

Chemical effects in 11-year solar cycle simulations with the Freie Universität Berlin Climate Middle Atmosphere Model with online chemistry (FUB-CMAM-CHEM)

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[1] The impact of 11-year solar cycle variations on stratospheric ozone (O_3) is studied with the Freie Universität Berlin Climate Middle Atmosphere Model with interactive chemistry (FUB-CMAM-CHEM). To consider the effect of variations in charged particle precipitation we included an idealized NO_x source in the upper mesosphere representing relativistic electron precipitation (REP). Our results suggest that the NO_x source by particles and its transport from the mesosphere to the stratosphere in the polar vortex are important for the solar signal in stratospheric O_3 . We find a positive dipole O_3 signal in the annual mean, peaking at 40–45 km at high latitudes and a negative O_3 signal in the tropical lower stratosphere. This is similar to observations, but enhanced due to the idealized NO_x source and at a lower altitude compared to the observed minimum. Our results imply that this negative O_3 signal arises partly via chemical effects.

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1. Introduction

[2] The impact of 11-year solar cycle variations on stratospheric O_3 is still under investigation. Observations show an in-phase variation of total column O_3 with the solar cycle of 1–2% [e.g., Zerefos *et al.*, 1997]. Recent observations suggest a dipolar solar O_3 signal with peaks of +4 to +5% near 60°N and 60°S at 40–45 km and a negative signal of 1–2% at 25–30 km near the equator [Lee and Smith, 2003]. However, uncertainties in the observations arise due to the short record. Modelling studies with 2D-Chemical Transport Models (CTMs) or parameterised O_3 chemistry [e.g., Huang and Brasseur, 1993; Haigh, 1994; Shindell *et al.*, 1999] underestimated the magnitude of the solar O_3 signal at 40–45 km [Hood, 2004]. More recent

chemistry-climate models (CCM) [Tourpali *et al.*, 2003; Rozanov *et al.*, 2004; Egorova *et al.*, 2004] could not simulate the relative minimum in the O_3 signal in the equatorial middle stratosphere.

[3] Different processes were suggested to influence O_3 in the equatorial stratosphere. Dynamical changes in the Brewer-Dobson (BD) circulation between solar min and max [Kodera and Kuroda, 2002] might be associated with changes in the transport of lower equatorial O_3 . An impact of the Quasi-Biennial Oscillation (QBO) and volcanic eruptions on the O_3 solar signal was shown by Lee and Smith [2003]. Another, so far neglected mechanism is variations in charged particle precipitation which influence NO_x chemistry and hence O_3 . The populations of the different types of particles depend on solar activity with opposing effects during a solar cycle. Medium-energy electrons associated with the auroral flux peak around solar max and produce NO_x mainly in the thermosphere, while enhanced populations of high-energy, relativistic electrons are observed near solar min and penetrate during REP events down into the mesosphere [Callis *et al.*, 1991, 2001]. In the wintertime polar vortex the enhanced NO_x may be carried by the BD-circulation into the stratosphere [e.g., Randall *et al.*, 1998], where it destroys O_3 . Energetic particles from solar proton events (SPE) lead to short-time increases in NO_x in the stratosphere, while galactic cosmic rays lead to enhanced NO_x in the lower stratosphere during solar min [Brasseur and Solomon, 1986].

[4] In this study we present the first CCM simulation of the 11-year solar cycle in which the effect of a NO_x source due to solar cycle dependent particle precipitation on O_3 was calculated using an interactive chemistry module. The goal was to address the potential effect of changes in particle precipitation on the solar O_3 signal for a particle type which was shown to vary clearly with the solar cycle, i.e. REP events [Callis *et al.*, 1991, 2001]. Please note that the effects of other particle types are neglected in this sensitivity study but would have to be included in a fully realistic simulation.

[5] Section 2 describes the model and the runs. Section 3 presents the solar annual mean temperature and O_3 response. In Section 4 we present an analysis of chemical effects on the solar signal. Section 5 provides conclusions.

2. Model and Experiments

[6] The simulations were performed with the Freie Universität Berlin Climate Middle Atmosphere Model with

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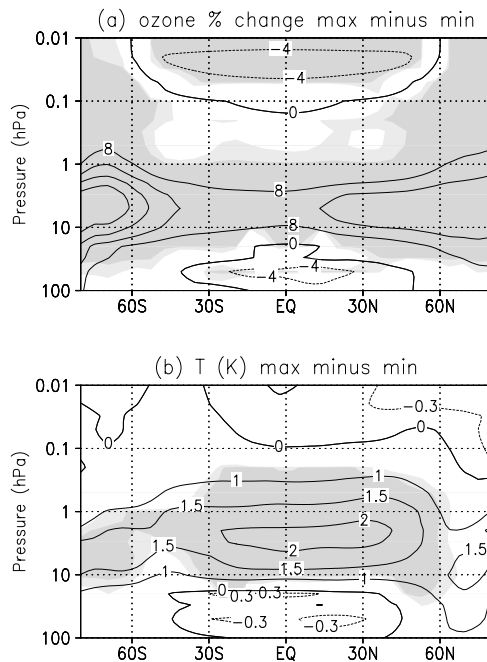


Figure 1. Zonal annual mean change in (a) O₃ [%] and (b) temperature [K] from solar min to max. Shadings denote the 95% and 99% significance levels (Students *t* test).

interactive chemistry (FUB-CMAM-CHEM). The basic model has been described by *Pawson et al.* [1998] and *Langematz* [2000]. The CCM version includes a semi-Lagrangian transport-scheme based on *Böttcher* [1996] and gravity wave drag [*Mieth et al.*, 2004]. The chemistry scheme is based on *Steil et al.* [2003]. To simulate downward fluxes from the thermosphere NO_x features a seasonal and latitude-dependent source polewards of 55° and above 73 km.

[7] The solar cycle simulations included 11-year variations in UV irradiance and in NO_x concentrations resulting from REP. In the chemical photolysis module, the top of atmosphere (TOA) radiative fluxes were modified from solar min to max by 53.87% for the Lyman- α line, and between 9.52% and 0.20% in the UV bands. In the radiation scheme, solar UV flux changes were calculated in 44 bands [*Matthes et al.*, 2004]. The spectral changes were derived from *Lean et al.* [1997]. The total solar irradiance (TSI) was adjusted by 0.1% between solar max and min. To simulate the effect of REP, the NO_x sources at 73–84 km were estimated to be higher (lower) by a factor 2 than mean conditions for the solar min (max) run (based on *Callis et al.* [1991]). Two equilibrium integrations of 15 years each were analyzed at solar min and solar max conditions.

3. Annual Mean O₃ and Temperature Response

[8] Figure 1 shows the simulated annual mean O₃ and temperature change from solar min to max. During solar max, O₃ (Figure 1a) decreases in the upper mesosphere at low and mid latitudes by up to 4% and increases at high latitudes by about 2–4%. Strong changes occur in the stratosphere with significant O₃ increases at solar max of about 10% in the tropics at 5 hPa (37 km) and up to 22% at high latitudes. In the lower tropical stratosphere O₃

decreases at solar max up to 6%. The observed pattern of the O₃ solar signal which shows a dipole structure with positive changes in upper stratosphere mid latitudes and negative changes in the equatorial lower stratosphere [*Lee and Smith*, 2003] is thus well reproduced. However, the simulated amplitude of the stratospheric O₃ signal is exaggerated and the altitude of the negative signal situated too low down. Compared to previous CCM simulations which tended to underestimate the upper stratospheric O₃ response and did not reproduce the weak O₃ signal in the lower/middle stratosphere, our results suggest that the inclusion of particle effects leads to an improvement of the simulated solar ozone signal.

[9] Consistent with the stratospheric O₃ increase at solar max the stratosphere warms significantly by up to 2 K at about 3 hPa (40 km) with a secondary maximum of 1.8 K at southern high latitudes around 7 hPa (35 km). This temperature increase is larger than in previous model simulations which used off-line calculated O₃ changes [*Matthes et al.*, 2003, 2004] or interactive chemistry modules [*Tourpali et al.*, 2003; *Egorova et al.*, 2004; *Rozanov et al.*, 2004]. In the low latitude upper stratosphere the simulated temperature change in our model compares well in magnitude and structure with the observed signal from NCEP/CPC data from 1980 to 1997 [*Hood*, 2004], although considerable uncertainties exist in the observations. *Scaife et al.* [2000] derived a smaller temperature signal of only 0.75 K. In the lower/middle subtropical stratosphere temperature decreases by 0.3 K at solar max in the model, although this is not significant.

4. Effects of Chemical Processes on the Solar Signal

4.1. Mesosphere

[10] In the upper mesosphere O₃ decreases equatorwards of 60° at solar max by 3–4% uniformly throughout the year. Figure 2 implies that this effect is due to the destruction from HO_x chemistry. As shown for northern hemisphere (NH) winter (December/January/February, DJF), H₂O (Figure 2a) decreases significantly in the upper mesosphere by up to 4% from solar min to max and leads to an increase in HO_x [HO_x = H + OH + HO₂] of more than 10% (Figure 2b) which destroys O₃. This result is in good agreement with *Egorova et al.* [2004], and with *Khosravi et al.* [2002], who attribute the negative O₃ signal from 70–80 km of 5–10% in their 2D-model to enhanced Lyman- α photolysis of H₂O. We also calculate rather large increases in H₂O₂ suggesting that increases due to HO₂ + HO₂ which form H₂O₂ at solar max are more important for H₂O₂ than its enhanced photolytic sink (not shown).

4.2. Stratosphere, High Latitudes in Winter

[11] In the mid to upper stratosphere the wintertime polar chemistry in both hemispheres is severely perturbed by the thermospheric NO_x parameterisation. E.g. in northern winter (DJF), the O₃ signal polewards of 60° is about 20% (not shown). This O₃ increase is consistent with a strong decrease of NO_x [NO_x = NO + NO₂ + NO₃ + 2(N₂O₅) + HO₂NO₂] in the whole stratosphere and mesosphere at solar max by more than 50% (Figure 3). At winter polar latitudes at 1 hPa (60 km) NO_x decreases from more than 100 ppbv at

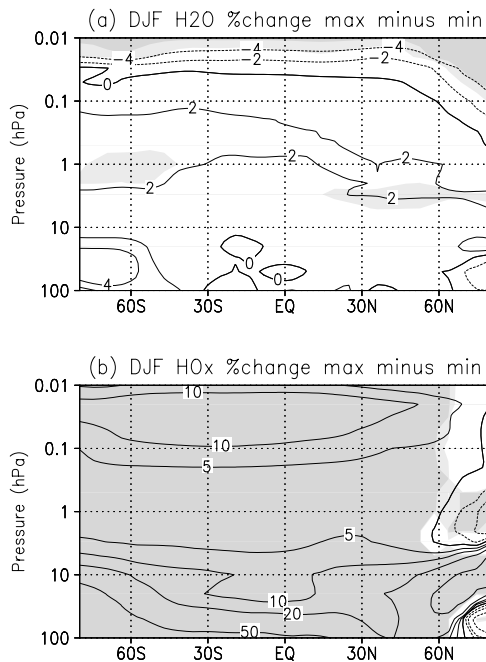


Figure 2. Zonal mean change in (a) H₂O [%] and (b) HO_x(= H + OH + HO₂) [%] in DJF from solar min to max. Shadings denote the 95% and 99% significance levels (Students *t* test).

solar min to 20 ppbv at solar max. NO_x source strengths were modified on the upper three layers (73–84 km) only. Thus, the extension of the strongest NO_x changes at wintertime high latitudes down into the lower stratosphere indicates the importance of downward transport by the BD circulation. Garcia *et al.* [1984] calculated NO_x changes of comparable magnitude due to auroral electrons which maximize during solar max and thus have a compensatory effect. NO_x changes of the order 50–60% extend into northern mid-latitudes in the lower mesosphere. These may be due to meridional transport by planetary waves, as was found by Siskind *et al.* [1997] in HALOE observations. The NO_x reservoirs N₂O₅ and HO₂NO₂, which can build up in the polar night in the absence of photolysis, were also suppressed in the stratosphere at solar max, associated with suppressed thermospheric NO_x sources (not shown).

4.3. Stratosphere, Low to Mid Latitudes

[12] The annual mean O₃ increase of 9–12% (Figure 1a) in the low latitude middle/upper stratosphere at solar max cannot be explained by reduced chemical O₃ destruction alone. While in the annual-mean, a 30% decrease in NO_x at 3 hPa over the equator reduces catalytic O₃ loss, this is balanced by enhanced O₃ destruction due to HO_x, ClO_x and O_x concentrations (not shown). The O₃ increase is rather due to enhanced irradiance at solar max which leads to enhanced O₂ photolysis and O₃ production in the upper stratosphere. This effect is reinforced by the more transparent upper mesosphere which features less O₃ during solar max.

[13] In the lower stratosphere, an enhanced release of O₃-destroying chemicals from their reservoirs dominates the O₃ signal. ClO increases by 20–40% in the lower stratosphere at low latitudes whereas its main reservoirs ClONO₂ and

Table 1. Annual Mean Ozone Loss dO₃/dt (ppt/day) Due to Four Dominant Chemical Reactions at 50 hPa Over the Equator, Calculated From 6 Hourly Chemical Fields

Reaction	dO ₃ /dt (ppt/day)	MIN	MAX	MAX-MIN
O ₃ + HO ₂		−297.74	−375.09	−77.35
NO ₂ + O ³ P		−137.35	−74.25	63.10
OH + HO ₂		−20.52	−26.96	−6.44
ClO + O ³ P		−27.76	−36.31	−8.55

HCl both decrease (not shown). The ClONO₂ decrease is favored directly by low thermospheric NO_x sources at solar max. Similarly HO_x increases by 20–50% (Figure 2b) whereas the HO_x reservoirs HNO₃ and HO₂NO₂ decrease by 20–30% and 10–20% respectively (not shown). J. L. Grenfell *et al.* (Chemical reaction pathways affecting stratospheric and mesospheric ozone, submitted to Journal of Geophysical Research, 2004, hereinafter referred to as Grenfell *et al.*, submitted manuscript, 2004) show that HO_x chemistry is an important chemical regulator of O₃ in the model at equatorial latitudes. Unlike the HO_x and ClO_x family members, NO_x species did not increase at solar max as these are controlled by the lower thermospheric NO_x sources at solar max. The lower NO_x contributes to the negative ozone signal as in the lower stratosphere NO_x in combination with the HO_x cycle causes O₃ production which is opposite to the NO_x effect on O₃ above 10 hPa [Brühl and Crutzen, 2000].

[14] Table 1 shows the annual mean change in O₃ in ppt/day at solar min and max from four chemical reactions which dominate chemical O₃ loss in the model at 50 hPa over the equator (Grenfell *et al.*, submitted manuscript, 2004) where the negative signal is strongest. During solar max an enhanced daily chemical O₃ destruction of $-77.35 + 63.10 - 6.44 - 8.55 = -29.24$ ppt/day occurs which is dominated by HO_x chemistry. This result suggests that chemical destruction contributes to the simulated O₃ decrease in the lower tropical stratosphere during solar max.

[15] In addition, the enhanced O₃ in the middle/upper stratosphere leads to stronger absorption of solar irradiance and increased short-wave heating rates of 0.2–0.4 K/day between 10 and 1 hPa (30–50 km) at solar max (not shown). As a result, less irradiance is available at the levels below for photolytic O₃ production.

5. Conclusions

[16] (1) We highlight the potential importance of the high-latitude thermospheric/mesospheric NO_x sources due

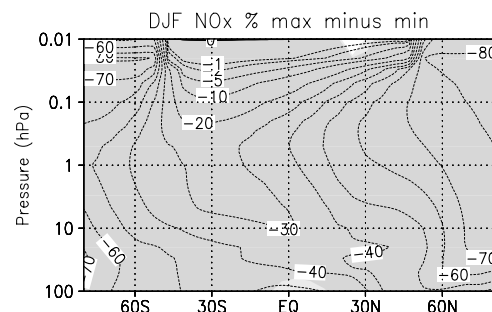


Figure 3. As Figure 2 but for NO_x (=NO + NO₂ + NO₃ + 2(N₂O₅) + HO₂NO₂) [%].

to variations in particle precipitation for the 11-year solar signal in O₃. Future work is required to quantify more accurately the various source strengths and chemical mechanisms. Including the REP mechanism which leads to NO_x enhancement during solar min reproduces the characteristic dipole in the solar O₃ signal of the middle atmosphere, although the magnitude is over-estimated. The inclusion of the other particle events which partially oppose the REP effect should lead to a more quantitative agreement of our model results with observations.

[17] (2) We confirm the importance of the downward transport of the thermospheric/mesospheric NO_x sources into the stratosphere in winter. The reduced NO_x sources during solar max lead to less catalytic O₃ destruction, and hence to an O₃ increase in the stratosphere.

[18] (3) We reproduce the negative O₃ signal observed at equatorial latitudes. This is associated at least partially with a chemical effect in which O₃-destroying chemicals are enhanced at solar max because their NO_x containing reservoirs are decreased. In the real atmosphere this chemical mechanism may operate in conjunction with the dynamical theories affecting O₃ in the lower stratosphere, like changes in the BD circulation and QBO interactions. A quantitative assessment of the chemical and dynamical contributions to the solar signal is deferred to a future simulation including a QBO.

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