2 + O \quad \lambda \leq 210\,nm. \tag{R4}$$

Increases in solar fluxes from solar minimum to solar maximum will reduce N_2O, particularly in the upper stratosphere. O(^{1}D) is only moderately affected by solar cycle variations since it is produced by photo-dissociation of ozone at longer wavelengths ($\lambda \leq 310$ nm) than for photodissociation of N_2O where flux variations are smaller. The result, in the upper stratosphere, is reduced NOx levels and reduced catalytic ozone loss through nitrogen reactions during solar maximum compared to solar minimum. The model calculations give N_2O reductions of 10 to 20% in the upper stratosphere, and NOx reductions of 5 to 10% compared to the values calculated during solar minimum. Active chlorine is enhanced by approximately 2% at solar maximum leading to a slight increase in the catalytic ozone loss through the chlorine cycle. This indicates smaller solar cycle amplification of catalytic ozone loss under current conditions than without human impact on the chlorine budget.

The effect of solar flux changes through the main chemical perturbations, reactions R1 and R4, is to increase ozone abundances in the upper stratosphere at solar maximum.

In the lower stratosphere NOx is converted to HNO_3 in a 3-body reaction with OH so that the impact of increased solar flux depends on ambient levels of OH (and thus the concentration of H_2O and CH_4). The solar signal in NOx may therefore be of the opposite sign in the lower and upper stratosphere (Kilifarska and Haigh, 2005).

3 Comparisons of observations with modelling

Three sets of satellite data and two sets of global chemistry model calculations (one 2-D and one 3-D) have been used to analyze the change in the stratospheric ozone from changes over the solar cycle.

3.1 Model studies

3.1.1 Oslo SCTM1

The 3-dimensional Oslo SCTM1 (Stratospheric Chemical Transport Model) is used in this study. For further references see Rummukainen et al. (1999). It is driven by off-line winds generated by the GISS GCM (Rind et al., 1988). The model has a horizontal resolution of 7.8°×10° latitude/longitude, with 21 layers from the surface up to 0.002 hPa (about 90 km). The stratospheric chemistry code is based on Stordal et al. (1985) including heterogeneous chemistry on polar stratospheric clouds (PSC) and aerosol particles (Isaksen and Stordal, 1986; Isaksen et al., 1990), with aerosol surface areas from SAGE II with 1989 as reference year (low aerosol loading). The modelling of PSCs is based on the NCEP temperature data and are allowed to occur poleward of 40° in the lower stratosphere. Photolysis rates are calculated online every 40 min with the Fast-J2 code for the stratosphere (Bian and Prather, 2002), and Fast-J (Wild et al., 2000) for the troposphere. Two runs have been done, one for solar maximum and one for solar minimum. For each model simulation the model has been run for 3 years to allow for spin up time. The emissions are kept at 1990 level in both runs, which means

that the changes seen in the simulations are due to changes in the solar radiation alone.

3.1.2 Imperial College 2-D model (ICSTM)

For comparison, and for our radiative forcing calculations, the ozone fields from experiments with a 2-D radiative-chemical-transport model are also used. This model is a classical Eulerian model extending from pole-to-pole with a resolution of 9.5° and from the surface to a height of ~95 km with a resolution of ~3.5 km. It contains detailed descriptions of radiation and chemistry, based on the family treatment of Law and Pyle (1993). Experiments were carried out in which solar irradiance (resolved into 171 spectral intervals between 121 and 700 nm) was prescribed for solar maximum and solar minimum situations as described by Haigh (1994).

3.2 Data analysis

3.2.1 Analysis SBUV/SAGE Aristotle University of Thessaloniki, Laboratory of Atmospheric Physics (AUTH)

The SAGE II ozone data, in the form of ozone mixing ratio (in parts per million), were derived and used to construct solar-cycle induced changes in 10° latitude belts from 60° S to 60° N. Even though the original data were retrieved from ground level and up to the altitude of 70 km, the many missing data and the volcanic aerosol data contamination force us to restrict the analysis to the altitudes range 20–55 km.

The adopted SBUV data are from the Version 8 Merged Ozone Data Sets, made available by the TOMS science team (http://code916.gsfc.nasa.gov/Data_services/ merged/). The SBUVv.8 merged data set provides nearly global coverage and consists of 25 years of ozone observations (1979–2003). The data are available as zonal means (every 5°) of profile ozone volume mixing ratio (ppm) at 15 pressure levels, ranging from 0.5 to 50 hPa.

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The seasonal cycle, the Quasi Biennial Oscillation (QBO), and the long-term linear trend (applicable for each month separately) were filtered out at each level and at all latitudes.

Composite differences (in % of solar minimum) of the annual mean ozone amount between the years of solar maximum and solar minimum were calculated from the filtered data. The years of solar maximum and solar minimum were defined using the F10.7 solar radio flux as a proxy for the 11-year solar cycle. Years with high stratospheric aerosol loading from volcanic eruptions were excluded from the analysis.

3.2.2 Analysis of SAGE data (NCAR)

The solar cycle variation in ozone derived from SAGE I (1979–1982) and SAGE II (1984–2005) data is based on the analysis in Randel and Wu (2007). Briefly, a multivariate regression analysis is applied to the SAGE I+II data, and solar cycle variability is modelled using the standard F10.7 radio flux as a solar proxy; additional terms in the regression include decadal trends (modelled using an effective stratospheric chlorine proxy), and QBO effects. SAGE measurements cover the approximate latitude range 55° N-S, and the vertical domain is 20–50 km. Details of the solar cycle variability in SAGE data, together with comparisons with the solar cycle in column ozone measurements, are discussed in Randel and Wu (2007).

3.3 Comparisons of observations with model results

Figure 2 shows the area-weighted vertical profiles of the ozone change over a solar cycle for 3 different latitude bands, 90° S–30° S, 30° S–30° N and 30° N–90° N. The results from the 3 sets of satellite data and the 2 models show many similarities in the vertical pattern. The general pattern is an increase in stratospheric ozone from solar minimum to solar maximum. With the exception of the SBUV data, modelled and observed ozone perturbations peak at approximately 40 km with maximum values around 2 to 3%. In the lower stratosphere, where ozone perturbations have the strongest impact on the

total ozone column, the modelled increase in ozone is of the order 1–2%, whereas the analyses based on the satellite data have weak or even negative ozone change in the lower stratosphere except for SBUV in the tropics. The best agreement between all data sets is obtained at northern latitudes, with the exception of SBUV data which show an increase above 40 km where the other data sets show a decrease in impact of solar variation. For all three latitude bands, the two model calculations agree reasonably well. The different behaviour of the SBUV data set is discussed also in Tourpali et al. (2007).

4 Radiative forcing

4.1 Description of radiation codes

The University of Oslo (UiO) radiative transfer schemes are an absorptivity/emissivity broad band model for thermal infrared radiation and the solar scheme is a multi-stream model using the discrete ordinate method (see Myhre et al. (2000) for more details). The broad band model includes two ozone absorption bands; at $9.6\,\mu$m and $14\,\mu$m. Meteorological data such as temperature, water vapour, and clouds are the same as in the SCTM.

The University of Reading (UoR) radiative transfer schemes used are those of Forster and Shine (1997), albeit the wavelength resolution in the UV and visible is enhanced to 1 nm. In the thermal infrared, a $10\,\text{cm}^{-1}$ resolution narrow band model is used. The climatology used for these calculations for temperature, water vapour and ozone is mostly from the ERA-40 analysis with cloud amounts, heights and optical depths are taken from Rossow and Schiffer (1999). Stratospheric adjustment of temperatures is calculated using the fixed-dynamical heating method. More information is given in Gray et al. (2007[1]) where a detailed study of the forcing (and stratospheric

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[1]Gray, L. J., Rumbold, S. T., and Shine, K. P.: Stratospheric Temperature and Radiative Forcing Response to 11-year Solar Cycle Changes in Irradiance and Ozone, J. Geophys. Res., submitted, 2007.

temperature change) due to solar-cycle induced changes in both irradiance and ozone (using the Randel and Wu (2007) data) is presented.

4.2 Forcing calculations

Table 1 shows radiative forcing due to changes in stratospheric ozone from the solar cycle variations described in Sect. 3. The net radiative forcing (sum of longwave, LW, and shortwave, SW) is weak for all five data sets and the 2 sets of radiative transfer schemes and ranges from −0.005 to 0.010 Wm^{-2}. This is small compared with a typical radiative forcing due to the change in TSI between solar maximum and minimum of about 0.2 Wm^{-2} (although we note that the ozone change may influence tropospheric climate via other mechanisms, such as changing the propagation of planetary waves, as a result of changes in stratospheric temperatures – Haigh, 1996, 1999). The LW forcing dominates the SW forcing thus giving a positive net radiative forcing for the two models, whereas for the satellite data sets the LW and SW are very similar, so that the sign of the net forcing varies between data sets. The ozone reduction in parts of the lower stratosphere in the two SAGE analyses is the main cause for the change in sign compared to the other data sets. For ozone changes in the lower stratosphere the LW forcing dominates over the SW forcing and the net radiative forcing will have the same sign as the ozone change; by contrast, in the upper stratosphere the SW forcing dominates over the LW forcing giving a net forcing of opposite sign of the ozone change (Forster and Shine, 1997; Hansen, et al., 1997). An important part of the cause of a strong LW radiative forcing in the lower stratosphere is the effect of adjustment in the stratospheric temperature which is part of the radiative forcing. Published estimates of the effect of ozone changes on solar radiative forcing have varied from −30% to +45% depending on the specified ozone and temperature profiles (see review by Haigh, 2007). With the ozone changes shown in Fig. 2 it can be seen that a significant ozone change is found in the upper and middle stratosphere. However, a significant part of the ozone change also occurs in the lower stratosphere. Therefore the weak net radiative forcing found here is a result of ozone changes in the whole stratosphere. Our

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results are consistent with those of Larkin et al. (2000) who found an ozone-induced SW radiative forcing of $-0.06\,\mathrm{Wm^{-2}}$ and LW effect in the range 0.03 to $0.11\,\mathrm{Wm^{-2}}$ using the ICSTM ozone data and temperatures calculated within a GCM run. However, the wide range in net ozone-induced radiative forcing found in that work (-0.03 to $+0.05\,\mathrm{Wm^{-2}}$) illustrates the sensitivity to the offset between LW and SW effects and thus, essentially, to the induced temperature changes.

The differences between the UoR and the UiO net radiative forcing are very small (less than $0.005\,\mathrm{Wm^{-2}}$) and largest for the ICSTM model in both absolute and relative terms (note however the forcing is close to zero). Mostly the differences between UoR and UiO are larger for the LW than for the SW but for all cases, the two radiation codes agree well on the net forcing.

In the UiO radiative forcing calculations, additional simulations with ozone changes from the solar cycle variations including changes in the troposphere have been performed based on the model simulations of ozone changes. The radiative forcing from these changes in ozone was in the range of 0.005–$0.02\,\mathrm{Wm^{-2}}$, which is at least of the same magnitude as the ozone changes in the stratosphere. For the ozone changes in the stratosphere the net forcing was small since the LW and SW forcing were of quite similar magnitude but of different sign. For ozone changes in the troposphere, the magnitude of the LW and SW forcing is of much smaller magnitude, but they have the same sign.

Finally, the UoR models were used to calculate the impact of the rather crude latitudinal resolution used here ($60°$) compared to $4°$, and also the impact of extending the ozone changes in the NCAR SAGE data down to 20 km (to an altitude region where the satellite data are more uncertain than at higher altitudes). The results are shown in Table 2. The impact of the degraded horizontal resolution is found to be very low; extending the calculations down to 20 km has, as expected, more of an effect on the LW forcing, and causes the net forcing to change in sign from negative (Table 1) to positive (Table 2); it, however, remains small.

5   Conclusions

The solar cycle impact on ozone based on 3 sets of ozone observation analysis and two model studies shows enhanced values at solar maximum due to enhanced UV radiation at wavelengths shorter than 300 nm. Best agreement is found in the datasets for northern latitudes and at peak altitudes around 40 km. The studies show more spread in the results in the lower stratosphere. Additional effects from changes in the dynamics over a solar cycle, which are not included here, could affect the results. Since, the differences between the observations and the models are relatively small this does not indicate significant dynamic effects. Solar cycle-induced changes in the upper stratospheric temperatures could affect the temperature dependent chemical reactions.

The weak radiative forcing found in this study (with a magnitude of $\sim$0.01 Wm^{-2} or less) is small compared to the forcing from the direct forcing from change in the solar output during a typical solar cycle of about 0.23 Wm^{-2} (Lean, et al., 1997). However, the sign of this ozone forcing is not well constrained because of the strong cancellation between the longwave and shortwave forcings, and varies amongst the different ozone data sets used here. Note that although the ozone change has a small impact on the radiative forcing compared to the solar-cylce irradiance change, its contribution to solar-cycle-induced stratospheric temperature change is likely to be much greater (Shibata and Kodera, 2005; Gray et al., 2007[1]).

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Table 1. Radiative forcing due to stratospheric ozone changes caused by changes in the solar output during a solar cycle for five different data sets, including the effect of stratospheric temperature adjustment. Model simulations preformed with 2 different set of radiation schemes at University of Reading (UoR) and University of Oslo (UiO) for longwave (LW), shortwave (SW) and net radiative forcing. The models are scaled by the factor 120/160 as described in Sect. 2.1.

Ozone data	UoR	UiO	UoR	UiO	UoR	UiO
	LW (Wm^{-2})		SW (Wm^{-2})		Net (Wm^{-2})	
AUTH SAGE	0.008	0.008	−0.013	−0.012	−0.005	−0.004
AUTH SBUV	0.036	0.034	−0.034	−0.033	0.001	0.001
NCAR SAGE	0.027	0.024	−0.027	−0.026	−0.001	−0.002
SCTM	0.051	0.050	−0.044	−0.043	0.007	0.007
ICSTM	0.043	0.048	−0.040	−0.041	0.004	0.008

Table 2. Adjusted radiative forcing from the UoR model using the NCAR SAGE data, but extending the calculations down to altitudes of 20 km, and comparing the impact of using a higher latitudinal resolution than in Table 1.

	LW (Wm^{-2})	SW (Wm^{-2})	Net (Wm^{-2})
4° latitudinal resolution	0.037	−0.033	0.004
60° latitudinal resolution	0.036	−0.031	0.004
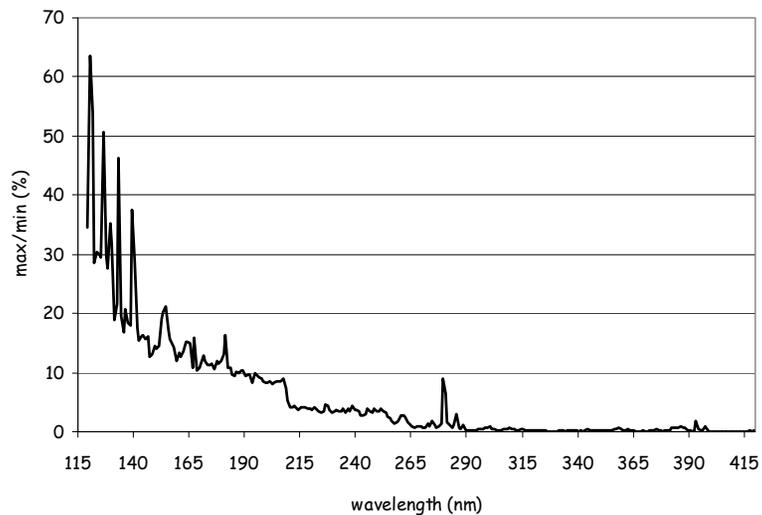
Fig. 1. The 11-year solar cycle variability at wavelengths 115 to 420 nm used in SCTM-1 model based on data from Lean et al. (1997).
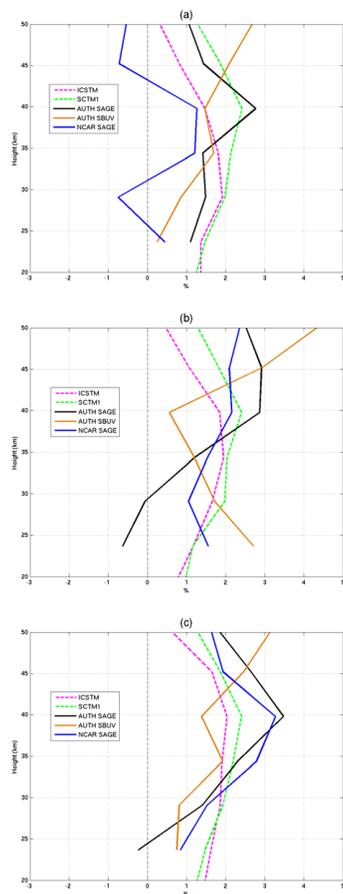
Fig. 2. Area-weighted vertical profiles of the ozone changes (in %) from solar minimum to solar maximum for 3 different latitude bands: **(a)** poleward of 30° S, **(b)** 30° S–30° N, **(c)** poleward of 30° N from 3 sets of satellite data and 2 models. The models are scaled by the factor 120/160 as described in Sect. 2.1.