Anonymous Referee #1

The manuscript discusses the responses of the stratospheric ozone response to the solar irradiance variability on the Sun rotation cycle time scale. The authors analyzed the satellite observations by MLS instrument and results obtained with LMDZ model in free running and specified dynamics modes. The subject of the manuscript is appropriate for ACP. Despite many attempts to characterize ozone response to solar irradiance variability several aspects of the problem still remain open. The manuscript is well written and structured, the figures and explanations are clear. The conclusions about the dependence of the ozone response uncertainty on the signal strength and necessary numbers of cycles could be of interest for the scientists working in this area. There are, however, some flaws which do not allow me to recommend immediate publication.

We thank the reviewer for reading the manuscript and providing helpful comments and suggestions. We address the raised issues in turn below.

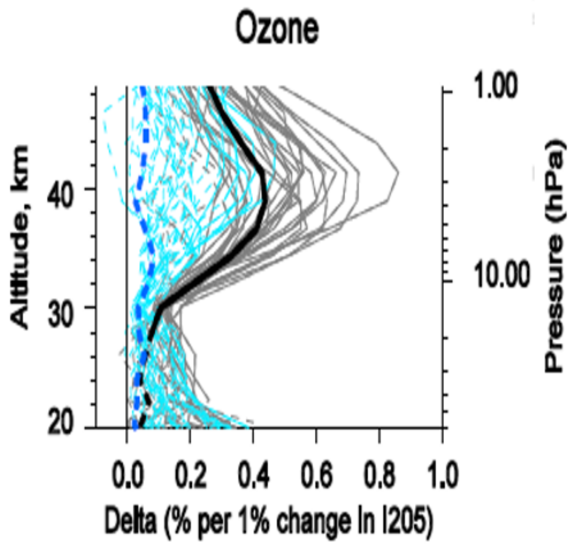
Our answers to comments and suggestions are written in blue and manuscript changes are written in italic type within double quotes ("*like this*").

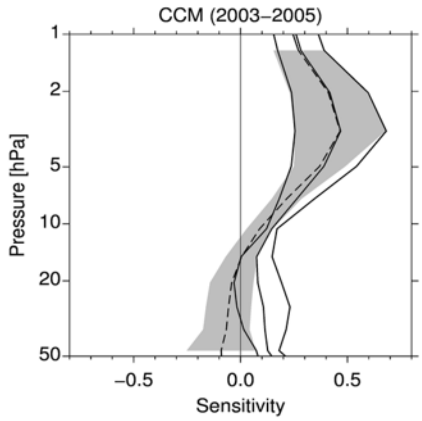
Major issues

1. It looks like the authors intentionally omitted the direct effects of the solar irradiance on the heating rates and temperature. After careful discussion of how important this process for the correct representation of the time lag between ozone and solar UV in the introduction they completely excluded this processes using not correct arguments. The importance of this process is clear even if we put aside the dynamical consequences of the direct radiative heating. I recall the importance of the direct heating was demonstrated in the recent paper by Sukhodolov et al., 2016.

We agree that neglecting the direct effect on heating rate must be considered carefully. To test the potential influence of the missing temperature feedback on ozone in our model setup, we performed additional analysis that we compared with recent model results of Sukhodolov et al. [2017].

In their recent study, Sukhodolov et al. [2017] performed an ensemble of 30 simulations for the period 2003-2005 with the SOCOL CCM model. In their experimental setup, they considered both (i) the effects of UV changes on the photolysis rates and (ii) the direct radiative effects on temperature which feeds back on ozone. Hence, their CCM formulation allows accounting for all photochemical and radiative effects. Their large number of simulations and the fact that they simulated the period 2003-2005 using NRLSSI data (like us) allows comparing with our CCM ensemble results in a fair way. Their UV-ozone cross-correlation analysis (see their Figure 3b) reveals a negative lag of 1 to 2 days in the upper stratosphere (above 3 hPa) which is consistent with the one we found in our study (see our Fig. 7c,f). When performing the UV-ozone cross-correlation analysis over the same period (i.e. 2003-2005), we also found a negative lag of 1 to 2 days. We also compared the analysis of the ozone sensitivity to F205 index (see below) over the period 2003-2005. Their ozone sensitivity profiles (left) are shown at optimum lag and ours (right) at optimum lag (solid) and zero lag (dashed). Results from both models compare very well, showing a maximum slightly larger than 0.4 at 3-4 hPa and decreases down to 0.25 near the stratopause. The very good agreement between the two CCMs, despite their different formulation, suggest that omitting the direct radiative heating does not have a strong effect on the ozone response and is therefore an acceptable approximation in our case.





Globally, the manuscript has been revised at several places to discuss the CCM configuration.

- More specifically in section 2.3, we modified the text as follows:

"Thus, the solar rotational cycle forcing is taken into account by using daily photolysis rates calculated by TUV in the photochemistry module of LMDz-Reprobus. *Note however that the direct effect on heating rates generated by UV variations associated with the 27-day rotational cycle is neglected: i.e. daily changes in the spectral irradiance are not considered by the model radiative scheme. As a consequence, part of the thermal and dynamical responses to the 27-day rotational cycle and hence their effect on ozone (through transport and temperature dependent chemical reactions, as described above) are missing. The impact of this approximation on our results seems to be small though, as discussed thereafter (sections 3 and 5). Note also that on timescales of the 11yr cycle, Swartz et al. (2012) found that their photolysis-only simulation captured almost all of the solar cycle effect on ozone.*"

- Discussion on results of Figure 6 (now Figure 7) has been extended as follows:

"*We now analyse the CCM ensemble results. The ensemble mean ozone sensitivity profiles (Figs. 7c and f) markedly differ with ozone sensitivity profiles derived from observations (Figs. 7a and d) and CTM (Figs. 7b and e) at the corresponding periods. These differences are particularly pronounced in the upper stratosphere (above ~5 hPa). On the other hand, despite the two different periods, the ensemble mean ozone sensitivity profiles show very similar features with positive sensitivity from 15 hPa to the stratopause and a maximum sensitivity of 0.4 at ~3 hPa (Figs. 7c and f). This maximum tropical sensitivity value and its altitude level is in good agreement with previous CCM estimates (e.g. Rozanov et al., 2006; Austin et al., 2007; Gruzdev et al., 2009; Kubin et al., 2011). The CCM ozone sensitivity analysis has also been repeated for the period 2003-2005 (not shown) to be directly comparable with the CCM results of Sukhodolov et al. (2017): we found very similar ozone sensitivity profiles.*"

- We added a full paragraph to discuss this aspect in section 5:

"*In our CCM experimental design, the direct radiative effect of UV on heating rates has been neglected leading to an underestimated temperature response to the 27day cycle. As a consequence, this may affect the ozone response by reducing the magnitude of the solar-induced temperature feedback on chemical reaction rates. Nevertheless, a detailed comparison of the ozone response in our analysis with results from previous independent CCM studies (Rozanov et al., 2006 ; Sukhodolov et al., 2017) revealed a very good correspondence, despite the fact that their* *experimental design included the direct radiative heating effect. This suggests that the feedback exerted by the solar-induced temperature fluctuations on ozone is modest, at least at the 27day time scales. This is in fact not surprising. In their recent study, Sukhodolov et al. (2017) shown that the atmospheric internal variability largely dominates the variability of the stratospheric temperature on 27day time scales, making the temperature response to the solar forcing difficult to identify and, hence, the influence of its feedback on ozone secondary. Nonetheless, we recognize that to quantify properly the impact of the neglected solar-induced temperature feedback on our results, additional CCM experiments including the direct radiative effect of UV on heating rate should be performed."*

- We added some details regarding the role of HOx on ozone in the upper stratosphere:

In the introduction, we added a short paragraph to describe the effect of HOx on ozone in the upper stratosphere and mesosphere: "*Fleming et al. (1995) further stressed the increasing importance with height of the solar-modulated HOx chemistry on the ozone response above 45 km. In the upper stratosphere and mesosphere, enhancement of HOx through photolysis of water vapour in Lyman-alpha line associated with an increasing solar irradiance contribute to destroy ozone. Above ~65 km and at zero-lag, the latter mechanism dominates over ozone production (i.e. by photolysis of oxygen) leading to a negative ozone-solar irradiance correlation. In the upper stratosphere and lower mesosphere (below 65 km), although ozone production dominates, increasing HOx at zero-lag contributes to the negative lag of the ozone response (Rozanov et al., 2006)."*

We also mention this mechanism in section 3.2 when we discuss the cross-correlation analysis shown in Figure 5: "*Above 3 hPa (~40 km), CCM cross-correlations of both periods (Fig. 5c,f) show a maximum at negative time lag (-2 days). As mentioned in the introductory section, this negative time lag can be induced by temperature feedback on ozone and by increasing HOx with solar irradiance which contributes to destroy ozone. While our model configuration allows to fully account for the HOx effect, the solar-induced temperature response is limited since the direct radiative heating effect is not included. The temperature response to the 27-days cycle is thus solely controlled by ozone production in the photolysis scheme. Although a temperature signal is found (not shown), it is small, reducing the likelihood for the solar-induced temperature feedback to be prominent in our experiments. Despite the approximation made in our model configuration, we notice however that the upper stratosphere negative lags compare very well with those found in CCM experiments of Sukhodolov et al. (2017) (see their Fig. 3) in which both HOx and solar-induced temperature feedback effects are fully included. Hence, this suggests that neglecting the direct effect on heating rates generated by UV variations has a limited effect on the ozone response, at least at 27-days timescale. More sensitivity experiments are required however to quantify accurately the impact of this approximation*."

2. It is also not clear why the authors used 3 year period to evaluate ozone response from the observation and model while they show that 3 year time period does not provide statistically robust results (uncertainty only below 50%). This should be somehow explained to the readers. The choice of the number of ensemble runs is also doubtful in the light of the obtained results. If the ensemble run is crucial it would be logical to estimate how many ensemble runs are necessary to reach some kind of convergence. Moreover, the obtained results with free running CCM will be more convincing if the analysis of the CCM runs without solar rotation variability is added.

Regarding the first part of the comment (about 3 year periods), section 3 of our paper, which considers the declining phases of cycles 22 and 23, actually serves as a case study where we compare observations and model results. It follows up on Bossay et al. [2015] which found some intriguing behavior in the response of the ozone to the 27-day solar cycle. Namely, that the correlation between the ozone and the forcing seems stronger when the amplitude of solar rotational fluctuations is small (more details are written in the introduction). In our study, we used in addition to observations, CTM and an ensemble of CCM simulations results. Section 3 allows illustrating clearly, with a concrete case, the effect of internal variability when retrieving the response of ozone to solar forcing at 27-day timescale.

To make the purpose of section 3 clearer, we revised the introductory part as follows:

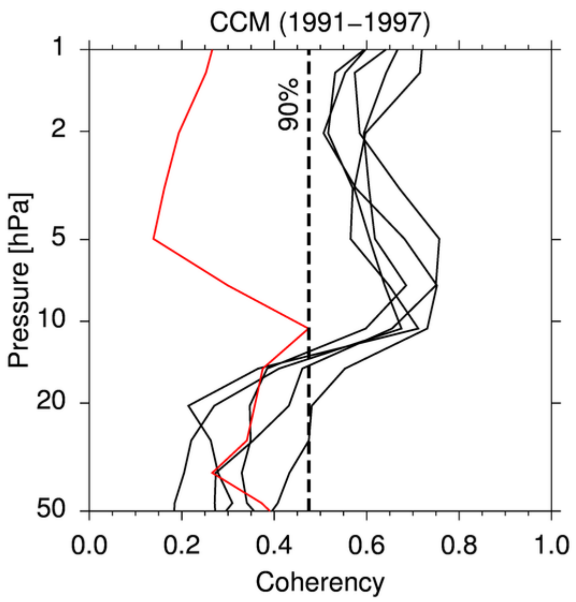
"As a first step, we follow up on the case study of Bossay et al. [2015] and make use of observations and modelling results comparison to provide a detailed picture of the ozone response to the solar rotational cycle during the declining phases of cycle 22 and cycle 23. We particularly aim to better understand the strong differences in the ozone response to solar rotational cycle found between the two periods. Two configurations of the LMDz-Reprobus chemistry climate model simulations are used, with specified dynamics (i.e. Chemistry Transport Model, or CTM) and in its free running mode (CCM). In the CTM configuration, temperature and wind fields calculated by the model are relaxed towards meteorological analysis; the dynamics is expected to be rather close to the reality, allowing direct comparisons with satellite observations for evaluating model chemical processes and its relevance to our study. In the CCM configuration, an ensemble of simulation is performed. Comparing the CCM ensemble results to CTM and observations during the declining phases of cycle 22 and cycle 23 allows to understand better the effect of internal dynamical variability on the ozone response. As a second step, we take advantage of the ensemble of CCM simulations and its large statistics to (i) assess the influence of the solar cycle phase on the ozone sensitivity to the rotational cycle and (ii) quantify the time window required for a robust estimation of the ozone sensitivity."

We also revised the introductory of section 3. part as follows:

"*In this section, we analyse the ozone response to the solar rotational cycle over the declining phase of solar cycles 22 and 23 in the observations and in the CTM and CCM model simulations. The analysis presented here follows up on Bossay et al. (2015) observational study. In particular, we aim to assess the model performances, understand better the differences in the results between the two solar declining phase periods and highlight the importance of internal dynamical variability*."

Regarding the second point (choice of the number of ensemble members), quantifying the number of ensemble members required to reach some kind of convergent was done in Sukhodolov et al. (2017). In our study, we address a similar problem in terms of size of time window (Figure 11) where a convergence is also reached.

Finally, as suggested by the reviewer, we added a figure (now Figure 5) to compare ozone/F205 coherency estimates of the unforced vs. forced experiment results.



"Figure 5. Vertical profile of the mean squared coherence between ozone and F205 averaged between 22 and 30 day periods and calculated for the time period 1991-1997. The black lines correspond to the results of individual ensemble members (five in total) and the red lines to the results of the experiment forced with constant solar forcing. The vertical dashed line indicates the 90% confidence limit."

We added the following part:

"To further test the robustness of the coherence signal, we perform an additional CCM simulation for the period 1991-1997 where the solar forcing is kept constant by using fixed (i.e. climatological) photolysis rates during the model simulation. Results are shown on Fig. 5. Below 15 hPa, the different experiments show no significant coherence between ozone and solar flux. Between 15 and 1 hPa, all forced experiments (black lines) reveal a similar and significant coherence signal while for the constant solar forcing experiment (red line), the coherence is weak and within the range of randomness. The absence of significant coherence found in the constant solar experiment confirms that the coherence found between F205 and stratospheric ozone is not fortuitous and primarily originates from photolysis processes"

Minor issues:

1. Page 7, line 1: It is clear since 2004 that two bands scheme cannot be used for the simulation of the atmospheric response to solar irradiance variability. It was confirmed in 2010,2011 and 2014. Several modifications of different complexity are known.

In its current version the LMDz-Reprobus radiative scheme has only 2 spectral bands resolved in the UV. This leads to an underestimation of the stratosphere temperature response to solar spectral variations (see also Figure 3.17 in chapter 3 of the CCMVal report). In the coming version of LMDz-Reprobus (prepared for CMIP6), the radiative scheme spectral resolution in the UV range has been improved.

2. Page 7, line 17: I do not understand how climatological temperature can be used in CCM. Please, elaborate. By the way the reference to TUV is confusing. If I am not mistaken in the cited 1999 paper the tropospheric version was described. Maybe it is better to use recent intercomparison of the codes where TUV showed excellent performance relative to reference models. I do not also understand whether or not daily NRL solar irradiance was used for the photolysis rate calculations.

In the off-line look-up tables, the temperature dependence of the absorption cross-section are computed using US standard atmosphere temperature profile in TUV, not in the CCM. This approximation has no significant impact on the results. We changed the sentence as follows: "*The temperature dependence of absorption cross-sections is calculated off-line in TUV using the US standard atmosphere*."

Regarding TUV, we now added the reference to Sukhodolov et al. (2016).

Finally, we clarified that daily NRL irradiance is used for photolysis rate calculation as follows: *"A separate photolysis look-up table is calculated every day using the daily NRLSSI as solar input"*

3. Page 7, line 21-24: I recommend to check carefully this sentence. I do not recall such a bold statement in the cited paper.

This sentence and the reference to Swartz et al. [2012] have been removed; the statements were indeed too strong. Instead, we now detail the potential effect of neglecting direct heating rates response on ozone (see also answer to comment 1):

"Thus, the solar rotational cycle forcing is taken into account by using daily photolysis rates calculated by TUV in the photochemistry module of LMDz-Reprobus. *Note however that the direct effect on heating rates generated by UV variations associated with the 27-day rotational cycle is neglected: i.e. daily changes in the spectral irradiance are not considered by the model radiative scheme. As a consequence, part of the thermal and dynamical responses to the 27-day rotational cycle and hence their effect on ozone (through transport and temperature dependent chemical reactions, as described above) are missing. The impact of this approximation on our results seems to be small though, as discussed thereafter (sections 3 and 5).*"

4. Page 12, line 9: I think this statement is not completely correct. In general, the magnitude of the rotational cycle depends on non-homogeneity of the dark/bright features distribution, which can be very small for very high level of solar activity.Fig.8 shows for example rather small variances in 2002, while the solar activity level is high.

We agree with the reviewer and modified the text as follows:

"The amplitude of the rotational cycle depends on the inhomogeneous brightness structure of the solar disc (i.e. distribution of sunspots and faculae). Given that the amount of sunspots and faculae increases with increasing solar activity, inhomogeneity in the brightness is likely to increase during solar maximum phases. One may thus expect minimum and maximum sensitivity during 11year solar maximum and minimum phases, respectively."

Anonymous Referee #2

Overall, this is a valuable comparison study of the ozone response to short-term solar UV variations in both observations and a state-of-the-art chemistry climate model. The analysis is detailed and the results offer plausible explanations for differing results obtained in observations covering different time periods. Final publication is certainly expected in ACP. However, I have some important comments that will require some revision.

We thank the reviewer for reading the manuscript and providing helpful comments and suggestions. We address the raised issues in turn below.

Our answers to comments and suggestions are written in blue and manuscript changes are written in italic type within double quotes ("*like this*").

Main Comments:

(1) In the description of the adopted CCM configuration in section 2.3 (p. 7), the authors say: "We do not take into account the direct effect on heating rates generated by UV variations because previous modelling studies have already shown that the stratospheric ozone response to solar variations is almost entirely driven by the effects of UV changes on the photolysis rates, in particular the photolysis of molecular oxygen (Swartz et al, 2012)." Even on the 11-year time scale when a steady-state approximation is allowed and both photolysis and radiative heating are accounted for, temperature feedback reduces the ozone response in the upper stratosphere at 2 hPa by about 30% compared to that calculated by considering changes in photolysis only (see Figure 2 of Swartz et al.). 30% is still a fairly large fraction and should not be neglected. On the 27-day time scale, it is more important to include radiative effects on temperature and their feedbacks on the ozone response for two reasons. First, on this time scale, the temperature response peaks at a positive phase lag. As reviewed in the Introduction (lines 5 to 14 on p. 3), the lagged temperature response significantly alters (reduces) the ozone response and shifts it to a negative phase lag in the upper stratosphere. Second, as also reviewed there, a dynamical component of the response is produced in the upper stratosphere which feeds back into the temperature response resulting in a larger effect on the ozone response than would be predicted by a 1D radiative-photochemical model. Therefore, please modify section 2.3 to note and discuss these issues and whether the neglect of the modeled temperature response (and its accompanying dynamical response) may lead to errors in the CCM results that would not be present in simulations done in the CTM mode (forced using observed dynamics and temperatures).

We agree that neglecting the direct effect on heating rate should be considered carefully. To test the potential influence of the missing temperature feedback on ozone in our model setup, we performed additional analysis that we compared with recent model results of Sukhodolov et al. [2017] (see the answer to comment 2 for details). The manuscript has been revised at several places to discuss the effect of the neglect (see also answer to specific comments).

More specifically in section 2.3, we modified the text as follows:

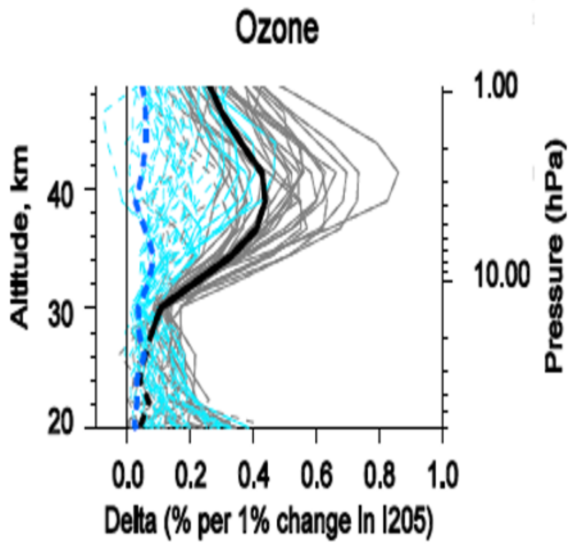
"Thus, the solar rotational cycle forcing is taken into account by using daily photolysis rates calculated by TUV in the photochemistry module of LMDz-Reprobus. *Note however that the direct effect on heating rates generated by UV variations associated with the 27-day rotational cycle is neglected: i.e. daily changes in the spectral irradiance are not considered by the model radiative scheme. As a consequence, part of the thermal and dynamical responses to the 27-day rotational cycle and hence their effect on ozone (through transport and temperature dependent chemical reactions, as described above) are missing. The impact of this approximation on our results seems to be small though, as discussed thereafter (sections 3 and 5). Note also that on timescales of the*

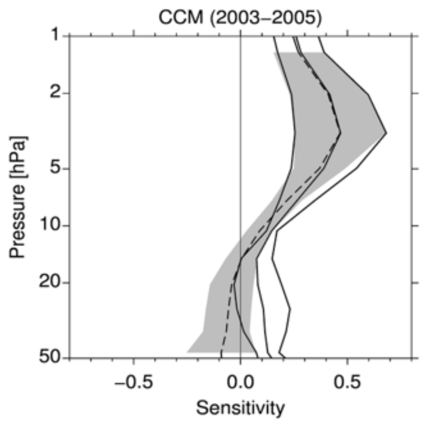
11yr cycle, Swartz et al. (2012) found that their photolysis-only simulation captured almost all of the solar cycle effect on ozone."

(2) Figure 6 compares the vertical profile of the ozone sensitivity to the solar UV (percent change in ozone for a 1 per cent change in solar UV at 205 nm) as derived from observations for two time periods, from the model using specified temperatures and dynamics (CTM), and from the model in a free-running mode (CCM). While the observational and CTM results agree fairly well, the mean CCM results show a much larger response in the upper stratosphere than is seen in either the observations or the CTM results. There is apparently no mention of this disagreement in the manuscript. In view of comment (1) above, it seems possible that part or all of the disagreement is due to neglect of the UV-induced temperature response in the CCM, which would modify both the amplitude and phase lag of the modeled ozone response. The sensitivity calculation is apparently at zero lag so it does not take into account the actual phase lag of the ozone response. Therefore, please modify the results and conclusions sections to consider the possibility that the chosen CCM configuration does not accurately simulate the net ozone response in the upper stratosphere (taking into account both the radiatively and the dynamically induced temperature response).

In a recent study, Sukhodolov et al. [2017] performed an ensemble of 30 simulations for the period 2003-2005 with the SOCOL CCM model. In their experimental setup, they considered both (i) the effects of UV changes on the photolysis rates and (ii) the direct radiative effects on temperature which feeds back on ozone. Hence, their CCM formulation allows accounting for all photochemical and radiative effects. Their large number of simulations and the fact that they simulated the period 2003-2005 using NRLSSI data (like us) allows comparing with our CCM ensemble results in a fair way. Their UV-ozone cross-correlation analysis (see their Figure 3b) reveal a negative lag of 1 to 2 days in the upper stratosphere (above 3 hPa) which is consistent with the one we found in our study (see our Fig. 7c,f). When performing the UV-ozone cross-correlation analysis over the same period (i.e. 2003-2005), we also found a negative lag of 1 to 2 days. We also compared the analysis of the ozone sensitivity to F205 index (see below) over the period 2003-2005. Their ozone sensitivity profiles (left) are shown at optimum lag and ours (right) at optimum lag (solid) and zero lag (dashed). Results from both models compare very well, showing a maximum slightly larger than 0.4 at 3-4 hPa and decreases down to 0.25 near the stratopause. The very good agreement between the two CCMs, despite their different formulation, suggest that omitting the direct radiative heating does not have a strong effect on the ozone response and is therefore an acceptable approximation in our case. Consequently, this model comparison suggests that the disagreement between the model and the observations in the upper stratosphere most likely comes from the dynamical variability, in line with our former interpretation. In 1991-1994, observational uncertainties (either in MLS or in the reanalysis used to nudge the model) may have an influence on the results. We will discuss this thoroughly in the revised version of the manuscript.

Nonetheless, we recognize that, ideally, the direct radiative heating should be considered. In its current version the LMDz-Reprobus radiative scheme has only 2 spectral bands resolved in the UV. Hence, this would lead anyway to an underestimation of the stratosphere temperature response to solar spectral variations (see also Figure 3.17 in chapter 3 of the CCMVal report). For the next version of LMDz-Reprobus, the radiative scheme spectral resolution in the UV range has been improved.





Globally, results and conclusions of the manuscript have been revised to discuss the CCM configuration => see answer to specific comments below (particularly comments 13, 15, 17, 18).

Specifically, discussion on results of Figure 6 (now Figure 7) has been extended as follows:

"*We now analyse the CCM ensemble results. The ensemble mean ozone sensitivity profiles (Figs. 7c and f) markedly differ with ozone sensitivity profiles derived from observations (Figs. 7a and d) and CTM (Figs. 7b and e) at the corresponding periods. These differences are particularly pronounced in the upper stratosphere (above ~5 hPa). On the other hand, despite the two different periods, the ensemble mean ozone sensitivity profiles show very similar features with positive sensitivity from 15 hPa to the stratopause and a maximum sensitivity of 0.4 at ~3 hPa (Figs. 7c and f). This maximum tropical sensitivity value and its altitude level is in good agreement with previous CCM estimates (e.g. Rozanov et al., 2006; Austin et al., 2007; Gruzdev et al., 2009; Kubin et al., 2011). The CCM ozone sensitivity analysis has also been repeated for the period 2003-2005 (not shown) to be directly comparable with the CCM results of Sukhodolov et al. (2017): we found very similar ozone sensitivity profiles.*"

Other Comments:

(3) Introduction, first paragraph, last sentence. "A thorough understanding and accurate quantification of the UV variability effect on the middle stratosphere from which the "top-down" theory stems, are thus necessary." If so, then why is the CCM configuration limited to only the photochemical ozone response? The thermal response and its associated dynamical response are the main components of the top-down mechanism for solar influences on the troposphere.

Indeed, our study examines the photochemical ozone response, not the more general thermal and dynamical responses. We modified the manuscript as follows:

"*A thorough understanding and accurate quantification of the UV variability effect on the middle stratosphere ozone are thus necessary.*"

(4) Section 2.1, line 11. Are you using the NRL SSI version 1 or version 2? It is fine if you are still using version 1 but it should be clarified. Version 2 is available from https://data.noaa.gov/dataset/noaa-climate-data-record-cdr-of-solar-spectral-irradiance-ssi-nrlssi-version-2

We are indeed using the NRL SSI model version 1. This is now clarified in the revised version of the manuscript in section 2.1:

"In our study, we use the solar spectral irradiance provided by the Naval Research Laboratory Solar Spectral Irradiance (NRLSSI) model version 1 (Lean, 2000; Wang et al., 2005)."

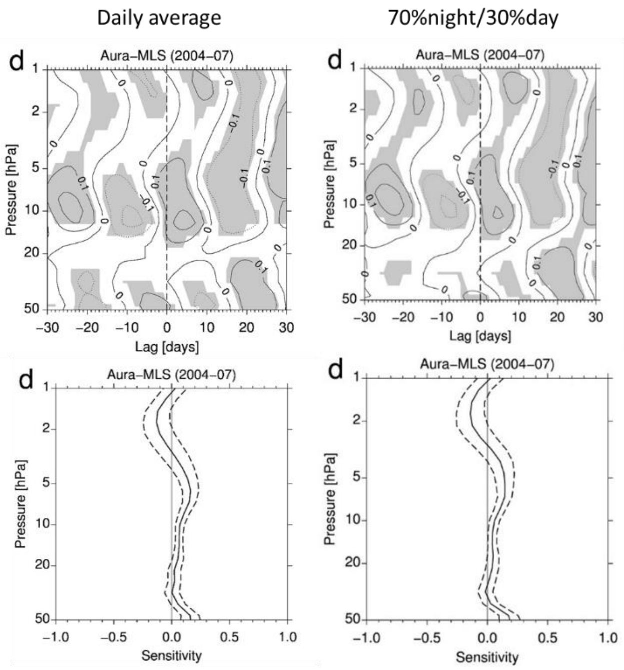
(5) Section 2.2, line 24. Please specify the pressure levels for ozone retrievals for the two MLS instruments. Which versions of the UARS MLS and AURA MLS data sets are being used for this analysis? Please reference more up-to-date descriptions of these data. The Version 5 UARS MLS data set is described by Livesey et al., JGR, v. 108, doi:10.1029/2002JD002273, 2003. Please give URLs where readers who wish to repeat the analysis can download the data. For example, the UARS MLS data are at https://mls.jpl.nasa.gov/uars/data.php.

We now clarified the pressure levels for ozone retrievals for versions 5 of UARS MLS and 4.2 of AURA MLS that are used in our study and give the references of the more up-to-date descriptions of the corresponding data (Livesey et al. 2003 for UARS MLS and Livesey et al. 2017 for AURA MLS). The URLs where data can be accessed are provided at the end of the paper section 6 (data availability).

Changes in the manuscript have been made in section 2.2:

"We used the version 5 UARS MLS dataset described Livesey et al., (2003). The ozone retrieval is based on 205 GHz radiances, provided onto 13 pressure levels in the range 100-1 hPa (100, 68.1, 46.4, 31.6, 21.5, 14.7, 10, 6.8, 4.6, 3.2, 2.2, 1.5 and 1 hPa) and has an average vertical resolution of 4 km in the stratosphere. The typical 1σ precision for ozone mixing ratio measurements is ~0.3 ppmv between 68 and 1 hPa."

(6) Section 2.2, line 31. If only 30% of the measurements are in the daytime, another problem arises, which is the ozone diurnal cycle. This cycle becomes important at roughly 2 hPa and above. Including 70% of measurements at night will therefore have the effect of reducing the estimated ozone response to solar UV variations at 2 hPa and above. This will not affect comparisons with the CTM and CCM provided that the model "measurements" also include both day and night data. Ideally, there should be 70% night and 30% day model data to allow an exact comparison. Please add text to explain this.



We tested the influence of an uneven distribution of night/day time measurements (see figure above). To do so, we reproduced the 2004-07 analysis Aura-MLS data but by resampling the data with the ratio of 70/30 of night/day time measurements and compared it to the daily average (which has a ratio of roughly 50/50). The results are fairly similar, indicating a limited diurnal effect on the unbalanced day/night sampling. Note that we repeated the analysis by considering only nighttime or daytime measurements (not shown) and did not find any significant difference.

The ozone diurnal cycle issue is now mentioned in section 2.2 :

"*Furthermore, the ozone diurnal cycle becomes important in the upper stratosphere, so that the results may be affected by the imbalance in daytime and night-time measurements used to construct daily time series. This issue will be discussed in section 3.2."*

(7) Section 2.3, line 24. 39 levels and 70 km lid means a resolution of less than 2 km. This is much better than the MLS vertical resolution, which is about 6 km. One should mention this before making direct comparisons in the following sections.

As clarified previously, the UARS MLS (Aura MLS) vertical resolution for ozone retrieval is 4 (3) km in the stratosphere. In the LMDz-Reprobus, model, the vertical resolution slowly varies with altitude, from 1 km in the upper troposphere to 3 km in the middle and upper stratosphere (i.e. above 10 hPa). Between 10 hPa and 1 hPa – i.e. the region we are focusing on - MLS and LMDz-Reprobus have roughly the same vertical resolution (~3 km).

We now clarify this in section 2.3:

"The model uses a classical hybrid σ-P coordinate in the vertical, has 39 vertical levels and a lid-height at ~70 km*. The model vertical resolution slowly decreases with height. In the middle and upper stratosphere (30-50 km or ~10-1 hPa) - focus of our study – the model vertical resolution reaches 3 km which is similar to the vertical resolution of UARS-MLS and Aura-MLS measurements in this altitude range. The model is integrated with a horizontal resolution of 3.75° in longitude and 1.9° in latitude.* The equations are discretized on a staggered and stretched latitude-longitude Arakawa-C grid."

(8) Figure 1. The units should be $W/m_2/nm$.

This has been corrected.

(9) Section 3.2, Figure 3. The periodogram of the MLS ozone measurements (Figure 3a,d) is done at 3.2 hPa. But, according to Livesey et al. (2003), the UARS MLS measurements were not retrieved at this level, only at 2.2 and 4.6 hPa. So, how are data obtained at 3.2 hPa?

For the version 5 of the UARS MLS measurements which is used in our study, the vertical retrieval grid over the stratosphere and lower mesosphere has been doubled compared to previous versions (i.e. v4 and before). Hence, the ozone measurements are provided at levels 2.2, 3.2, and 4.6 and not only 2.2 and 4.6 (see also Table 6 in Livesey et al. 2003). See also answer to comment #5.

(10) Section 3.2, lines 10-12. Please note that the lack of an obvious solar rotational signal in the MLS data considered here is partly because the measurements were obtained during the declining phases of solar activity using a limb sounding instrument, whose measurements are spatially and temporally sparse. The ozone signal is more easily detectable and repeatable in daily zonal means of nadir-viewing backscattered ultraviolet measurements under solar maximum conditions when solar UV variations are stronger and more coherent. The CTM simulations are also affected by the relatively weak solar rotational UV variations during the selected time periods.

We agree that the solar rotational signal should be more easily detectable during maximum phases of solar activity than during declining phases and this may contribute to the difficulty in the identification of a prominent peak in the power spectrum. Note however that we repeated the MLS power spectrum analysis during the maximum phase of solar cycle 24 (2012-2015) and we still did not identify a clear peak (over this period, the MLS sampling is large since almost 800 profiles are retrieved each day). Regarding daily zonal means of nadir-viewing backscattered ultraviolet measurements (like SBUV), it may be indeed easier to detect but we did not find any reference where this is clearly shown in power spectrum analysis (only in coherency as in our study).

We modified section 3.2 accordingly:

"*This illustrates the difficulty in detecting solar rotational signals in the observations, as well as in a single ensemble member over these 3year periods. Note that we additionally computed periodograms in observations during solar maximum phases (i.e. 2012-2015) where 27day fluctuations in the solar forcing are stronger than during the declining phase (not shown). The results were however similar and no clear peak at 27 days could be identified. Hence, the absence of a distinctive rotational signal suggests the presence of strong and rather random ozone variability of non-solar origin which makes the ozone rotational signal very difficult to detect and estimate*."

(11) P. 9, Figure 4. Normally, a cross-spectral analysis should yield phase estimates as well as coherency estimates. There is no mention of phase on p. 9 so it must be assumed that the coherency estimates are at zero lag. But the cross-correlation functions in Figure 5 show that the phase lags are not constant with altitude and are not always zero. They tend to be somewhat negative in the upper stratosphere and become positive in the middle and lower stratosphere. The ozone-UV sensitivities shown in Figure 6 are also presumably at zero lag. This differs from previous observational studies (e.g., Hood and Zhou, 1998) which calculated sensitivities at the so-called optimum lag, i.e., the lag where the correlation maximizes. Please add text to explain that these calculations are being done at zero lag and why this lag is chosen.

We made the choice not to provide the phase lag with the coherency since we calculate the cross-correlation afterwards which provides basically the same information and both are consistent.

The sensitivities are indeed shown at lag 0 and not at optimum lags. Optimum lags are in fact not simple to define as they may vary between observations and models results, between two different periods of observations, or between two ensemble members. Alternatively, we could choose one reference optimum lag vertical profile upon which the sensitivity would be calculated. But similarly, this poses the problem of defining the most accurate reference profile; shall we use observational or model results? Hence we opt for the lag 0 as a common reference. Finally, note that we tried both but it did not affect the results and conclusions as the sensitivity profiles shown in the answer to comment #2 reveal.

Section 3.2 has been modified accordingly:

"In previous studies, ozone sensitivity profiles were either calculated at optimum lags where the correlation coefficient maximizes (e.g. Hood and Zhou, 1998) or at zero lag (e.g. William et al., 2001; Austin et al., 2007). Both alternatives were tried but given the limited effect on the results and conclusions, we elected to show only ozone sensitivity profiles using a common time frame, hence at zero lag. Results are shown on Fig. 7."

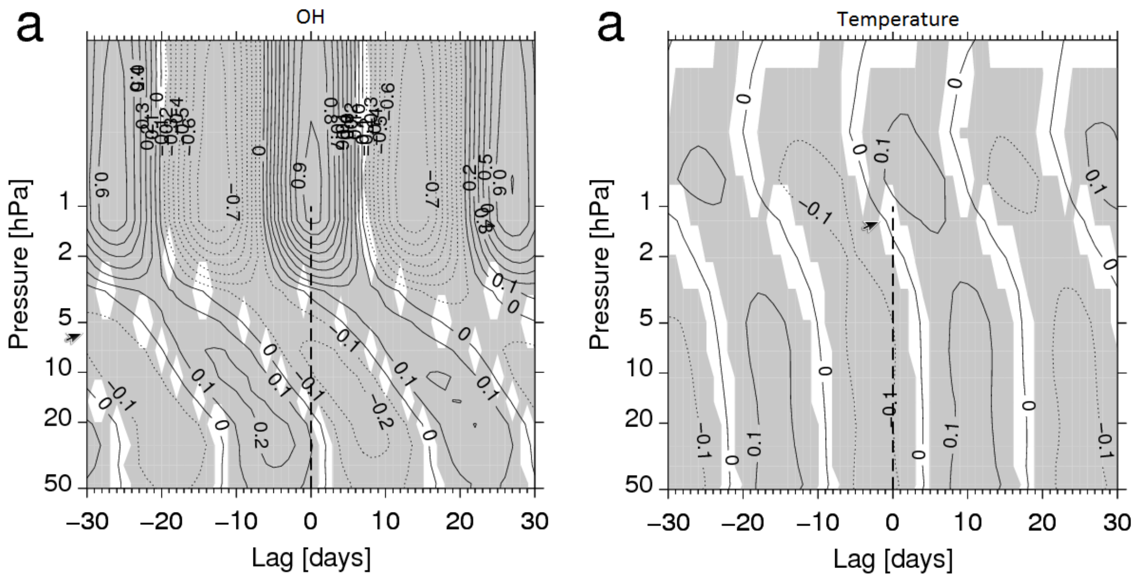
(12) P. 10, line 24. Typo: Seizing? Caption to Figure 3: from the runs ensemble?

We changed "*Seizing*" for "*Marked*" and corrected caption 3: "*The middle panels (b and e) represent the ozone Lomb-Scargle periodograms for CTM simulation and the bottom panels (c and f) the average periodogram of the CCM ensemble. The dotted envelop (c and f) indicates the 2σ standard deviation of the ensemble of CCM simulations."*

(13) P. 11, top of page. The CCM results shown in Figure 5c,f are characterized by negative lags near the stratopause. What is the cause of these negative lags? Is it feedback from a temperature response caused only by increased radiative heating associated with the ozone response (holding

direct UV heating changes constant)? Or, is it increased photolysis of water vapor in the lower mesosphere and resulting destruction of ozone by odd hydrogen? Or both? Can this be diagnosed?

Below are shown the cross-correlation analysis (as in Fig. 5) for the hydroxyl and the temperature for the CCM ensemble mean. The hydroxyl response is highly consistent with the results of Sukhodolov et al. (2017) (see their Figure 3). The temperature response is also consistent. Thus even in absence of the direct UV heating, an "indirect" temperature response is produced by the ozone response. These two effects may thus contribute to the negative lags near the stratopause. Note the particularly good agreement in the upper stratosphere ozone phase lag with Sukhodolov et al. (2017) which suggests that the OH contribution is more important than the temperature one because in our case the temperature response is strongly reduced: the temperature sensitivity in our experiments yields a maximum of 0.035 in the upper stratosphere instead of 0.1 in Sukhodolov et al. (2017).



We modified the manuscript as follows:

- In the introduction, we added a short paragraph to describe the effect of HOx on ozone in the upper stratosphere and mesosphere: "*Fleming et al. (1995) further stressed the increasing importance with height of the solar-modulated HOx chemistry on the ozone response above 45 km. In the upper stratosphere and mesosphere, enhancement of HOx through photolysis of water vapour in Lyman-alpha line associated with an increasing solar irradiance contribute to destroy ozone. Above ~65 km and at zero-lag, the latter mechanism dominates over ozone production (i.e. by photolysis of oxygen) leading to a negative ozone-solar irradiance correlation. In the upper stratosphere and lower mesosphere (below 65 km), although ozone production dominates, increasing HOx at zero-lag contributes to the negative lag of the ozone response (Rozanov et al., 2006).*"
- We also mention this mechanism in section 3.2 when we discuss the cross-correlation analysis shown in Figure 5: "*Above 3 hPa (~40 km), CCM cross-correlations of both periods (Fig. 5c,f) show a maximum at negative time lag (-2 days). As mentioned in the introductory section, this negative time lag can be induced by temperature feedback on ozone and by increasing HOx with solar irradiance which contributes to destroy ozone. While our model configuration allows to fully account for the HOx effect, the solar-induced temperature response is limited since the direct radiative heating effect is not included. The temperature response to the 27-days cycle is thus solely controlled by ozone production in the photolysis scheme. Although a temperature signal is found (not shown), it is small, reducing the*

likelihood for the solar-induced temperature feedback to be prominent in our experiments. Despite the approximation made in our model configuration, we notice however that the upper stratosphere negative lags compare very well with those found in CCM experiments of Sukhodolov et al. (2017) (see their Fig. 3) in which both HOx and solar-induced temperature feedback effects are fully included. Hence, this suggests that neglecting the direct effect on heating rates generated by UV variations has a limited effect on the ozone response, at least at 27-days timescale. More sensitivity experiments are required however to quantify accurately the impact of this approximation."

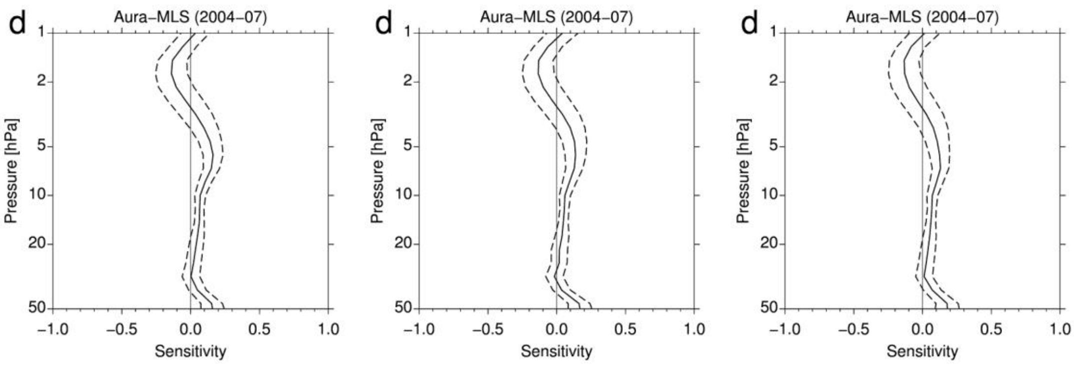
(14) P. 11, bottom of page. In addition to not mentioning the anomalously large CCM response in the upper stratosphere, there is also no mention here of the likely effect of the ozone diurnal cycle in reducing the ozone response in the upper stratosphere relative to that measured earlier from backscattered ultraviolet instruments, which operated only in the daytime. This difference is emphasized in Hood and Zhou [1998] for example.

The CCM response is indeed anomalously large compared to observations and CTM results. It is however not anomalously large when comparing with Sukhodolov et al. (2017) (see also answer to comment 2). Section 3.2 has been modified accordingly.

Regarding the diurnal cycle, we tested its influence on the results over the period covered by Aura-MLS (i.e. 2004-2007). The Aura-MLS instrument measures in the tropics at one fixed night local time (~0142 LST) and one fixed day local time (~1342 LST). To mimic an irregular sampling with respect to the local time, we repeated the Aura-MLS analysis as follows:

1. We initially build the ozone time series using daytime measurements only (1095 days in total)
2. Out of these 1095 days, we select N days randomly where daytime measurements are replaced by nighttime measurements only.
3. We then compute the ozone sensitivity.

The procedure was repeated for N=100 (i.e. 91% of daytime measurements), N=500 and N=1000 (i.e. 9% of daytime measurements) (see figure below) and the results reveal very minor differences between the various sensitivity profiles. Hence this analysis suggests that the diurnal cycle has a small effect on the ozone solar rotational signal.



We added a paragraph to discuss ozone diurnal cycle at the end of section 3.2:

"As mentioned in the section 2.2, the results based on UARS-MLS measurements may be affected by the imbalance between night and daytime sampling due to the ozone diurnal cycle becoming significant in the upper stratosphere. To test the influence of the ozone diurnal cycle, we repeated all the analysis performed in this section by mimicking an irregular sampling over the period covered by Aura-MLS (i.e. 2004-2007). Each day, ~800 ozone vertical profiles of the Aura-MLS instrument are evenly retrieved in the tropics [20S-20N] at two fixed local times: one at night (~0142 LST) and

one during daytime (~1342 LST). We initially build the ozone time series using daytime measurements only (1095 days in total). Among these 1095 days, we selected N days randomly where daytime measurements were replaced by night time measurements. We then repeated the spectral, correlation and regression analysis. The procedure was performed for various values of N, from N=100 (i.e. 91% of daytime measurements) to N=1000 (i.e. 9% of daytime measurements). The results (not shown) revealed almost no dependence to N, suggesting that the diurnal cycle has a small effect on the ozone solar rotational signal."

(15) Section 4. While it is useful to carry out these analyses, one must question whether the CCM in its chosen configuration (no direct solar UV heating changes) is ideal for this purpose. Also, a time window with a length of 10 years includes both solar maximum periods (when 27-day UV variations are strong and numerous) as well as solar minimum periods (when these variations are weak and sparse). Could it therefore be possible that a shorter time window of 3 years centered on a strong solar maximum (e.g., that in 1979-82) could yield more reliable results than a 10-year window which includes mostly non-maximum solar conditions?

Regarding the suitability of the chosen configuration (see also answer to major comment #2), paragraphs have now been added at several places in section 3.2 where we discuss our results by comparing with previous independent CCM studies (i.e. Rozanov et al., 2006 ; Sukhodolov et al., 2017). We further mention this in the concluding paragraph of section 3:

"*Moreover, the fine correspondence of our results with those based on independent previous chemistry-climate modelling experiments (e.g. Rozanov et al., 2006; Sukhodolov et al., 2017) emphasizes the relevance of our experimental model setup (i.e. despite neglecting the direct effect on heating rates) to examine the ozone response to 27day solar variations.*"

Regarding the discussion of the length of the time window, we agree that it strongly depends on the amplitude of solar rotational fluctuations and, hence, the phase of the solar cycle. We examined this further with the solar maximum of cycle 23 with the model and found that choosing a 10year time window length still leads to the more accurate and robust estimates of the ozone sensitivity in comparison with restricting the analysis to one solar maximum period only. Of course, the strongest the solar maximum and its associated 27day solar fluctuations are, the shorter the time window size can be. But if a best option has to be given, our results seem to suggest that a long time window is preferable.

We modified the main text of section 4 to make this point clearer:

"*These uncertainty ranges also strongly depend on the amplitude of the solar rotational variations and hence the phase of the 11year solar cycle; we find that during solar maximum of cycle 23, minimum of cycle 22, a minimum time window size of 2, 5 years, respectively, is required for the standard deviation to drop under 50%. To obtain a standard deviation lower than 20%, we however found that randomly choosing a 10year time window length performs better than restricting the analysis to short but solar maximum period only (i.e. solar 23). These results suggest that long time series are preferable to estimate accurately the ozone sensitivity to solar rotational fluctuations in observations.*"

(16) Minor corrections: In the abstract, lines 23-24, neither nor should be either or. P.13, line 10. anti-correlation should be inverse correlation.

This has been corrected.

(17) P. 15, lines 11,12: "Applying the same spectral analysis to the average of the CCM ensemble simulations allows reducing the 'masking' effect by random dynamical variability, so that the rotational signal in ozone can be more easily identified and estimated." However, the negative aspect of this approach is that the CCM may not perfectly simulate the actual ozone response to

short-term UV variations, partly because of the neglect of the direct radiative effet of the UV variations in the model, and their secondary dynamical effects.

We added a full paragraph to discuss this aspect in section 5:

"In our CCM experimental design, the direct radiative effect of UV on heating rates has been neglected leading to an underestimated temperature response to the 27day cycle. As a consequence, this may affect the ozone response by reducing the magnitude of the solar-induced temperature feedback on chemical reaction rates. Nevertheless, a detailed comparison of the ozone response in our analysis with results from previous independent CCM studies (Rozanov et al., 2006 ; Sukhodolov et al., 2017) revealed a very good correspondence, despite the fact that their experimental design included the direct radiative heating effect. This suggests that the feedback exerted by the solar-induced temperature fluctuations on ozone is modest, at least at the 27day time scales. This is in fact not surprising. In their recent study, Sukhodolov et al. (2017) shown that the atmospheric internal variability largely dominates the variability of the stratospheric temperature on 27day time scales, making the temperature response to the solar forcing difficult to identify and, hence, the influence of its feedback on ozone secondary. Nonetheless, we recognize that to quantify properly the impact of the neglected solar-induced temperature feedback on our results, additional CCM experiments including the direct radiative effect of UV on heating rate should be performed."

(18) P. 15, lines 18-21: "Analysis of the CCM ensemble simulations suggest that the differences mostly originate from the dynamical variability." Usually, internal dynamical variability in a model is larger than in observations so it is not clear that a single model run is equivalent to a single sample of observations (or a single run of the CTM). The large spread in the ensemble mean sensitivity profile could also reflect a less complete simulation of the upper stratospheric dynamical response to short-term solar UV variations.

See answer to comment #17.

Anonymous Referee #3

The paper uses models and satellite observations to investigate the response of tropical stratospheric ozone to short-term solar UV variations due to the 27-day solar rotation. This is a topic that has been investigated in a number of previous studies but is still not resolved; therefore an updated study will be of interest to journal readers. The stated goals of the present paper are to "(i) assess the influence of the solar cycle phase on the ozone sensitivity to the rotational cycle and (ii) quantify the time window required for a robust estimation of the ozone sensitivity".

While the paper does address the stated topics, the focus is drawn elsewhere by awkward organization and extraneous material. The authors do not make it clear why they include Section 3, which is longer than the section addressing their goals. Section 3 does not contribute to the focus of the paper and, in my view, does not contribute to the understanding of the solar response.

My most serious concern about the paper is in regard to the implicit assumptions in the text. At a number of places, the authors have assumed that a solar response is present even when they do not see a signal of it in their analysis or that the response is larger than what they find from the analysis. The text then describes why and how the signal has been masked. This is dangerous; if you do not find a signal of a response, the first explanation should be that there is no response. Even if processes that might mask a signal are present, it is not appropriate to conclude that this masking is the reason for not finding a signal that is "known" to be present based on prior assumptions. A more appropriate way to say it would be: if there is a response, it is too weak to detect.

We thank the reviewer for reading the manuscript and providing helpful comments and suggestions. We address the raised issues in turn below.

Our answers to comments and suggestions are written in blue and manuscript changes are written in italic type within double quotes ("*like this*").

Section 3 is a case study which follows up on Bossay et al. [2015] study using, in addition to observations, CTM and an ensemble of CCM simulations results. This part allows illustrating clearly, with a concrete case, the effect of internal variability when retrieving the response of ozone to solar forcing at 27-day timescale. Another interesting point of Section 3 is that it allows comparing model and observations and validating the model in light of the very good agreement when comparing the CTM and observational results in 2004-2007. We however agree that in the initial version of the manuscript, Section 3 was not sufficiently motivated. The manuscript has now been revised accordingly (See answer to major comment 1 below).

Regarding the second main concern of the reviewer, we agree that more caution is required when describing results where no signal is found (namely Figure 3 where a power spectrum analysis of raw ozone time series is performed). The manuscript has been revised accordingly.

The description of figure 3 and 4 has been changed in order not to presume of a solar rotational signal:

"The two periodograms of MLS ozone measurements (Fig. 3a and Fig. 3d) reveal no prominent peak in the range of the 20-30 days period, *suggesting an absence of a solar rotational signal in ozone*. More prominent peaks are found at longer periods although they are not consistent between the two periods. The large peak found at the 35day period for 1991-94 corresponds to the yaw-maneuver period of the MLS instrument as described previously (Froidevaux et al., 1994; Hood and Zhou, 1998). Similarly to observations, the periodograms of CTM results (Fig. 3b and Fig. 3e) do also not exhibit a distinctive solar rotational peak; there are some minor peaks between 20 and 30 days and their amplitudes are smaller in 2004-07 than in 1991-94. *The analysis has been repeated at lower pressure-height levels (e.g. 10 hPa, not shown) and led to the same conclusions. Overall, the raw power spectrum analysis of observations and CTM results in the middle and upper tropical* *stratosphere does not allow identifying an ozone signal associated with the solar forcing fluctuations at rotational timescales for the two periods considered here.*

[…]

We further examine the relationship between stratospheric ozone and solar rotational cycle by performing cross-spectrum analysis between stratospheric ozone and F205. *Despite the absence of a solar rotational peak in the ozone power spectrum derived from observations and CTM results*, cross-spectrum analysis should help identifying coherent variability modes between the solar forcing and tropical ozone. Figure 4 presents the vertical profile of the magnitude-squared coherence (hereinafter referred as coherence) between F205 and tropical stratospheric ozone from MLS observations (a and d), CTM model results (b and e) and CCM model results (c and f)."

We also made correction throughout the manuscript to avoid overstatements regarding the ozone response to F205 when this is not so clear.

It is however very clear that there is a response, as the cross-spectrum analysis reveals. In this regard, we now added a new Figure to the manuscript to show the difference in the ozone/F205 coherency between forced and unforced CCM experiments.

Major Comments

1. As indicated above, I did not see the purpose of Section 3. Since you consider two fairly short 3-year periods, the analyses do not have any bearing on the questions raised about variations of the response with timing within the 11-year solar cycle or the dependence on the length of the analysis period.

Given that our analysis focuses on the 27-day cycle, one would expect 3 years to be sufficient to characterize the ozone response on rotational timescales because it represents about 40 cycles. Our results show it is not sufficient.

The first part of our paper, which considers the declining phases of cycles 22 and 23, actually serves as a case study where we compare observations and model results. It is a follow-up of the study of *Bossay et al*. [2015] which found some intriguing behavior in the response of the ozone to the 27-day solar cycle. Namely, that the correlation between the ozone and the forcing seems stronger when the amplitude of solar rotational fluctuations is small (more details are written in the introduction). We also found in the literature large differences between observational studies. To understand better this apparent contradiction and the variability of the ozone response to solar rotational fluctuations found in different observational studies, we need more than only one realization in order to improve the statistics and better understand the role of other sources of ozone variability. We thus used an ensemble of CCM simulations.

To make the purpose of section 3 clearer, we revised the introductory part as follows:

"As a first step, we follow up on the case study of Bossay et al. [2015] and make use of observations and modelling results comparison to provide a detailed picture of the ozone response to the solar rotational cycle during the declining phases of cycle 22 and cycle 23. We particularly aim to better understand the strong differences in the ozone response to solar rotational cycle found between the two periods. Two configurations of the LMDz-Reprobus chemistry climate model simulations are used, with specified dynamics (i.e. Chemistry Transport Model, or CTM) and in its free running mode (CCM). In the CTM configuration, temperature and wind fields calculated by the model are relaxed towards meteorological analysis; the dynamics is expected to be rather close to the reality, allowing direct comparisons with satellite observations for evaluating model chemical processes and its relevance to our study. In the CCM configuration, an ensemble of simulation is performed. Comparing the CCM ensemble results to CTM and observations during the declining phases of cycle

22 and cycle 23 allows to understand better the effect of internal dynamical variability on the ozone response. As a second step, we take advantage of the ensemble of CCM simulations and its large statistics to (i) assess the influence of the solar cycle phase on the ozone sensitivity to the rotational cycle and (ii) quantify the time window required for a robust estimation of the ozone sensitivity."

We also revised the introductory of section 3. part as follows:

"*In this section, we analyse the ozone response to the solar rotational cycle over the declining phase of solar cycles 22 and 23 in the observations and in the CTM and CCM model simulations. The analysis presented here follows up on Bossay et al. (2015) observational study. In particular, we aim to assess the model performances, understand better the differences in the results between the two solar declining phase periods and highlight the importance of internal dynamical variability*."

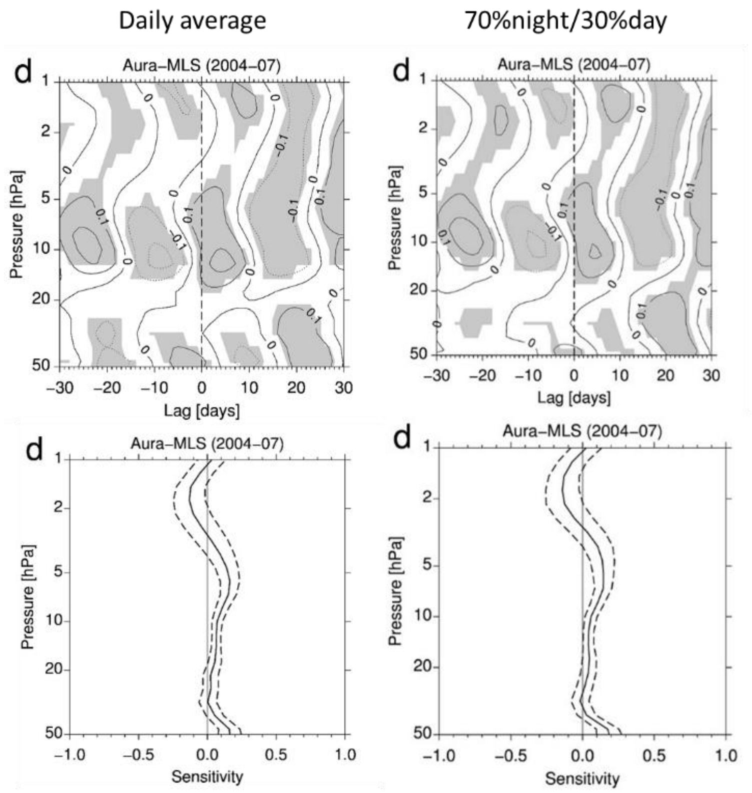
2. Perhaps the response diagnosed from MLS is intended as validation for your model simulations. In this case, why ignore the 9.5 years of MLS/Aura observations that have been taken since 08/2007? As you later find using model simulations, a 3-year period does not give results that are very robust and so is not convincing as validation.

In light of the observation/CTM results comparison in 2004-2007, we argue that it is rather convincing as validation: model and observations show almost the same results on the several diagnostics we applied. We also performed extra analysis over the 2003-2005 period to compare with Sukhodolov et al. [2017] which also reveal very consistent results. This is now clarified in the Section 3 of the manuscript.

In our study, it is very important to use short periods as it is what was done in previous efforts. We can thus compare our results more easily and provide explanation on the different discrepancies that have been found in these previous studies.

Further note that the prolonged solar minimum after 2007 which lasted for almost 5 years has an exceptionally weak 27-day component. As a consequence, it is not the most appropriate period to examine ozone response to 27-day solar flux variations. There is again another maximum from 2011-onwards that could be investigated and we did it but it does not bring any further elements. In this regard, we do not intend to extend our CTM and CCM ensemble simulations by ~10 years that would turn out being very expensive.

3. It seems that both satellite data (in Section 3) and model output (throughout) are zonally and latitudinally averaged over each day, including both day and night. There is a mention of local time issues (Section 2.2) but you then decide not to use any local time or day/night information in your analysis. There is evidence that this should be considered: 1) why else would the MLS/UARS ozone variations show a prominent peak at the yaw period?; 2) it is known that local time variations in the response of ozone in the upper stratosphere to solar variability are not negligible (e.g. Li et al., Earth and Space Science, doi:10.1002/2016EA000199).



We tested the influence of an uneven distribution of night/day time measurements (see figure above). To do so, we reproduced the 2004-07 analysis Aura-MLS data but by resampling the data with the ratio of 70/30 of night/day time measurements and compared it to the daily average (which has a ratio of roughly 50/50). The results are fairly similar, indicating a limited diurnal effect on the unbalanced day/night sampling. Note that we repeated the analysis by considering only nighttime or daytime measurements (not shown) and did not find any significant difference.

The ozone diurnal cycle issue is now mentioned in section 2.2:

"Furthermore, the ozone diurnal cycle becomes important in the upper stratosphere, so that the results may be affected by the imbalance in daytime and night-time measurements used to construct daily time series. This issue will be discussed in section 3.2."

The following paragraph has been added in section 3.2:

"As mentioned in the section 2.2, the results based on UARS-MLS measurements may be affected by the imbalance between night and daytime sampling due to the ozone diurnal cycle becoming significant above 2 hPa. To test the influence of the ozone diurnal cycle, we repeated all the analysis performed in this section by mimicking an irregular sampling over the period covered by Aura-MLS (i.e. 2004-2007). Each day, ~800 ozone vertical profiles of the Aura-MLS instrument are evenly retrieved in the tropics [20S-20N] at two fixed local times: one at night (~0142 LST) and one during daytime (~1342 LST). We initially build the ozone timeseries using daytime measurements only (1095 days in total). Among these 1095 days, we selected N days randomly where daytime measurements were replaced by nighttime measurements. We then repeated the spectral, correlation and regression analysis. The procedure was performed for various values of N, from N=100 (i.e. 91% of daytime measurements) to N=1000 (i.e. 9% of daytime measurements). The results (not shown) revealed almost no dependence to N, suggesting that the diurnal cycle has a small effect on the ozone solar rotational signal."

Regarding the Li et al (2016) paper, the problematic is quite different. The orbit of SBUV drifts relatively to the diurnal cycle on the same timescale as the solar signal which is investigated (decadal). So, in this case, there is an artificial decadal fluctuation simply created by the drift which aliases the “real” decadal signal of solar origin. This type of problem does not apply in our case, though.

In UARS-MLS, the yaw maneuver creates an artificial periodicity of 36 days in zonally average data. Furthermore, the non-fixed local time measurements may introduce spurious variations in the temporal evolution of the daily zonal mean (due to the diurnal cycle) in the upper stratosphere. This may partly affect the observed signals in the upper stratosphere for the period 1991-94.

4. Although some spread should be expected, if I see a response peaking at 22 days (as you show in Figure 4a), I would automatically assume that it has no relation to the solar rotation. The signal in that particular panel is near zero at 27 days. It seems shaky to interpret it as driven by the solar flux variations. Can you provide more justification for your interpretation?

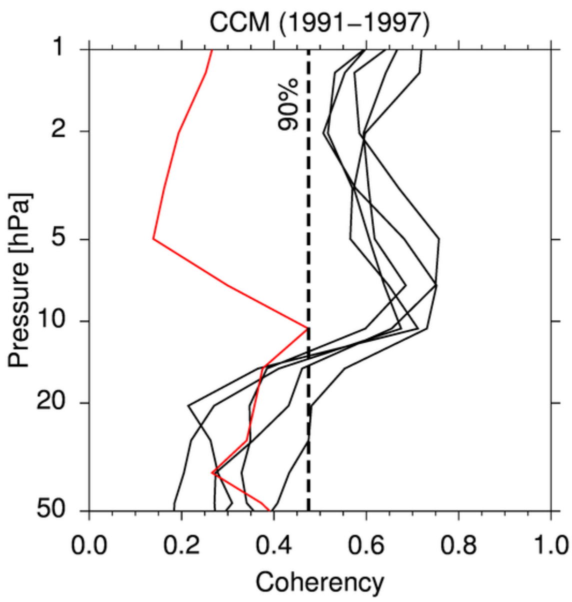
Active regions are not always located at the same longitude on the Sun. Furthermore, the Sun’s rotational period depends on the latitude (i.e. differential rotation). For these reasons, we do not expect a thin peak at 27 days, but a rather broad peak near 27 days. Wavelet analysis of solar forcing time series (Figure 3c) shows that solar forcing fluctuations are strong in the band 20-30 days, hence it is not shocking to see a large patch of coherency between 20 and 27 days. The most convincing evidence of a link between the solar flux variations and the ozone response is the high coherency between both: it means that the UV forcing and ozone vary together.

See also answer to the next comment (#5) for further justification.

5. (“additional CCM simulation where the solar flux is kept constant”) Since you show that the CCM responses vary considerably between realizations, a single simulation is not a very useful. Alternatives would be to perform additional realizations and/or to perform a similar case (fixed solar flux) with the CTM, particularly for a longer period (>10 years).

The CCM realization with constant solar flux is performed in the CCM configuration from 1/1/1991 to 30/11/1997. To be fair, we compared the coherency in this experiment with the coherency in the five individual ensemble members over the same period. Below, we plotted the vertical profiles of the spectral coherency around 27 days between the F205 and ozone for the (dashed) constant solar experiment and (solid) the five individual ensemble members. It is clear that although the spectral coherency varies among the different ensemble members, the results are extremely similar above the 10 hPa level and very robust. If we remove the forcing, there is no coherency signal (dashed line). Given the very consistent results between the five different ensemble members, we are confident in saying that the coherency signal is due to the forcing and is not fortuitous. We now modified the manuscript as follows.

We added the following Figure:



"Figure 5. Vertical profile of the mean squared coherence between ozone and F205 averaged between 22 and 30 day periods and calculated for the time period 1991-1997. The black lines correspond to the results of individual ensemble members (five in total) and the red lines to the results of the experiment forced with constant solar forcing. The vertical dashed line indicates the 90% confidence limit."

We added the following part in the main text:

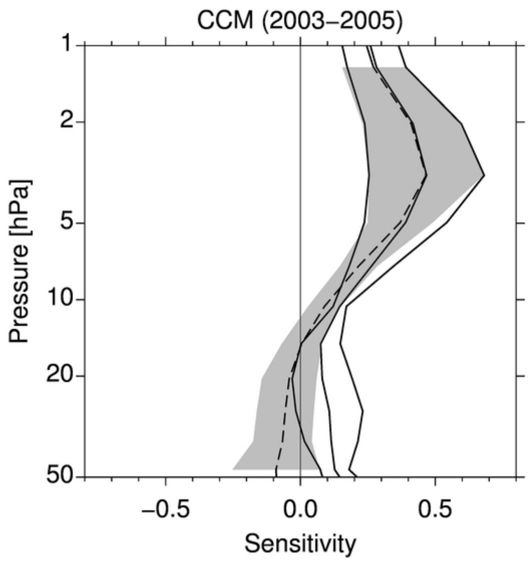
"To further test the robustness of the coherence signal, we perform an additional CCM simulation for the period 1991-1997 where the solar forcing is kept constant by using fixed (i.e. climatological) photolysis rates during the model simulation. Results are shown on Fig. 5. Below 15 hPa, the different experiments show no significant coherence between ozone and solar flux. Between 15 and 1 hPa, all forced experiments (black lines) reveal a similar and significant coherence signal while for the constant solar forcing experiment (red line), the coherence is weak and within the range of randomness. The absence of significant coherence found in the constant solar experiment confirms that the coherence found between F205 and stratospheric ozone is not fortuitous and primarily originates from photolysis processes"

6. I am having trouble reconciling Figure 4c, f with Figure 6 and 7. Figure 4 indicates largest signal at ~10 hPa and near zero signal at ~0.3 hPa while Figures 6-7 indicates the opposite. Even though sensitivity (Fig 6-7) is different from absolute response (Fig4), these do not appear to be consistent. Please explain.

Please note that:

1/ none of our analysis goes up to the lower mesosphere (i.e. 0.3 hPa). We will however assume that the reviewer meant 3 hPa.

2/ in the upper stratosphere, there is not near zero coherency signal at 3 hPa in Figure 4c and f (the signal is just weaker and only in Figure 4f).



On Figure above, we plotted the ensemble mean and spread of the ozone sensitivity to F205 index over the period 2003-2005 to compare our results with those of Sukhodolov et al. (2017) (see also answer to reviewer #2). The ozone is shown at optimum lag (where the correlation maximizes, solid) and zero lag (as in our analysis throughout the paper, dashed). This analysis shows that - particularly clearly when we examine the optimum lag sensitivity profile – the sensitivity does not maximize at 10 hPa, but it corresponds to the pressure level where the sensitivity average value is the most stable (as revealed by the strongly reduced standard deviation). Hence, coherency results and sensitivity are consistent.

7. Your conclusion (p. 15, l. 18) that "the differences mostly originate from the dynamical variability" is an important one that should be brought out more prominently.

We insist on this conclusion at many places in the manuscript (abstract, results part, conclusion). The various corrections in the manuscript also allow insisting further on the importance of the internal variability.

Specific Comments

1. (p. 2) "the life span of a single satellite instrument is generally far less than one solar cycle" This has been true for a few but just as many last for a time span comparable to a solar cycle.

We changed the sentence to:

"*Furthermore, the life span of a single satellite instrument is generally shorter than (comparable to in some cases, e.g. MIPAS-ENVISAT, MLS-Aura) one solar cycle*"

2. (p. 3, l. 24) Why "nonlinear"? Any dynamics will affect ozone.

The term nonlinear has been removed.

3. (p. 7) The description of simulated solar flux variation was confusing. You imply that the variations are included in your photolysis lookup table but I could not tell exactly how. Is the table recalculated every day? Is there a separate table for each value of your solar flux parameter? Please be explicit. Also, the impact of the solar variation on heating rate is not clear. Reading between the lines, I guess what you mean is that the heating will respond to the increased or decreased ozone but that, in the heating part of the calculation, the solar flux is kept constant. Is that what you mean? And one other comment: O2 should also be included on your list of radiatively active gases.

We now provide more details on the fact that the photolysis separate look-up table is calculated every day from daily NRLSSI solar flux. We added the following sentence:

"A separate photolysis look-up table is calculated every day using the daily NRLSSI as solar input"

We also now provide more explanation on the effect of neglecting UV variations associated with the 27-day rotational cycle on heating rates. The following paragraph has been added:

"Note however that the direct effect on heating rates generated by UV variations associated with the 27-day rotational cycle is neglected: i.e. daily changes in the spectral irradiance are not considered in the CCM radiative scheme. As a consequence, part of the thermal and dynamical responses to the 27-day rotational cycle and hence their effect on ozone (through transport and temperature dependent chemical reactions, as described above) are missing. The impact of this approximation on our results seems to be small though, as discussed thereafter (sections 3 and 5). Note also that on timescales of the 11yr cycle, Swartz et al. (2012) found that their photolysis-only simulation captured almost all of the solar cycle effect on ozone."

Finally, we added O_2 in the list of radiatively active gases.

4. (Figure 3) There is a mismatch between the level shown in the figure (~3 hPa) and the level where you see a response in Figure 4 (~10 hPa). Perhaps this is related the mismatch between the SAGE/SBUV results, which contributed to the conclusions in the Hood paper you cite to choose the level of maximum response, and other data and models (e.g. see discussion by Dhomse et al., 2016). Also, a better label for the area where you see a signal would be middle stratosphere, not upper.

The pressure level at ~3 hPa level is the one we used throughout the manuscript as it corresponds to the level where the maximum sensitivity response is found (not only in our study but also in many other observational and model based study). Note that we repeated the power spectrum analysis at 10 hPa and similar conclusions were reached. This has been now clarified in the main text:

"The analysis has been repeated at lower pressure-height levels (e.g. 10 hPa, not shown) and led to the same conclusions."

Regarding the fact that the stronger coherency signal is found at 10 hPa while the maximum sensitivity is found at 3 hPa is explained in the detail in the answer to major comment 6.

5. (p. 9, l. 9) "This explains why ..." This could be the explanation but, as you show later, you are not using enough data to determine a robust signal. It would be safer to say that "This could contribute etc.". Since you cannot see a signal in the observational analysis, it is not appropriate to assume that the response is there but the signal is masked. There may not be a response.

We now changed the text as follows:

"This illustrates the difficulty in detecting solar rotational signals in the observations, as well as in a single ensemble member over these 3year periods."

6. (p. 10, l. 28) "The absence of correlation signal in the middle and lower stratosphere in the observations is consistent with the large noise present in the ozone dataset at these altitudes" As in the comment above, this is misleading since it implies that there is an ozone response but it is masked. You have not shown that a response exists.

There is a signal found in the CTM in the lower stratosphere (below 10 hPa). This signal is however not found in observations. We reformulated the text as follows:

"The fact that the correlation signal in the middle and lower stratosphere (below 10 hPa) is found in the CTM but not in the observations may partly arise from the large noise present in the MLS-UARS ozone dataset at these altitudes (not shown)".

7. (p. 13, l. 10) “overall anti-correlation” All I see is that there is a period when F205 variance is low and sensitivity variance is high. The curves are otherwise not related and, even in this period, do not follow a similar evolution. Coincidence of one perturbation is not enough to deduce anti-correlation.

We reformulated as follows:

“This is further supported by the apparent inverse relationship which is found between the F205 index variance (Fig. 8b) and the ozone sensitivity variance (Fig. 8d)”

Editorial comment

“increasing (decreasing)” and similar construction is grammatically incorrect and very confusing, especially since elsewhere you use parentheses in their legitimate use to define or clarify, e.g. “solar forcing index (F205)”.

We modified the following sentences:

“The phase lag vertical profile between the ozone response and the solar forcing was found to be negligible at about 40 km and gradually increasing/decreasing, below/above that altitude”

“After removing outlier values, 85% and 93% of the 1095day ozone time series of the periods 1991-94 and 2004-07, respectively, are kept for the analysis”

“Given that the amplitude of the rotational cycle increases with increasing solar activity, one may thus expect minimum and maximum sensitivity during 11year solar maximum and minimum phases, respectively.”

Sensitivity of the tropical stratospheric ozone response to the solar rotational cycle in observations and chemistry-climate model simulations

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Abstract. The tropical stratospheric ozone response to solar UV variations associated with the rotational cycle (~27 days) is analysed using MLS satellite observations and numerical simulations from the LMDz-Reprobus chemistry-climate model. The model is used in two configurations, as a chemistry-transport model (CTM) where dynamics are nudged toward ERA-Interim reanalysis and as a chemistry-climate model (free-running) (CCM). An ensemble of five 17year simulations (1991-2007) is performed with the CCM. All simulations are forced by reconstructed time-varying solar spectral irradiance from the Naval Research Laboratory Solar Spectral Irradiance model. We first examine the ozone response to the solar rotational cycle during two 3year periods which correspond to the declining phases of solar cycle 22 (10/1991-09/1994) and solar cycle 23 (09/2004-08/2007) when the satellite ozone observations of the two Microwave Limb Sounders (~~MLS~~ UARS MLS and ~~MLS~~ Aura MLS) are available. In the observations, during the first period, ozone and UV flux are found to be correlated between about 10 and 1 hPa with a maximum of 0.29 at ~5 hPa; the ozone sensitivity (% change in ozone for 1% change in UV) peaks at ~0.4. Correlation during the second period is weaker and has a peak ozone sensitivity of only 0.2, possibly due to the fact that the solar forcing is weaker during that period. The CTM simulation reproduces most of these observed features, including the differences between the two periods. The CCM ensemble mean results comparatively show much smaller differences between the two periods, suggesting that the amplitude of the rotational ozone signal estimated from MLS observations or the CTM simulation is strongly influenced by other (non-solar) sources of variability, notably dynamics. The analysis of the ensemble of CCM simulations shows that the estimation of the ensemble mean ozone sensitivity does not vary significantly ~~n~~either with the amplitude of the solar rotational fluctuations, ~~n~~or with the size of the time window used for the ozone sensitivity retrieval. In contrast, the uncertainty of the ozone sensitivity estimate significantly increases during periods of decreasing amplitude of solar rotational fluctuations (also coinciding with minimum phases of the solar cycle), and for decreasing size of the time window analysis. We found that a minimum of 3year and 10year time window is needed for the 1σ uncertainty to drop below 50% and 20%, respectively. These uncertainty sources may explain some of the discrepancies found in previous estimates of the ozone response to the solar rotational cycle.

1 Introduction

The thermal structure and the composition of the middle atmosphere are sensitive to fluctuations in the incoming solar radiation, which in turn can affect the Earth's surface climate variability (Gray et al., 2010). These solar variations are dominated by the 11year solar magnetic activity cycle and the solar rotational cycle, also called 27day solar cycle. Changes in total solar irradiance (TSI) over an 11year solar cycle are typically lower than 0.1%, that correspond to 1 W m^{-2} change for a reference value of 1360.8 ± 0.5 W m^{-2} (Kopp and Lean, 2011). Such small variations in the total energy input are not expected to have a significant impact on climate, compared for instance to the variations of anthropogenic origin, and thus air-sea coupling mechanisms have been proposed that act to amplify the small solar initial perturbations (e.g. Meehl et al., 2008). Another possible amplification mechanism, also known as "top-down" (Kodera and Kuroda, 2002), operates through changes in the spectral solar irradiance (SSI) - in particular in the ultraviolet (UV) range - that directly modulate the stratospheric temperatures and ozone concentrations. These perturbations induce dynamical changes in the stratosphere, which may in turn affect the tropospheric circulation through stratosphere-troposphere couplings (e.g. Gerber et al., 2012). A thorough understanding and accurate quantification of the UV variability effect on the middle stratosphere ozone, from which the "top-down" theory stems, are thus necessary.

Solar irradiance fluctuations strongly depend on the wavelength range and their relative amplitudes tend to increase sharply with decreasing wavelengths (Lean, 2000). In the UV range, the variability over the course of the 11year solar cycle is of about 8% at 200 nm. Several observational and modeling studies have examined the impact of 11year UV variability on stratospheric ozone and temperature (e.g. Hood, 2004; Soukharev and Hood, 2006; Randel and Wu, 2007; Austin et al., 2008; Remsberg et al., *2008*; Gray et al., 2009; Remsberg, 2014; Dhomse et al., 2016). These studies found a change associated with 11year solar cycle in the range of 2 to 5% in ozone mixing ratio, which maximizes near 40 km. Maycock et al. (2016) recently compared the ozone 11year solar cycle signal of several different satellite records and found substantial differences. One inherent issue of the observational investigation of the 11year cycle ozone response is the fact that only three complete periods of the 11year solar cycle have been covered by satellite observations so far. Furthermore, the life span of a single satellite instrument is generally far lessshorter than (comparable to in some cases, e.g. ENVISAT MIPAS, Aura MLS) one solar cycle and instrumental biases between different ozone profile data sets complicate statistical analysis of decadal variations (Fioletov, 2009; Dhomse et al., 2016). In this regard, a suitable alternative for understanding better the direct effect is to examine the ozone response on Sun's rotational timescale (i.e. about 27 days). Although the irradiance fluctuations during the rotational cycle are on average smaller than during the 11year solar cycle, there are many more rotational cycles than 11year cycles, improving considerably the statistics.

A number of observational studies has been carried out to determine the effects of the solar rotational cycle on stratospheric ozone, generally at low-latitudes (i.e. tropical region) based on the analysis of satellite observations (e.g. Hood, 1986; Eckman, 1986b; Keating et al., 1985; 1987; Hood et al., 1991; Fleming et al., 1995; Hood and Zhou, 1998; 1999; Fioletov, 2009; Dikty et al., 2010). These studies have shown that the sensitivity of tropical ozone to the solar rotational cycle maximizes at about

40 km (or ~3 hPa) and varies from 0.2 to 0.6% for a 1% change in solar UV radiation index, typically taken as the irradiance at the 205 nm wavelength. It was further shown that the phase lag of the tropical stratospheric ozone response varies with the altitude. The phase lag vertical profile between the ozone response and the solar forcing was found to be negligible at about 40 km and gradually increasing/ (decreasing,) below/ (above) that altitude. The phase lag was estimated to be approximately 4 days at 30 km and -2 days at 50 km (e.g. Hood, 1999; and references therein).

Simulations with numerical models of various complexities have been performed to understand the influence of the rotational cycle on ozone variability. One-dimensional photochemical-radiative model experiments (e.g. Hood, 1986; Eckman, 1986a; Brasseur et al., 1987) allowed identifying the importance of temperature/ozone couplings and reproducing the gross features of the observed ozone response. In particular, they found that the negative phase lag between the solar forcing and the ozone response in the upper stratosphere originated from the strong influence of the temperature feedback on ozone response through the temperature dependent chemical reactions (Brasseur et al., 1987). They however noticed that including the solar induced temperature changes alone was not sufficient to adequately reproduce the observed magnitude and phase lag of the ozone response and suggested that atmospheric dynamical variability – which is not simulated in 1-D models - may also have a sizeable influence (Hood, 1986; Brasseur et al., 1987). The latter issue has later been addressed with two-dimensional models which revealed better agreement with observations (Brasseur, 1993; Fleming et al., 1995; Chen et al., 1997). Fleming et al. (1995) further stressed the increasing importance with height of the solar-modulated HOx chemistry on the ozone response above 45 km. In the upper stratosphere and mesosphere, enhancement of HOx through photolysis of water vapour in Lyman-alpha line associated with an increasing solar irradiance contribute to destroy ozone. Above ~65 km and at zero-lag, the latter mechanism dominates over ozone production (i.e. by photolysis of oxygen) leading to a negative ozone-solar irradiance correlation. In the upper stratosphere and lower mesosphere (below 65 km), although ozone production dominates, increasing HOx at zero-lag contributes to the negative lag of the ozone response (Rozanov et al., 2006).

Using a large ensemble (nine 1year long runs) of chemistry-climate model (CCM) simulations, Rozanov et al. (2006) found that the ensemble mean ozone sensitivity to the solar rotational irradiance changes was in very good agreement with observational data. They however pointed out – despite an identical solar forcing for each experiment - a large scatter in maximum ozone sensitivities that could vary by a factor of almost 10 between the two most distant ensemble members. A large variability in ozone sensitivity was similarly found in an ensemble of three transient CCM simulations (1960-2005) (Austin et al., 2007). Bossay et al. (2015) analysed satellite observations of two periods of 3 years during the declining phases of cycles 22 and 23 (i.e. 1991-1994 and 2004-2007) and found that the solar rotational signal in stratospheric ozone time series strongly varies from one year to another. These results suggest that the background dynamical state and variability of the atmosphere contribute to masking the solar rotational signal in ozone (Gruzdev et al., 2009).

In addition to the nonlinear dynamics, the intensity of the solar forcing naturally modulates the solar rotational signal in ozone. When the solar rotational fluctuations are well marked with large amplitudes, notably around the maxima of 11year cycles (e.g. Rottman et al., 2004), ozone response and correlation are expected to be the largest. This has been supported by observational (e.g. Hood, 1986; Zhou et al., 2000; Fioletov, 2009; Ditky et al., 2010) as well as modeling (Kubin et al., 2011) studies which demonstrated a better identification of the ozone signal associated with enhanced rotational forcing fluctuations. This relationship has however been challenged by contradictory results. Hood and Zhou (1998) analysed ~~MLS/~~UARS MLS ozone data for the 1991-1994 period and found a correlation two times stronger during the last half of the period, i.e. when the rotational forcing fluctuations are reduced. They suggested that it might have been the result of an artefact of either instrumental or geometric (local time coverage) origin that may have affected the earliest part of the ~~MLS/~~UARS MLS ozone record more than the later part. In their recent observational study which compared the declining phases of cycle 22 and cycle 23, Bossay et al. (2015) further showed that even though the amplitude of solar rotational fluctuations of the 205 nm flux was by far the largest during the first year of both periods, the correlation with tropical ozone was found to be maximum the subsequent years.

The ozone sensitivity response to the solar rotational forcing has also been suggested to vary with the intensity of the forcing. We recall that the "sensitivity" is a quantity expressed as % changes in ozone (or any other variable of interest) per % change of the forcing (here specifically solar). Hence, the sensitivity is normalized by the amplitude of the forcing and may not be expected to change strongly with the amplitude of the forcing, or at least not as much as the absolute amplitude of the ozone response which directly depends on the amplitude of the forcing. Grudzev et al. (2009) used an idealized solar rotational forcing in their model (prescribed as a sinusoidal 27day oscillation) and found a significant reduction of the ozone sensitivity when applying an enhanced solar forcing amplitude (3 times the standard amplitude). Reciprocally, in the CCM experiments of Kubin et al. (2011), the ozone sensitivity seemed to be enhanced during periods of weak 27 day cycles. Finally, the observational study of Bossay et al. (2015) also hints at an opposite relationship between the solar rotational irradiance fluctuations and the ozone sensitivity. Given the strong influence of the dynamical background state on the variability of estimated ozone sensitivity and the rather shortness of the considered time windows of analysis, they recognized that it was not possible to conclude to a systematic effect. All these results thus highlight the uncertainty regarding the influence of the forcing intensity on ozone sensitivity and on the length of the time window required for an accurate and robust estimation of the ozone rotational signal.

In the present study, we examine the sensitivity of the tropical stratospheric ozone response to the rotational cycle by comparing satellite observations and chemistry climate model experiments to understand better the origin of the discrepancies - and sometime contradictory results - in the estimation of the ozone response to the solar rotational cycle found in previous studies. As a first step, we follow up on the case study of Bossay et al. [2015] and make use of observations and modelling results comparison to provide a detailed picture of the ozone response to the solar rotational cycle during the declining phases of cycle 22 and cycle 23. We particularly aim to better understand the strong differences in the ozone response to solar rotational cycle found between the two periods. Two configurations of the LMDz-Reprobus chemistry climate model simulations are used, with specified dynamics (i.e. Chemistry Transport Model, or CTM) and in its free running mode (CCM). In the CTM configuration, temperature and wind fields calculated by the model are relaxed towards meteorological analysis; the dynamics is expected to be rather close to the reality, allowing direct comparisons with satellite observations for evaluating model chemical processes and its relevance to our study. In the CCM configuration, an ensemble of simulation is performed.

Comparing the CCM ensemble results to CTM and observations during the declining phases of cycle 22 and cycle 23 allows to understand better the effect of internal dynamical variability on the ozone response. As a second step, we take advantage of the ensemble of CCM simulations and its large statistics In particular, we aim to (i) assess the influence of the solar cycle phase on the ozone sensitivity to the rotational cycle and (ii) quantify the time window required for a robust estimation of the ozone sensitivity. Two configurations of the LMDz-Reprobus chemistry climate model simulations are used, with specified dynamics (i.e. Chemistry Transport Model, or CTM) and in its free running mode (CCM). In the CTM configuration, temperature and wind fields calculated by the model are relaxed towards meteorological analysis; the dynamics is expected to be rather close to the reality, allowing direct comparisons with satellite observations for evaluating model chemical processes and its relevance to our study. Then, an ensemble of CCM simulations is performed to examine the ozone sensitivity as a function of the solar cycle phase and the size of the analysis time window. Performing an ensemble of simulations allows us to assess better the statistical robustness of the results.

Observational datasets, and model configurations and simulations are described in section 2. Section 3 presents comparisons between satellite observations and model (CTM and CCM) simulations of the ozone response to the solar rotational cycle. Section 4 focuses on CCM results to examine the influence of (i) the solar activity fluctuations and (ii) the length of the time window in the estimation of the ozone sensitivity to the solar rotational cycle. The main findings are summarized in section 5. Note that for the sake of simplicity, the first period (10/1991-09/1994) during cycle 22 will be referred thereafter as 1991-94 period and the second period (09/2004-08/2007) during cycle 23 will be referred as 2004-07 period.

2 Data and model description

2.1 The 205 nm solar flux (or F205)

The solar proxy used in regressions analyses is the UV solar irradiance at 205 nm. This wavelength is chosen because it is important for the ozone chemical budget throughout the stratosphere. The 205 nm wavelength is included in the Herzberg continuum region (200-242 nm) that is positioned between two strong absorption bands: the Schumann–Runge band of molecular oxygen and the Hartley band of ozone (Brasseur and Solomon, 2005). In the Herzberg continuum, atmospheric absorption is relatively low and hence solar UV radiation penetrates deeply in the atmosphere, down to the lower stratosphere, where it photolysis molecular oxygen (O_2) to produce O_3. The 205 nm flux, called thereafter F205, has been commonly used in previous studies because it is a very good proxy for characterizing solar variability in the UV domain.

In our study, we use the solar spectral irradiance provided by the Naval Research Laboratory Solar Spectral Irradiance (NRLSSI) model version 1 (Lean, 2000; Wang et al., 2005). NRLSSI is an empirical model which aims to reconstruct long-term SSI over the wavelength domain 120-100,000 nm. It uses historical estimates of faculae brightening and sunspot darkening to extend in time wavelength-dependent parameterizations of SSI derived from satellite measurements and model. Shortwards of 400 nm, the SSI is derived from UARS/SOLSTICE observations (Rottman et al., 2001) through a multiple regression analysis with respect to a SOLSTICE reference spectrum. The regression analysis includes a facular brightening and a sunspot darkening time-dependent term. Above 400 nm the SSI is reconstructed by adding the irradiance changes caused by the presence and the characteristics of faculae and sunspots (see Lean (2000) for details) to a quiet Sun intensity spectrum, i.e., defined by the absence of faculae and sunspots. The intensity spectrum of the quiet Sun is a composite compiled from space-based observations made by UARS/SOLSTICE (120-401 nm) and SOLSPEC/ATLAS-1 (401-874 nm) (Thuillier et al., 1998), and a theoretical spectrum at longer wavelengths (Kurucz, 1991).

2.2 Microwave Limb Sounder ozone satellite observations

We use the stratospheric ozone measurements from the two Microwave Limb Sounder (MLS) instruments on-board UARS (cycle 22) and Aura (cycle 23).

MLS UARS MLS was launched on 12 September 1991, into a 57° inclination and a 585 km altitude orbit and was operational until 1994. Waters (1989; 1993) describe in detail the microwave limb-sounding technique. We used the version 5 UARS MLS dataset described Livesey et al., (2003). The ozone retrieval is based on 205 GHz radiances, provided onto 13 pressure levels in the range 100-1 hPa (100, 68.1, 46.4, 31.6, 21.5, 14.7, 10, 6.8, 4.6, 3.2, 2.2, 1.5 and 1 hPa) and has an average vertical resolution of 4 km in the stratosphere.. The typical 1σ precision for ozone mixing ratio measurements is ~0.35 ppmv at 0.46 hPa, 0.3 ppmv at 1-4.6 hPa and 0.2 ppmv at 10-46 hPa between 68 and 1 hPa. As shown in Hood and Zhou (1998), an artificial 36day periodicity, caused by the UARS yaw manoeuvre cycle (Froidevaux et al., 1994), is seen in zonally averaged MLS UARS MLS data at all latitudes and increasing with height. To remove this artefact, Hood and Zhou (1998) suggested restricting zonal averaging ozone profiles to daytime measurement near a single local time. They however recognized that the ratio of daytime measurements per day would be too low (around 30%), resulting in very large sampling errors and time gaps in the zonal averages. Furthermore, ozone diurnal cycle becomes important in the upper stratosphere so that the results may be affected by the imbalance in daytime and night-time measurements used to construct daily time series. This issue will be discussed in section 3.2.

MLS Aura MLS was launched on 15 July 2004 into a sun-synchronous near-polar orbit around 705 km. Detailed information on the MLS Aura MLS instrument is given in Waters et al. (2006). In brief, MLS Aura MLS observes a large suite of atmospheric parameters by measuring millimeter and submillimeter-wavelength thermal emission from Earth's limb with seven radiometers covering five broad spectral regions (118, 190, 240, 640 GHz and 2.5 THz). The "standard product" of ozone is retrieved from radiance measurement near the 240 GHz. Here, we used version 4.2 of the MLS Aura MLS ozone product (Livesey et al., 2017). The MLS Aura MLS fields of view point forward in the direction of orbital motion and vertically scan the limb in the orbit plane, resulting in a data coverage from 82°N to 82°S latitude on every orbit. MLS Aura MLS provides continuous daily sampling of both polar regions without temporal gaps from yaw maneuvers that occurred with MLS UARS MLS. The MLS Aura MLS limb scans are synchronized to the Aura orbit, with 240 scans per orbit at essentially fixed latitudes. This results in about 3500 scans per day, with an along-track separation between adjacent retrieved profiles of 1.5° great circle angle. Ozone profiles are provided onto 25 pressure levels in the range 100-1 hPa (100, 82.5, 68.1, 56.2, 46.4, 38.3, 31.6, 26.1, 21.5, 17.8, 14.7, 12.1, 10, 8.2, 6.8, 5.6, 4.6, 3.8, 3.2, 2.6, 2.1, 1.8, 1.5, 1.2 and 1 hPa) with an average vertical resolution of 3 km in the stratosphere.~~The vertical resolution is about 3 km in the upper troposphere and stratosphere and about 4-6 km in the mesosphere.~~ The 1σ precision for ozone mixing ratio measurements is about 0.1 to 0.3 from 46 hPa to 0.5 hPa.

For our study, daily stratospheric ozone profiles averaged over the tropical band [20°S,20°N] are used. Among the 1095 days of each period, 121 and 38 days of ozone data are missing for the period 1991-94 and 2004-07, respectively. For each height level of the vertical profile, the outliers of the corresponding ozone time series are removed by excluding data which take absolute values beyond 2 standard deviations of the deaseasonnalized time series. After removing outlier values, 85% and ~~(~~93%~~)~~ of the 1095day ozone time series of the periods 1991-94 and ~~(~~2004-07, respectively,~~)~~ are kept for the analysis.

2.3 The LMDz-Reprobus model

The LMDz-Reprobus model is a Chemistry-Climate Model resulting from the coupling between the extended version of the General Circulation Model LMDZ5 (Sadourny and Laval, 1984; Le Treut et al., 1994; 1998; Lott et al., 2005; Hourdin et al., 2006; 2013) and the chemistry module of the Reprobus stratospheric chemistry-transport model (Lefèvre et al., 1994; Lefèvre et al., 1998). LMDZ was developed at the Laboratoire de Météorologie Dynamique (LMD). The dynamical part of the code is based on a finite-difference formulation of the primitive equations of meteorology (Sadourny and Laval, 1984). The model uses a classical hybrid σ-P coordinate in the vertical, has 39 vertical levels and a lid-height at ~70 km. The model vertical resolution slowly decreases with height. In the middle and upper stratosphere (30-50 km or ~10-1 hPa) - focus of our study – the model vertical resolution reaches 3 km which is similar to the vertical resolution of UARS MLS and Aura MLS measurements in this altitude range. The model is integrated with a horizontal resolution of 3.75° in longitude and 1.9° in latitude. The equations are discretized on a staggered and stretched latitude-longitude Arakawa-C grid.

The Reprobus chemistry model (Jourdain et al., 2008; Marchand et al., 2012) calculates the chemical evolution of 55 atmospheric species and includes a comprehensive description of the stratospheric chemistry (Ox, NOx, HOx, ClOx, BrOx and CHOx). It uses 160 gas-phase reactions and 6 heterogeneous reactions on sulfuric acid aerosols and PSCs. Absorption cross-sections and kinetics data are based on the 2011 Jet Propulsion Laboratory (JPL) evaluation (Sander et al., 2011). In the troposphere, where the chemistry is not explicitly treated, the model is relaxed towards a monthly varying climatology (annual cycle) of O_3, CO and NOx computed by the TOMCAT chemical-transport model (Law et al., 1998; Savage et al., 2004).

The solar component of the radiative scheme of LMDZ5 is based on an improved version of the two bands scheme developed by Fouquart and Bonnel (1980) and the thermal infrared part of the radiative code is taken from Morcrette et al. (1986). While this scheme is crude, note that the thermal component of the solar forcing (e.g. changes in net heating from solar changes only, keeping chemical composition unchanged) does not exhibit a dependency on wavelength as strong as photolysis component of the solar forcing. Nonetheless, the use of a simple two bands radiation code tends to underestimate the temperature response when compare to other radiations models with the same solar irradiance fluctuations (CCMVal, 2010; Forster et al., 2011). The radiative scheme takes into account the radiative active species H_2O, CO_2, O_3, O_2, N_2O, CH_4, CFC-11 and CFC-12.

The photolysis rates used in Reprobus are pre-calculated off-line with the Tropospheric and Ultraviolet Visible (TUV) model (Madronich and Flocke, 1999; Sukhodolov et al., 2016) and then tabulated in a look-up table for 101 altitudes, 7 total ozone columns and 27 solar zenith angles. TUV calculates in spherical geometry the actinic flux, scattering and absorption through the atmosphere by the multi-stream discrete ordinate method of (Stamnes et al., 1988). The spectral domain extends from 116 to 850 nm. Calculations of photolysis rate are performed on a 1 nm wavelength grid, except in the regions relevant for solar cycles (rotational and 11-year solar cycles). In these spectral regions, the resolution is largely increased to accurately describe the spectral features in the solar flux or in the absorption cross-sections: the wavelength resolution increases up to 0.01 nm in the Schumann-Runge bands of O_2. At this resolution, the absorption by O_2 can be considered to be treated line-by-line. Moreover, the temperature dependent polynomial coefficient determined by Minschwaner et al. (1992) is used. The temperature dependence of absorption cross-sections is taken into account using climatological temperature profilescalculated off-line in TUV using the US standard atmosphere. The albedo considered for the computation of photolysis rates is set to a globally average value of 0.3 with solar zenith angle varying from 0 to 95°. For each sunlit grid point, the actual photolysis rates used by LMDz-Reprobus are then interpolated in the table according to those parameters (solar zenith angle, ozone column, altitude). Thus, tThe solar rotational cycle forcing is taken into account by using daily photolysis rates calculated by TUV in the photochemistry module of LMDz-Reprobus. A separate photolysis look-up table is calculated every day using the daily NRLSSI as solar input. Note however that the direct effect on heating rates generated by UV variations associated with the 27day rotational cycle is neglected: i.e. daily changes in the spectral irradiance are not considered in the CCM radiative scheme. As a consequence, part of the thermal and dynamical responses to the 27day rotational cycle and hence their effect on ozone (through transport and temperature dependent chemical reactions, as described above) are missing. The impact of this approximation on our results seems to be small though, as discussed thereafter (sections 3 and 5). Note also that on timescales of the 11yr cycle, Swartz et al. (2012) found that their photolysis-only simulation captured almost all of the solar cycle effect on ozone.

We do not take into account the direct effect on heating rates generated by UV variations because previous modelling studies have already shown that the stratospheric ozone response to solar variations is almost entirely driven by the effects of UV changes on the photolysis rates, in particular the photolysis of molecular oxygen (Swartz et al., 2012).

LMDz-Reprobus is used in two configurations. The first one is the free-running model configuration (i.e. CCM) that accounts for all the interactions between chemistry, dynamics and radiation. LMDz-Reprobus is additionally used in its nudged version (i.e. CTM) where transport and dynamics are nudged towards temperatures and winds from the 6 hourly ECMWF model outputs (ERA-interim (Dee et al., 2011)). As the dynamics is specified and is close to observations, the CTM configuration allows a fair comparison with MLS observations. The CTM configuration is used over the two 3year periods of MLS ozone measurements, as analysed in Bossay et al. (2015). In the CCM configuration, we perform an ensemble of five simulations of 17 years each (from 1991 to 2007). As for the observations, we use the daily stratospheric ozone profiles averaged over the tropical band [20°S,20°N].

3 Ozone response to the solar rotational cycle during the declining phase of solar cycles 22 and 23

In this section, we analyse the ozone response to the solar rotational cycle over the declining phase of solar cycles 22 and 23 in the observations and in the ~~different~~ CTM and CCM model ~~configurations~~simulations. ~~Note that the present modelling study~~ The analysis presented here ~~partly~~ follows ~~on~~ up on ~~from~~ Bossay et al. (2015) observational study. In particular, we aim to assess the model performances, understand better the differences in the results between the two solar declining phase periods and highlight the importance of internal dynamical variability. ~~As such, few of their results are shown next to our model results for comparison.~~

3.1 The rotational cycle in UV irradiance

Figure 1 shows the solar UV variability represented by F205 from 1985 to 2008 with the two periods of interest highlighted in red which correspond to the declining phase of solar cycles 22 and 23. F205 is a good indicator of the NRLSSI solar forcing prescribed in CTM and CCM simulations. Thereafter, F205 is used as the UV index in the regression analysis of the solar signal in stratospheric ozone from MLS observations and model simulations.

The Fast Fourier Transform (FFT) power spectra of the two F205 declining periods time series are shown on Fig. 2 (top panel). For both periods, the high frequency spectrum is dominated by a strong peak centred around 27 days corresponding to the main solar rotational periodicity. The broadness of the peaks indicates that the solar rotational cycle is not regular and cover a rather wide frequency domain. A small secondary peak is also found at ~13.5 days which corresponds to the first harmonic of the rotational cycle and to the presence on the Sun surface of two sunspots which rotate with the same period but are separated by about 180° in longitude (e.g. Bai, 2003; Zhang et al., 2007). The time-resolved power spectral density derived from the continuous wavelet transforms (CWT, (Torrence and Compo, 1998)) of the two F205 time series are shown on Fig. 2 (bottom panels). CWT spectral analysis reveals that the solar rotational component strongly varies in time for both declining periods. Overall, the rotational component decreases over the declining solar activity periods and even can sporadically disappear for several months (e.g. late boreal summer 1993, spring 2006 and winter/spring 2006/2007). In addition, the solar rotational fluctuations are stronger during the first period than the second period (see Fig. 1 and 2). As the solar rotational forcing is stronger during the first period, one might expect the solar signal in ozone to be clearer.

3.2 Observed and modelled ozone response to the rotational cycle

We first examine potential rotational periodicities in upper stratospheric tropical ozone by carrying out a spectral analysis of daily stratospheric ozone time series averaged over the tropical band [20°S-20°N]. Figure 3 shows the normalized Lomb-Scargle periodograms (well adapted for non-continuous series, Lomb (1976); Scargle (1982)) of tropical stratospheric ozone from observations (Figs. 3a,d), CTM (Figs. 3b,e) and CCM results (Figs. 3c,f), calculated for the declining period of cycle 22 (Figs. 3a,b,c) and cycle 23 (Figs. 3d,e,f). Periodograms are shown for the 3.2 hPa (~40 km) pressure level, close to the altitude where the ozone solar signal maximizes (Hood, 1986).

The two periodograms of MLS ozone measurements (Fig. 3a and Fig. 3d) reveal no prominent peak in the range of the 20-30 days period, ~~indicating that~~suggesting an absence of ~~the~~a solar rotational signal in ozone~~ is weak~~. More prominent peaks are found at longer periods although they are not consistent between the two periods. The large peak found at the 35day period for 1991-94 corresponds to the yaw-maneuver period of the MLS instrument as described previously (Froidevaux et al., 1994; Hood and Zhou, 1998). Similarly to observations, the periodograms of CTM results (Fig. 3b and Fig. 3e) does also not exhibit a distinctive solar rotational peak; there are some minor peaks between 20 and 30 days and their amplitudes are smaller in 2004-07 than in 1991-94. The analysis has been repeated at lower pressure-height levels (e.g. 10 hPa, not shown) and led to the same conclusions. Overall, the raw power spectrum analysis of observations and CTM results in the middle and upper tropical stratosphere does not allow identifying an ozone signal associated with the solar forcing fluctuations at rotational timescales for the two periods considered here.

In contrast, the periodogram averaged over the five CCM simulations exhibits a distinctive peak centred at 27 days for 1991-94 (Fig. 3c). This peak is less pronounced and centred at 25 days for 2004-07 (Fig. 3f), presumably because of the smaller amplitude of solar rotational fluctuations and hence model forcing in 2004-07 (see Fig. 2). However, the 2σ standard deviation (i.e. spread of the ensemble simulations) associated with these peaks is very large, indicating the presence of a strong high frequency (periods < 50 days) natural variability in ozone in this region. This ~~explains~~ illustrates~~why~~ the difficulty in detecting solar rotational signals in the~~ can be hardly detectable in the~~ observations, as well as in a single ensemble member over these 3year periods. Note that we additionally computed periodograms in observations during solar maximum phases (i.e. 2012-2015) where 27day fluctuations in the solar forcing are stronger than during the declining phase (not shown). The results were however similar and no clear peak at 27 days could be identified. Hence, ~~T~~the absence of a distinctive rotational signal ~~in MLS data and CTM 3year time series~~ suggests the presence of strong and rather random ozone variability of non-solar origin which makes the ozone rotational signal very difficult to detect and estimate.

We further examine the relationship between stratospheric ozone and solar rotational cycle by performing cross-spectrum analysis between stratospheric ozone and F205. Despite ~~the weak magnitude~~the absence of a~~the ozone~~ solar rotational peak~~signal~~ in the ozone power spectrum derived from observations and CTM results, cross-spectrum analysis should help identifying coherent variability modes between the solar forcing and tropical ozone. Figure 4 presents the vertical profile of the magnitude-squared coherence (hereinafter referred as coherence) between F205 and tropical stratospheric ozone from MLS observations (a and d), CTM model results (b and e) and CCM model results (c and f).

A strong and statistically significant coherence is found for ~~MLS~~ UARS MLS (1991-94) between 20 and 28 days and between about 10 and 1 hPa with a maximum of about 0.7 at the 22day period around 6 hPa. In contrast, the coherence for ~~MLS~~ Aura MLS (2004-07) is generally weaker with only a small patch of significant coherence at the 90% confidence level. The coherence fields from the CTM results resemble those of the observations and reproduce the main features during the two periods. The main difference between observed and CTM signals is that the coherence patch extends farther to lower levels in the CTM (down to 15 hPa) and covers longer periods (20 to 33 days at ~10 hPa). For the 1991-94 period, the CTM results also overestimate the coherence around 13.5 days compared to observations.

The general features in the coherence fields from CCM results are also consistent with those of the observations. However, the area of statistical significant coherence around the 27day period is wider in the CCM results. In addition, the coherence patch does not extend as low as the CTM results. The differences observed between the MLS coherence fields of the two periods are also reasonably well reproduced in the CCM coherence results. As for the CTM fields in 1991-94, CCM results reveal a secondary area of significant signal centred at about 13.5day period and extends almost throughout the stratosphere. For 2004-07, there is no significant signal around 13-14 days in all the coherence fields. This is consistent with the UV forcing (Fig. 2) exhibiting a stronger 13.5day period component in 1991-94.

To further test the robustness of the coherence signal, we perform an additional CCM simulation for the period 1991-1997 where the solar forcing is kept constant by using fixed (i.e. climatological) photolysis rates during the model simulation. Results are shown on Fig. 5. Below 15 hPa, the different experiments show no significant coherence between ozone and solar flux. Between 15 and 1 hPa, all forced experiments (black lines) reveal a similar and significant coherence signal while for the constant solar forcing experiment (red line), the coherence is weak and within the range of randomness. The ~~total~~ absence of significant coherence found in the constant solar experiment ~~is simulation~~ ~~(not shown)~~ confirms that the coherence found between F205 and stratospheric ozone is not fortuitous and primarily originates from photolysis processes. We can also note that the reduced coherence for 2004-07 may be expected because the solar rotational fluctuations are smaller during that period compared to 1991-94 (Fig. 2). To summarize these first steps in our analysis, we find that, despite the weak magnitude of the signal, the upper stratosphere tropical ozone concentration fluctuates coherently with UV variability at solar rotational timescales.

To focus on periodicities relevant to the solar rotational cycle (13.5 and 27 days), all the time series are now filtered using the digital filter that has been commonly used in previous solar rotational studies (e.g; Hood, 1986; Chandra, 1986; Keating et al., 1987; Hood and Zhou, 1998 and Zhou et al., 2000). The filtering procedure consists of smoothing data with a 7day running mean which removes short-term fluctuations. Linear trend and mean value are also removed from these smoothed time series. Finally, a 35day running mean is subtracted from the data, removing long-term fluctuations (e.g. seasonal, semi-annual, annual and QBO variations). The overall procedure is more or less equivalent to a 7-35 days band-pass filter in the frequency domain. The vertical extent and temporal evolution of the tropical ozone response to the solar rotational cycle are examined by calculating the cross-correlations between filtered F205 and ozone in observations and model results. Results are shown in Fig. 6~~5~~. For 1991-94, the observations exhibit a cross-correlation peak at 0.28 on the 4.6 hPa level with no time lag (Fig. ~~5~~6a). This maximum value is close to the maximum of 0.35 found by Hood and Zhou (1998) on the same pressure level. Furthermore, the overall variation of the time lag with altitude shown in Fig. 6~~5~~ is similar to that found in previous studies (Hood, 1986; Brasseur et al., 1987; Brasseur, 1993; Hood and Zhou, 1998) with a negative lag above 3-4 hPa (ozone "leading" the solar flux) and a positive lag below (ozone lagging the solar flux). As mentioned in the introduction, the negative lag in the upper stratosphere result of the influence of the temperature feedback on the ozone response through the temperature dependent chemical reactions. For 2004-07, the cross-correlation pattern (Fig. 6~~5~~d) is more distorted and weaker than for 1991-94 (Fig.

65a). The cross-correlation maximum (0.2) is smaller than for 1991-94 and is found at 10 hPa with a time lag of +5 days (ozone lagging solar flux).

Although the cross-correlation fields for the CTM and CCM simulations appear smoother and with larger statistically significant (shaded) areas than for the MLS data, most of the general features present in the MLS cross-section fields appear consistently reproduced by the simulations in the two model configurations. Seizing Marked differences between the CTM and the observations are found in 1991-94 though. The high correlation area (with a maximum of 0.4 at 7 hPa and a positive time lag of 3 days) expanding throughout the middle stratosphere (between 30 and 10 hPa) in the CTM (Fig. 65b) is not detected found in observations (Fig. 65a). Overall, the main area of significant correlation appears also lifted upward in the observations (Fig. 65a) compared to the CTM (Fig. 65b). The absence of The fact that the correlation signal in the middle and lower stratosphere (below 10 hPa) is found in the CTM inbut not in the observations is consistent may partly arise withfrom the large noise present in the UARS MLS ozone dataset at these altitudes (not shown). In contrast, the results for the period 2004-07 reveal a particular good agreement throughout stratosphere between the observations (Fig. 65d) and the CTM (Fig. 65e), where the maximum is found at the same altitude (10 hPa), time lag (+4 days) and with the same amplitude (0.2). CCM results show a maximum of correlation also at 10 hPa and at the same time lag but with a higher value (0.3). In addition to the area of statistical significance which increases when examining CCM results, we notice a strong reduction of the difference in the response between both periods. This suggests that averaging over the five ensemble members allows to reduce the effect of the non-solar random variability in the signal estimation and hence to identify more robustly the solar signal. Nevertheless, for 2004-07, we note a weaker correlation and a reduced downward propagation of its extension which is likely due to a weaker rotational UV forcing compared to 1991-94 (Figs. 1 and 2).

Above 3 hPa (~40 km), CCM cross-correlations of both periods (Fig. 6c,f) show a maximum at negative time lag (-2 days). As mentioned in the introductory section, this negative time lag can be induced by temperature feedback on ozone and by increasing HOx with solar irradiance which contributes to destroy ozone. While our model configuration allows to fully account for the HOx effect, the solar-induced temperature response is limited since the direct radiative heating effect is not included. The temperature response to the 27-days cycle is thus solely controlled by ozone production in the photolysis scheme. Although a temperature signal is found (not shown), it is small, hence reducing the likelihood for the solar-induced temperature feedback to be prominent in our experiments. Despite the approximation made in our model configuration, we notice however that the upper stratosphere negative lags compare very well with those found in CCM experiments of Sukhodolov et al. (2017) (see their Fig. 3) in which both HOx and solar-induced temperature feedback effects are fully included. Hence, this suggests that neglecting the direct effect on heating rates generated by UV variations has a limited effect on the ozone response, at least at 27-days timescale. More sensitivity experiments are required however to quantify accurately the impact of this approximation.

In addition to correlation analysis, ozone response to solar UV flux changes can also be measured in terms of sensitivity, i.e. percentage change in ozone per 1% change in solar UV. Considering ozone sensitivity instead of ozone absolute change allows in principle to analyse an ozone signal that does not depend on the magnitude of the solar rotational forcing, assuming implicitly that the relationship between the solar forcing index (F205) and the ozone response is linear. We derive the ozone sensitivity on different pressure levels by linear regression of the filtered ozone time series on one independent variable, F205. In previous studies, ozone sensitivity profiles were either calculated at optimum lags where the correlation coefficient maximizes (e.g. Hood and Zhou, 1998) or at zero lag (e.g. William et al., 2001; Austin et al., 2007). Both alternatives were tried but given the limited effect on the results and conclusions, we elected to show only ozone sensitivity profiles using a common time frame, hence at zero lag. Results are shown on Fig. 7~~6~~.

For the 1991-94 period, the observational (~~MLS~~ UARS MLS) sensitivity peaks at 0.4 (0.4% of ozone change for 1% change in F205) near 4-5 hPa (35 km), consistent with the results of Hood and Zhou (1998) (Fig. 7~~6~~a). For the 2004-07 period, the shape of the observational (~~MLS~~ Aura MLS) sensitivity profile is distorted and the sensitivity peaks at only 0.2 around 5 hPa (Fig. ~~6~~7d); it is consistent with a peak value of 0.15 derived at the same level shown in Dikty et al. (2010) for a similar period (2006-07) but with a different instrument (ENVISAT SCIAMACHY). In the middle stratosphere, the sensitivity profile calculated from the CTM results for the period 1991-94 (Fig. 7~~6~~b) is consistent with the MLS sensitivity profile (Fig. 7~~6~~a); the CTM sensitivity profile peaks at 4-5 hPa with a value slightly lower (0.3) than that derived from the MLS observations. Discrepancies between CTM and observational sensitivities are more pronounced in the upper stratosphere. In the CTM, above the peak, the sensitivity suddenly drops around 3 hPa to values close to 0 (Fig. 7~~6~~b), while in the observation the sensitivity gradually decreases from 3-4 hPa to the stratopause region (around 1hPa) (Fig. 7~~6~~a). Below 10 hPa, we also note that the sensitivity profile errors are larger in the observations than in the CTM. This is consistent with the absence of solar-ozone correlation signal at these altitudes in the observations (Fig.6~~5~~a) and, inversely, the clear solar-ozone correlation signal in the CTM (Fig. 6~~5~~b). For 2004-07, the CTM sensitivity profile appears to be highly consistent with observations throughout the stratosphere, in accordance with the previous coherence and correlation analyses (Figs. 4 and 6~~5~~).

We now analyse the CCM ensemble results. The ensemble mean ozone sensitivity profiles (Figs. 7c and f) markedly differ with ozone sensitivity profiles derived from observations (Figs. 7a and d) and CTM (Figs. 7b and e) at the corresponding periods. These differences are particularly pronounced in the upper stratosphere (above ~5 hPa). On the other hand, despite the two different periods, the ensemble mean ozone sensitivity profiles ~~for the two periods are~~ show very similar features with positive sensitivity from 15 hPa to the stratopause and a maximum sensitivity of 0.4 at ~3 hPa (Figs. 7~~6~~c and f). This maximum tropical sensitivity value and its altitude level is in good agreement with previous CCM estimates (e.g. Rozanov et al., 2006; Austin et al., 2007; Gruzdev et al., 2009; Kubin et al., 2011). The CCM ozone sensitivity analysis has also been repeated for the period 2003-2005 (not shown) to be directly comparable with the CCM results of Sukhodolov et al. (2017): we found very similar ozone sensitivity profiles. The ensemble spreads (i.e. 2σ standard deviation calculated over the five CCM simulations for each 3year period, dashed line) are of the same order for both periods (Figs. 7~~6~~c and f). They are also very large, indicating important variations from one ensemble member to another, which are most likely due to differences in dynamical variability. Similar conclusions have been reached in previous CCM studies (e.g. Rozanov et al., 2006; Austin et al., 2007). This may partly explain the strong differences in ozone sensitivity found between the two periods in the observations and the CTM simulation. In a sense, each 3year observed period can be viewed as a single realization of an ensemble.

As mentioned in Section 2.2, the results based on UARS MLS measurements may be affected by the imbalance between night and daytime sampling due to the ozone diurnal cycle becoming significant in the upper stratosphere. To test the influence of the ozone diurnal cycle, we repeated all the analysis performed in this section by mimicking an irregular sampling over the period covered by Aura MLS (i.e. 2004-2007). Each day, ~700 ozone vertical profiles of the Aura MLS instrument are evenly retrieved in the tropics [20S-20N] at two fixed local times: one at night (~0142 LST) and one during daytime (~1342 LST). We initially build the ozone time series using daytime measurements only (1095 days in total). Among these 1095 days, we selected N days randomly where daytime measurements were replaced by night time measurements. We then repeated the spectral, correlation and regression analysis. The procedure was performed for various values of N, from N=100 (i.e. 91% of daytime measurements) to N=1000 (i.e. 9% of daytime measurements). The results (not shown) revealed almost no dependence to N, suggesting that the diurnal cycle has a small effect on the ozone solar rotational signal.

Overall, our results demonstrate that the LMDz-REPROBUS model produces an ozone response to the solar rotational cycle that is consistent with observations, especially when the dynamical variability is accounted for in the analysis. The results of our ensemble of transient CCM simulations further support the importance of atmospheric internal variability in modulating or masking the solar signal in ozone at solar rotational time scales. Moreover, the fine correspondence of our results with those based on independent previous chemistry-climate modelling experiments (e.g. Rozanov et al., 2006; Sukhodolov et al., 2017) emphasizes the relevance of our experimental model setup (i.e. despite neglecting the direct effect on heating rates) to examine the ozone response to 27day solar variations. In the following, we exploit the ensemble simulation to examine thoroughly the temporal variability of the ozone sensitivity to the rotational cycle.

4 Temporal variability of the ozone response sensitivity

4.1 Does ozone sensitivity to the rotational cycle depend on the amplitude of the solar fluctuations?

Results from CCM studies of Gruzdev et al. (2009) and Kubin et al. (2011) suggested that ozone sensitivity seems to decrease with increasing amplitude of the rotational cycle. The amplitude of the rotational cycle depends on the inhomogeneous brightness structure of the solar disc (i.e. distribution of sunspots and faculae). Given that the amount of sunspots and faculae increases with increasing solar activity, inhomogeneity in the brightness is likely to increase during solar maximum phases. ~~Given that the amplitude of the rotational cycle increases with increasing solar activity, o~~One may thus expect minimum and ~~(~~maximum~~)~~ sensitivity during 11year solar maximum and ~~(~~minimum~~)~~ phases, respectively. Next, we test this hypothesis by dividing 15 years (1991-2005) of the CCM simulations into five 3year windows corresponding to the four different phases of the 11year solar cycle (*i.e.* maximum, minimum, descending, ascending phases). These time windows are highlighted with different colours in the insert panel of Fig. 8~~7~~a. Figures 8~~7~~b-f show, for each 3year time window, the ensemble mean sensitivity profiles and the associated 2σ ensemble spread. The ensemble mean for a specific 3year window is calculated by first computing the ozone sensitivity over this specific 3year interval for each of the five ensemble members and then averaging theses five sensitivities; we define the ensemble spread as the ensemble 2σ standard deviation. Note that, in total, 15 years of model data are taken into account for the calculation of the ensemble mean sensitivity.

Whatever the solar cycle phase considered (Fig. 8~~7~~a), all the mean sensitivity profiles have similar shapes with a maximum at around 3 hPa, consistent with observed and modelled sensitivity profiles during solar declining phase (Fig. 7~~6~~). The most pronounced difference is the maximum sensitivity which varies between 0.3 (green) and 0.5 (red). Overall, the ensemble mean sensitivity profiles appear to vary little from a 3year window to another. Thus, the model ensemble mean ozone sensitivity seems to be rather independent of the level of solar activity (Fig. ~~7~~8a), at least when 15 years of model data are considered in total. In comparison, the model ensemble spread is clearly more sensitive to the 11year solar cycle phase than the ensemble mean. The ensemble spread is found to be generally smaller during high solar activity periods. It is not surprising. The estimation of the ozone sensitivity is expected to be less affected by the noise and more robust when the solar rotational fluctuations are stronger: the amplitude of the ozone response is much greater, improving the signal-to-noise ratio. We also notice that the ensemble spread is smaller during the maximum phase of cycle 22 (black) than that of cycle 23 (green). It is consistent with the results of Fioletov (2009) observational study that also shows a stronger rotational periodicity in the upper stratosphere tropical ozone during the maximum phase of the solar cycle 22 than the maximum phase of the cycle 23.

Although the rotational cycle amplitude varies with the phase of the 11year solar cycle, the relationship is not systematic as revealed by the wavelet analysis of Fig. 2. In the following, the ensemble mean ozone sensitivity and its spread are examined as a function of the amplitude of the solar rotational cycle fluctuations using sliding time windows. The analysis focuses on the 3 hPa level where the maximum sensitivity is found (Fig. 8~~7~~). Figure 9~~8~~ compares the temporal evolution (from 01/01/1991 to 31/12/2005) of the variance of the filtered F205 time series (Fig. 9~~8~~b) with the ensemble mean (Fig. 9~~8~~c) and variance (Fig. 9~~8~~d) of the ozone sensitivity derived from the five CCM simulations. Each point of the time series is obtained by first calculating the ozone sensitivity for each ensemble member over a 1year time window and then computing the ensemble mean and its variance over the five simulations. The time window is then shifted by 1 month and the same procedure is repeated. This gives a total of 168 1year time slices (14 years x 12 months).

The mean ozone sensitivity time series (Fig. 9~~8~~c) on 1year time window strongly fluctuates from 0 to 0.6 around an average value of ~0.4, consistent with the value of the ensemble mean sensitivity profiles at 3 hPa (Fig. 8~~7~~). These fluctuations increase during the minimum phase of the solar cycle in 1995-1998, indicating a larger uncertainty in the estimation of ozone sensitivity during low solar activity periods. This is further supported by the ~~overall~~ apparent inverse ~~anti-correlation~~ relationship which is found between the F205 index variance (Fig. 9~~8~~b) and the ozone sensitivity variance (Fig. 9~~8~~d). Hence, the accuracy of the ozone sensitivity estimate to solar rotational cycle is degraded when solar rotational fluctuations are small, and reciprocally.

Finally, note that the low-frequency (i.e. decadal scales) variability of the ensemble mean ozone sensitivity (Fig. 9~~8~~c) appears also to be anti-correlated with the F205 index variance (Fig. ~~8~~9b). In the following, we test further the robustness of the relationships found here which link the solar rotational variability to the ensemble mean and spread of ozone sensitivity

Figure 10~~9~~ shows the regression analysis of the ensemble mean (Fig. 10~~9~~a) and spread (Fig. 10~~9~~b) of ozone sensitivity (i.e. dependent variables) on the solar rotational variance (i.e. explanatory variable). We assess the statistical significance of the regression slope using a block bootstrapping technique to account for the autocorrelation in the residuals that can lead to an underestimation of the standard error (Mudelsee, 2014). The bootstrap procedure is carried out as follows. The original residuals are first obtained by subtracting the original fitted model (i.e. derived from the linear regression) to the dependent variable. The original residual time series is then segregated into moving blocks of length *L* (see e.g. schematic p74 in Mudelsee (2014)) that are randomly resampled to reconstruct a synthetic residual time series of the same size as the original one. Adding this synthetic residual time series to the original fitted model allows creating a new synthetic time series (so-called bootstrap sample) to which the linear regression is applied to derive a synthetic slope value. For each value of *L*, this procedure is repeated 10,000 times in order to construct a distribution of synthetic slopes (Poulain et al., 2016). Finally, we estimate, from this distribution, the likelihood (*p-value*) for the slope to be greater than - or equal to - 0 (i.e. null hypothesis). Note that since *L* is not known a priori, the calculation is repeated for *L*=1, 2, 3, ..., 10,…, 20, etc. and the largest *p-value* is retained.

Figure 109a reveals no significant negative trend between the mean ozone sensitivity and the F205 variance. Although the linear regression hints at increasing mean ozone sensitivity for decreasing F205 variance, the likelihood for the slope to be positive or equal to zero cannot be excluded statistically ($p > 0.10$). In addition, a non-significant correlation coefficient of -0.19 between the mean ozone sensitivity and the F205 variance is found. This is not the case for the spread of ozone sensitivity, which significantly increases with decreasing high-frequency (short-term) F205 variability (Fig. 109b). This trend further intensifies for the lowest F205 variance values (black and purple dots), corresponding to the phase of the solar cycle with the lowest activity (see insert panel on Fig. 109b). This quantitative analysis hence confirms that the accuracy of the ozone sensitivity estimation increases when the F205 fluctuations are large. We similarly tested the dependence of the mean ozone sensitivity and its spread to the absolute value of F205 (shown in the insert of Fig. 98a), an indicator of solar activity. Results are not shown here for brevity. Although we obtain results consistent with those based on the F205 variance (which is expected given the close connection between solar cycle activity and solar rotational fluctuations), the statistical significance is found to be less pronounced, suggesting a closer link with the amplitude of the fluctuations of the rotational solar cycle rather than the absolute values of F205.

4.2 Influence of the size of the time window analysis

Finally, the robustness of the estimated ozone sensitivity is examined with respect to the size of the time window. The procedure is as follows. For each ensemble simulation (of maximum size t_{max}=15 years), a time window of a given size, say Δt, (Δt is comprised between 1 and 15 years) sliding by a 1year step is used to resample the ozone 15year time series and create $n_{ensemble,windows}(\Delta t)$ $(= t_{max}-\Delta t+1)$ shorter time series of size Δt. Given that the ensemble contains five simulations, the total number of samples for a given Δt is thus $n_{windows}=5 \times n_{ensemble,\,windows}(\Delta t)$ (i.e. 75, 45, 5 samples for 1, 7, 15year time windows, respectively). For each time window size, the ozone sensitivity to F205 is estimated per individual sample. Finally, the mean ozone sensitivity and its spread are derived by calculating the average and the standard deviation over all samples.

Figure 110a shows the ozone sensitivity profiles when a 1year time window is considered. In agreement with the previous ensemble mean ozone sensitivity profiles calculated for 3year time windows and at different solar cycle phases (Figs 76 and

87), a maximum mean sensitivity of 0.4 is found near 3 hPa. The ozone sensitivity spread (dashed envelop) is larger though and even expands towards negative values, demonstrating that a 1year window is not at all long enough to estimate robustly the ozone sensitivity. Figure 110b focuses on the 3 hPa pressure level, where the sensitivity peaks, and reveals that, as expected, the longer the time window is, the smaller the spread is. Figure 110c shows the coefficient of variation of the ozone sensitivity (1σ standard deviation normalized by the mean and expressed in percent) as a function of the size of the time window. It is found that a minimum time window size of 3 years or 10 years is required for the standard deviation to drop under 50% or 20%, respectively, of the mean sensitivity (i.e. ~0.4). These uncertainty ranges additionally also strongly depend on the amplitude of the solar rotational variations and hence the phase of the 11year solar cycle; we find that during solar maximum of cycle 23, minimum of cycle 22, a minimum time window size of 2, or 5 years, respectively, is required for the standard deviation to drop under 50%. To obtain a standard deviation lower than 20%, we however found that randomly choosing a 10year time window length performs better than restricting the analysis to short but solar maximum period only (i.e. solar 23). These results suggest that long time series are preferable to estimate accurately the ozone sensitivity to solar rotational fluctuations in observations.These results demonstrate that long time series are required for an accurate estimation of the ozone sensitivity to solar rotational fluctuations in observations. It is very likely that some, if not most, of the discrepancies between estimates of the ozone sensitivity found in previous studies originate from differences in the periods and lengths of the considered time windows.

5 Summary and concluding remarks

In this paper, we examined the tropical stratosphere ozone response to the solar rotational cycle in satellite observations and simulations of the chemistry-climate model LMDz-Reprobus. We first focused our analysis on the case study of two 3year periods associated with the declining phases of solar cycles 22 and 23. The solar rotational fluctuations are stronger during the first period than the second period. We found that, although the solar rotational signature in the UV forcing is reasonably well marked during both periods, the amplitude of ozone variations expected at the corresponding timescales (i.e. ~27 days), in observational records and individual model realizations, does not significantly differ from the noise. Nonetheless, UV and ozone fluctuations show a statistical significant coherence in the middle and upper tropical stratosphere (above ~30 km, or 10 hPa) at the solar rotational timescales. These results hence suggest that ozone significantly responds to the solar rotational variations but the signal is partly masked by other sources of ozone variability at these timescales, most likely of dynamical origin. Applying the same spectral analysis to the average of CCM ensemble simulations allows reducing the 'masking' effect by random dynamical variability, so that the rotational signal in ozone can be more easily identified and estimated.

Lag correlations and linear regressions have then been used to characterize the vertical profile of the ozone response to the solar rotational cycle in the observations and the model during the same periods. Although these results are consistent with estimates of previous studies (Hood, 1986; Brasseur et al., 1987; Brasseur, 1993; Hood and Zhou, 1998) and a reasonable agreement is found between the MLS observations and the CTM experiments, significant differences are found between the two periods. This may be attributed to differences in solar UV forcing or in dynamical variability between the two periods. Analysis of the CCM ensemble simulations suggest that the differences mostly originate from the dynamical variability. The large spread in the ensemble mean sensitivity profile calculated for 3year intervals reflects the 'masking' effect of non-solar dynamical variability in the estimation of the solar rotational signal in ozone and may certainly explain some inconsistencies found in previous studies.

In our CCM experimental design, the direct radiative effect of UV on heating rates has been neglected leading to an underestimated temperature response to the 27day cycle. As a consequence, this may affect the ozone response by reducing the magnitude of the solar-induced temperature feedback on chemical reaction rates. Nevertheless, a detailed comparison of the ozone response in our analysis with results from previous independent CCM studies (Rozanov et al., 2006 ; Sukhodolov et al., 2017) revealed a very good correspondence, despite the fact that their experimental design included the direct radiative heating effect. This suggests that the feedback exerted by the solar-induced temperature fluctuations on ozone is modest, at least at the 27day time scales. This is in fact not surprising. In their recent study, Sukhodolov et al. (2017) shown that the atmospheric internal variability largely dominates the variability of the stratospheric temperature on 27day time scales, making the temperature response to the solar forcing difficult to identify and, hence, the influence of its feedback on ozone secondary. Nonetheless, we recognize that to quantify properly the impact of the neglected solar-induced temperature feedback on our results, additional CCM experiments including the direct radiative effect of UV on heating rate should be performed.

Given the good representation of the ozone response to the solar rotational cycle calculated by our model in the CTM configuration, wNext, we take advantage of the ensemble of five CCM simulations to test whether the ozone sensitivity depends on the phase of the 11year solar cycle. Considering an ensemble of simulations allows in particular to reduce the masking effect induced by the dynamical random variability. Our results suggest that the level of solar activity does not have an impact on the expected value (i.e. ensemble mean) of the ozone sensitivity. However, the ensemble spread decreases during high solar activity periods, making the ozone sensitivity retrieval easier and more robust, e.g., during the maximum phase of the 11year solar cycle.

The ensemble mean ozone sensitivity and its spread have been additionally examined as a function of the amplitude of (i) the solar rotational cycle fluctuations (shown) and (ii) the phase of the 11year solar cycle (not shown). Here again, no robust dependence of the ensemble mean ozone sensitivity against each of the two variable is found when the results of the five 15year simulations are averaged. Although the results hint at a slightly negative trend, i.e. increasing ensemble mean ozone sensitivity for decreasing rotational fluctuations (or 11year solar cycle activity), neither the slopes nor the correlation coefficients are statistically significant. Hence, our results could not confirm previous findings of Grudzev et al. (2009) or Kubin et al. (2011) who, using model experiments, suggested an increased ozone sensitivity with decreasing solar rotational fluctuations. Nevertheless, it must be noted that the conclusions of Gruzdev et al. were reached by carrying out experiments with a solar rotational forcing that had an amplitude 3 times larger than a realistic one. Further model experiments, considering for instance longer simulations and/or stronger forcing, would help to address this issue more thoroughly.

In contrast with the ensemble mean ozone sensitivity, as expected, the ensemble spread ozone sensitivity shows a clear increase with decreasing solar rotational cycle fluctuations. The negative trend further intensifies during the period with very low solar rotational fluctuations, corresponding here to the period of minimum solar activity between the end of the solar cycle 22 and the beginning of the solar cycle 23 (i.e. 1994-1997). These findings are consistent with the results of Fioletov (2009) who showed a noticeable difference in the estimate of the ozone sensitivity profile in 1994-1998 by comparison with other periods. Hence, when the solar rotational fluctuations are small, the 'masking' effect of dynamical variability becomes more prominent and makes the estimate of the ozone sensitivity less accurate.

Finally, we demonstrate that, while the mean ozone sensitivity (e.g. ~0.4 at 3 hPa) is more or less independent of the size of the time window (tested from 1 to 15 years) when the results of the five 15year simulations are analysed and averaged, the accuracy of its estimate improves dramatically with increasing size of the time window. We found that, on average, a minimum time window size of 3 years (corresponding to ~40 solar rotational cycles) is needed for the 2σ uncertainty to drop below 100%. More concretely, this means that if the ozone sensitivity to solar rotational fluctuations is derived over only three successive years of observations (or of a single model realization), there is a 95% likelihood for the estimate to take any value in the range [0-0.8] at 3 hPa. The error in the sensitivity estimation also strongly depends on the amplitude of the solar rotational fluctuations and is thus linked to the solar activity. For a constant uncertainty threshold, the higher the solar activity is, the shorter the required time window length is. We finally find that a minimum of 10 years of data is required for the 1σ uncertainty in the ozone sensitivity estimate to drop under 20%.

Overall, it is likely that the discrepancies in the estimated value of ozone sensitivity found in previous studies originate from differences in the length of time windows that were used for analysis and in the level of solar activity associated with these periods. Both parameters significantly influence the accuracy of solar rotational signal estimates. In this regard, it is likely that similar issues have also affected the accuracy in the estimation of ozone response to the 11year solar signal. The estimation is expected to be even more difficult because observational time series cover a very limited number of 11-year cycles and there are other well-known sources of decadal variability in the atmosphere and climate system. Maycock et al. (2016) recently found very large discrepancies in the estimation of the ozone response the 11year cycle using various satellite datasets which cover different time periods of different length.

6 Data availability

~~MLS~~ UARS MLS and ~~MLS~~ Aura MLS satellite data are publicly available at https://earthdata.nasa.gov/ after registration. LMDz-Reprobus data used in this study are available upon request to the corresponding author (remi.thieblemont@latmos.ipsl.fr).

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References

Austin, J., Hood, L., and Soukharev, B.: Solar cycle variations of stratospheric ozone and temperature in simulations of a coupled chemistry-climate model, *Atmos. Chem. Phys.*, 7(6), 1693–1706, 2007.

Austin, J., Tourpali, K., Rozanov, E., Akiyoshi, H., Bekki, S., Bodeker, G., Brühl, C., Butchart, N., Chipperfield, M., Deushi, M., Fomichev, V. I., Giorgetta, M. A., Gray, L., Kodera, K., Lott, F., Manzini, E., Marsh, D., Matthes, K., Nagashima, T., Shibata, K., Stolarski, R. S., Struthers, H., and Tian, W.: Coupled chemistry climate model simulations of the solar cycle in ozone and temperature, *J. Geophys. Res*., 113, D11306, doi:10.1029/2007JD009391, 2008.

Bai, T.: Hot spots for solar flares persisting for decades: longitude distributions of flares of cycles 19-23, *Astrophys. J.*, 585(2), 1114, 2003.

Bossay, S., Bekki, S., Marchand, M., Poulain, V., and Toumi, R.: Sensitivity of tropical stratospheric ozone to rotational UV variations during the declining phases of solar cycles 22 and 23, *J. Atmos. Solar-Terr. Phys.*, 130-131, 10.1016/j.jastp.2015.05.014, 2015

Brasseur, G., De Rudder, A., Keating, G., and Pitts, M.: Response of middle atmosphere to short-term solar ultraviolet variations: 2. theory, *J. Geophys. Res*., 92(D1), 903–914, 1987.

Brasseur, G.: The response of the middle atmosphere to long-term and short-term solar variability: A two-dimensional model, *J. Geophys. Res*., 98(D12), 23,079–23,090, 1993.

Brasseur, G. and Solomon, S.: *Aeronomy of the middle atmosphere: chemistry and physics of the stratosphere and mesosphere*, vol. 32, Springer Science & Business Media, 2005

CCMVal, S., SPARC report on the evaluation of chemistry–climate models. V. Eyring, T.G. Shepherd, and D. W. Waugh, eds., SPARC rep. 5, wcrp-132, Tech. rep., WMO/TD-1526, 434 pp.[Available online at www. sparc-climate. org/publications/sparc-reports/.], 2010

Chen, L., London, J., and Brasseur, G.: Middle atmosphere ozone and temperature responses to solar irradiance variations over 27day periods, *J. Geophys. Res.*, 102, 29957–29979, 1997.

Dee, D.P., Uppala, S.M., Simmons, A.J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., and

Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Q. J. Roy. Meteor. Soc.*, 137, 553–597, 2011.

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

Dikty, S., Weber, M., von Savigny, C., Sonkaew, T., Rozanov, A., and Burrows, J.P.: Modulations of the 27-day solar rotation signal in stratospheric ozone from scanning imaging absorption spectrometer for atmospheric cartography (SCIAMACHY) (2003–2008), *J. Geophys. Res.*, 115(D1), 2010

Eckman, R. S.: Response of ozone to short-term variations in the solar ultraviolet radiance. 1. A theoretical model, *J. Geophys. Res.*, 91, 6695–6704, 1986a.

Eckman, R. S.: Response of ozone to short-term variations in the solar ultraviolet irradiance: 2. observations and interpretation, *J. Geophys. Res*., 91(D6), 6705–6721, 1986b.

Fioletov, V.: Estimating the 27-day and 11-year solar cycle variations in tropical upper stratospheric ozone, *J. Geophys. Res*., 114(D2), 2009.

Fleming, E. L., Chandra, S., Jackman, C.H., Considine, D.B., and Douglass, A.R.: The middle atmospheric response to short and long term solar UV variations: Analysis of observations and 2d model results, *J. Atmos. Solar-Terr. Phys.*, 57(4), 333–365, 1995.

Forster, P. M., Fomichev, V. I., Rozanov, E., Cagnazzo, C., Jonsson, A. I., Langematz, U., Fomin, B., Iacono, M. J., Mayer, B., Mlawer, E., Myhre, G., Portmann, R. W., Akiyoshi, H., Falaleeva, V., Gillett, N., Karpechko, A., Li, J., Lemennais, P., Morgenstern, O., Oberländer, S., Sigmond, M., and Shibata, K.: Evaluation of radiation scheme performance within chemistry climate models, *J. Geophys. Res.-Atmos*., 116, D10302, doi:10.1029/2010JD015361, 2011.

Fouquart, Y. and Bonnel, B.: Computations of solar heating of the earth's atmosphere- a new parameterization, *Beitraege zur Physik der Atmosphaere*, 53, 35–62, 1980.

Fröhlich, C. and Lean, J.: Solar Radiative Output and its Variability: Evidence and Mechanisms. *Astron. and Astrophys. Rev*., 12, pp. 273-320, 2004, doi: 10.1007/s00159-004-0024-1, 2004.

Froidevaux, L., Waters, J.W., Read, W.G., Elson, L.S., Flower, D.A., and Jarnot, R.F.: Global ozone observations from the UARS MLS: An overview of zonal-mean results, *J. Atmos. Sci.*, 51(20), 2846–2866, 1994.

Gerber, E. P., Butler, A., Calvo, N., Charlton-Perez, A., Giorgetta, M., Manzini, E., Perlwitz, J., Polvani, L. M., Sassi, F., Scaife, A. A., Shaw, T. A., Son, S.-W., and Watanabe, S.: Assessing and understanding the impact of stratospheric dynamics and variability on the earth system, *B. Am. Meteorol. Soc*., 93, 845–859, doi: 10.1175/bAms-d-11-00145.1, 2012.

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. Atmos. Sci.* 66, 2402–2417, 2009.

Gray, L. J., Beer, J., Geller, M., Haigh, J. D., Lockwood, M., Matthes, K., Cubasch, U., Fleitmann, D., Harrison, G., Hood, L., Luterbacher, J., Meehl, G. A., Shindell, D., van Geel, B., and White, W.: Solar influence on climate, *Rev. Geophys*., 48, RG4001, doi:10.1029/2009RG000282, 2010.

Gruzdev, A., Schmidt, H., and Brasseur, G.: The effect of the solar rotational irradiance variation on the middle and upper atmosphere calculated by a three-dimensional chemistry-climate model, *Atmos. Chem. Phys.*, 9(2), 595–614, 2009.

Hood, L.L.,:Coupled stratospheric ozone and temperature responses to short-term changes in solar ultraviolet flux: An analysis of NIMBUS 7 SBUV and SAMS data, *J. Geophys. Res*., 91(D4), 5264–5276, 1986.

Hood, L. L., Huang, Z., and Bougher, S. W.: Mesospheric effects of solar ultraviolet variations: Further analysis of SME IR ozone and Nimbus 7 SAMS temperature data, *J. Geophys. Res.*, 96, 12989–13002, 1991.

Hood, L., and Zhou, S.: Stratospheric effects of 27-day solar ultraviolet variations: An analysis of UARS MLS ozone and temperature data, *J. Geophys. Res*., 103(D3), 3629–3638, 1998.

Hood, L. L. and Zhou, S.: Stratospheric effect of 27-day solar ultraviolet variations: The column ozone response and comparison of solar cycles 21 and 22, *J. Geophys. Res.*, 104, 26473–26479, 1999.

Hood, L.L.: Effects of short-term solar UV variability on the stratosphere, *J. Atmos. Solar-Terr. Phys.*, 61, 45– 51, 1999.

Hood, L. L.: Effects of solar UV variability on the stratosphere, *Solar variability and its effects on climate*, pp. 283–303, 2004.

Hourdin, F., Musat, I., Bony, S., Braconnot, P., Codron, F., Dufresne, J.L., Fairhead, L., Filiberti, M.A., Friedlingstein, P., Grandpeix, J. Y., Krinner, G., Levan, P., Li, Z. X., and Lott, F.: The LMDZ4 general circulation model: Climate performance and sensitivity to parametrized physics with emphasis on tropical convection, *Clim. Dynam*., 27, 787–813, 2006.

Hourdin, F., Foujols, M.-A., Codron, F., Guemas, V., Dufresne, J.-L., Bony, S., Denvil, S., Guez, L., Lott, F., Ghattas, J., Braconnot, P., Marti, O., Meurdesoif, Y., and Bopp, L.: Impact of the LMDZ atmospheric grid configuration on the climate and sensitivity of the IPSL-CM5A coupled model, *Clim. Dyn.*, 40: 2167. doi:10.1007/s00382-012-1411-3, 2013.

Jourdain, L., Bekki, S., Lott, F., and Lefèvre, F.: The coupled chemistry-climate model LMDz-Reprobus: description and evaluation of a transient simulation of the period 1980–1999, *Ann. Geophys*., 26(6), 1391–1413, 2008.

Keating, G. M., Brasseur, G. P., Nicholson III, J. Y., and De Rudder, A.: Detection of the response of ozone in the middle atmosphere to short-term solar ultraviolet variations, *Geophys. Res. Lett*., 12, 449–452, 1985.

Keating, G., Pitts, M., Brasseur, G.P., and De Rudder, A.: Response of middle atmosphere to shortterm solar ultraviolet variations: 1. observations, *J. Geophys. Res*., 92(D1), 889–902, 1987.

Kodera, K. and Kuroda, Y.: Dynamical response to the solar cycle. *J. Geophys. Res*. 107, 4749 2002.

Kopp, G., and Lean, J.L.: A new, lower value of total solar irradiance: Evidence and climate significance, *Geophys. Res. Lett*, 38(1), 2011.

Kubin, A., Langematz, U., and Brühl, C.: Chemistry climate model simulations of the effect of the 27-day solar rotational cycle on ozone, *J. Geophys. Res*., 116, D15301, doi:10.1029/2011JD015665, 2011.

Kurucz, R. L.: New Opacity Calculations, in *NATO Advanced Science Institutes* (ASI) *Series C*, vol. 341, edited by L. Crivellari, I. Hubeny, and D. G. Hummer, p. 441, 1991.

Law, K., Plantevin, P., Shallcross, D., Rogers, H., Pyle, J., Grouhel, C., Thouret, V., and Marenco, A.: Evaluation of modeled O_3 using measurement of ozone by airbus in-service aircraft (MOZAIC) data, *J. Geophys. Res.*, 103(D19), 25,721–25,737, 1998.

Le Treut, H., Li, Z., and Forichon, M.: Sensitivity study of the LMD GCM to greenhouse forcing associated with two different cloud water parametrizations, *J. Clim.*, 7, 1827–1841, 1994.

Le Treut, H., Forichon, M., Boucher, O., and Li, Z.-X.: Sulfate aerosol indirect effect and co2 greenhouse forcing: Equilibrium response of the LMD GCM and associated cloud feedbacks, *J. Clim.*, 11(7), 1673–1684, 1998.

Lean, J.: Evolution of the sun's spectral irradiance since the maunder minimum, *Geophys. Res. Lett*, 27(16), 2425–2428, 2000

Lefevre, F., Brasseur, G., Folkins, I., Smith, A., and Simon, P.: Chemistry of the 1991–1992 stratospheric winter: Three-dimensional model simulations, *J. Geophys. Res*., 99(D4), 8183–8195, 1994.

Lefèvre, F., Figarol, F., Carslaw, K.S., and Peter, T.: The 1997 arctic ozone depletion quantified from three-dimensional model simulations, *Geophys. Res. Lett*, 25(13), 2425–2428, 1998.

Livesey, N.J., Read, W.G., Froidevaux, L., Waters, J.W., Pumphrey, H.C., Wu, D.L., Santee, M.L., Shippony, Z., and Jarnot, R.F.: The UARS Microwave Limb Sounder version 5 dataset: Theory, characterization and validation, *J. Geophys. Res.*, 108, (D13) 4378, doi:10.1029/2002JD002634, 2003.

Livesey, N. J., Read, W., Wagner, P. A. Froidevaux, L., Lambert, A., Manney, G. L., Millán Valle, L., Pumphrey, H. C., Santee, M. L., Schwartz, M. J., Wang, S., Fuller, R. A., Jarnot, R. F., Knosp, B. W., and E., M.: Version 4.2x Level 2 data quality and description document, JPL D-33509 Rev. C, availablefromhttp://mls.jpl.nasa.gov, 2017.

Lomb, N. R.: Least-squares frequency analysis of unequally spaced data, *Ap. Space Sci.*, 39(2), 447–462, 1976

Lott, F., Fairhead, L., Hourdin, F., and Levan, P.: The stratospheric version of LMDz: dynamical climatologies, arctic oscillation, and impact on the surface climate, *Clim. Dyn.*, 25(7-8), 851–868, 2005.

Madronich, S., and Flocke, S.: The role of solar radiation in atmospheric chemistry, in *Environmental photochemistry*, pp. 1–26, Springer, 1999.

Marchand, M., Keckhut, P., Lefebvre, S., Claud, C., Cugnet, D., Hauchecorne, A., Lefevre, F., Lefebvre, M.-P., Jumelet, J., Lott, F., Hourdin, F., Thuillier, G., Poulain, V., Bossay, S., Lemennais, P., David, C., and Bekki, S.: Dynamical amplification of the stratospheric solar response simulated with the Chemistry-Climate Model LMDz-Reprobus, *J. Atmos. Solar-Terr. Phys*., 75–76, 147–160, 2012.

Maycock A. C., Matthes, K., Tegtmeier, S., Thiéblemont, R., and Hood, L.: The representation of solar cycle signals in stratospheric ozone – Part 1: A comparison of satellite observations, *Atmos. Chem. Phys.*, doi:10.5194/acp-2015-882, 2016.

Meehl, G. A., Arblaster, J.M., Branstator, G., and van Loon, H.: A coupled air-sea response mechanism to solar forcing in the pacific region, *J. Clim.*, 21(12), 2883–2897, 2008.

Minschwaner, K., Anderson, G., Hall, L., and Yoshino, K.: Polynomial coefficients for calculating O_2 schumann-runge cross sections at 0.5 cm^{-1} resolution, *J. Geophys. Res*., 97(D9), 10,103–10,108, 1992.

Morcrette, J.-J., Smith, L., and Fouquart, Y.: Pressure and temperature dependence of the absorption in longwave radiation parametrizations, *Beiträge zur Physik der Atmosphäre*, 59(4), 455–469, 1986.

Mudelsee, M.: *Climate Time Series Analysis: Classical Statistical and Bootstrap Methods*. Second edition. Springer, Cham Heidelberg New York Dordrecht London, 2014.

Poulain, V., Bekki, S., Marchand, M., Chipperfield, M.P., Khodri, M., Dhomse, S., Bodeker, G.E., Toumi, R., De Maziere, M., Pommereau, J.-P., Pazmino, A., Goutail, F., Plummer, D., Rozanov, E., Mancini, E., Akiyoshi, H., Lamarque, J.-F., and Austin, J.: Evaluation of the inter-annual variability of stratospheric chemical composition in chemistry-climate models using ground-based multi species time series, *J. Atmos. Sol-Terr. Phys.*, 145, Pages: 61-84, doi: 10.1016/j.jastp.2016.03.010, 2016.

Randel, W. J., and Wu, F.: A stratospheric ozone profile data set for 1979–2005: Variability, trends, and comparisons with column ozone data, *J. Geophys. Res*., 112(D6), 2007.

Remsberg, E. E., Marshall, B. T., Garcia-Comas, M., Krueger, D., Lingenfelser, G. S., Martin-Torres, F. J., Mlynczak, M. G., Russell III, J. M., Smith, A. K., Zhao, Y., Brown, C. W., Gordley, L. L., Lopez-Gonzalez, M., Lopez-Puertas, M., She, C.-Y., Taylor, M. J., and Thompson, R. E.: Assessment of the quality of the Version 1.07 temperature versus pressure profiles of the middle atmosphere from TIMED/SABER, *J. Geophys. Res*., 113, D17101, doi:10.1029/2008JD010013, 2008.

Remsberg, E. E.: Decadal-scale responses in middle and upper stratospheric ozone from SAGE II version 7 data, *Atmos. Chem. Phys*., 14, 1039–1053, doi:10.5194/acp-14-1039-2014, 2014.

Rottman, G., Woods, T., Snow, M., and DeToma, G.: The solar cycle variation in ultraviolet irradiance, *Adv. Space Res*., 27, 1927{1932, doi:10.1016/S0273-1177(01)00272-1., 2001.

Rottman, G., Floyd, L., and Viereck, R.: Measurements of the solar ultraviolet irradiance, *Solar Variability and Its Effects on Climate*, pp. 111–125, 2004.

Rozanov, E., Egorova, T., Schmutz, W., and Peter, T.: Simulation of the stratospheric ozone and temperature response to the solar irradiance variability during sun rotation cycle, *J. Atmos. Solar-Terr. Phys.*, 68(18), 2203–2213, 2006

Sadourny, R., and Laval, K.: January and July performance of the LMD general circulation model, *New Perspectives in Climate Modeling*, 1, 173–197, 1984.

Sander, S., Friedl, R.R., Barkern, J., Golden, D., Kurylo, M., Wine, P., Abbat, J., Burkholder, J., Moortgart, C., Huie, R., and Orkin, R. E.: Chemical kinetics and photochemical data for use in atmospheric studies, Technica Report, NASA/JPL Publication, California, USA, Evaluation No. 17, JPL Publication 10-6, 2011.

Savage, N., Law, K., Pyle, J., Richter, A., Nüß, H., and Burrows, J.: Using GOME n°2 satellite data to examine regional differences in tomcat model performance, *Atmos. Chem. Phys.*, 4(7), 1895–1912, 2004.

Scargle, J. D.: Studies in astronomical time series analysis. ii-statistical aspects of spectral analysis of unevenly spaced data, *Astrophys. J.*, 263, 835–853, 1982.

Soukharev, B., and Hood, L.: Solar cycle variation of stratospheric ozone: Multiple regression analysis of long-term satellite data sets and comparisons with models, *J. Geophys. Res*., 111(D20), 2006.

Stamnes, K., Tsay, S. Wiscombe, W., and Jayaweera, K.: A numerically stable algorithm for discrete-ordinate-method radiative transfer in multiple scattering and emitting layered media, *Appl. Opt*., 27, 2502–2509, 1988.

Sukhodolov, T., Rozanov, E., Ball, W. T., Bais, A., Tourpali, K., Shapiro, A. I., Telford, P., Smyshlyaev, S., Fomin, B., Sander, R., Bossay, S., Bekki, S., Marchand, M., Chipperfield, M P., Dhomse, S., Haigh, J. D., Peter, T., and Schmutz, W.: Evaluation of the simulated photolysis rates and their response to solar irradiance variability, *J. Geophys. Res*., 121, 6066–6084, 2016.

Sukhodolov, T., Rozanov, E., Ball, W.T., Peters, T., Schmutz, W., Modeling of the middle atmosphere response to 27-day solar irradiance variability, *J. Atmos. Sol-Terr. Phys*., 152–153, 50–61, 2017.

Swartz, W. H., Stolarski, R.S., Oman, L.D., Fleming, E.L., and Jackman, C.H.: 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, 2012.

Thuillier, G., Herse, M., Simon, P.C., Labs, D., Mandel, H., Gillotay, D., and Foujols, T.: The Visible Solar Spectral Irradiance from 350 to 850 NM as Measured by the SOLSPEC Spectrometer During the Atlas I Mission, *Solar Phys*., 177, 41–61, 1998.

Torrence, C., and Compo, G.P.: A practical guide to wavelet analysis, *Bull. Am. Meteor. Soc*., 79(1), 61–78, 1998.

Wang, Y.-M., Lean, J.L., and Sheeley, Jr N. R.: Modeling the Sun’s magnetic field and irradiance since 1713. *Astrophys. J*. 625, 522–538, 2005.

Waters, J.: Microwave limb-sounding of Earth’s upper atmosphere, *Atmos. Res*., 23(3), 391– 410, 1989.

Waters, J.: Microwave limb sounding, in *Atmospheric Remote Sensing by Microwave Radiometry*, vol. 1, p. 383, 1993.

Waters, J. W., Froidevaux, L., Harwood, R. S., Jarnot, R. F., Pickett, H. M., Read, W. G., Siegel, P. H., Cofield, R. E., Filipiak, M. J., Flower, D. A., Holden, J. R., Lau, G. K., Livesey, N. J., Manney, G. L., Pumphrey, H. C., Santee M. L., Wu, D. L., Cuddy, D.T., Lay, R. R., Loo, M. S., Perun, V. S., Schwartz, M. J., Stek, P. C., Thurstans, R. P., Boyles, M. A., Chandra, K. M., Chavez, M. C., Chen, G. S., Chudasama, B. V., Dodge, R., Fuller, R. A., Girard, M. A., Jiang, J. H., Jiang, Y. B., Knosp, B. W., LaBelle, R. C., Lam, J. C., Lee, K. A., Miller, D., Oswald, J. E., Patel, N.C., Pukala, D. M., Quintero, O., Scaff, D. M., Van Snyder, W., Tope, M. C., Wagner, P. A., and Walch, M. J.: The Earth Observing System Microwave Limb Sounder (EOS MLS) on the Aura satellite, *IEEE Transactions on Geoscience and Remote Sensing*, 44, 1075–1092, 2006.

Zhang, L., Cui, Y., He, Y., He, H., Du, Z., Li, R., and Wang, H.: Longitudinal distribution of major solar flares during 1975–2005, *Adv. Space Res*., 40(7), 970–975, 2007.

Zhou, S., Miller, A.J., and Hood, L.L.: A partial correlation analysis of the stratospheric ozone response to 27-day solar UV variations with temperature effect removed, *J. Geophys. Res*., 105(D4), 4491–4500, 2000.

Figures

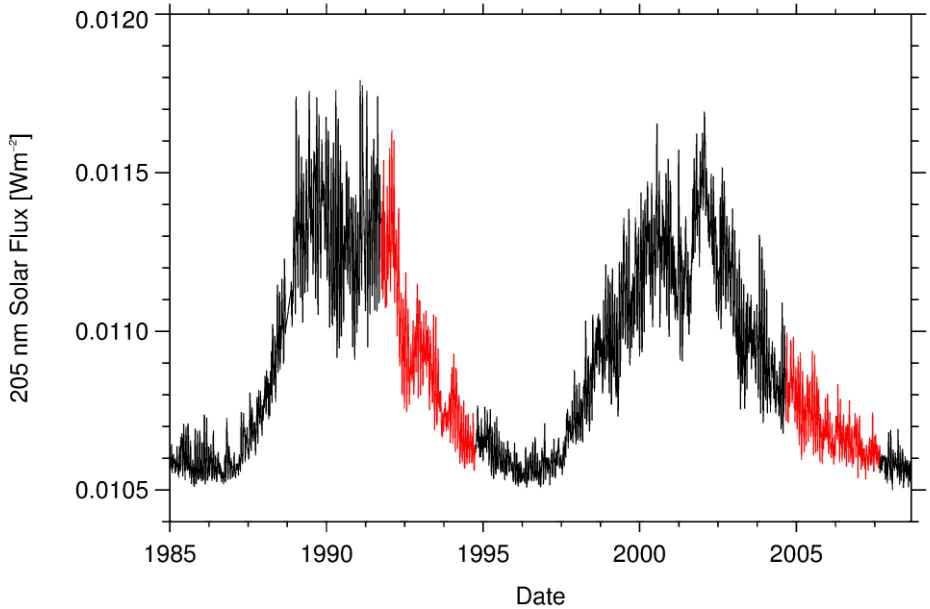


Figure 1: Temporal evolution of daily F205 from NRLSSI model over solar cycles 22 (1985-1996) and 23 (1996-2008). The two 3year periods considered here (1991-94 and 2004-07) are highlighted in red.

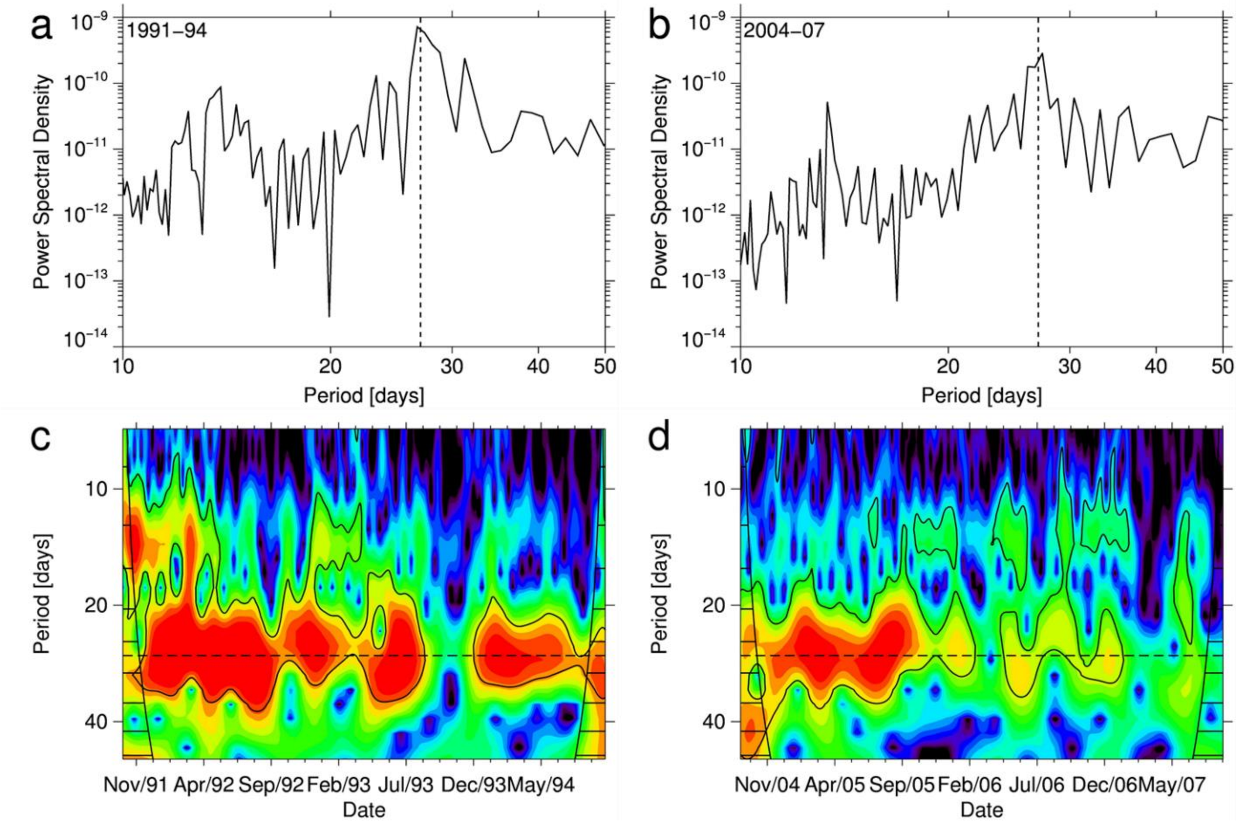
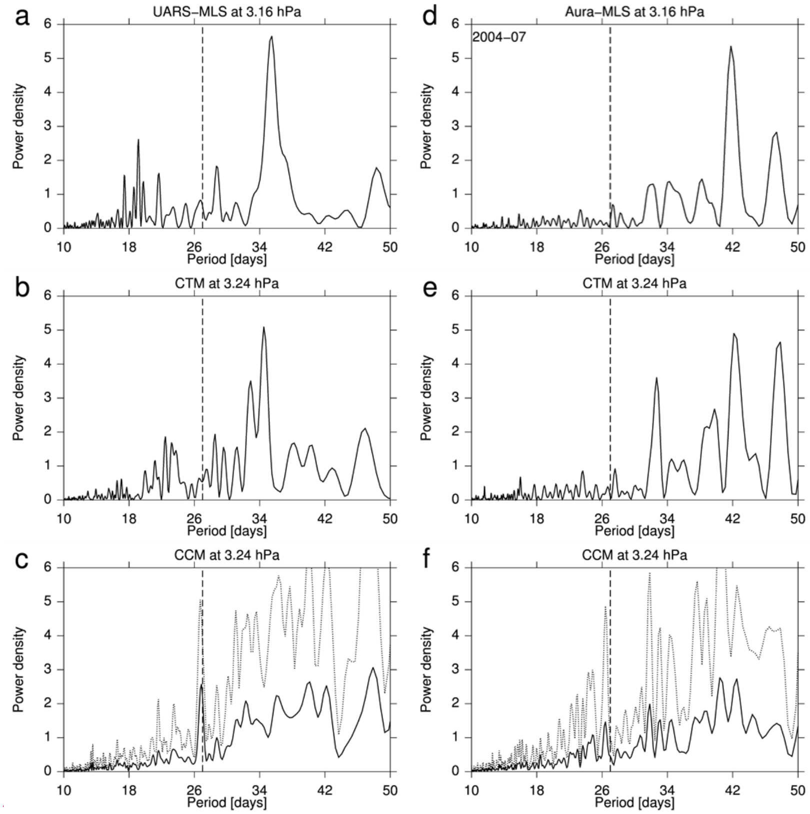


Figure 2: (Top) F205 FFT power spectra (from NRLSSI model) for the (a) 1991-94 and (b) 2004-07 period. (Bottom) Time-resolved power spectra densities (or scalogram) estimated from continuous wavelet transform (CWT) for the (c) 1991-94 and (d) 2004-07 period. The vertical, horizontal, dashed lines on (a,b), (c,d), indicate the 27day period. The cone of influence, i.e. limit beyond which scalogram should not be interpreted, is marked by horizontal solid stripes. The solid contour lines represent the 95% confidence level.



a
UARS−MLS at 3.16 hPa
b
CTM at 3.24 hPa
c
CCM at 3.24 hPa
d
Aura−MLS at 3.16 hPa
2004−07
e
CTM at 3.24 hPa
f
CCM at 3.24 hPa
Power density
Period [days]

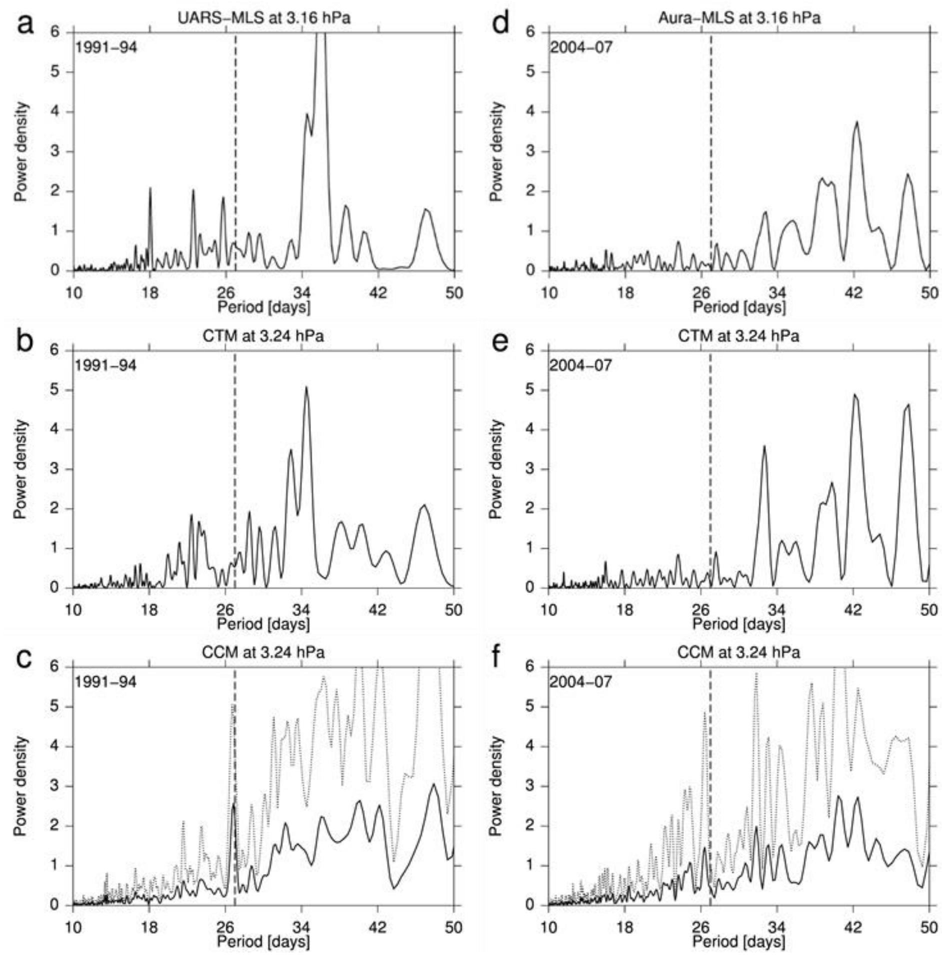


Figure 3: Ozone Lomb-Scargle periodograms for the (left) 1991-94 and (right) 2004-07 periods. The top panels represent ozone Lomb-Scargle periodograms from (a) ~~MLS~~ UARS MLS and (d) ~~MLS~~ Aura MLS observations. The middle panels (b and e) represent the ozone Lomb-Scargle periodograms for CTM simulation and the bottom panels (c and f) ~~from the runs ensemble of the~~the average periodogram of the CCM ensemble~~simulations~~. The dotted envelop (c and f) indicates the 2σ standard deviation of the ensemble of CCM simulations.

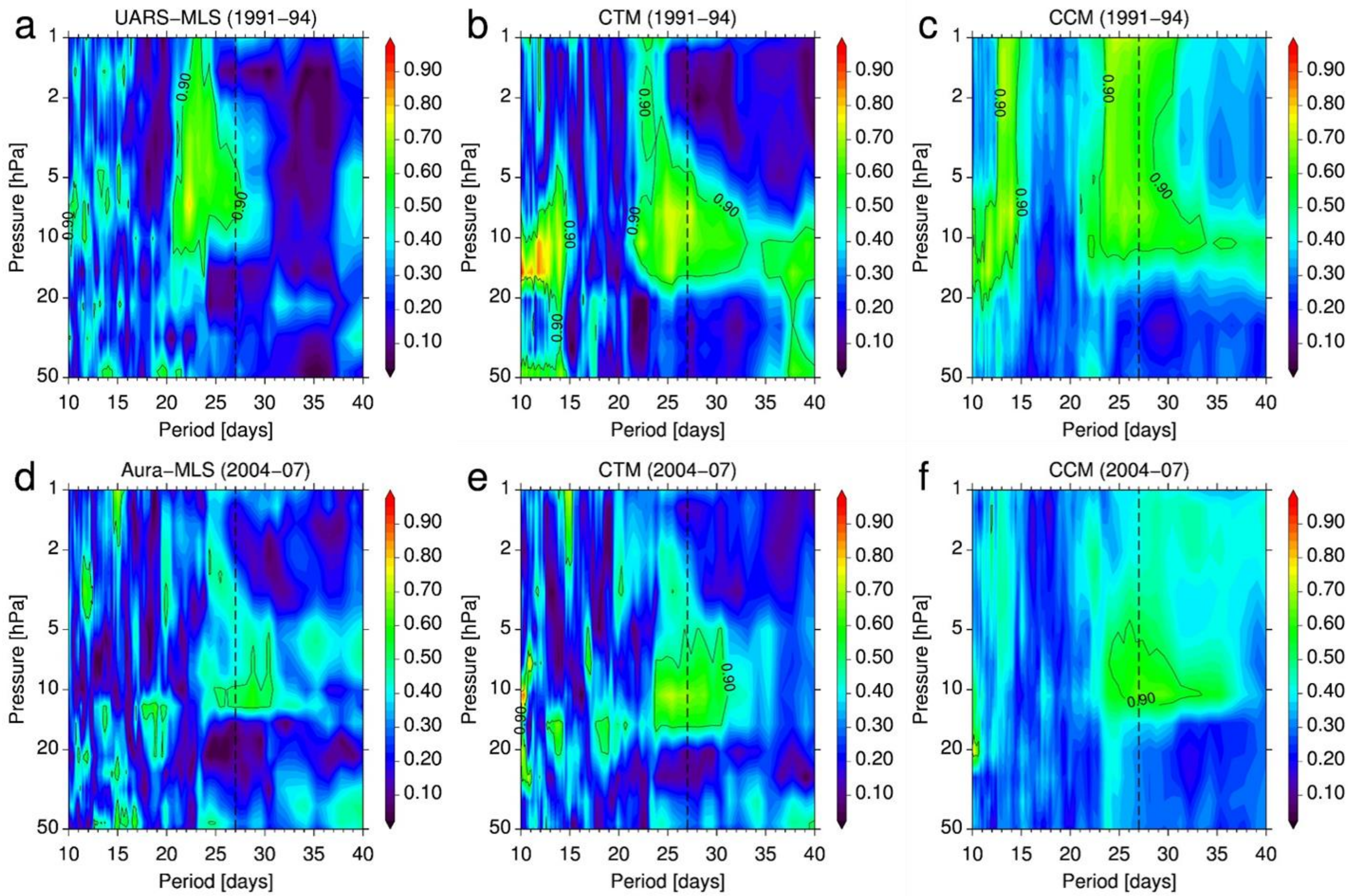


Figure 4: Mean squared coherence between ozone and F205 as a function of period (days) and pressure level (hPa) for the (top) 1991-94 and (bottom) 2004-07 period and for (a, d) MLS observations, (b,e) CTM and (c,f) CCM simulations. Black contour lines indicate the 90% confidence level and the vertical dashed black lines indicate the 27day period.

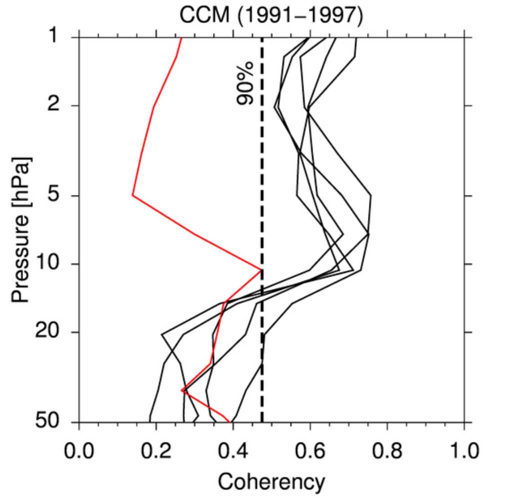


Figure 5. Vertical profile of the mean squared coherence between ozone and F205 averaged between 22 and 30 day periods and calculated for the time period 1991-1997. The black lines correspond to the results of individual ensemble members (five in total) and the red line to the results of the experiment forced with constant solar forcing. The vertical dashed line indicates the 90% confidence limit.

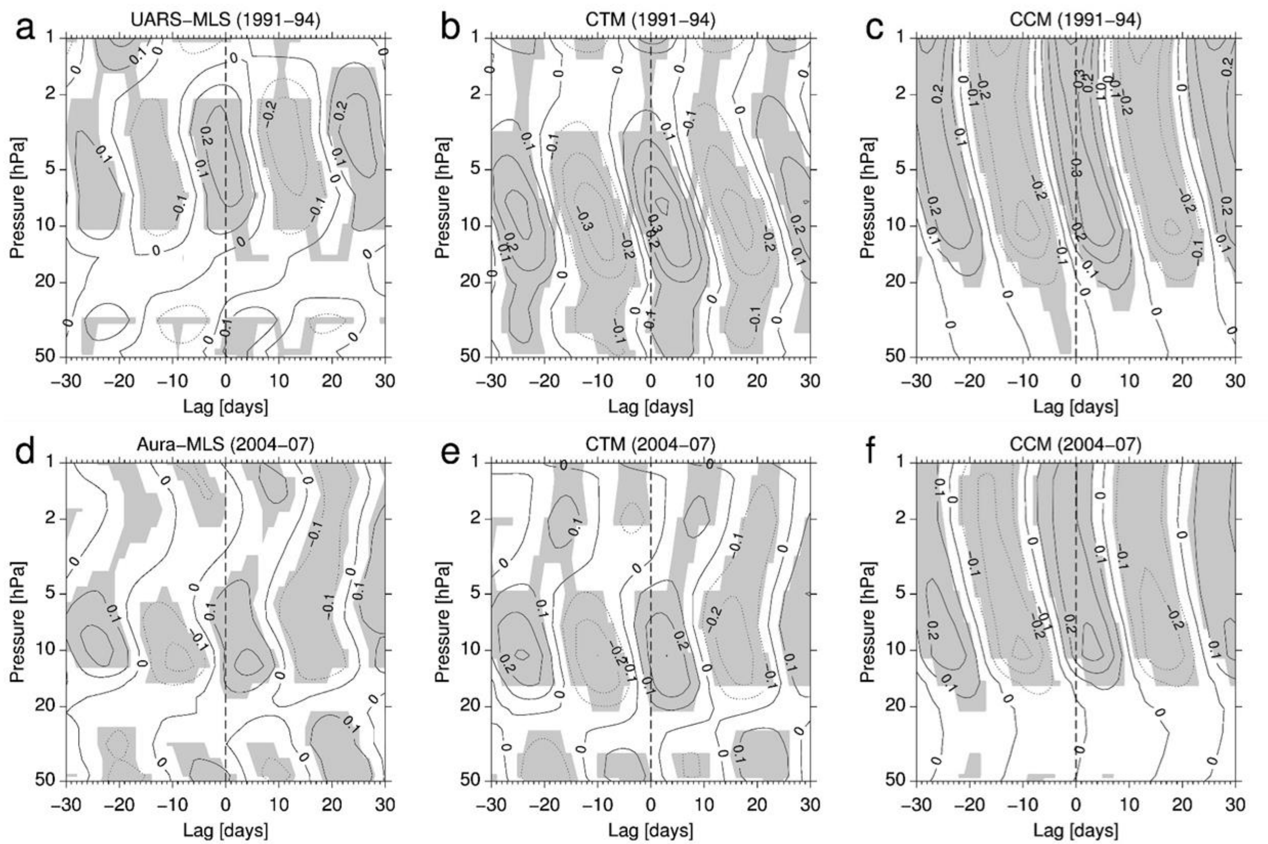


Figure 6~~5~~: Cross-correlation between digitally filtered (see main text) ozone and F205 as a function of time lag (in days) and pressure level (hPa) for the (top) 1991-94 and (bottom) 2004-07 periods. (a,d), (b,e) and (c,f) panels show cross-correlation between F205 and MLS observations, CTM and CCM simulations, respectively. Shading represents areas with 95% confidence level.

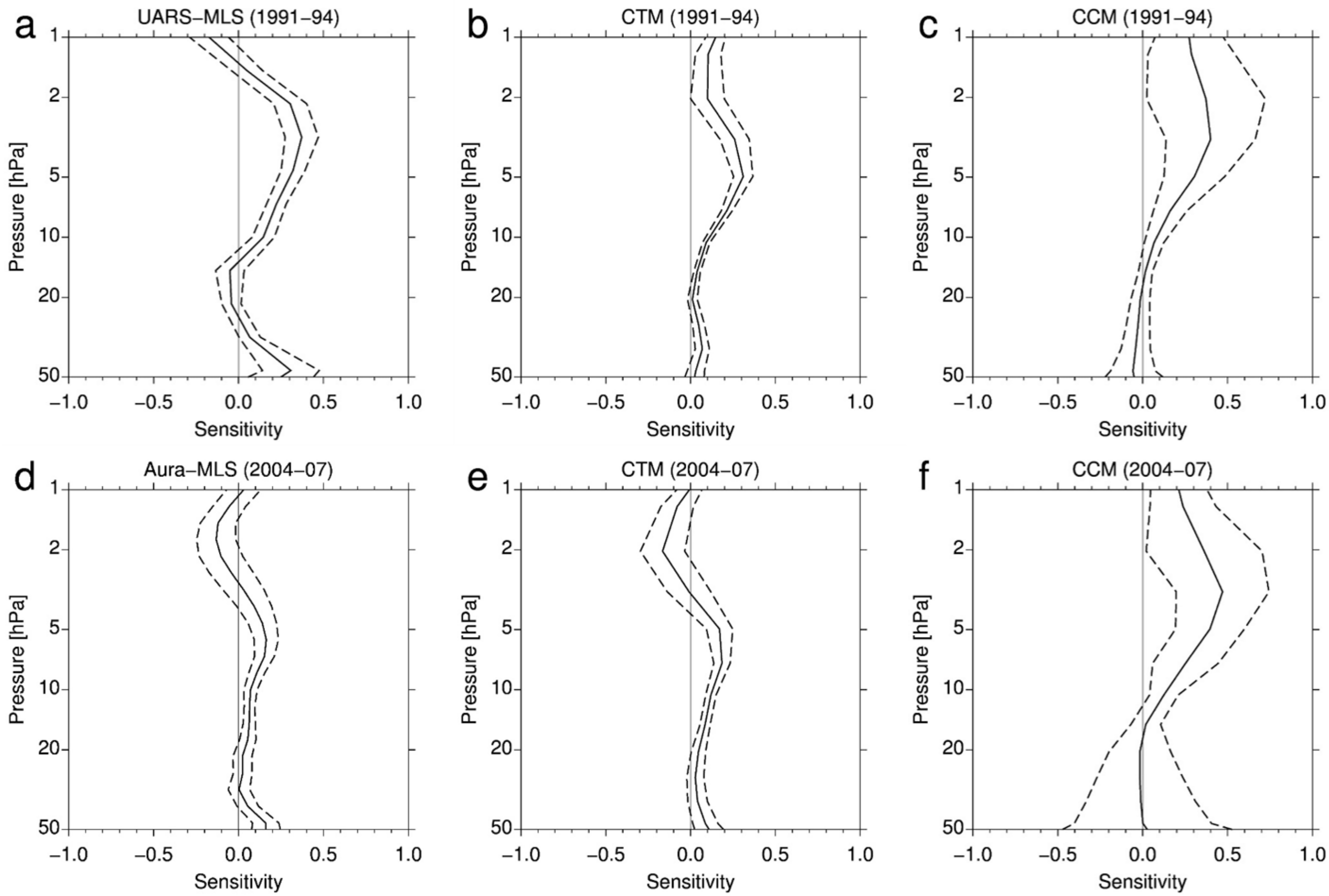


Figure 7~~6~~: Vertical profile of ozone sensitivity to F205 (% change in ozone for 1% change in F205) at lag 0 for the (top) 1991-94 and (bottom) 2004-07 periods. Results are shown for (a) ~~MLS~~ UARS MLS, (d) ~~MLS~~ Aura MLS, (b, e) CTM simulations and (c,f) CCM ensemble simulations. (a,b,d,e) The dashed envelop indicates the 2σ standard error of the regression estimates. (c,f) The dashed envelop indicates the 2σ range.

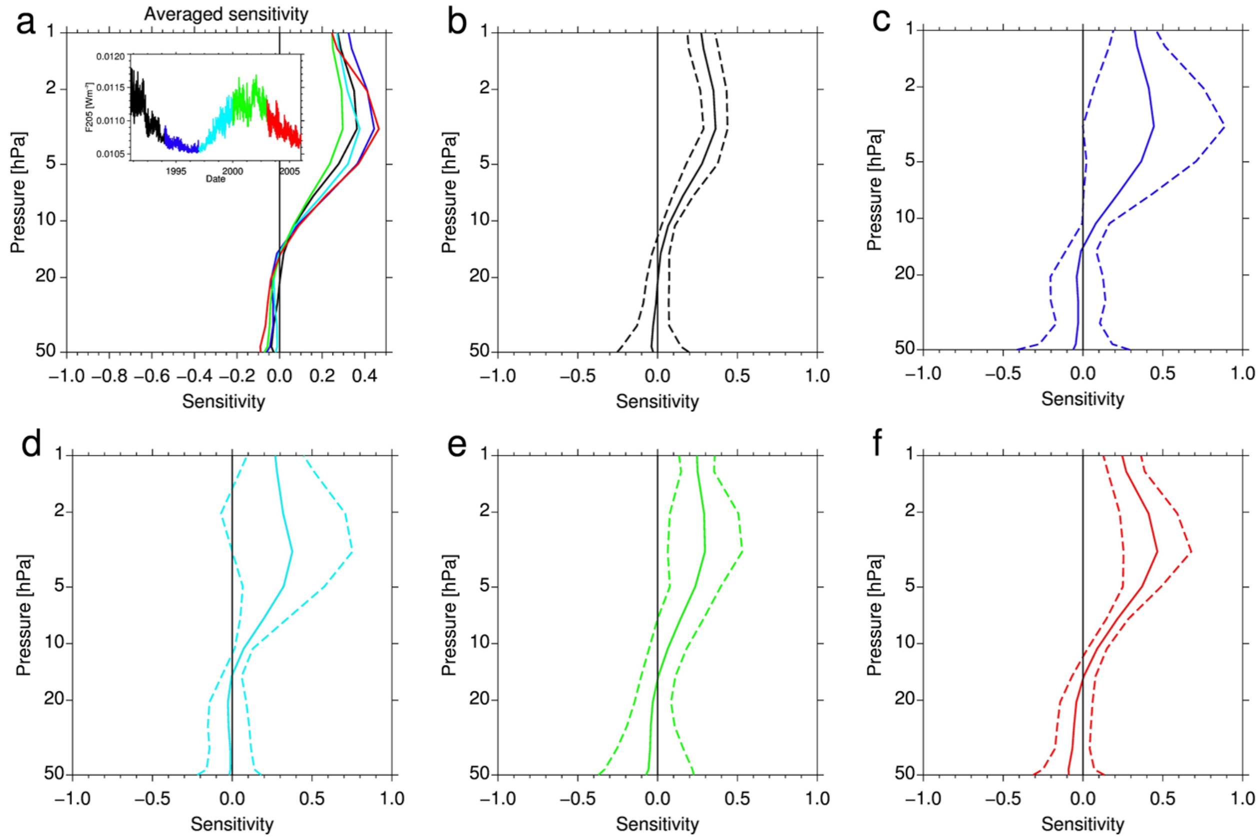


Figure ~~87~~8: (a) CCM ensemble ozone sensitivity profile at lag 0 for each of the 3year period. Each period and its corresponding colour is shown in the insert plot (a). CCM ensemble mean ozone sensitivity profile and its 2σ range are shown for each individual 3year periods: (b) 07/1990-06/1993, (c) 07/1993-06/1996, (d) 07/1996-06/1999, (e) 07/1999-06/2002, (f) 07/2002-06/2005.

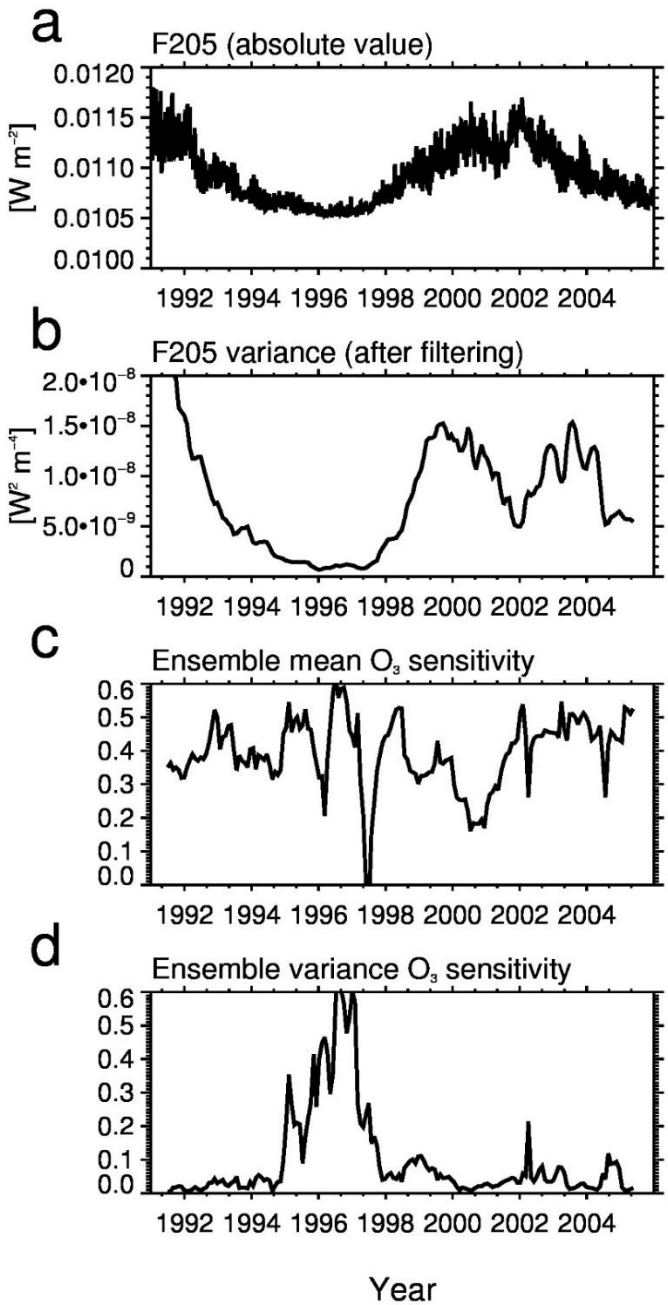


Figure 98: Digitally filtered (b) F205 variance time series, (c) ensemble mean ozone sensitivity and (d) ozone sensitivity ensemble variance time series at 3hPa computed over a 1year running window. Each window is sliding for one month at each step. (a) The F205 index time series is reproduced on the top panel for clarity.

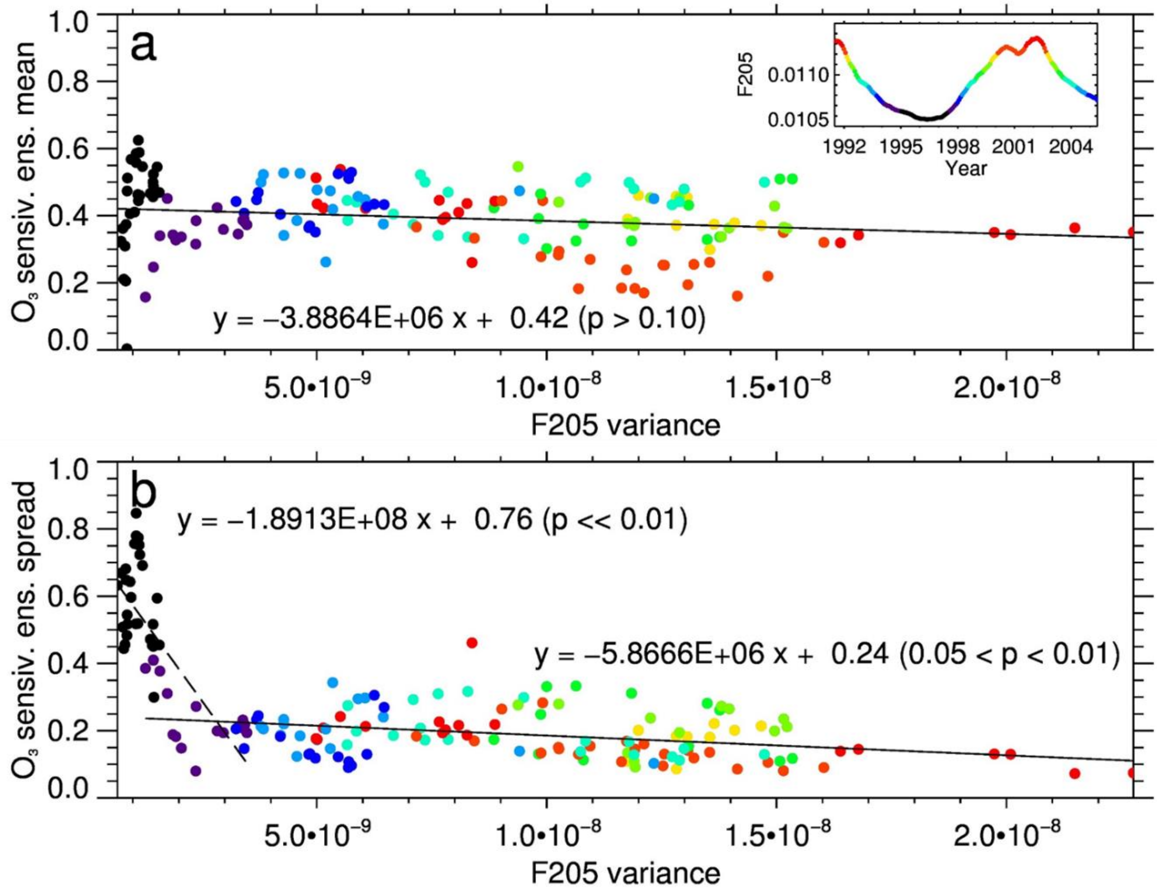


Figure 109: Scatter plots of the CCM ensemble (a) mean ozone sensitivity and (b) its spread (1σ) versus the F205 variance. Dots are coloured with respect to the value of the F205 flux, shown in the insert plot of panel a. Least square linear regression fits are superimposed (solid and dashed segments) together with their equation and the statistical significance of the slope value (in brackets, see text for details). The correlation coefficients are (a) -0.19 (p>0.10), (b, dashed) -0.76 (p<0.05) and (b, solid) -0.36 (p<0.10).

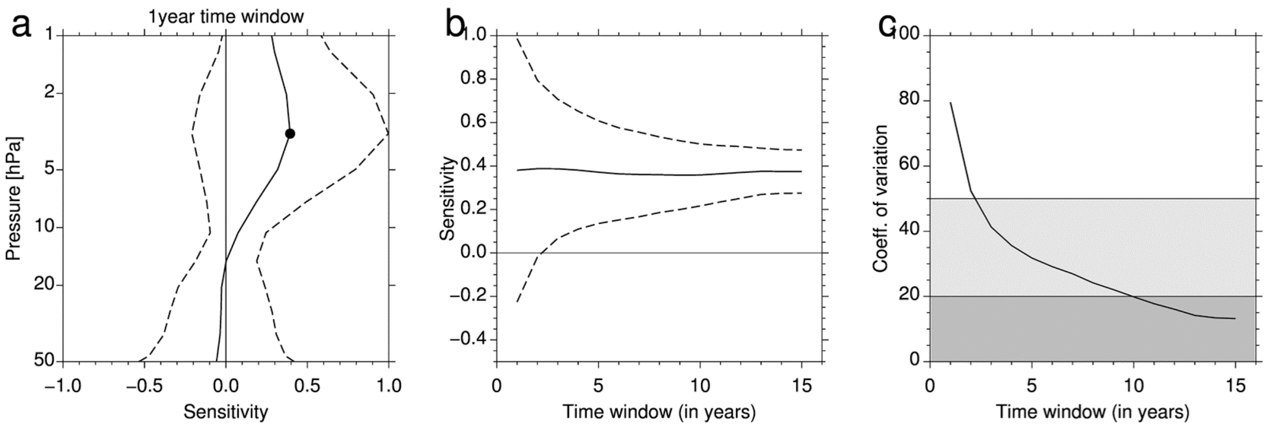


Figure 11~~0~~: (a) CCM mean ozone sensitivity profile over the 1991-2005 period computed for a 1-year time window (see text for details on calculations). (b) Mean ozone sensitivity at 3 hPa (dot on (a)) as a function of the size of the time window. The dashed lines on (a) and (b) represent the 2σ spread. (c) Coefficient of variation (in %) of the ozone sensitivity as a function of the size of the time window. Intervals with values lower than 50% and 20% are highlighted by the gray shaded areas.