

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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Received 17 February 2005; revised 22 April 2005; accepted 31 May 2005; published 2 July 2005.

[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.

Citation: Langematz, U., J. L. Grenfell, K. Matthes, P. Mieth, M. Kunze, B. Steil, and C. Brühl (2005), Chemical effects in 11-year solar cycle simulations with the Freie Universität Berlin Climate Middle Atmosphere Model with online chemistry (FUB-CMAM-CHEM), *Geophys. Res. Lett.*, 32, L13803, doi:10.1029/2005GL022686.

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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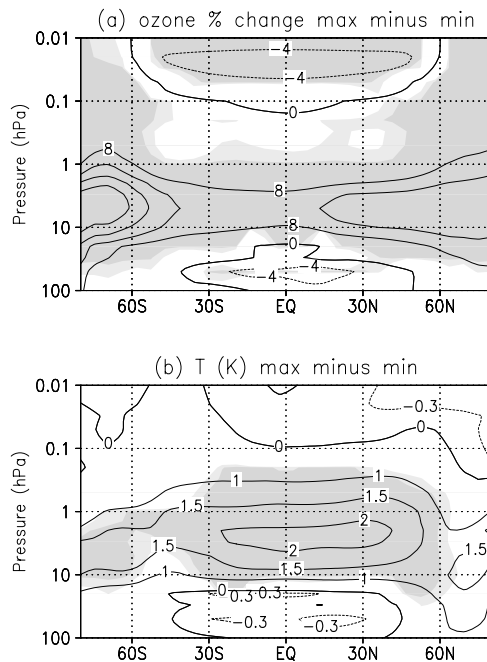


Figure 1. Zonal annual mean change in (a) O_3 [%] and (b) temperature [K] from solar min to max. Shadings denote the 95% and 99% significance levels (Students t test).

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 O_3 and Temperature Response

[8] Figure 1 shows the simulated annual mean O_3 and temperature change from solar min to max. During solar max, O_3 (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 O_3 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 O_3

decreases at solar max up to 6%. The observed pattern of the O_3 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 O_3 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 O_3 response and did not reproduce the weak O_3 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 O_3 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 O_3 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 O_3 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), H_2O (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 [$HO_x = H + OH + HO_2$] of more than 10% (Figure 2b) which destroys O_3 . This result is in good agreement with *Egorova et al.* [2004], and with *Khosravi et al.* [2002], who attribute the negative O_3 signal from 70–80 km of 5–10% in their 2D-model to enhanced Lyman- α photolysis of H_2O . We also calculate rather large increases in H_2O_2 suggesting that increases due to $HO_2 + HO_2$ which form H_2O_2 at solar max are more important for H_2O_2 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 O_3 signal polewards of 60° is about 20% (not shown). This O_3 increase is consistent with a strong decrease of NO_x [$NO_x = NO + NO_2 + NO_3 + 2(N_2O_5) + HO_2NO_2$] 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

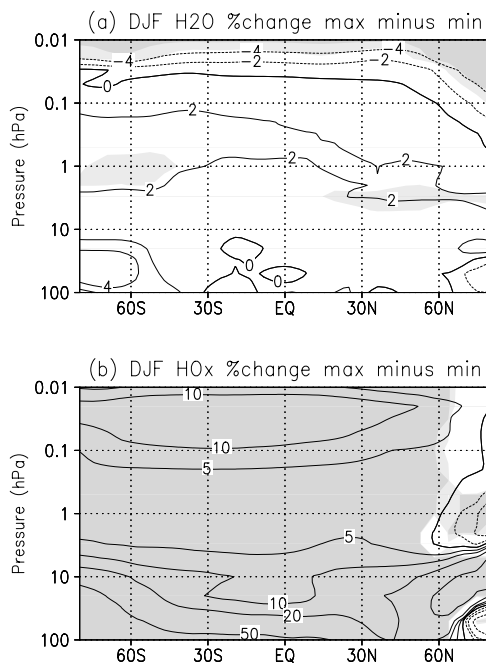


Figure 2. Zonal mean change in (a) H₂O [%] and (b) HO_x(= H + OH + HO₂) [%] in DJF from solar min to max. Shadings denote the 95% and 99% significance levels (Students *t* test).

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

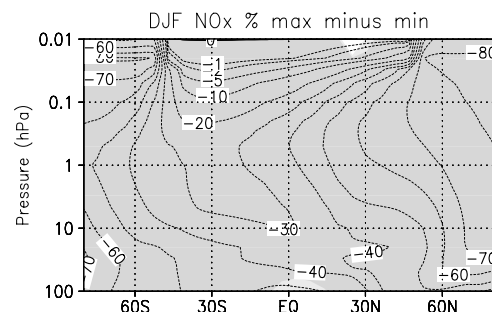


Figure 3. As Figure 2 but for NO_x (=NO + NO₂ + NO₃ + 2(N₂O₅) + HO₂NO₂) [%].

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.

[19] **Acknowledgments.** We thank John McConnell, Cora Randall, and Kleareti Tourpali for useful discussions. JLG was supported under the project MESA (01 LG 0001) funded by the German Ministry for Research and Education, KM and MK under the European project SOLICE (EVK2-2002-00543). The GCM simulations were performed at the Konrad-Zuse-Zentrum für Informationstechnik, Berlin.

References

- Böttcher, M. (1996), A semi-Lagrangian advection scheme with modified exponential splines, *Mon. Weather Rev.*, *124*, 716–729.
- Brasseur, G., and S. Solomon (1986), *Aeronomy of the Middle Atmosphere*, 452 pp., Springer, New York.
- Brühl, C., and P. J. Crutzen (2000), NO_x-catalyzed ozone destruction and NO_x activation at midlatitudes to high latitudes as a main cause of the spring to fall ozone decline in the Northern Hemisphere, *J. Geophys. Res.*, *105*, 12,163–12,168.
- Callis, L. B., D. N. Baker, J. B. Blake, J. D. Lambeth, R. E. Boughner, M. Natarajan, R. W. Klebesadel, and D. J. Gorney (1991), Precipitating relativistic electrons: Their long-term effect on stratospheric odd nitrogen levels, *J. Geophys. Res.*, *96*, 2939–2976.
- Callis, L. B., M. Natarajan, and J. D. Lambeth (2001), Solar-atmospheric coupling by electrons (SOLACE): 3. Comparisons of simulations and observations, 1979–1997, issues and implications, *J. Geophys. Res.*, *106*, 7523–7539.
- Egorova, T., E. Rozanov, E. Manzini, M. Haberreiter, W. Schmutz, V. Zubov, and T. Peter (2004), Chemical and dynamical response to the 11-year variability of the solar irradiance simulated with a chemistry-climate model, *Geophys. Res. Lett.*, *31*, L06119, doi:10.1029/2003GL019294.
- Garcia, R. R., S. Solomon, R. G. Noble, and D. W. Rusch (1984), A numerical response of the middle atmosphere to the 11-year solar cycle, *Planet Space Sci.*, *32*, 423–441.
- Haigh, J. D. (1994), The role of stratospheric ozone in modulating the solar radiative forcing of climate, *Nature*, *370*, 544–546.
- Hood, L. L. (2004), Effects of solar UV variability on the stratosphere, in *Solar Variability and Its Effects on Climate*, *Geophys. Monogr. Ser.*, edited by J. Pap and P. Fox, pp. 283–304, AGU, Washington, D.C.
- Huang, T. Y. W., and G. P. Brasseur (1993), Effect of long-term solar variability in a two-dimensional interactive model of the middle atmosphere, *J. Geophys. Res.*, *98*, 20,413–20,427.
- Khosravi, R., G. Brasseur, A. Smith, D. Rusch, S. Walters, S. Chabrilat, and G. Kockarts (2002), Response of the mesosphere to human-induced perturbations and solar variability calculated by a 2-D model, *J. Geophys. Res.*, *107*(D18), 4358, doi:10.1029/2001JD001235.
- Kodera, K., and Y. Kuroda (2002), Dynamical response to the solar cycle, *J. Geophys. Res.*, *107*(D24), 4749, doi:10.1029/2002JD002224.
- Langematz, U. (2000), An estimate of the impact of observed ozone losses on stratospheric temperature, *Geophys. Res. Lett.*, *27*, 2077–2080.
- Lean, J. L., G. J. Rottman, H. L. Kyle, T. N. Woods, J. R. Hickey, and L. C. Puga (1997), Detection and parameterization of variations in solar mid- and near-ultraviolet radiation (200–400 nm), *J. Geophys. Res.*, *102*, 29,939–29,956.
- Lee, H., and A. K. Smith (2003), Simulation of the combined effects of solar cycle, quasi-biennial oscillation, and volcanic forcing on stratospheric ozone changes in recent decades, *J. Geophys. Res.*, *108*(D2), 4049, doi:10.1029/2001JD001503.
- Matthes, K., K. Kodera, J. D. Haigh, D. T. Shindell, K. Shibata, U. Langematz, E. Rozanov, and Y. Kuroda (2003), GRIPS Solar Experiments Intercomparison Project: Initial results, *Pap. Meteorol. Geophys.*, *54*, 71–90.
- Matthes, K., U. Langematz, L. L. Gray, K. Kodera, and K. Labitzke (2004), Improved 11-year solar signal in the Freie Universität Berlin Climate Middle Atmosphere Model (FUB-CMAM), *J. Geophys. Res.*, *109*, D06101, doi:10.1029/2003JD004012.
- Mieth, P., J. L. Grenfell, U. Langematz, and M. Kunze (2004), Sensitivity of the Berlin Climate Middle Atmosphere Model (FUB-CMAM) to different gravity-wave drag parameterisations, *Ann. Geophys.*, *22*, 1–21.
- Pawson, S., U. Langematz, G. Radek, U. Schlese, and P. Strauch (1998), The Berlin troposphere-stratosphere-mesosphere GCM: Sensitivity to physical parametrizations, *Q. J. R. Meteorol. Soc.*, *124*, 1343–1371.
- Randall, C. E., D. W. Rusch, R. M. Bevilacqua, K. W. Hoppel, and J. D. Lumpe (1998), Polar Ozone and Aerosol Measurement (POAM) II stratospheric NO₂, 1993–1996, *J. Geophys. Res.*, *103*, 28,361–28,371.
- Rozanov, E. V., M. E. Schlesinger, T. A. Egorova, B. Li, N. Andronova, and V. A. Zubov (2004), Atmospheric response to the observed increase of solar UV radiation from solar minimum to solar maximum simulated by the University of Illinois at Urbana-Champaign climate-chemistry model, *J. Geophys. Res.*, *109*, D01110, doi:10.1029/2003JD003796.
- Scaife, A. A., J. Austin, N. Butchart, S. Pawson, M. Keil, J. Nash, and I. N. James (2000), Seasonal and interannual variability of the stratosphere diagnosed from UKMO TOVS analyses, *Q. J. R. Meteorol. Soc.*, *126*, 2585–2604.
- Shindell, D., D. Rind, N. Balachandran, J. Lean, and P. Lonergan (1999), Solar cycle variability, ozone and climate, *Science*, *284*, 305–308.
- Siskind, D. E., J. T. Bacmeister, M. E. Summers, and J. M. Russell III (1997), Two dimensional model calculations of nitric oxide transport in the middle atmosphere and comparison with Halogen Occultation Experiment data, *J. Geophys. Res.*, *102*, 3527–3545.
- Steil, B., C. Brhl, E. Manzini, P. J. Crutzen, J. Lieleveld, P. J. Rasch, E. Roeckner, and K. Krger (2003), A new interactive chemistry-climate model: 1. Present-day climatology and interannual variability of the middle atmosphere using the model and 9 years of HALOE/UAARS data, *J. Geophys. Res.*, *108*(D9), 4290, doi:10.1029/2002JD002971.
- Tourpali, K., C. J. E. Schuurmans, R. van Dorland, B. Steil, and C. Brhl (2003), Stratospheric and tropospheric response to enhanced solar UV radiation: A model study, *Geophys. Res. Lett.*, *30*(5), 1231, doi:10.1029/2002GL016650.
- Zerefos, C. S., K. Tourpali, B. R. Bojkov, D. S. Balis, B. Rognerund, and I. S. A. Isaksen (1997), Solar activity-total column ozone relationships: Observations and model studies with heterogeneous chemistry, *J. Geophys. Res.*, *102*, 1561–1570.

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