Anonymous Reviewer #1

1 General Comments

This paper is well structured and generally well written. It addresses an important question: Is stratospheric ozone sensitive enough to (multi-annual) solar flux variations to allow discrimination between significantly different solar flux datasets (i.e. SORCE versus NRL or SATIRE-S)?

This question is approached through a sensitivity study using the 3D CTM SLIMCAT, which is driven by realistic wind fields from ECMWF. This is an excellent idea, but the paper will be useful only if sufficient details are given about the SLIMCAT model to help understand the differences with previous modelling studies on this topic. Hence great care must be given to the description of the model and its set-up, which are serious shortcomings here – especially regarding the importance of the solar emission line at Lyman-alpha. Due to the low lid of SLIMCAT (0.1 hPa), I also doubt the validity of the results presented in the Upper Stratosphere-Low Mesosphere (USLM); they are probably too influenced by upper boundary conditions which are not described at all.

This review assumes that even though their variations with the 11-year solar cycle were overlooked, SLIMCAT accounts for two major processes in ozone photochemistry above ~10 hPa: the absorption of solar radiation by O2 above the model lid, and the solar Lyman-alpha line. If that is not the case, we recommend to reject the paper as SLIMCAT would not be able to separate properly the dynamical and photochemical influences on ozone variations. Otherwise, the excellent agreement between the SLIMCAT runs and MLS or SABER observations certainly deserves publication and I recommend publishing this paper after major modifications, as described in the comments below.

## We thank the reviewer for his/her insightful comments. We re-checked our model setup for Lyman-alpha photolysis parameterization and lower lid issues. We are confident that above mentioned issues are correctly accounted for in our simulations. Additional discussion and references are included in the revised manuscript.

2 Major Comments

2.1 Solar Lyman-alpha line

The solar emission line at Lyman-alpha (121.59 nm) matches a deep minimum in the O2 absorption cross-section, allowing this line to penetrate the middle atmosphere much lower than the neighbour wavelengths. In view of the large and well-documented variations of the solar flux at Lyman-alpha, this is a key process that must be included in any modelling study of the impact of solar flux variations on ozone. The existence of Lyman-alpha is not mentioned in this paper, so it is impossible to know if (and how) it was taken into account by the model. Section 3 should describe the intensity of solar Lyman-alpha used in the different runs, as well as the parameterization used to account for the variation of the O2 cross-section over the emission line.

If the solar flux at Lyman-alpha is kept to one constant value in all model runs (e.g. a value typical of solar minimum), then an additional experiment should be run with this flux set to a
very different value (e.g. a value typical of solar maximum). The results of this experiment should then be added in the figures and discussion. Note that this could well change the conclusions of the paper. If, on the other hand, the 11-year variations of Lyman-alpha are already taken into account, these variations should be clearly described for each model run and the consequence stated in the discussion – i.e. the impossibility to discriminate between two solar flux datasets if they do not cover wavelengths shorter than the Schumann-Runge Bands (SRB).

## We agree with the reviewer that Lyman-alpha changes were not discussed and not shown in Figure 1. We do have O2 photolysis changes associated with Lyman-alpha in our simulations (except the simulations with SORCE solar fluxes) and details/references are now provided. SATIRE-S is available from 115nm to 0.16mm whereas NRL-SSI ranges from 120nm to 0.1mm. Except for Lyman-alpha, our SLIMCAT simulations only include flux variation at and beyond the Schumann Runge Bands (172 nm). Figure 1 is modified in a revised draft showing Lyman-alpha changes between December 2004 and December 2007 for NRL-SSI (~12.1%) and SATIRE-S (~11.3%). The text has been modified accordingly. In the revised manuscript we also added a paragraph explaining that Schumann-Runge continuum is not included as most of the O2 photolysis in this region occurs well above the lower mesosphere (Brasseur and Solomon, Figure 4.3). However, O2 and O3 photolysis changes at and beyond the Schumann-Runge Bands (above 176nm) are included in our simulations.

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#### 2.2 Upper lid and boundary conditions

The variations of ozone in the USLM are discussed a lot in this study, while the model lid is the same as ERA-Interim i.e. at 0.1 hPa, right in the middle of the lower mesosphere. Hence I expect that in the USLM the model results are very sensitive to the Upper Boundary Conditions (UBC) used for ozone itself, even if they are specified as a vertical flux. This is indeed a major weakness of all ERA-driven CTM’s to study the USLM.

In view of the excellent comparison with observations, I recommend the following approach to get around this difficulty: describe precisely, in Section 3, the UBC used for ozone in each model run; perform a sensitivity test with a very different UBC to show the vertical range where the results are significantly influenced by this choice of UBC; exclude this vertical range from the other figures and from the discussion.

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### The SLIMCAT top level is approx. 0.1 hPa. Above 0.1 hPa the model uses standard atmospheric profiles to determine overhead density (e.g. slant column to determine O2 absorption). There are no upward/downward mass fluxes through the top of the top level, so tracers are not overwritten. At the top level (~0.1 hPa) tracers are transported into all the neighbouring grids except upwards. At this level, O3 is very short lived (~minutes), so this does not affect O3 fields. This discussion is added in the revised manuscript. However, some of the longer lived tracers might be influenced by this assumption. An example is CH4 which is not completely destroyed up to this level, so one might expect some CH4 build-up in USLM region at high latitudes. Therefore, we did an additional run similar to Run A, but model output sampled at Atmospheric Chemistry Experiment (ACE) measurements collocations. Our comparison there shows very good agreement in modelled and ACE measured O3 (Figure R1) as well as CH4 (R2). We also did profile by profile comparison and did not find any significant biases in the high-latitude USLM region. This analysis shows that upper lid or boundary conditions do not affect our tracer distribution in the USLM.
**Figure R1:** Comparison between co-located SLIMCAT and ACE ozone profiles for tropics (left), NH mid-latitudes (middle) and NH high latitudes (right). SLIMCAT data is shown with solid line and ACE data with symbols. Red and blue colours represent sunrise and sunset measurements, respectively. The number of sunrise and sunset measurements for each latitude band are also given. Horizontal bars indicate the standard deviation of ACE measurements at a given altitude.

**Figure R2:** Same as R1 but for methane.
2.3 Description of the radiative transfer model

The paper will be useful only if sufficient details are given about the SLIMCAT model to help understand the differences with previous modelling studies on this topic. Besides the realistic winds (and lower model lid), a key component to explain these results is the radiative transfer model used by SLIMCAT to compute the photodissociation and solar heating rates. There is not any description of this module in the paper.

This should be corrected with the following information (and appropriate references):

- What radiative transfer model is used by SLIMCAT? What is its wavelength range and resolution? Does it use the same vertical grid as SLIMCAT itself?

  ## The revised manuscript includes a reference for radiative transfer model which uses Delta-Eddington approximation for solar radiation that is used in the NCAR community climate model (Briegleb, 1992).

- What parameterization is used for the absorption by the SRB of O2 (175–205nm)? Fig.1 shows that the different solar flux variations seen by SORCE cover precisely this special wavelength region.

  ## We use the scheme of Minschwaner et al (1993). A reference has been added.

- How is the absorption of solar radiation above 0.1 hPa taken into account?

  ## See reply to comment 2.1. We calculate the O2 column from the air density and use a standard O3 profile.

- Is the same radiative transfer model used for solar heating rates and for photodissociation rates? If not, what are the differences?

  ## See replies to comments above. These are different schemes. The radiative transfer and calculation of vertical transport using heating rates is described in Chipperfield (2006).

3 Minor comments

- Section 4 states that the SORCE dataset used here is “the available SORCE data that was used in Haigh et al (2010)” but no reference describes directly this dataset, nor the instrument itself. If no peer-reviewed paper is available on this topic, please cite a technical report or at least a web page describing this dataset. This should be part of a new paragraph in Section 2, which currently does not describe SORCE.

  ## We have added (Harder et al., 2009 and Harder et al., 2010). In the revised manuscript, we also included new simulations with updated SORCE data. The paragraph is modified as “The SORCE mission is described by Rottman (2005). For the SORCE fluxes used here, we combine data from two of the instruments on board SORCE: the SOLar STellar Irradiance Comparison Experiment (SOLSTICE; Mcclintock et al. 2005); and the Spectral Irradiance Monitor (SIM; Harder et al. 2009, 2010). We wish to make a direct comparison with Haigh et al. (2010) and thus use the same dataset for most of our runs. It is based on SOLSTICE (version 10) below 200 nm and on SIM intermediate-release version, J. Harder, pers comm 2010) for wavelengths above 200 nm; we label this dataset SORCE_1. As the use of SIM data below 310 nm is no longer recommended, so we also included two test runs using the currently available SORCE data. These data are labelled SORCE_2 and use SOLSTICE (version 12) and SIM (version 17) data for wavelengths below and above 310 nm, respectively”
• On Fig. 2, SABER shows a diurnal variation of ozone in the 30-45 km range (3.5 ppmv) while SLIMCAT finds none. This variation (0.2 ppmv/6 ppmv) may be negligible but is worth mentioning. What does MLS show on this topic?

## MLS differences have been added in the revised version. We have looked into this day-night difference. At these altitudes in the stratosphere $[O_3] \gg [O]$ and the observations show that $[Ox]$ ($O_3 + O$) during the day is larger than at night. In the submitted paper the SLIMCAT night-day difference was based on output at 12 UT and 0 UT. In fact, this night-day difference shows a variation with time and is larger a few hours later. In the revised paper we plot the SLIMCAT data near 1:30 and 13.30 UT (which is closer to MLS times) and which does capture some of this diurnal variation (nb the mesospheric comparisons do not change). We assume that this variation arises because the production of Ox (from O2 photolysis) peaks more strongly in the middle of the day, compared to loss which proceeds more evenly through the day. The SLIMCAT-SABER-MLS comparison in the mid stratosphere is discussed in the revised version.

• On Fig. 3: how are computed these ozone anomalies? In other words: they are departing from what monthly mean climatology?

## In the revised manuscript we have added a sentence “Monthly mean anomalies are calculated by subtracting climatological monthly mean values from monthly means.”

• Section 4 (p. 9, line 8, while discussing Fig. 4): “middle stratospheric enhancements can be simulated with NRL (or SATIRE), fixed and SORCE solar fluxes”. I do not understand the “fixed” part, as run C_FIX is not used on Fig. 4. That is a pity since otherwise this sentence would be an important conclusion. If possible, please include results from run C on fig. 4.

## Differences from run C_FIX as well as one with new SLIMCAT simulations using updated SORCE data have been added.

• P. 9, line 28: Figure 3h does not exist.

## Corrected.

• P. 10, line 6 mentions “a fixed dynamics simulation with SORCE fluxes” but no such simulation is reported neither in Section 3 nor in Table 1.

## Here we meant that we are using identical meteorological fields in Runs D_SORCE2004 and E_SORCE2007 (or D_SORCE2004_1 and E_SORCE2007_1 in the revised version). So we modified the sentence to be “a fixed (or identical) dynamics simulation with different SORCE fluxes”.

• Fig. 6 does not show any model results using SORCE. Please do it or explain why it is not done.

## As mentioned earlier, the simulations with SORCE fluxes use identical meteorological fields, so they cannot be different to Figure 5.

• P.11, line 1 (while discussing Fig. 6): “our simulations and SABER show O3 differences of nearly 4%”. I see a rather different result, with SABER observing maximum differences of 2% much like run C_FIX but twice lower than runs A_NRL or B_SATIRE.

## We have modified this as “our simulations and SABER show about 2-4 % O3 differences….“.
• Conclusions: the last paragraph is quite unclear – does it refer to Fig. 7a or 7b? Fig. 7a does not cover 2001-2010 while Fig. 7b does not include SBUV nor SAGE, and it is not possible to compare both since 7a uses SMAX-SMIN while 7b uses a regression model.

## The paragraph draws an overall conclusion from Figure 7 for the general period 2001-2010. Figure 7a shows our current understanding of the solar signal, while Figure 7b shows solar signal for 2001-2010. However, due to very short time series presenting composite solar response for this period would not be realistic, therefore we present results from the regression analysis.

• The key sentence from the abstract (“Ozone changes in the lower mesosphere cannot be used to discriminate...”) should be part of the conclusions.

## We agree with the reviewer, so we have added the following sentence in the Conclusions section “Overall, our simulations suggest that out of phase solar signal in the lower mesospheric O3 during recent solar cycle cannot be used to distinguish between the various solar flux data sets due to large uncertainties and the shorter time span of the observations.”

However, we also believe that some key results must be included in the abstract as well, so we kept abstract unchanged.