



RESEARCH LETTER

10.1002/2016GL069958

Key Points:

- Updated SAGE v7.0 data shows improved O3-T anti-correlation in the upper stratosphere
- Reduced solar signal in the upper stratosphere in SAGE v7.0 data is consistent with model and HALOE data
- Large uncertainties in observational (and reanalysis) data, hinders establishing the true nature of solar signal in the stratospheric ozone

Supporting Information:

- Supporting Information S1
- Figure S1
- Figure S2

Correspondence to:

S. S. Dhomse,
S.S.Dhomse@leeds.ac.uk

Citation:

Dhomse, S. S., M. P. Chipperfield, R. P. Damadeo, J. M. Zawodny, W. T. Ball, W. Feng, R. Hossaini, G. W. Mann, and J. D. Haigh (2016), On the ambiguous nature of the 11 year solar cycle signal in upper stratospheric ozone, *Geophys. Res. Lett.*, 43, 7241–7249, doi:10.1002/2016GL069958.

Received 8 JUN 2016

Accepted 13 JUN 2016

Accepted article online 16 JUN 2016

Published online 12 JUL 2016

©2016. The Authors.

This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

On the ambiguous nature of the 11 year solar cycle signal in upper stratospheric ozone

S. S. Dhomse^{1,2}, M. P. Chipperfield^{1,2}, R. P. Damadeo³, J. M. Zawodny³, W. T. Ball⁴, W. Feng^{1,5}, R. Hossaini¹, G. W. Mann^{1,5}, and J. D. Haigh⁶

¹School of Earth and Environment, University of Leeds, Leeds, UK, ²National Centre for Earth Observation, University of Leeds, Leeds, UK, ³NASA Langley Research Center, Hampton, Virginia, USA, ⁴PMOD/WRC, Davos, Switzerland, ⁵National Centre for Atmospheric Science, University of Leeds, Leeds, UK, ⁶Grantham Institute and Blackett Laboratory, Imperial College, London, UK

Abstract Up to now our understanding of the 11 year ozone solar cycle signal (SCS) in the upper stratosphere has been largely based on the Stratospheric Aerosol and Gas Experiment (SAGE) II (v6.2) data record, which indicated a large positive signal which could not be reproduced by models, calling into question our understanding of the chemistry of the upper stratosphere. Here we present an analysis of new v7.0 SAGE II data which shows a smaller upper stratosphere ozone SCS, due to a more realistic ozone-temperature anticorrelation. New simulations from a state-of-art 3-D chemical transport model show a small SCS in the upper stratosphere, which is in agreement with SAGE v7.0 data and the shorter Halogen Occultation Experiment and Microwave Limb Sounder records. However, despite these improvements in the SAGE II data, there are still large uncertainties in current observational and meteorological reanalysis data sets, so accurate quantification of the influence of solar flux variability on the climate system remains an open scientific question.

1. Introduction

Over the 11 year solar cycle, although total solar irradiance (TSI) changes by less than 0.1%, flux changes of up to 100% occur in the ultraviolet (UV) region of the solar spectrum. Hence, the so-called “top-down” mechanism is widely accepted as a plausible explanation for solar-climate interaction [e.g., *Gray et al.*, 2010]. In this mechanism, increased UV radiation enhances ozone production via O₂ photolysis, thereby altering middle atmosphere (stratosphere and mesosphere) temperatures. This modifies the upward propagation of planetary waves [*Kodera and Kuroda*, 2002], which drive the middle atmospheric circulation, and also influence the tropospheric circulation [e.g., *Haigh*, 1996].

Consequently, this mechanism suggests that the direct effect of solar flux changes should be detectable in the ozone profile, especially in the upper stratosphere. However, accurate quantification of the impact of the 11 year solar variability on stratospheric ozone (and hence on climate) is very challenging. First, stratospheric ozone concentrations are also controlled by various chemical and dynamical processes such as the quasi-biennial oscillation (QBO), the meridional circulation, and stratospheric aerosol, some of which are nonlinearly coupled [e.g., *Holton and Tan*, 1980; *World Meteorological Organization*, 2015]. Second, there are very few good quality satellite-based ozone profile observations with retrieval errors less than the 11 year solar cycle signal (SCS). Third, large volcanic eruptions during the declining phases of solar maxima, such as El Chichon and Mount Pinatubo, could have caused aliasing effects [e.g., *Lee and Smith*, 2003]. Fourth, modeling the effects of solar variability on climate is also difficult due to large uncertainties in the solar flux measurements [e.g., *Ermolli et al.*, 2013].

Despite these difficulties, our present understanding of the SCS in stratospheric ozone is that it has a “double-peak” structure with maxima in the tropical lower (~22 km) and upper stratosphere (~50 km) and a negligible SCS in the tropical middle stratosphere. This understanding is based on the analysis of the longest record of ozone profile data (October 1984 to August 2005) from the Stratospheric Aerosol and Gas Experiment (SAGE) II satellite instrument [e.g., *Soukharev and Hood*, 2006; *Randel and Wu*, 2007].

An analysis of another high-quality, but relatively short (1991–2005), satellite data set from the Halogen Occultation Experiment (HALOE) instrument also shows a double-peaked SCS, but the primary peak is shifted upward to the mid stratosphere (35 km altitude) with a second peak in the lower mesosphere [e.g., *Remsberg*, 2008]. Also, HALOE data do not show any significant ozone SCS in the upper stratosphere (~45 km). *Dhomse et al.* [2011] showed that the estimated SCS using a 3-D chemical transport model (CTM) forced with National

Research Laboratory (NRL) solar fluxes showed better agreement with the HALOE-derived SCS than the one derived from the then current SAGE v6.2 data. Furthermore, 18 models participated in Chemistry-Climate Model Validation Activity Phase 2 for chemistry-climate models with some having sophisticated representations of solar flux variability. However, none of these models could simulate a large positive SCS in upper stratospheric ozone (see Figure 8.11 in *Stratospheric Processes and their Role in Climate (SPARC)* [2010]). Hence, in general, the inability of models to simulate “SAGE (v6.2)” and “solar backscatter ultraviolet (SBUV)-type” SCS in the upper stratospheric ozone has been generally attributed to deficiencies in model chemistry and dynamics [e.g., *SPARC, 2010 (Chapter 8); Hood et al., 2015*]. In contrast, most of the models simulated smaller “HALOE-type” SCS.

Additional complications arise from the fact that recent solar flux measurements from the Solar Radiation and Climate Experiment (SORCE) satellite show significantly different UV variability [Ermolli et al., 2013]. Recent observations suggest that solar cycle 24 (2008 onward) has the smallest number of sunspots in the last 100 years (e.g., <http://solarscience.msfc.nasa.gov/predict.shtml>). Some studies such as Haigh et al. [2010], Merkel et al. [2011], and Swartz et al. [2012] used earlier versions of SORCE solar fluxes in chemical models and produced a negative SCS in the upper stratosphere/lower mesosphere, which showed reasonable agreement with Microwave Limb Sounder (MLS) and Sounding of the Atmosphere using Broadband Emission Radiometry (SABER)-derived SCS in stratospheric ozone. Hence, they suggested that the stratospheric ozone SCS during recent solar cycle 23 (1996–2008) was significantly different to previous solar cycles and can only be simulated if SORCE solar fluxes are used. However, using a 3-D CTM with either SORCE or NRL-solar spectral irradiance (SSI) solar fluxes, Dhomse et al. [2013] could simulate ozone changes that agreed reasonably well with the MLS and SABER data over this period. There is also now some evidence that SORCE SSI strongly overestimates UV solar cycle variability [Ermolli et al., 2013].

The SCS in the upper stratosphere/lower mesosphere is attributed to photochemistry while that in the mid-lower stratosphere is attributed to dynamical coupling. However, there is a lack of understanding of even the photochemical (upper stratospheric/lower mesospheric) SCS during earlier solar cycles (21, 22, and 23). Here we use the TOMCAT/SLIMCAT 3-D CTM to simulate long-term ozone changes using different solar fluxes and realistic meteorological data. We also present the estimated SCS profile using different satellite instrument (SAGE II v6.2 and v7.0, HALOE v19, and MLS v4.2) data sets. In particular, we investigate differences between the upper stratospheric SCS from SAGE II and other satellite data sets. A brief description of the satellite data is provided in the supporting information.

2. Model Simulations

We use the TOMCAT/SLIMCAT 3-D CTM [Chipperfield, 1999, 2006] at a horizontal resolution of $5.6^\circ \times 5.6^\circ$ with 32 σ - p levels from the surface to ~ 60 km. The model includes a detailed stratospheric chemistry and is forced with 6-hourly ERA-Interim reanalysis data. The model setup used for the control simulation is similar to our recent studies such as Chipperfield et al. [2015], Dhomse et al. [2015], and Hossaini et al. [2015]. Here we present results from three simulations with different solar fluxes with prescribed stratospheric aerosol fields. In run A_NRL the model used solar fluxes from the NRL-SSI model and aerosol surface area density data from Arfeuille et al. [2013]. As NRL fluxes are only available until 2011, we extended them until 2013 by regressing them on the $F_{10.7\text{ cm}}$ flux for each spectral bin. Runs B_SAT and C_SOR were similar to A_NRL but used Spectral and Total Irradiance Reconstructions (SATIRE) [Yeo et al., 2014] and SORCE solar fluxes extrapolated to cover the analysis period (eSORCE; see the supporting information), respectively. Importantly, eSORCE is constructed using SOLSTICE v13 and SIM v20 at wavelengths less than/greater than 247 nm, respectively, as described by Ball et al. [2016]. In absolute terms, NRL TSI is about 4 Wm^{-2} higher [Kopp and Lean, 2011] than either SATIRE or eSORCE (1361 Wm^{-2}) so all the fluxes are scaled to identical long-term (1979–2013) mean TSI (1365 Wm^{-2}).

3. Results and Discussion

3.1. Ozone in the Upper Stratosphere

Figure 1 compares tropical (20°S – 20°N) monthly mean ozone volume mixing ratios (VMRs) from the model simulations and satellite data (SAGE II: 1985–2005, HALOE: 1991–2005, and Atmospheric Chemistry Experiment (ACE): 2004–2013) at 35, 40, 45, and 50 km. The modeled time series are shown after removing the effects of

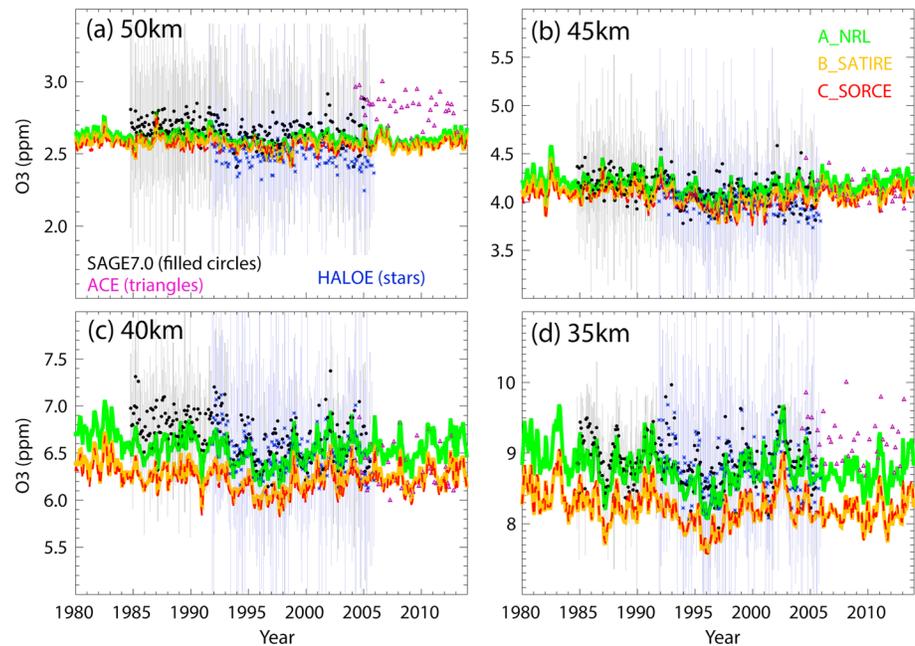


Figure 1. Tropical (20°S – 20°N) monthly mean ozone anomalies (ppm) from SAGE II v7.0 (black), HALOE v19 (blue), and ACE v3.5 (indigo) at (a) 50 km, (b) 45 km, (c) 40 km, and (d) 35 km. To reveal absolute differences, the long-term mean value is added back to each time series. Monthly mean values from ACE, HALOE, and SAGE II are calculated by combining both sunrise and sunset measurements. The vertical error bars (light grey) indicate the root-mean-square errors of all the SAGE and HALOE observations at a given altitude. Ozone VMRs from model runs A_NRL (green), B_SAT (orange), and C_SOR (red) are also shown.

inhomogeneities in ERA-Interim data after 1998 using a step function in a regression model with 12-monthly harmonics [see *Dhomse et al.*, 2011].

Overall, the modeled ozone shows little difference between the simulations, although at the altitudes plotted values from run C_SOR are lower than A_NRL and B_SAT. Although modeled ozone does show some biases against satellite data, there are significant biases in the observational data itself. Detailed analysis of the differences between SAGE v6.2 and SAGE v7.0 are discussed in *Damadeo et al.* [2013]. The number of profiles used to calculate tropical monthly mean and calculated monthly means using SAGE v6.2 and v7 is also shown in Figures S1 and S2 in the supporting information. Note that monthly means are calculated only if there are more than 20 profiles in a given latitude band (20°S – 20°N) with retrieval errors less than 300%. Additionally, HALOE and SAGE data show significant biases in ozone concentrations in the upper stratosphere with an anomaly from mid-1993 to mid-1994. This anomaly is the consequence of using monthly mean values in the presence of diurnal variability as the sunset measurements were disproportionately affected by SAGE II scan-mirror adjustments due to a battery problem in 1993–1994, causing uneven sampling. At 45 km there is better agreement between model and observations. During the overlap period much reduced biases are observed between HALOE and SAGE II (1992–2005).

A key feature in Figure 1 is that the monthly mean data from occultation instruments (ACE, HALOE, and SAGE) show much larger variability than the model. This is likely due to the limited spatial and temporal coverage of the occultation instruments and the variable sampling of the diurnal as well as seasonal cycle (see *Damadeo et al.* [2014] and Figure S1), as well as the large dynamical variability in ozone. Again, this suggests difficulties in establishing the true nature of the SCS when using a limited and unevenly sampled number of ozone profiles, even if the data are of high quality. Another important aspect of Figure 1 is that in absolute terms, at these altitudes A_NRL seems to show better agreement with most of satellite data, whereas both B_SAT and C_SOR are about 5% lower than A_NRL. There are negligible ozone differences between B_SAT and C_SOR.

As noted above, using a 3-D model *Dhomse et al.* [2011] could simulate a positive SCS (SAGE-SBUV type) in upper stratospheric ozone only if they used unrealistic meteorology, due to the temperature dependence of ozone destruction rates. For more details on the well-known ozone-temperature inverse relationship in the upper stratosphere see *Stolarski et al.* [2012], and references therein. In Figure 2 we analyze the representation

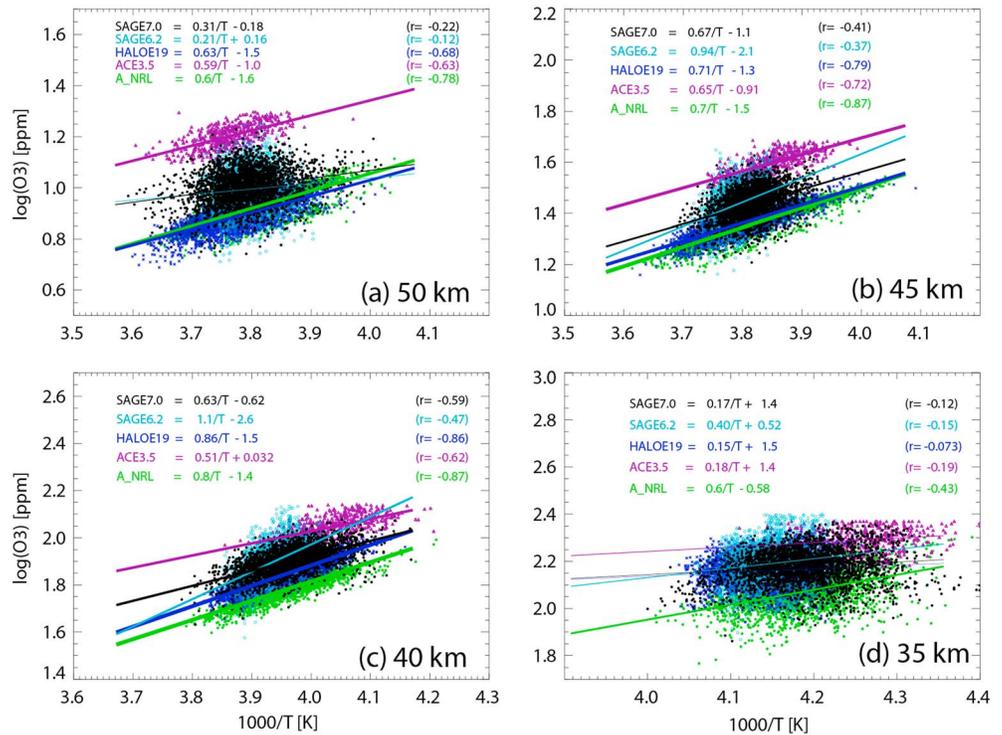


Figure 2. Variation of $\log(O_3)$ VMR versus $1000/T$ at (a) 50 km, (b) 45 km, (c) 40 km, and (d) 35 km for tropical (20°S – 20°N) sunset measurements from ACE (2006–2010), HALOE (1996–2000), and SAGE II (v7.0 and v6.2, 1996–2000). The model sunset values from run A_NRL sampled at SAGE II co-locations for the 1996–2000 time period are also shown. The legends indicate the parameters to the linear regression fits; the thicknesses of the regression lines are scaled to the magnitude of the correlation coefficients (r).

of this relationship in individual ACE, HALOE, and SAGE (v6.2 and v7.0) sunset profile data at 50, 45, 40, and 35 km. Data are shown for a 5 year period (1996–2000 for HALOE, SAGE, and model as well as 2006–2010 for ACE) to avoid complications of the correlation changing with time, e.g., due to changing atmospheric chlorine.

Overall, ozone and ERA-Interim temperature from the model, ACE, and HALOE show a significant negative correlation above 35 km. However, notable differences occur in SAGE II data sets. SAGE v6.2 shows only a weak T dependence at 50 km, but strong values at 45 and 40 km. Nevertheless, in all cases the correlation values are small. In contrast, SAGE v7.0 shows T dependencies, which are more consistent with the other observations and the model with larger correlations than v6.2, thereby enhancing our confidence in this newer version of SAGE II data. However, there are some inhomogeneities in Modern Era Retrospective Analysis for Research and Applications upper stratospheric temperature (used for SAGE processing) due to drifting of the NOAA satellite platform carrying the stratospheric sounding unit (SSU) instruments [Wang *et al.*, 2012], causing relatively small correlations even for SAGE v7.0 data. The effects of drifts in SSU temperatures on the SCS estimation are also discussed in Mitchell *et al.* [2014].

3.2. Ozone Solar Response

We now analyze the solar response in tropical stratospheric ozone using a multivariate linear regression (MLR) analysis. Damadeo *et al.* [2014] discussed the potential pitfalls in applying MLR to sparsely sampled data sets that have trends in sampling (e.g., from precession and degradation of orbit). The non-uniform temporal, spatial, and diurnal sampling aliases into the various terms and impacts the regression analysis. For these reasons, to analyze the SAGE II data we apply the model from that study. For the other data sets we use a slightly adapted MLR model to that used in Dhomse *et al.* [2011]. Briefly, this model contains the effective equivalent stratospheric chlorine (EESC) loading, two QBO terms (30 hPa and 10 hPa), $F_{10.7\text{ cm}}$ flux, ENSO index, and stratospheric aerosol loading (as in Dhomse *et al.* [2008]). EESC and QBO terms are represented by 12, 6, 4, and 3 month harmonics.

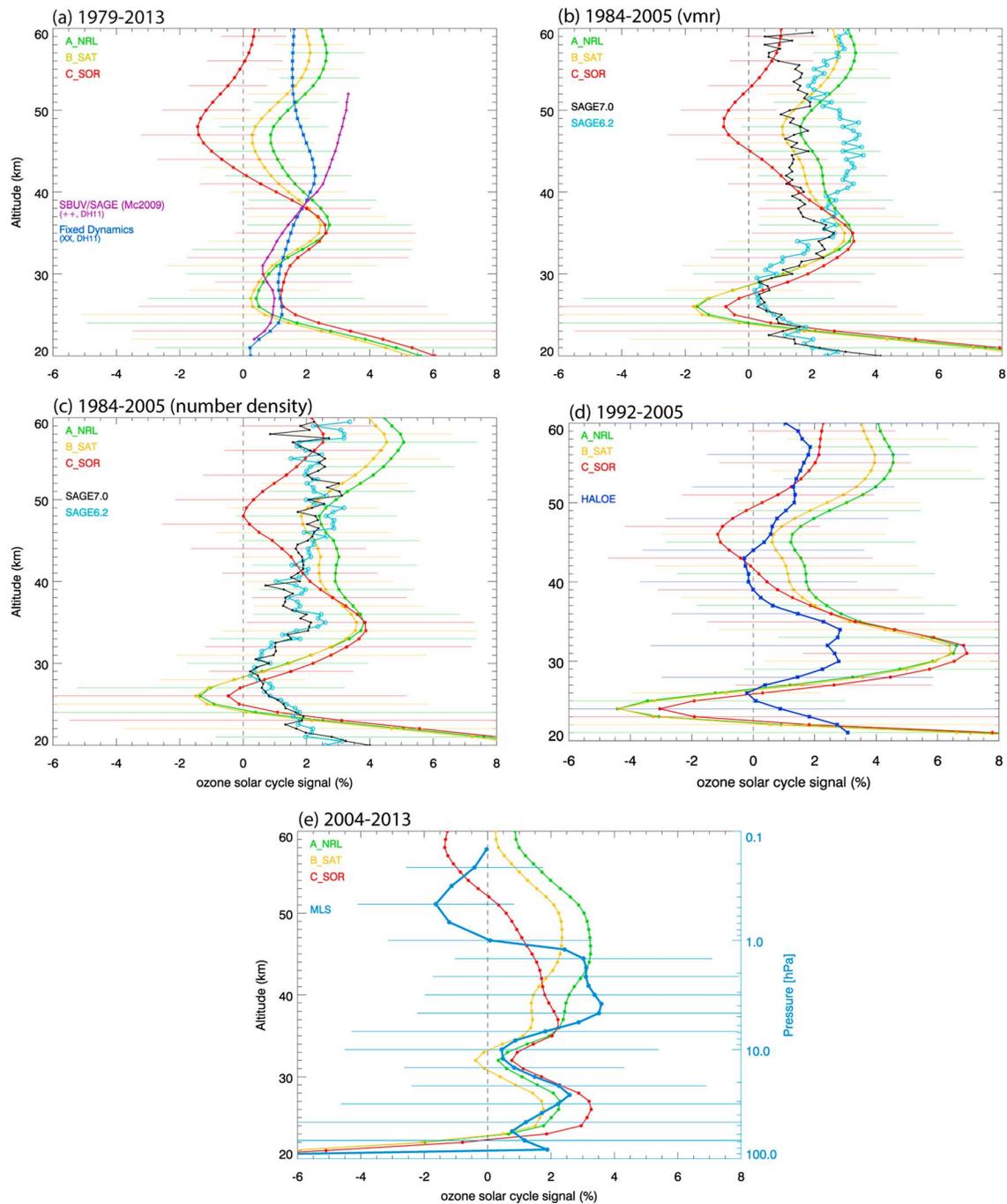


Figure 3. (a) Vertical profiles of the regression coefficients or estimated solar cycle signal (SCS) in stratospheric ozone VMR from runs A_NRL (green), B_SAT (orange), and C_SOR (red) for 1979–2013 period. The estimated SCS using SBUV/SAGE data (1979–2005 [from *McLinden et al., 2009*]) and a 3-D model simulation with fixed dynamics, as presented in *Dhomse et al. [2011]*, are also shown. The horizontal lines show 2- σ error bars. To improve the clarity error bars are shown every 4 km. (b and c) Estimated SCS from SAGE II (1984–2005) v7.0 (black) and v6.2 (light blue) data calculated from VMR (solid lines) and number densities (dashed lines), respectively, using the recently developed regression model described in *Damadeo et al. [2014]*. No error bars are shown for SAGE data. The modeled VMR and number density SCS for the 1984–2005 period are also shown. (d) Same as Figure 3a but for HALOE v19 (blue) and model VMR data for 1992–2005. (e) Same as Figure 3a but for MLS (light blue) and model data for 2004–2013.

The estimated SCS from satellite data set and model simulations are shown in Figure 3. A typical double-peak-structured SCS derived from SAGE-based data [*Randel and Wu, 2007*] and SBUV/SAGE data [*McLinden et al., 2009*] is shown in Figure 3a. The estimated SCSs from runs A_NRL and B_SAT are slightly different to the equivalent runs presented in *Dhomse et al. [2011]* due to an updated photolysis scheme in the model

and use of ERA-Interim forcing fields. Although this study is focused on the upper stratosphere, we can note that there is significant reduction in the lower stratospheric SCS in all the simulations, as the effect of volcanic aerosol on estimation of SCS is less than 2% in our present simulations due to improvements in simulating chemical ozone changes after the volcanic eruption [Dhomse *et al.*, 2015]. However, it is also important to note that radiative heating and subsequent changes in Brewer-Dobson (BD) circulation are probably not well simulated in ERA-Int due to the lack of a detailed aerosol microphysics module [e.g., Dhomse *et al.*, 2014] in the European Centre for Medium-Range Weather Forecasts reanalysis system.

An important point to note from Figure 3a is that even with identical (ERA-interim) dynamical forcing, there are significant differences in SCS from the updated model and satellite-based (SBUV/SAGE) ozone profiles in the upper stratosphere. All model simulations show a small (or even negative in case of C_SOR) SCS near 45–50 km (stratopause), whereas the SBUV/SAGE signals are +2–4% at this altitude. Interestingly, although there appear to be significant differences in A_NRL and B_SAT ozone concentrations shown in Figure 1, the estimated SCS from B_SAT is almost similar to A_NRL. The large negative SCS simulated by C_SOR near 50 km is different to any other model run or satellite-based estimate for this period. This analysis clearly shows that via photochemistry, NRL and SATIRE solar fluxes give nearly similar SCS, but eSORCE solar fluxes give more negative SCS.

Figures 3b and 3c show the estimated SCS for SAGE II v6.2 and v7.0 for the 1984–2005 period derived when SAGE measurements are converted to VMR or native number density observations are used. Note that the regression model used for SAGE II data is the one described in Damadeo *et al.* [2014]. Above 30 km, the SCS from v7.0 number densities is about 0.5% less than v6.2, which improves the agreement with the model. However, when converted to VMRs the SCS differences diverge to more than 2%. This shows that even with same measurements with different retrieval algorithms can give significantly different SCS in stratospheric ozone and that conversion between number density and VMR needs to be performed carefully. Overall, the estimated SCSs from both A_NRL and B_SAT show much better agreement with the SAGE v7.0 data when VMRs are used and are significantly different to the v6.2 data, which must be due to various improvements in SAGE v7.0 data [Damadeo *et al.*, 2013] and use of the more sophisticated regression model [Damadeo *et al.*, 2014]. A key feature of Figure 3b is that if we use VMR, the estimated SCSs from both A_NRL and B_SAT show remarkable agreement with SAGE v7.0 data throughout the midupper stratosphere.

Figures 3d and 3e show the estimated SCS in the tropical stratosphere from HALOE (1992–2005) and Aura-MLS (2004–2013) data sets, respectively. Again, there is better agreement in estimated SCS from HALOE, A_NRL, and B_SAT [Dhomse *et al.*, 2011]. Most importantly, in the upper stratosphere (~45–50 km), the weak SCS observed in SAGE II (Figure 3b) is visible for the HALOE period. Interestingly, a negative SCS just above the stratopause is seen in the MLS data (Figure 3e) but is not present in any model simulation. However, due to some issues with MLS data during earlier periods (e.g., Figure 3 in Dhomse *et al.* [2013]) and shorter temporal coverage, a detailed analysis of MLS is not presented here. Interestingly, all the simulations show somewhat enhanced SCS in the tropical middle stratosphere. This could be due to the fact that model simulations show higher correlation between ozone and temperature (Figure 2d) than the observational data sets and there is inherent positive SCS in ERA-Interim temperatures [e.g., Dhomse *et al.*, 2011].

Assuming that ERA-Interim represents realistic stratospheric dynamics, we can analyze the effects of different solar fluxes over different time periods. Figure 4 shows zonal mean latitude-height cross sections of the modeled SCS for 35 year (1979–2005), SAGE II (1984–2005), HALOE (1992–2005), and MLS (2004–2013) time periods from runs A_NRL, B_SAT, and C_SOR, diagnosed using the same regression model as Figure 3. As shown in Figure 3a, all of the model simulations show a positive SCS (up to 3%) in the middle stratosphere over the full 35 year period. This is close to the transition region where ozone concentrations are controlled by both photochemistry and dynamics. Hence, this suggests that part of positive midstratospheric ozone SCS must be a direct result of enhanced ozone production via increased UV radiation. In the lowermost mesosphere (near 50–55 km) all the simulations show a slight negative SCS at midlatitude and high latitude that is likely a direct result of increased ozone losses via the HO_x cycle (increased CH₄ and H₂O loss under increased UV radiation). Again, run C_SOR shows a negative lower mesospheric SCS even at low latitudes. So differences seen in the upper stratospheric SCS are directly associated with the differences in the solar fluxes as photochemistry controls ozone concentrations at those altitudes. For 1979–2013 time period, nearly all the simulations show about –0.5% SCS near the stratopause (~50 km) in the tropics, but C_SOR shows an even more negative

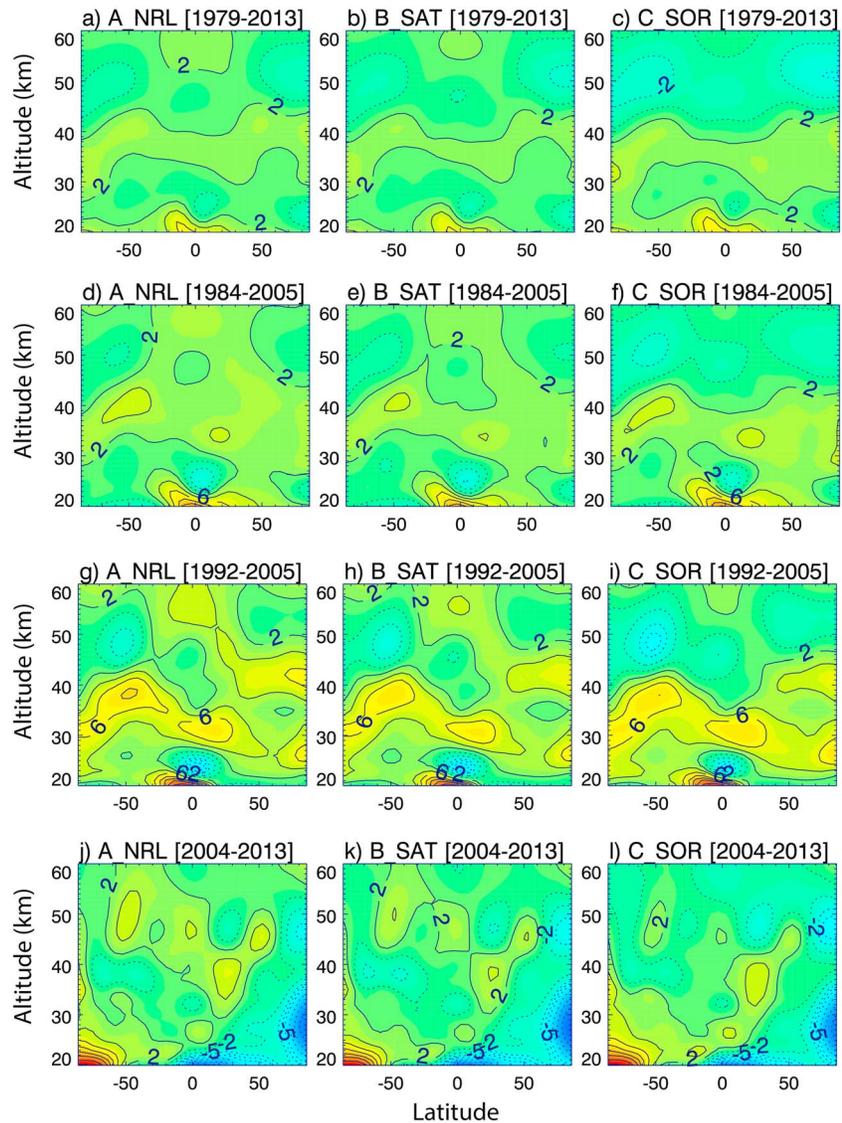


Figure 4. Estimated peak-to-peak amplitude (with respect to mean for a given time period) of the SCS in stratospheric ozone (in %) from model simulations (left column) A_NRL, (middle column) B_SAT, and (right column) C_SOR for different time periods. The horizontal rows compare results (top to bottom rows) for complete 35 year (1979–2013), SAGE II (1984–2005), HALOE (1992–2005), and MLS (2004–2013) time periods. Contour interval is 2%.

value (−4%) at higher altitudes. A slightly positive SCS in the tropical lower mesosphere is not visible in C_SOR. Similar differences are observed for other time periods.

For the SAGE II period (1984–2005; Figure 4, second row), the modeled SCS is similar to that for 1979–2013, except that a slight increase in SCS magnitude is observed at all altitudes. In the upper stratosphere, both A_NRL and B_SAT show almost negligible SCS near stratopause but somewhat positive SCS in the lower mesosphere. C_SOR gives negative SCS throughout upper stratosphere and lower mesosphere. For the HALOE period (1992–2005; Figure 4, third row), runs A_NRL and B_SAT show a positive SCS at north hemisphere (NH) high latitudes near 45 km and negative SCS in the south hemisphere high latitudes. In general, estimated SCS from C_SOR is more negative compared to A_NRL or B_SAT and differences are largest at higher latitudes.

Interestingly, for the MLS time period (2004–2013; Figure 4, fourth row), all three simulations seem to show a “V”-structured SCS with a positive signal in the low-latitude and midlatitude stratosphere. The positive SCS is almost double in magnitude in the NH midlatitude and low latitude, suggesting a much enhanced BD circulation during solar maximum 23 [see also Mahieu *et al.*, 2014]. In summary, Figure 4 shows that very different

ozone SCS can be derived over periods, where different satellite data sets are available. As expected in the mid stratosphere and low stratosphere all the simulations simulate a similar SCS because at this altitude ozone is under dynamical control, and they use identical dynamical forcing. However, in the upper stratosphere, models using NRL and SATIRE solar fluxes simulate nearly similar SCS, but with eSORCE fluxes the model simulates negative SCS in the tropical upper stratosphere. However, the estimated SCS from SAGE v7.0 and HALOE v19 for tropical latitudes (Figure 3) is negligible or even positive for the 1984–2005 and 1992–2005 time periods. Hence, our simulations suggest that SORCE-based reconstructed solar fluxes (eSORCE) are not useful for studying earlier solar cycles.

4. Summary and Conclusions

We have used a 3-D CTM to diagnose the 11 year solar signal in the upper stratospheric ozone with different solar flux data sets. Simulated ozone is compared against three satellite instruments ACE, HALOE, and SAGE II. In absolute terms, there are only small differences in modeled ozone from the various simulations. However, much larger differences in observational data reduce our confidence in the estimated SCS using any single satellite data set. The main causes for these differences are different measurement techniques and retrieval algorithms. We find that the recently updated SAGE II v7.0 shows a reduced SCS compared to v6.2 near the stratopause region, improving agreement with model and HALOE data. SAGE II v7.0 also shows improvements in the well-known ozone-temperature anticorrelation in the upper stratosphere, although this correlation is still weaker than that in ACE and HALOE data sets.

Hence, a reduced upper stratospheric SCS and improved ozone temperature relationship in SAGE II 7.0 indicates that the upper stratospheric SCS is likely smaller than previous estimates [e.g., Soukharev and Hood, 2006]. An analysis of MLS data over the recent solar cycle 23 period also shows negligible or even negative SCS in the upper stratosphere.

Also, we find that in absolute terms, upper middle stratospheric ozone concentrations from simulation A_NRL are about 5% larger than either B_SAT or C_SOR. However, there is very good agreement in estimated SCS from A_NRL and B_SAT, whereas C_SOR consistently shows more negative SCS in the upper stratosphere/lower mesosphere. This highlights that even if there are small differences between simulated ozone concentrations (B_SAT and C_SOR), small variations in relative solar fluxes can give significantly different SCS. Even with recent updates SORCE-type (e.g., eSORCE) solar fluxes may not be able to simulate SCS over previous solar cycles. It seems that the only possibility for SORCE-type solar fluxes to be correct is to have a negative SCS in the upper stratospheric temperatures, but reanalysis data sets do not support this hypothesis.

Furthermore, assuming that ERA-Interim gives a realistic representation of stratospheric dynamics for the last 35 years, our model simulates a different type of SCS for different time periods (even in the upper stratosphere), which is also consistent with differences in the amplitude of the 11 year sunspot cycle. However, inhomogeneities in reanalysis data cannot be ignored. Hence, we argue that large uncertainties in observational ozone data, time varying anthropogenic emissions, changes in amplitude of sunspot cycle, and issues with reanalysis data sets still hamper our efforts to establish the true nature of SCS in the ozone throughout the stratosphere.

Acknowledgments

This work was supported by the NERC SOLCLI (NE/D002753/1) and MAPLE (NE/J008621/1) projects. We thank the NASA/NOAA for the MLS and HALOE data. Model simulations were performed on the Archer and Leeds Arc1 HPC systems.

References

- Arfeuille, F., B. P. Luo, P. Heckendorn, D. Weisenstein, J. X. Sheng, E. Rozanov, M. Schraner, S. Brönnimann, L. W. Thomason, and T. Peter (2013), Modeling the stratospheric warming following the Mt. Pinatubo eruption: Uncertainties in aerosol extinctions, *Atmos. Chem. Phys.*, *13*, 11,221–11,234, doi:10.5194/acp-13-11221-2013.
- Ball, W. T., J. D. Haigh, E. V. Rozanov, A. Kuchar, T. Sukhodolov, F. Tummon, A. V. Shapiro, and W. Schmutz (2016), High solar cycle spectral variations inconsistent with stratospheric ozone observations, *Nat. Geosci.*, *9*, 206–209, doi:10.1038/ngeo2640.
- Chipperfield, M. P. (1999), Multiannual simulations with a three-dimensional chemical transport model, *J. Geophys. Res.*, *104*, 1781–1805, doi:10.1029/98JD02597.
- Chipperfield, M. P. (2006), New version of the TOMCAT/SLIMCAT off-line chemical transport model: Intercomparison of stratospheric tracer experiments, *Q. J. R. Meteorol. Soc.*, *132*, 1179–1203, doi:10.1256/qj.05.51.
- Chipperfield, M. P., S. S. Dhomse, W. Feng, R. L. McKenzie, G. J. M. Velders, and J. A. Pyle (2015), Quantifying the ozone and ultraviolet benefits already achieved by the Montreal Protocol, *Nat. Commun.*, *6*, 7233, doi:10.1038/ncomms8233.
- Damadeo, R. P., J. M. Zawodny, L. W. L. Thomason, and N. Iyer (2013), SAGE version 7.0 algorithm: Application to SAGE II, *Atmos. Meas. Tech.*, *6*, 3539–3561, doi:10.5194/amt-6-3539-2013.
- Damadeo, R. P., J. M. Zawodny, and L. W. Thomason (2014), Reevaluation of stratospheric ozone trends from SAGE II data using a simultaneous temporal and spatial analysis, *Atmos. Chem. Phys.*, *14*, 13,455–13,470, doi:10.5194/acp-14-13455-2014.

- Dhomse, S. S., M. P. Chipperfield, W. Feng, W. T. Ball, Y. C. Unruh, J. D. Haigh, N. A. Krivova, S. K. Solanki, and A. K. Smith (2013), Stratospheric O₃ changes during 2001–2010: The small role of solar flux variations in a chemical transport model, *Atmos. Chem. Phys.*, *13*, 10,113–10,123, doi:10.5194/acp-13-10113-2013.
- Dhomse, S. S., et al. (2014), Aerosol microphysics simulations of the Mt. Pinatubo eruption with the UM-UKCA composition-climate model, *Atmos. Chem. Phys.*, *14*(20), 11,221–11,246.
- Dhomse, S. S., M. P. Chipperfield, W. Feng, R. Hossaini, G. W. Mann, and M. L. Santee (2015), Revisiting the hemispheric asymmetry in mid-latitude ozone changes following the Mount Pinatubo eruption: A 3-D model study, *Geophys. Res. Lett.*, *42*, 3038–3047, doi:10.1002/2015GL063052.
- Dhomse, S., M. Weber, and J. Burrows (2008), The relationship between tropospheric wave forcing and tropical lower stratospheric water vapor, *Atmos. Chem. Phys.*, *8*, 471–480, doi:10.5194/acp-8-471-2008.
- Dhomse, S., M. P. Chipperfield, W. Feng, and J. D. Haigh (2011), Solar response in tropical stratospheric ozone: A 3-D chemical transport model study using ERA reanalyses, *Atmos. Chem. Phys.*, *11*, 12,773–12,786, doi:10.5194/acp-11-12773-2011.
- Ermolli, I., et al. (2013), Recent variability of the solar spectral irradiance and its impact on climate modelling, *Atmos. Chem. Phys.*, *13*, 3945–3977, doi:10.5194/acp-13-3945-2013.
- Gray, L. J., et al. (2010), Solar influences on climate, *Rev. Geophys.*, *48*, RG4001, doi:10.1029/2009RG000282.
- Haigh, J. D. (1996), The impact of solar variability on climate, *Science*, *272*, 981–984, doi:10.1126/science.272.5264.981.
- Haigh, J., A. Winning, R. Toumi, and J. Harder (2010), An influence of solar spectral variations on radiative forcing of climate, *Nature*, *467*, 696–699, doi:10.1038/nature09426.
- Holton, J., and H. Tan (1980), The influence of the equatorial quasi-biennial oscillation on the global circulation at 50 mb, *J. Atmos. Sci.*, *37*, 2200–2208.
- Hood, L. L., et al. (2015), Solar signals in CMIP-5 simulations: The ozone response, *Q. J. R. Meteorol. Soc.*, *141*(692), 2670–2689.
- Hossaini, R., M. P. Chipperfield, S. A. Montzka, A. Rap, S. Dhomse, and W. Feng (2015), Efficiency of short-lived halogens at influencing climate through depletion of stratospheric ozone, *Nat. Geosci.*, *8*, 186–190, doi:10.1038/ngeo2363.
- Kodera, K., and Y. Kuroda (2002), Dynamical response to the solar cycle, *J. Geophys. Res.*, *107*(D24), 4749, doi:10.1029/2002JD002224.
- Kopp, G., and J. L. Lean (2011), A new, lower value of total solar irradiance: Evidence and climate significance, *Geophys. Res. Lett.*, *38*, L01706, doi:10.1029/2010GL045777.
- Lee, H., and A. 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.
- Mahieu, E., et al. (2014), Recent Northern Hemisphere stratospheric HCl increase due to atmospheric circulation changes, *Nature*, *515*, 104–107, doi:10.1038/nature13857.
- McLinden, C. A., S. Tegtmeier, and V. Fioletov (2009), Technical Note: A SAGE-corrected SBUV zonal-mean ozone data set, *Atmos. Chem. Phys.*, *9*(20), 7963–7972.
- Merkel, A. W., J. W. Harder, D. R. Marsh, A. K. Smith, J. M. Fontenla, and T. N. Woods (2011), The impact of solar spectral irradiance variability on middle atmospheric ozone, *Geophys. Res. Lett.*, *38*, L13802, doi:10.1029/2011GL047561.
- Mitchell, D. M. D., et al. (2014), Signatures of naturally induced variability in the atmosphere using multiple reanalysis datasets, *Q. J. R. Meteorol. Soc.*, *141*, 2011–2031, doi:10.1002/qj.2492.
- Randel, W., and F. Wu (2007), A stratospheric ozone profile data set for 1979–2005: Variability, trends, and comparisons with column ozone data, *J. Geophys. Res.*, *112*, D06313, doi:10.1029/2006JD0073339.
- Remsberg, E. (2008), On the response of Halogen Occultation Experiment (HALOE) stratospheric ozone and temperature to the 11-year solar cycle forcing, *J. Geophys. Res.*, *113*, D22304, doi:10.1029/2008JD010189.
- Soukharev, B. E., and L. L. Hood (2006), Solar cycle variation of stratospheric ozone: Multiple regression analysis of long-term satellite data sets and comparisons with models, *J. Geophys. Res.*, *111*, D20314, doi:10.1029/2006JD007107.
- SPARC (2010), Chemistry-Climate Model Validation (CCMVal), edited by V. Eyring, T. G. Shepherd, and D. W. Waugh, World Climate Research Programme, WCRP-30 WMO/TD- No. 40, SPARC Report No. 5.
- Stolarski, R. S., A. R. Douglass, E. E. Remsberg, N. J. Livesey, and J. C. Gille (2012), Ozone temperature correlations in the upper stratosphere as a measure of chlorine content, *J. Geophys. Res.*, *117*, D10305, doi:10.1029/2012JD017456.
- Swartz, W. H., R. S. Stolarski, L. D. Oman, E. L. Fleming, and C. H. Jackman (2012), Middle atmosphere response to different descriptions of the 11-yr solar cycle in spectral irradiance in a chemistry-climate model, *Atmos. Chem. Phys.*, *12*, 5937–5948, doi:10.5194/acp-12-5937-2012.
- Wang, L., C. Z. Zou, and H. Qian (2012), Construction of stratospheric temperature data records from stratospheric sounding units, *J. Clim.*, *25*(8), 2931–2946.
- WMO (2015), Assessment for decision maker: Scientific assessment of ozone depletion: 2014 Global Ozone Research and Monitoring Project, *Rep. 56*, Geneva, Switzerland.
- Yeo, K. L., N. A. Krivova, S. K. Solanki, and K. H. Glassmeier (2014), Reconstruction of total and spectral solar irradiance from 1974 to 2013 based on KPVT, SoHO/MDI, and SDO/HMI observations, *Astron. Astrophys.*, *570*, A85, 1408.1229.