Answers to comments by anonymous referees to manuscript acp-2020-273: Jia et al., Is there a direct solar proton impact on lower stratospheric ozone?

We would like to thank all the anonymous referees for dedicating their time to the comments and the discussion. We have made our conclusions much more carefully accordingly. Some further analysis and discussion were added as well. Please find our answers and responses to the comments below.

Anonymous referee #1

Specific comments:
Title and page 1, line 11: on lower stratospheric ozone depletion. I would say that "lower stratosphere" would mean tropopause (8-12 km) to about 20-25 km. However lower stratosphere is not a well-defined term, so just clarify in this sentence which altitudes you focus on (10-30 km?)

The lower stratospheric ozone is clarified as altitudes 10-30 km now.

Page 2, line 12 "most advanced climate models are now including EPP forcing" better change this to "many advanced chemistry-climate models : : :" because a) I doubt that this is really done by "most" models, and b) you need atmospheric chemistry to include EPP forcing. Climate models include atmospheric dynamics and ocean coupling, but not necessarily interactive atmospheric chemistry. The term is either chemistry-climate model or composition-climate model to clarify that you need atmospheric chemistry as well.

Modified. Thank you.

Page 2, line 14: please provide a reference for 10% alpha

The specific number of alpha in SPE can be found, for example, in the book 'Health Physics in the 21st Century' published in 2008, by Joseph John Bevelacqua. This number is irrelevant to the paper, we have deleted this information instead of adding a reference.

Page 2, line 15 and 16: . . . tens to hundreds of MeV . . . at altitudes of 35âAT90 km.... I think what you mean is that solar proton events affect the atmosphere mainly˘ in the altitude region of 35-90 km, but what you say is that protons and alpha of 10-1000 MeV mainly deposit their energy in 35-90 km. This is not correct. Protons with energies of 10 MeV release most of their energy around 70 km, protons with energies of 100 MeV release their energy in 30-40 km, protons with energies of 1000 MeV release their energy below 20 km (Turunen et al., 2009, Fig 3; Wissing and Kallenrode, 2009, Fig. 2). Soft protons and electrons may also contribute to affect altitudes above 70 km, and the fluxes of protons larger than 100 MeV are low in many solar proton events, though events with a very hard spectrum, with large fluxes of > 100 MeV protons, exist. One example is the very strong ground-level enhancement of January 2005 (e.g., Jackman et al., 2011, see also Table 1 in Gopalswamy et al., 2005). Please be more precise.

The sentence has been modified to 'Such high-energy particles mainly affect the atmosphere at altitudes of 35-90 km, …'

Page 2, Line 25 "large events", and line 27 "very extreme events", please specify what you mean by those terms. Presumably fluxes of protons at, or larger than, some specified energy range.

The terms are specified now.

Page 3, line 22: 300 MeV protons mostly affect the altitude range around 25 km (Turunen et al., 2009; Wissing and Kallenrode, 2009). As 300 MeV is the upper limit of proton energies considered in your model runs, you can therefore only investigate the impact of direct proton forcing at altitudes above ~25 km. As you don't implement proton energies able to reach altitudes below 25 km, you can not make any statements on the impact of proton ionization on altitudes below 25 km based on these model experiments. You could presumably use the model experiments to investigate possible dynamical feedbacks onto the lower stratosphere below 25 km to proton ionization above this altitude though. Page 3, line 27-30: . . . protons > 300 MeV are not included .. as the contribution of > 300 MeV protons to direct ozone loss . . . would likely be neg- ligible due to the small fluxes at such high energies . . . to summarize: you don't include those proton energies because they likely have no impact, do model experiments without those proton energies, analyse the model experiments, and conclude that there is no impact in those altitudes? That is circular reasoning. See my comment above: you can not draw any conclusions of a direct impact of proton ionization on altitudes below 25 km on the basis of these model experiments.

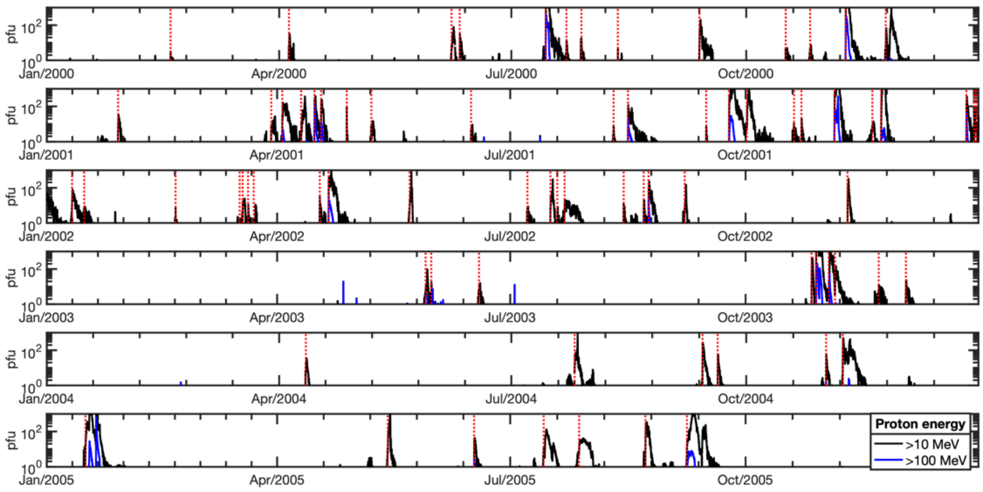
Agreed. The model certainly cannot detect proton caused changes below 25 km without sufficient particle input. We have stressed the statement more carefully in sect. 2.2, so that the readers are aware of this limitation: The sentence "We also stress that protons >300 MeV are not included in the simulation, as the contribution of >300 MeV protons to direct ozone loss in the lower stratosphere would likely be negligible due to the relatively small fluxes at such high energies (Jackman et al., 2011)" is modified to "We also stress that protons >300 MeV are not included in the simulation. 300 MeV protons mostly affect the atmosphere at around 25 km (Turunen et al., 2009; Wissing and Kallenrode, 2009). As 300 MeV is the upper limit of proton energies considered in our model simulation, the WACCM-D simulation presented here can therefore only investigate the impact of direct proton forcing at altitudes above ~25 km".

Page 4, Figure 1: the figure caption states that ionization rates used for this figure are derived at 1 hPa, that is, about 45 km – around the stratopause. If you want to investigate the impact of those events on the lower stratosphere below 30 km altitude, this is not a very useful quantity. Ionization rates at 10 hPa (~30 km) or even lower would be much more relevant here. I would suggest that you either exchange this figure with 10 hPa, or show both 10 hPa and 1 hPa.

Agreed. We have kept the original figure as the upper panel, and added the ionization rate at ~12 hPa (pressure level in ionization rate data) as the lower panel in Figure 1. The annotation is adjusted accordingly.

Page 4, line 20, and page 3, line 35: the SPE onset time is defined as the time when 5-min average proton fluxes with energies > 10 MeV are greater than 10 pfu. Why base the analysis on protons of comparably low energies (10 MeV protons mainly affect ~70 km altitudes, see above) if you want to analyse the impact on the lower stratosphere below 30 km? > 100 MeV would be more relevant. Even if you argue that you do not want to exclude soft-spectrum SPEs with large fluxes, the onset time of the event may vary for different energies.

We agree that using > 100 MeV proton fluxes could be more accurate and relevant for our study. The SPE onset was chosen in this way to keep consistency with Denton et al. The results from individual SPE studies also showed that our conclusion will not be influenced by changing the definition of SPE onset. The figure below shows the GOES pfu of >10 MeV protons (black line) and > 100 MeV protons (blue line) respectively, while the red line represents our current SPE onsets. The timing of the events does not change significantly. We argue that defining SPE onset using 10 MeV protons or 100 MeV protons is not very critical.



Support figure: GOES proton fluxes example with energy threshold of 10 MeV and 100 MeV in 2000-2005.

Page 5, line 13-14: the spatial distribution of events is similar in summer and winter, but the amplitudes are larger during winter. This may also be a purely statistical effect due to the much lower number of events (19 compared to 49), as outliers have a larger impact in small sample sizes.

Indeed. We address the statistical effect in the manuscript now.

Page 5, line 24: "the signatures above and below are not related to the epoch time" what you actually see is that the signatures already appear a considerable time before the event. So you argue that they are not related to the event. This is not necessarily true. Solar proton events are not completely isolated events. There often are series of solar proton events separated by a few (up to 27) days, as clearly seen, e.g., in Figs. 3, A2 and A3. If the first event in a series is strong, then the superposed epoch gives a response before the event. You can clearly see this in the right panel of Figure 2 at 60-70 km. Solar proton events are also often preceded by strong flares which may or may not have an impact on the atmosphere, and occur during periods of strong geomagnetic activity, with geomagnetic storms before, during or after the event. You can't exclude a significant response solely on the basis of the timing alone.

Agreed. As is suggested by reviewer 2, we have now re-calculated the statistical response by keeping the first event only, when several events happened within 10 days. This will partly exclude the possible response before epoch time from previous SPEs in a nearby time frame. Nevertheless, *"the signatures above and below are not related to the epoch time"* is removed from the manuscript.

Page 6, line 3-4: However, there was no robust ozone loss below 30 km in the WACCM- D simulations; considering the limit of proton energies in the model experiments, one would not expect a direct impact of proton ionization in these model results below 25 km. However, the WACCM-D simulations could be used to analyse the observed response of MLS ozone in a more rigorous way, by doing the analysis of WACCM- D ozone with exactly the same sampling as done in MLS data – that is using the same number of events, and the same time-period for the baseline annual cycle. As WACCM-D runs are carried out in the specified dynamics mode, any dynamical variations of ozone including ozone hole chemistry, should be reproduced by the model very well, but any direct proton impacts below 25 km would not be reproduced at all, so significant differences between model and MLS response might indicate a direct proton impact. However, if results are very similar, this would indicate no significant (on average) proton impact. This would provide a more rigorous test also than comparing the individual events in the Appendix figures, and I urge the authors to do such an analysis.

This is a great idea. When re-analyzing ozone's responses to SPE in Figs 3 and 4, WACCM-D profiles were output at Aura Microwave Limb Sounder (MLS) observation times and locations. Climatology from MLS and WACCM-D are calculated from the same time period (2nd August 2004 – 31th December 2012). We provide a comparison result between MLS and WACCM-D for the individual case study in January 2005 as the new Fig.5 in the manuscript, the discussion is added accordingly.

We keep analyzing statistical response result from MLS using all possible SPEs for statistical reasons in the manuscript. A similar epoch analysis using MLS and WACCM-D simulation at MLS observation time and location during their overlapping period, as is suggested by the reviewer, is added to the supplement as Fig. S1.

We would like to point out an inconsistency between ozone from MLS and the used model below 30 km. We added a comparison of WACCM-D simulation at MLS time and location, and MLS daily ozone anomalies in the polar cap in the supplement (Fig. S4). WACCM-D model overestimates northern polar cap ozone by 10% to > 20% below 30 km in January-April. This is consistent with SD-WACCM ozone vs MLS ozone results reported in Froidevaux et al., 2019. Such difference may implicate a transport-related issue in the model (Froidevaux et al., 2019), thus, weaken our confidence of the reviewer's idea that '*significant differences between model and MLS response might indicate a direct proton impact*'.

Page 6, lines 6-7: Despite the fact that WACCM-D epoch analysis . . . around 20 km . . . it is a good idea to look at individual events, but that there is no response of WACCM-D results at 20 km is the totally wrong argument here, because WACCM-D only includes proton energies > 300 MeV. A better argument would be the low number of events, and high variability of stratospheric ozone, influenced, e.g., also by SSWs or heterogeneous chemistry on PSC surfaces, particularly during winter. Please rewrite this sentence accordingly.

Amended. We appreciate the suggestion.

Page 6, section 4 and Figures 3, 4: you select events here based on large proton fluxes > 10 MeV. However, if you really want to look at impacts on the lower stratosphere, it would make more sense to select for > 300 MeV fluxes. You could also select for ground-level events, however, based on the list provided by Gopalswamy et al 2005, this would presumably leave you with a list of 1 in the MLS time-period – January 2005, which really seems to have been exceptional (is there an update for 2005-now?).

We have ground-level data till April 2017. In the MLS time period, there is no event that is comparable with the January 2005 event. There is a smaller event in December 2012, right in between two SPE events that I used. We didn't see visible ozone abnormal that are at a 95% confidential level. January 2005 is quite exceptional and is worth to be checked more carefully in the future.

Page 6, line 17-18: if you want to compare the variation in the MLS and WACCM-D events, it would be better to use the same period – the MLS data period – for both MLS and WACCM-D. Else differences in the anomalies might also be due to differences in the background period.

We agree. The results in Fig. 3 (from MLS) and Fig. 4 (from WACCM-D) are adjusted by using data at the overlapping period, i.e., 2nd August 2004 – 31th December 2012 (Currently our WACCM-D results are till the end of 2012 only). Moreover, to make it more comparable, the WACCM-D profiles used in Fig. 4 are also changed to the WACCM-D profile outputs at MLS observation time and location, as mentioned in the response to comment on Page 6 line 3-4. Figs. 5 (i.e., Fig. 6 after paper modification), A2 and A3 are kept the same, that is to say, these results are calculated as described in the discussion paper Page 6, line 17-18. Result description in Sect. 4 are revised accordingly.

Page 7, line 6 to page 8, line 16: this analysis on the reasons for strong ozone anomalies not related to SPEs is very useful and concise. I also agree to your conclusion as stated in lines 15-16 of page 8, that these variations contribute to the robust anomalies below 30 km as seen in Fig. 2. In particular, the significant negative ozone anomaly in 10-30 km starting well before the event onset is clearly influenced strongly by the anomalously cold late winter/spring in early 2011, whose impact on lower stratospheric ozone is well documented (e.g., Sinnhuber et al., 2011). However, I think you should go one step further and redo the superposed epoch analysis excluding those events in cold winters (that is, in winter 2010/2011, 2015/2016 and 2019/2020), and also those events where an SSW occurred within the epoch period. While this would reduce the number of events, it would also reduce the background variability, and thus hopefully provide more robust results.

This is a good suggestion. We agree that the background variability will be reduced if days with unstable polar conditions are excluded (e.g., cold winters and SSWs). However, we have to consider that for a statistical ozone study, removing data using such selecting method will introduce bias to the background variation. For instance, if the removed data are not balanced from eastly/westly QBO years, bias from QBO signal will be brought in. To avoid that, we need to discuss eastly/westly QBO years separately. With the current amount of SPE events we have, it is not necessary to go to such complicated selecting criteria yet.

Page 9, lines 11-14: you should stress here that the MLS anomalies you observe are (contrary to the analysis in Jackman et al 2011) not due to seasonal changes. The changes you observe during and after the January 2005 GLE may be unique within the MLS timeseries; however, so apparently is the event itself, at least in terms of the highest energies (compare to Tab 1 in Gopalswamy et al 2005). It may be comparable in terms of the > 10 MeV fluxes compared to the Oct-Nov 2003 SPEs, but in terms of the highest energies, fluxes were apparently much larger – more than an order of magnitude in terms of the GLE intensity. So the ozone changes observed during this event below 20 km altitude might indicate that ozone losses related to SPEs in these altitudes may be possible for events with very hard spectra (GLEs). I agree with you that more research needs to be done on this before a robust conclusion can be drawn on this, but I don't think you can dismiss this on the basis that no other event shows something similar. It appears to be a fairly unique event.

Thank you for the comment. We have now stressed the contravention with Jackman et al., 2011 regarding the January 2005 event. The conclusion is modified as well to note that there is a possibility of lower stratospheric ozone's response to protons with highest energies.

Page 9, lines 36-39: I do not agree that you can draw the conclusion that "bases on our analysis . . . SPE do not cause direct lower stratospheric ozone anomalies" based on the evidence you have provided. I agree that you provided evidence that some of the significant negative ozone anomalies are not due to a direct ozone impact but to other forcings, most obviously in March 2011. However, you do not provide a similar convincing explanation for the January 2005 event, which was exceptional in containing a very hard spectrum, and thus provides the most likely candidate of an impact on the lower stratosphere from the events sampled here. This of course does not prove that such an impact exists for very hard spectra ground-level events in general, or even during this event. However, you can't just disregard it, either; there clearly is a need for further analysis on this topic. You can conclude that solar proton events with large fluxes at > 10 MeV do not necessarily provide a large impact below 30 km altitude, if they don't have a very hard spectrum with high fluxes at > 200 MeV as well. However, you can't say anything definite about hard-spectra solar proton events here because you did not explicitly test for this.

Agreed. The conclusion is modified accordingly.

Anonymous referee #2

The manuscript is a response to two previous studies by Denton et al. (2018a,b) which reported up to 10% average decrease of ozone at ~20 km following solar proton events. Applying the same method (superposed epoch analysis) on different ozone observations (MLS, Aura), the current study arrive at a different conclusion: "SPE do not cause direct lower stratospheric ozone anomalies", which is corroborated by both observed and modeled case studies. The paper is well written and logically organized. Nevertheless, the manuscript still holds the potential for improvement, both in regard to the methods applied and the subsequent discussion.

Major revisions
1) Selection of events for the superposed epoch analysis:
The superposed epoch analysis and case studies suffer from the lack of isolated SPEs. Several years have multiple SPEs occurring days apart. That implies that the period before the zero epoch time is already influenced by SPEs. Despite lower statistics, it would be more accurate to select one event, possible the first, within "the time frame". The same argument applies to the case studies, where one period could be marked with several onsets to avoid reproduction of the "same figure". Further, Figure 2 and the case studies would be more informative if the estimated ionization rates were added.

Agreed. We have now re-calculated the statistical response by keeping the first event when several events happened within a 'time frame' of 10 days. With this limitation, the selected SPE events went down from 49 to 35.

We would like to keep the case-study figures as they are. We think the figures are capable to explain the close-by SPE onset themselves. Although there is a reproduction of almost the 'same figure', the event onset is rather clear than being squeezed into one figure. It allows us to better mark particle fluxes information as well.

We agree that the ionization rates will be useful information. The ionization rates are added to Fig 2. We are able to provide the ionization rates for WACCM-D case studies in the appendix as Fig. A4. We also provided a statistical response comparison between MLS and WACCM-D during their overlapping time in the supplement, the corresponding ionization rate average is added as the right panel of new Fig. S1.

2) Ozone anomalies in respect to climatology:
Ozone anomalies are evaluated in respect to the climatology. The case studies demonstrates, as pointed out in the discussion, that the year to year dynamical variability is larger than the potential ozone impact. These conditions make it impossible to conclude that SPEs has zero impact on ozone. It is only possible to conclude that it is less than the year to year dynamical variability. It also demonstrates that the climatology is not necessarily a good reference frame to evaluate the SPE-impact. E.g. one of the strongest SPE, with onset 2012.03.07, has a strong positive ozone anomaly before the event which becomes less positive after the event. Hence it might be a reduction compared to the pre-storm values. Also, in Figure 2 (the superposed epoch analysis) single years such as January 2012 is evident as a significant positive anomaly below 40 km. Hence, I speculate if the SPE impact would be better represented as a change relative to the ~20 days preceding the event. (Alternatively, events dominated by extreme dynamical anomalies such as January 2012 should be excluded from the superposed epoch analysis.)

Using ~20 days before the event as the background instead of the climatology is an interesting point. We agree this is probably a better option if the epoch frame is a rather short period. However, for an epoch time frame of 90 days, it is quite dangerous to be used as the background to detect SPE impact, since the ozone annual variation (especially in the stratosphere) is very large. We think climatology is

the right choice for a three months epoch time frame. Other dynamical variations are larger than the potential SPE impact is a fact that cannot be avoided. Using ~20 days before the event as background could potentially induce an artificial signal caused by the natural decay of the dynamical variations as well.
For the alternative suggestion, please see our response to reviewer 1's comment on Page 7, line 6 to page 8, line 16.

3) Proton energy range in the WACCM model:
For the model runs, only protons with <300 MeV are included in the ionization rates. The respective energy range is therefore insufficient to account for the direct impact at ~20 km (e.g. Turunen et al., 2009). Without a complete energy range impacting 20 km, the discussion and the subsequent conclusion should reflect this limitation. It should also be noted that the 2005.01.16 case study are more pronounced in the observations compared to the model. This is particular true below 30 km, which might imply that the model might underestimate the ionization rates, transport or chemical processes.

Agreed. We now stress the limitation caused by proton < 300 MeV in Sect. 2.2, discussion and the conclusion section.

By accepting both reviewer 1 and 2's suggestion, we added a comparison of WACCM-D (at MLS measurement time and location) and MLS ozone anomalies in the polar cap in the supplement (Fig. S4). We point out that WACCM-D model overestimates northern polar cap ozone by 10% to > 20% below 30 km in January-April. This is consistent with SD-WACCM ozone vs MLS ozone results reported in Froidevaux et al., 2019. Such difference may implicate a transport-related issue in the model (Froidevaux et al., 2019).

4) Ozone chemistry:
Would you expect the same chemistry to impact ~20 km altitude as ~70 km? Is it still only EPP produced NOx and HOx than deplete ozone as described in the introduction, or are the chemical pathways of more complex deep into the lower stratosphere? E.g. Jackman et al. (2000) suggest that enhanced NOx values can lead to enhanced formation of the chlorine and bromine reservoir species ClONO2 and BrONO2, slowing down the 'ozone hole' formation chemistry in cold polar winters.

Yes, the chemistry is different in the lower stratosphere. We do expect an increased O_3 by increasing NO_x at this altitude. While we keep open to any kind of ozone changes (either increase or decrease) related to SPEs, we have added the statement into the manuscript in paragraph 3 of the introduction. We appreciate the comment.

Minor revisions:
1 Introduction: Define altitude range of upper and lower stratosphere Line 12: define altitude range of lower stratosphere Line 13: Define acronym when "superposed epoch analysis" are first written (Line 12) Introduction: define altitude range of upper and lower stratosphere Line 18: "at many altitudes" be more precise Line 35-37: Outline where the results from WACCM is coming
2 Data sets Line 3: remove Microwave Limb Sounder as acronym is already defined Line 1/7 page 4: add (~50 km) the first time you write ~1 hPa
References, page 14, line 4: remove hyphen

Modified accordingly. Thank you.

References

Froidevaux, L., Kinnison, D. E., Wang, R., Anderson, J., and Fuller, R. A.: Evaluation of CESM1 (WACCM) free-running and specified dynamics atmospheric composition simulations using global multispecies satellite data records, Atmos. Chem. Phys., 19, 4783–4821, https://doi.org/10.5194/acp-19-4783-2019, 2019.

Is there a direct solar proton impact on lower stratospheric ozone?

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Abstract. We investigate Arctic polar atmospheric ozone responses to Solar Proton Events (SPEs) using MLS satellite measurements (2004–now) and WACCM-D simulations (1989–2012). Special focus is on lower stratospheric (10–30km) ozone depletion that has been proposed earlier based on superposed epoch analysis (SEA) of ozonesonde anomalies (up to 10% ozone decrease at ~20 km). ~~Superposed Epoch Analysis~~ SEA of the satellite dataset provides no solid evidence of any average SPE impact on the lower stratospheric ozone, although at the mesospheric altitudes a statistically significant ozone depletion is present. In the individual case studies, we find only one potential case (January 2005) in which the lower stratospheric ozone level was significantly decreased after the SPE onset (in both model simulation and MLS observation data). However, similar decreases could not be identified in other SPEs of similar or larger magnitude. Despite the model can only detect direct proton effect above 25 km due to the input proton energy threshold 300 MeV, we find a very good overall consistency between SPE-driven ozone anomalies derived from the WACCM-D model simulations and the Aura MLS satellite data. The simulation results before the Aura MLS era indicate no significant effect on the lower stratospheric ozone either. ~~As a conclusion, the SPE has a zero direct impact on the lower stratospheric ozone.~~

1 Introduction

In the near-Earth space, solar wind charged particles are guided by the Earth's magnetic field and are able to precipitate into the middle and upper atmosphere in the polar regions. Such kind of precipitation creates the spectacular aurora, but also produces considerable amounts of HO_x (H, OH, HO_2) and NO_x (N, NO, NO_2) through ion-neutral chemistry (e.g. Verronen and Lehmann, 2013). HO_x and NO_x increases lead to ozone loss through catalytic reactions in the mesosphere and upper stratosphere, respectively (Sinnhuber et al., 2012). Moreover, in polar winter, NO_x has a long chemical lifetime due to limited photodissociation by solar radiation. NO_x produced by energetic particle precipitation (EPP) in the mesosphere-lower thermosphere is transported down to the stratosphere by the Brewer-Dobson circulation inside the polar vortex (Funke et al., 2014), causing depletion of upper stratospheric ozone (Damiani et al., 2016). A number of studies have confirmed EPP's remarkable role in ozone depletion directly during large EPP events (e.g. Funke et al., 2011) and indirectly due to descending NO_x (e.g., Randall et al., 2007). Thus, many advanced chemistry-climate models are now including EPP forcing, in order to correctly represent the ozone distribution in the polar stratosphere and mesosphere (Matthes et al., 2017; Stone et al., 2018).

Solar proton events (SPE) are one of the main types of EPP. During SPE, particles (mainly protons~~, ~10% alpha~~) with energies from tens to hundreds of MeV precipitate into the atmosphere at geomagnetic latitudes larger than 60° for days. Such high-energy particles mainly affect the atmosphere at altitudes of 35–90 km, providing direct ionization forcing on the polar middle atmosphere. Large SPEs have been studied since the 1960s until today using satellite observations and model simulation. In addition to tens of percent of ozone loss observed at altitudes above 35 km (Jackman 2001, Seppälä et al. 2004, Verronen et al. 2006), a strong SPE can reduce total ozone by 1–3% for months after the event (Jackman 2011, 2014).

Recently, Denton et al. (2018 a, b) presented statistical studies of average ozone changes from 191 SPEs between 1989–2016 using ozonesonde measurements. Superposed epoch analysis of ozone anomalies at polar stations (Sodankylä, Ny-Ålesund, and Lerwick) indicated that SPEs occurring during winter are causing ozone decrease by 5–10%, on average, at 20 km altitude. This effect is not produced in the current models because SPE-induced ionisation rates are insignificant at this altitude even during largest events with high proton energies from 300–20000 MeV (Jackman et al., 2011), and Denton et al. included also a large number of very small SPEs in their analysis. Such ozone decreases have not been observed in the case studies of very extreme (particles with energies >10MeV are greater than 10 000 particle flux units) SPEs, e.g., the 2003 'Halloween' event, from either simulation or satellite observation (Funke et al. 2011 and references therein). Recently, statistical analysis based on simulations has found no evidence of such low-altitude ozone impact (Kalakoski et al. 2020). Moreover, from the chemical aspect, we also rather expect ozone increase at lower stratosphere due to the enhanced NOx interfering with chlorine-driven catalytic ozone loss (Jackman et al., 2008).

Here we investigate the proposed SPE-induced direct depletion on lower stratospheric (10–30 km) ozone using ozone data from the Microwave Limb Sounder (MLS) instrument aboard the Aura satellite and the Whole Atmosphere Community Climate Model (WACCM-D) simulations. We proceed to evaluate ozone changes at altitudes 10–70 km caused by SPEs both statistically (superposed epoch analysis) and individually (case by case). The MLS ozone data, WACCM-D atmospheric simulation, and SPE data sets are presented in Sect. 2. In order to cross-check ozone depletion at 20 km reported based on the ozonesonde data, statistical ozone responses from MLS satellite measurements are firstly provided in Sect. 3. Following that, MLS and WACCM-D ozone changes after individual SPEs are given in Sect. 4. Finally, we summarize our results and conclusions in Sect. 5.

2 Data sets

2.1 O_3 profile measurements by MLS

~~The Microwave Limb Sounder~~ MLS onboard the Earth Observing System (EOS) Aura satellite measures ozone emission at 240 GHz, providing ozone volume mixing ratios at 55 pressure levels since 15 July 2004 (Waters et al., 2006). Vertical profiles are retrieved from the MLS observations with a 165 km horizontal spacing at altitudes between 8 and 90 km, a spatial resolution of 500 km × 500 km (along-track × across-track), and a vertical resolution of ~3.2 km. In this work, we use version 4.2 ozone data measured at 261 - 0.02 hPa (~10–70 km) to calculate the daily averaged ozone density profile at northern high latitudes (60°-90°N). Readers who are interested in the MLS data quality are referred to Livesey et al. (2018).

2.2 O_3 from WACCM-D simulations

WACCM is a global circulation model, including fully coupled dynamics and chemistry. Here, we use version 4 of the WACCM with resolution of 1.9° latitude by 2.5° longitude, with 88 vertical levels reaching from surface to 6×10^{-6} hPa (≈140 km). Overview of the model and the description of climate and variability in long-term simulation was presented by Marsh et al. (2013), with details of model physics in MLT (mesosphere - lower thermosphere region) and the response of the model to radiative and geomagnetic forcing during solar maximum and minimum described by Marsh et al. (2007). The simulation results presented here are from WACCM-D, a variant of WACCM with more detailed set of lower ionospheric chemical reactions, aimed at better reproduction of observed effects of EPP on MLT neutral composition (Verronen et al. 2016, Andersson et al. 2016).

We use SD-WACCM-D specified dynamics configuration, with Modern-Era Retrospective Analysis for Research and Applications (MERRA) (Rienecker et al., 2011) meteorological fields to force dynamics at every time step up to about 50 km. Simulation covers years 1989-2012, and uses forcings from auroral electrons (E<10keV), solar protons (< 300 MeV), and galactic cosmic rays for energetic particle precipitation. The SPE ionization rates are based on proton flux measurements from the Geostationary Operational Environmental Satellites (GOES) (see e.g. Jackman et al., 2011, for the calculation method). The WACCM-D SPE effects on neutral species are compared to satellite observations in Andersson et al (2016). Note that WACCM-D has not been validated below 20 km. Nevertheless, in Andersson et al (2016) the HNO_3 response above 15 km to single SPE onset was reasonable compared to MLS data. We also stress that protons with energy over 300 MeV are not included in the simulation. 300 MeV protons mostly affect the atmosphere at around 25 km (Turunen et al., 2009; Wissing and Kallenrode, 2009). As 300 MeV is the upper limit of the proton energies considered in our model simulation, the WACCM-D simulation presented here can therefore only investigate the impact of direct proton forcing at altitudes above 25 km. For more details of the simulation setup, see Kalakoski et al. (2020).

2.3 Solar proton events

The data of solar proton events (SPEs) used in this study is based on NOAA GOES proton flux observations. Fig. 1 presents 261 SPEs recorded from 1975 to date, including their onset time, fluxes detected in space, approximated time of duration, and average ionization rates to the atmosphere at two altitudes. Here, the onset of a SPE is defined as the time when 5-min average proton fluxes with energies >10MeV are greater than 10 Particle Flux Units (1 pfu = 1 particle /cm^2/s/sr) at the geosynchronous orbit. For the estimation of SPE duration and its impact on the atmosphere, we use the daily average ion pair production rates at ~1 hPa (~46 km, upper panel) and ~12 hPa (~29 km, lower panel). These ionization rates are calculated from GOES proton flux observations using the energy deposition methodology described in, e.g., Jackman et al. (2011). The SPE durations presented here were calculated as the period when the ionization rates at ~1 hPa / 12 hPa are larger than 2 ion pair/cm^3/s before the next event happens. The ionization rates to the atmosphere were then the average ionization rates at 1 hPa / 12 hPa during this period. Our study used 49 events that occurred after the launch of Aura MLS (July 2004–now) and 177 events that occurred in WACCM-D simulation period (Jan 1989–Dec 2012) to evaluate the ozone changes following SPEs. It is clearly demonstrated in Fig. 1 that these SPEs are more frequent near solar maximum years. Majority of the events are with flux less than 400 pfu, and their impacts to the atmosphere below 1 hPa are small. It is worth to mention that although these SPEs seem to have no preference in occurring season, their seasonal distribution varies by months and should be considered during the interpretation (Fig. A1).

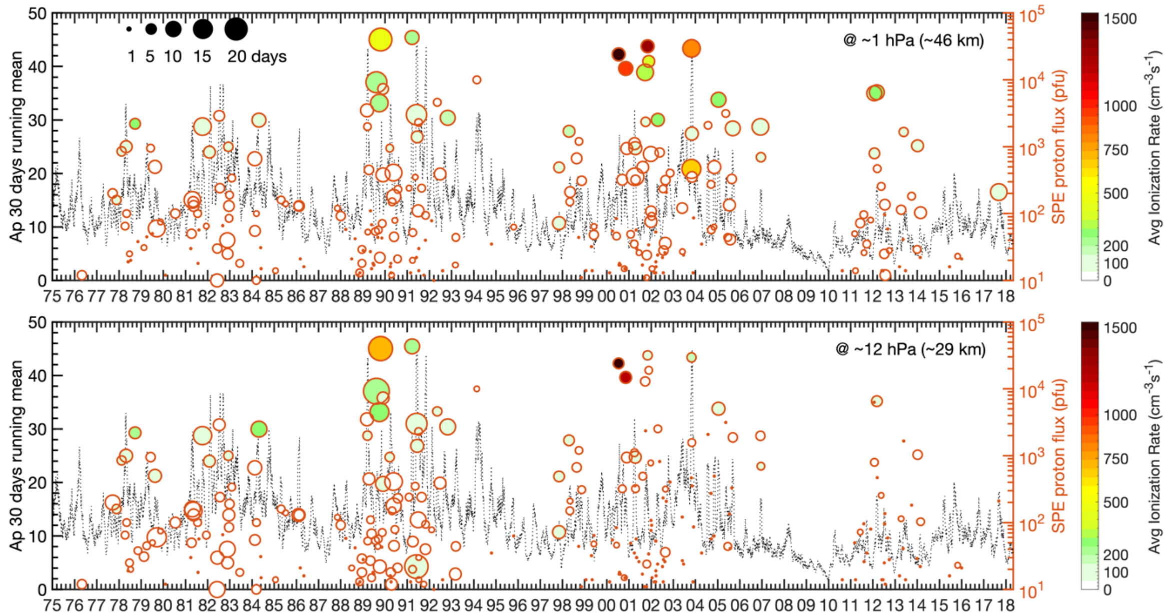

Figure 1. Onset time of SPEs and their proton fluxes since 1975. The filled colors are the average ionization rate during each SPE at ~1hPa (upper panel) and ~12 hPa (lower panel), while the size of the markers represents the approximate duration time of the SPEs obtained from the daily mean ionization rate at the two altitudes. The black dotted line in the background is the 30-day mean of the daily geomagnetic activity Ap-index.

3 Statistical O_3 response from MLS

Similar to the method used by Denton et al. (2018 b), we applied a superposed epoch analysis to the MLS daily ozone anomalies. The superposed epoch analysis, also referred to as compositing in geophysics, is used to acquire variation of a time series before and after an event or a chain of certain kind of events. The point of time when the event begins is the epoch time. In this case, the epoch times are the onset times of individual SPEs during MLS operating period. All available ozone data were binned as a function of epoch time and altitude, with temporal resolution of one day. The pre- and post-epoch spans used here are 30 and 60 days, respectively. For the selected sets of SPEs, all the binned ozone data sets were averaged to represent the effect of the SPEs. This method excludes natural ozone variations that are larger than the span-scale. Since SPE-driven effects are expected to take place on daily to monthly time scales, variations caused by e.g. QBO can be excluded. However, seasonal variations must be excluded before using superposed epochs. Thus, the daily profile climatology calculated from the ozone data was subtracted from the daily ozone data. Different from Denton. et al., to make sure SPEs are 'isolated' from the previous events, events that happened within 10 days of the previous SPE were excluded.

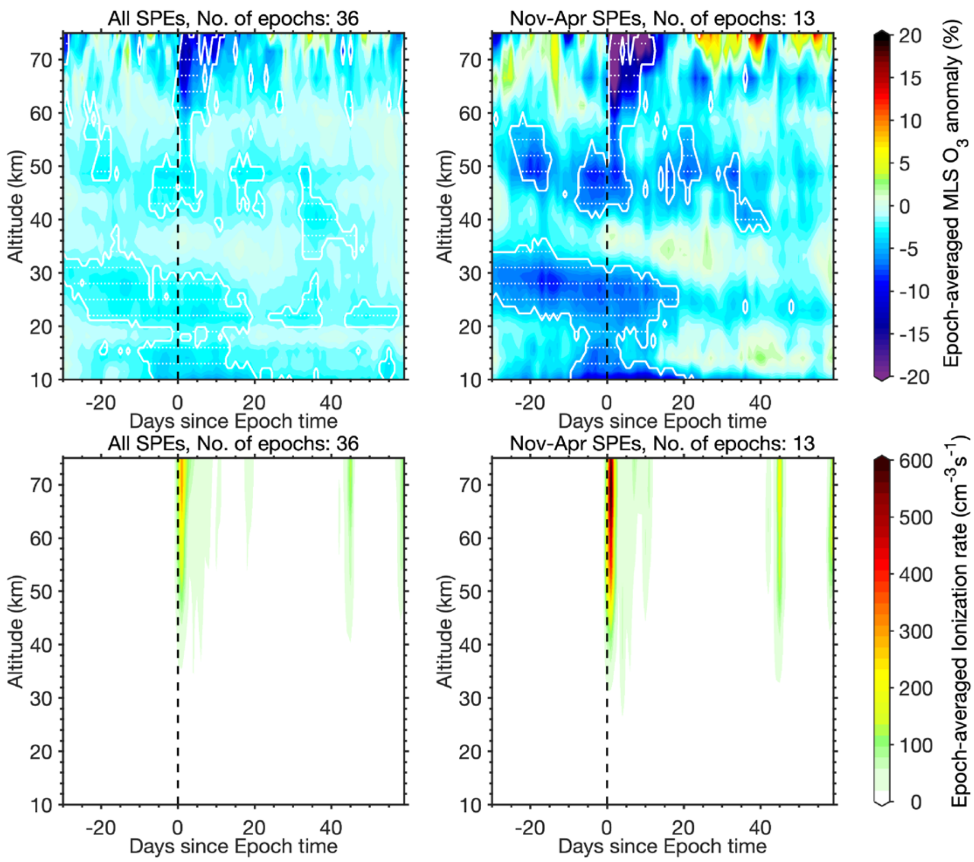

Figure 2. Epoch-averaged MLS ozone anomalies (relative in %) (upper panels) and the corresponding daily ionization rates (lower panels) in the northern polar region (60°-90°N) along with geopotential altitude for a total of 35 'isolated' SPE epochs (left panel) and 13 'isolated' winter SPE epochs (right panel). The black dashed line represents the epoch time, i.e., onset of SPEs. The white thick line area corresponds to the epoch-averaged anomalies with >95% confidence after the Monte Carlo test.

In order to test the statistical significance of the obtained results, a Monte Carlo test was implemented. Instead of using SPE onset as epoch times, the analysis was rerun using 2000 random sets of epoch times. SPE-epoch averaged variations larger than 95% of the 2000 randomized results are considered significant and robust (reported as >95% confidence), suggesting that these extracted signatures are likely related to SPE.

Fig. 2 shows the superposed epoch of MLS northern polar ozone anomalies and the corresponding daily ionization rates for all 'isolated' SPEs (35 out of 49 events, left panels) and for the ones occurred in winter (Nov-Apr) (right panels) within the instrument's operational period. Robust averaged anomalies (>95% confidence) are presented within the white thick lines. Spatial distribution of statistically robust anomalies is similar in all-SPE epochs and winter-SPE epochs. The depletion is more pronounced for winter epochs. This, of course, could be a statistical effect due to the much lower number of events used in the study, but is also expected due to two facts: 1) ozone recovery is slower due to less production from O_2 photodissociation; 2) Largest SPEs with flux >1000 pfu that cause more ozone depletion happen to occur in NH winter. Among all the SPEs during MLS measurement period, ~3/4 of big SPEs are in NH wintertime (see Fig. A1). In both upper panels, closely following the SPE onset, very pronounced ozone depletion appears above 50 km for over 5 days. This is the direct ozone loss caused by the SPE-induced HO_x enhancement. The number of extreme SPEs is relatively small, which explains the absence of the long-lasting ozone depletion that would be expected between 40–50 km from enhanced amounts of NO_x. While the upper stratospheric ozone depletion signature is not seen in the statistical average, 5-10% decrease of ozone is present below 30 km, including ozone loss around 20 km similar to that reported by Denton et al. However, we cannot exclude the possibility that the whole robust variation in the stratosphere is more related to other phenomena in the northern polar cap, e.g. to changes in the strength of polar vortex or related chemical effects. We will discuss this in more detail in Sect. 4.

A superposed epoch analysis of WACCM-D ozone anomalies from SPEs during 1989–2012 has been reported by Kalakoski et al. (2020), thus we will not repeat it here. In their results, the epoch-averaged WACCM-D ozone anomalies showed the same robust depletion at above 50 km. Since their analysis included also the very large SPEs that occurred 1989–2004 (see Fig. 1 in this study, or list of largest 15 SPEs in Tab. 1, Jackman et al., 2008), long-term ozone depletion in the upper stratosphere was clearly detected as well. However, there was no robust ozone loss below 30 km found in the WACCM-D simulations.

4 O_3 response to individual SPEs

Considering the limited number of SPE events during MLS era, and the high variability of stratospheric ozone, influenced, e.g. by SSWs or heterogeneous chemistry on PSC surfaces, particularly during winter, in this section we analyse ozone responses to individual SPEs.

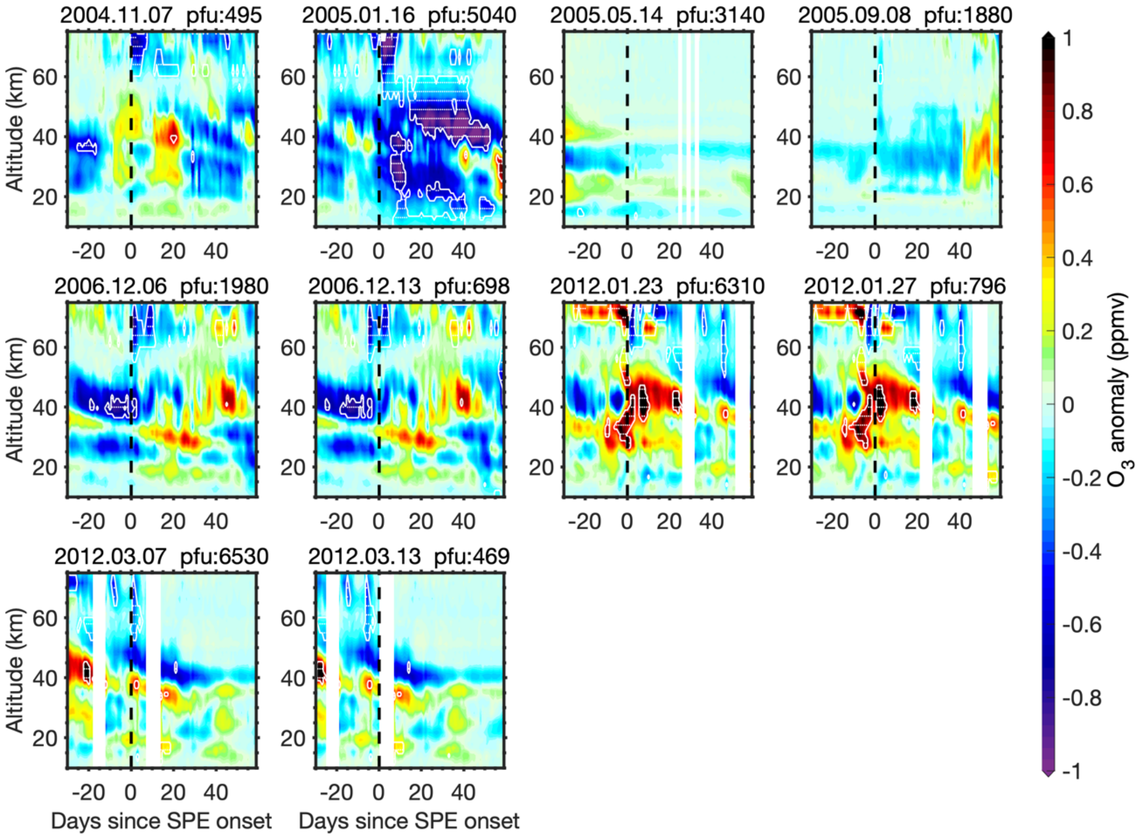

Figure 3. MLS ozone anomalies (in ppmv) along with altitude at 30 days before and 60 days after individual big SPEs (proton fluxes >400 pfu) in July 2004–December 2012. The white thick line area demonstrates ozone anomalies with >95% confidence after the Monte Carlo test.

Similar to the analysis presented in Sect. 3, ozone anomalies presented here were calculated by subtracting daily climatology from daily averaged ozone data from MLS and WACCM-D. For Figs 3 and 4, to make the results from MLS and WACCM-D simulation comparable, WACCM-D daily ozone was calculated using simulation profiles at MLS observation time and location. The climatology from MLS and WACCM-D were derived from their overlapping time period to guarantee a comparable background. For Figs. 6, A2 and A3, the subtracted daily mean climatology from MLS and WACCM-D were derived from the MLS data period and the WACCM-D simulation period, respectively. Then, instead of applying superposed epoch analysis on multiple SPEs, ozone anomalies are presented 30 days before and 60 days after onset of individual SPE. For estimating the statistical significance of the ozone anomalies found in the individual SPEs, we applied a similar Monte Carlo approach as in the case of SEA, i.e., the variance of 6000 random ‘onset’ times was used as a measure for a significant anomaly. It is worth noting, however, that this method recognizes all ‘statistically significant’ anomalies larger than the random background variation, whether the anomaly is due to SPE or, for instance, due to exceptional dynamical/chemical anomalies, which have a similar occurrence probability as SPEs.

Anomalies following all individual SPEs can be found in Figs. A2 and A3. In general, SPEs with proton fluxes < 400 pfu cause neither visible daily ozone depletion in the mesosphere (below 75 km), nor in other altitudes. Ozone changes following individual SPEs are more pronounced during winter. Figs. 3 and 4 demonstrate MLS and WACCM-D ozone variations following SPEs with proton fluxes > 400 pfu in July 2004 – end of 2012. Both the ozone variations and the robust signatures from these two different data sets are very consistent. After 2004, three large winter SPEs, i.e., January 2005, September 2005 and March 2012, produced clear upper stratospheric ozone loss. Ozone depletion is most pronounced following the January 2005 event. For this event, we also observe a robust lower stratospheric ozone loss from MLS following SPE for the first time: ozone is depleted by ~1 ppmv (~15%) at 20–35 km and by ~0.15 ppmv (>20%) below 15km 5 days after SPE onset.

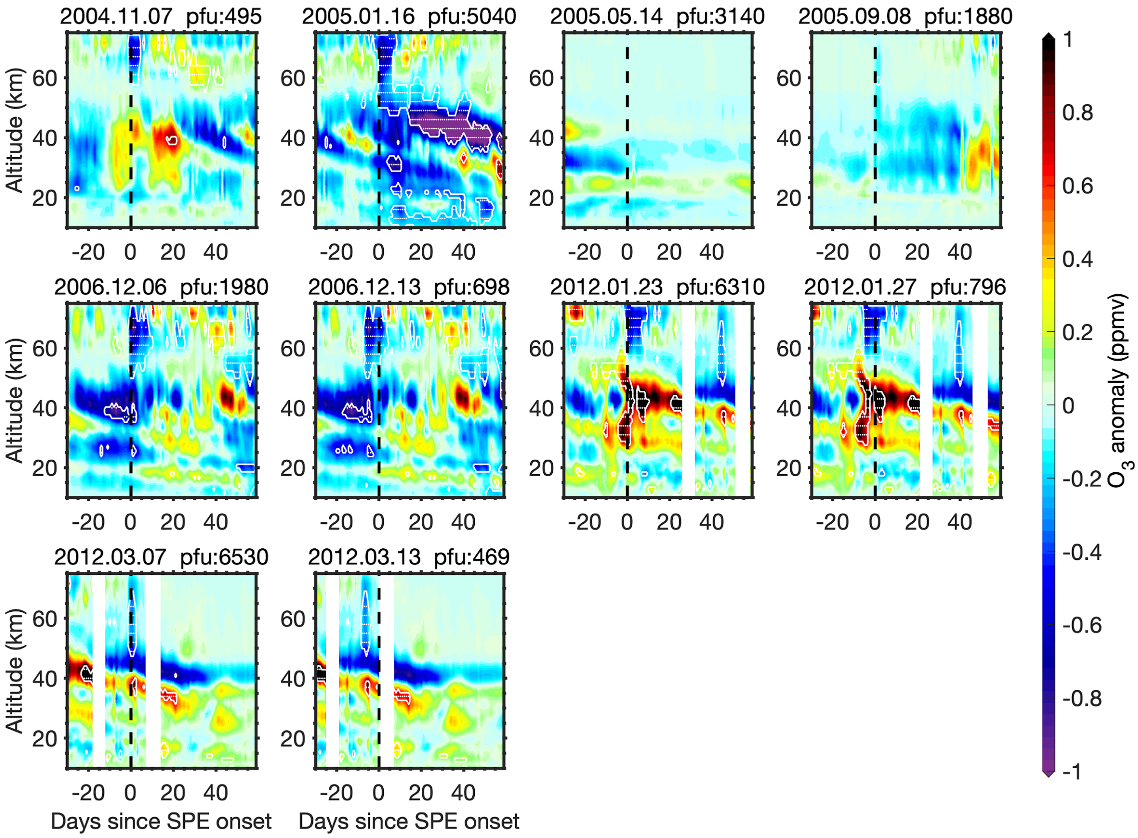

Figure 4. Same as Fig. 3 but for ozone anomalies from WACCM-D simulation at MLS measurement time and location.

Overall, wintertime ozone variation below 35 km is rather complicated. Year-to-year variability of stratospheric polar ozone is mostly controlled by dynamical and chemical processes, both are essentially coupled to temperature changes. Factors that modify polar temperature, e.g., sudden stratospheric warming (SSW) and El Niño–Southern Oscillation (ENSO), are essentially planetary wave perturbations that modulate the strength of polar vortex. The probabilities of major SSWs and, on the other hand, springs with extremely strong polar vortex are at similar levels as the one of SPEs. Thus, ozone variations by these events will be seen as robust signatures in our study as well, yet they do not necessarily coincide with onsets of SPEs with proton fluxes >400 pfu and >10000 pfu, respectively. The large SPE in January 2012 (Figs. 3 and 4) is severe enough to destroy stratospheric ozone. However, the stratospheric ozone anomalies at that time were dominated by dynamical ozone enhancement from SSW in 17th Jan 2012 (Päivärinta et al., 2016). One of the most pronounced examples of extreme strong polar vortex impact is the well-reported ozone depletion during spring of 2011, which can be observed in ozone anomalies around the two small SPEs that occurred in March 2011 (see Fig. A2). The lower stratospheric polar vortex was the strongest (in either hemisphere) in the previous 32 years (Manney et al., 2011). Large volume of polar stratospheric clouds (PSCs) converted chlorine reservoirs to ozone-destroying species, leading to extraordinary low ozone in the stratosphere (Pommereau et al., 2018). Similarly, robust anomaly seen after January 2016 SPE can be explained by cold 2015–2016 winter anomaly. We are confident to exclude SPE's influence on the anomaly in both cases because: firstly, the signal is not following SPE onset, secondly these SPEs are such small events that ozone loss was not observed, not even in the mesosphere. These robust non-SPE signals are included in the superposed-epoch analysis performed in Sect. 3, contributing to the robust anomalies below 30 km in Fig. 2.

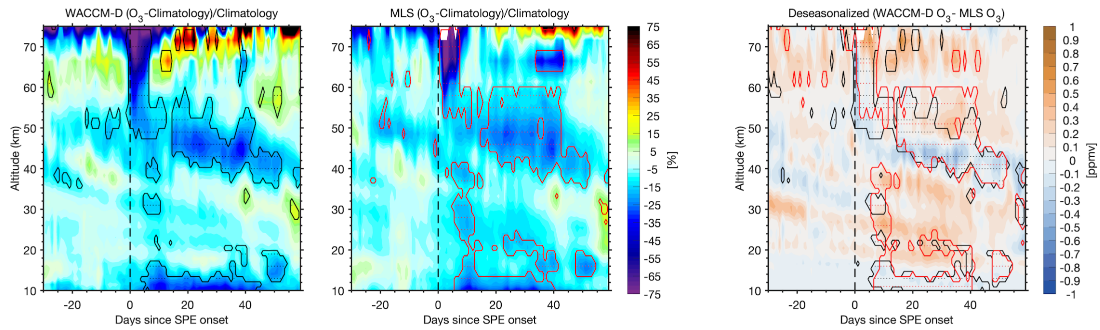

Figure 5. WACCM-D (left panel) and MLS (middle panel) relative ozone anomalies along with altitude at 30 days before and 60 days after SPE on 16th January 2005. The WACCM-D simulation used here are the profiles at MLS measurement time and locations. The climatology is calculated using data between July 2004 – December 2012 for both MLS and WACCM-D. The black/red thick line area demonstrates relative ozone anomalies with >95% confidence after the Monte Carlo test. Right panel is ozone differences between WACCM-D and MLS during this time frame (deseasonalize means that the seasonal difference showed in Fig. S4 is removed). The black/red thick line area demonstrates direct ozone anomalies with >95% confidence after the Monte Carlo test from WACCM-D and MLS data, respectively.

Identify sources of the robust ozone anomaly below 35 km following the SPE beginning on 16th Jan 2005 is difficult. With a moderate cold winter temperature causing more ozone loss, coincident of robust dynamical ozone changes following the SPE exists. Meanwhile, an extremely large (over 270%) ground level enhancement (GLE) of neutrons occurred during the SPE period on 20 January 2005 (Jackman et al., 2011). Ionization rate reached 500 cm^{-3}s^{-1} at 30 km for one day due to the very high energy protons (300-20 000 MeV) that caused the GLE (Usoskin et al., 2011). Jackman et al. (2011) carried out a detailed study of January 2005 SPE's influence on the northern polar atmosphere using WACCM3 simulation, and reported an ozone column decrease of less than 0.01% by GLE protons, while the ozone changes below 50 km observed in MLS data are seasonal changes. The MLS ozone anomalies we observe are, on contrary to the analysis in Jackman et al. 2011, not due to seasonal changes. To identify whether the anomalies are due to direct SPE effect or not, relative ozone response from MLS and WACCM-D simulation in MLS observation time and location to 16th Jan 2005 SPE are compared in Fig. 5. As WACCM-D simulation are carried out in the specified dynamics mode, any dynamical variations of ozone including ozone chemistry, are expected to be reproduced by the model well. But any direct proton impacts below

25 km would not be reproduced at all since protons with energy > 300 MeV are not included in the model input. So significant differences between model and MLS response might indicate a direct proton impact. As shown in Fig. 5, ozone responses below 20 km are very similar between results derived from these two data sources, indicating no significant proton effect. We do see some difference between 20–30 km, which might demonstrate a possible direct proton effect. However, we would like to point out that compared to MLS, WACCM-D holds an > 20% overestimation of northern polar cap ozone below 30 km in January–April (see right panel of Fig.5, Fig. S4 in the supplement, and Fig. 1 in Froidevaux et al., 2019). Such differences may implicate a transport-related issue in the model (Froidevaux et al., 2019), therefore weaken our confidence to confirm the robust signal difference between MLS and WACCM-D at 20–30 km as the evidence of direct SPE impact. Readers who are interested in the ionization rate of this case is referred to Fig. A4.

Nonetheless, in our study the robust MLS ozone destruction signature in the lower stratosphere following the January 2005 SPE is extremely unique, not only when compared to other SPEs cases after 2004, but also when large and extreme SPEs before 2004 are included (see the WACCM-D simulation result presented in Fig. 6). Further research needs to be done to confirm the dynamical/chemical factors that led to ozone destruction below 35 km in January 2005.

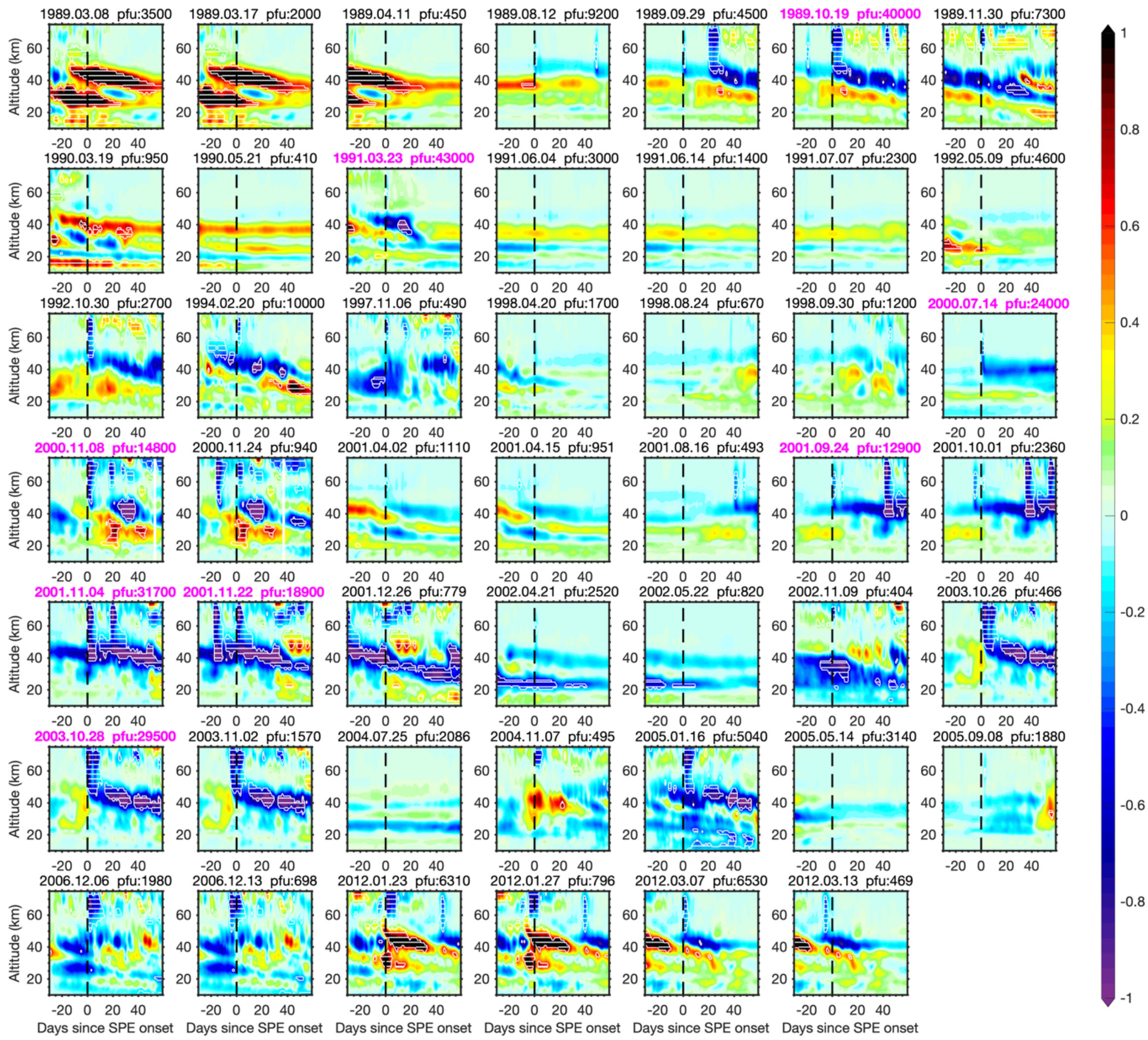

Figure 6. Same as Fig. 4 but for all simulated WACCM-D ozone anomalies (not only collocated with MLS measurement) before and after individual big SPEs (proton fluxes >400 pfu) since 1989. Extreme SPEs (proton fluxes >10000 pfu) are marked with bold magenta titles.

5 Conclusions

Recent studies have reported up to 10% average decrease of lower stratospheric ozone at 20 km altitude following solar proton events (SPE). However, mechanisms which could cause such a large low-altitude impact are not clear. We used the Aura MLS satellite ozone datasets from 2004 to date and WACCM-D model simulations from 1989–2012 to analyse SPE-driven ozone changes. In our approach, stratospheric and mesospheric daily ozone anomalies (10–70 km) were examined over the epochs of SPEs by applying 1) a Superposed Epoch Analysis (SEA) for all the cases and 2) a case-by-case analysis for individual events. Statistical significance of the anomalies found in the ozone levels was estimated by employing a Monte Carlo approach.

Arctic polar ozone destruction in the mesosphere and upper stratosphere can be directly observed from satellite measurement anomaly, when following SPEs in September–April with proton fluxes >400 pfu and >1000 pfu, respectively. We observe 5–10% ozone destruction below 30 km altitude in MLS SEA results. However, the depletion appears before the epoch time, i.e. SPE onset. We argue that such lower stratospheric ozone losses are rather caused by unusually stable and strong polar vortex, together with sufficient ozone depleting reservoirs of chlorine. In the case by case study, we find a very good overall consistency between SPE-driven ozone anomalies derived from the WACCM-D model simulations and the Aura MLS data. Despite the model can only detect direct proton effect above 25 km due to the input proton energy threshold 300 MeV, the good consistency enables us to generalise the study also to the SPEs before the Aura MLS era. From 1989 to date, robust lower stratospheric ozone decrease after SPEs was observed only once in ozone anomaly, i.e. following the January 2005 SPE. Ozone was depleted by ~1 ppmv (~15%) at 20–35 km and by ~0.15 ppmv (>20%) below 15km 5 days after SPE onset. We further investigated this case by comparing WACCM-D and MLS data. Since WACCM-D is not expected to observe direct SPE impact below 25km, a consistent ozone depletion below 15 km demonstrated that direct SPE impact is less likely to be the reason for this robust ozone loss. The source of ozone loss over 20km, however, is not fully confirmed. We state that the exact mechanisms of the suggested lower stratosphere impact are currently unclear. The simulation results indicate that even for the strongest SPEs in our record, there is no significant effect on the lower stratospheric ozone as such.

Although it remains unclear to what degree the lower ozone decrease in January 2005 was caused by the SPE, and how much due to natural variability, we suspect that the observed, statistically significant lower stratospheric ozone impact is most likely by chance coincident with the SPE onset. We encourage further research on January 2005 SPE case to solidly confirm the EPP/dynamical/chemical factors that led to ozone destruction below 35 km.

Appendix

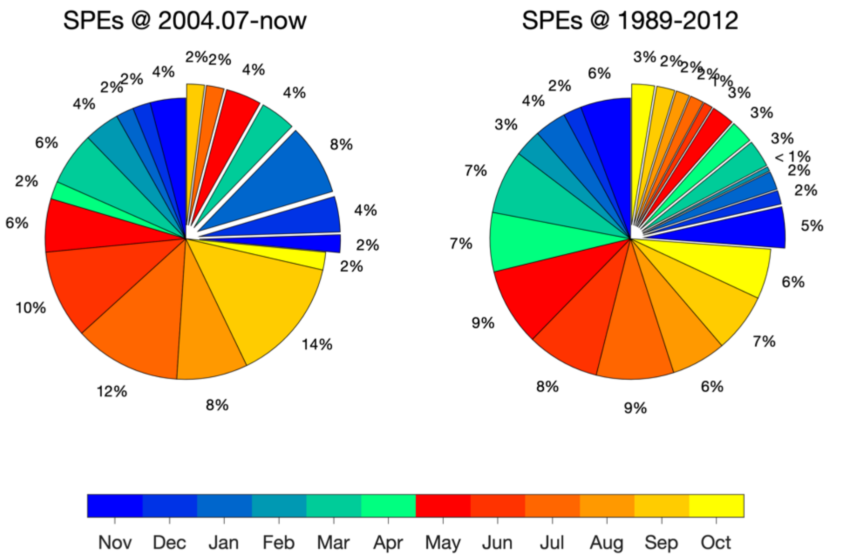

Figure A1. SPE's seasonal distribution for those with fluxes >400 pfu (exploded parts of the pie chart) and the ones with fluxes <400 pfu (regular parts of the pie chart). Left panel demonstrates the cases in between MLS measurement period (2004.07– now). Right panel shows the cases during WACCM-D simulation (1989–2012).

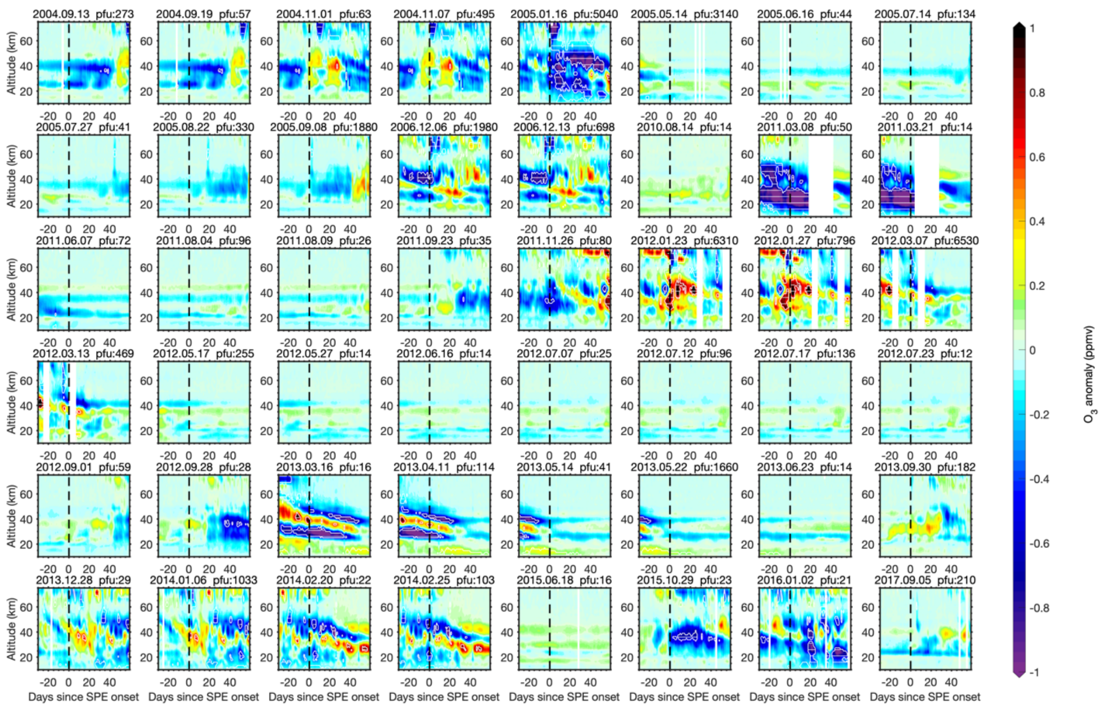

Figure A2. Same as Fig. 3 but after all individual SPEs since July 2004.

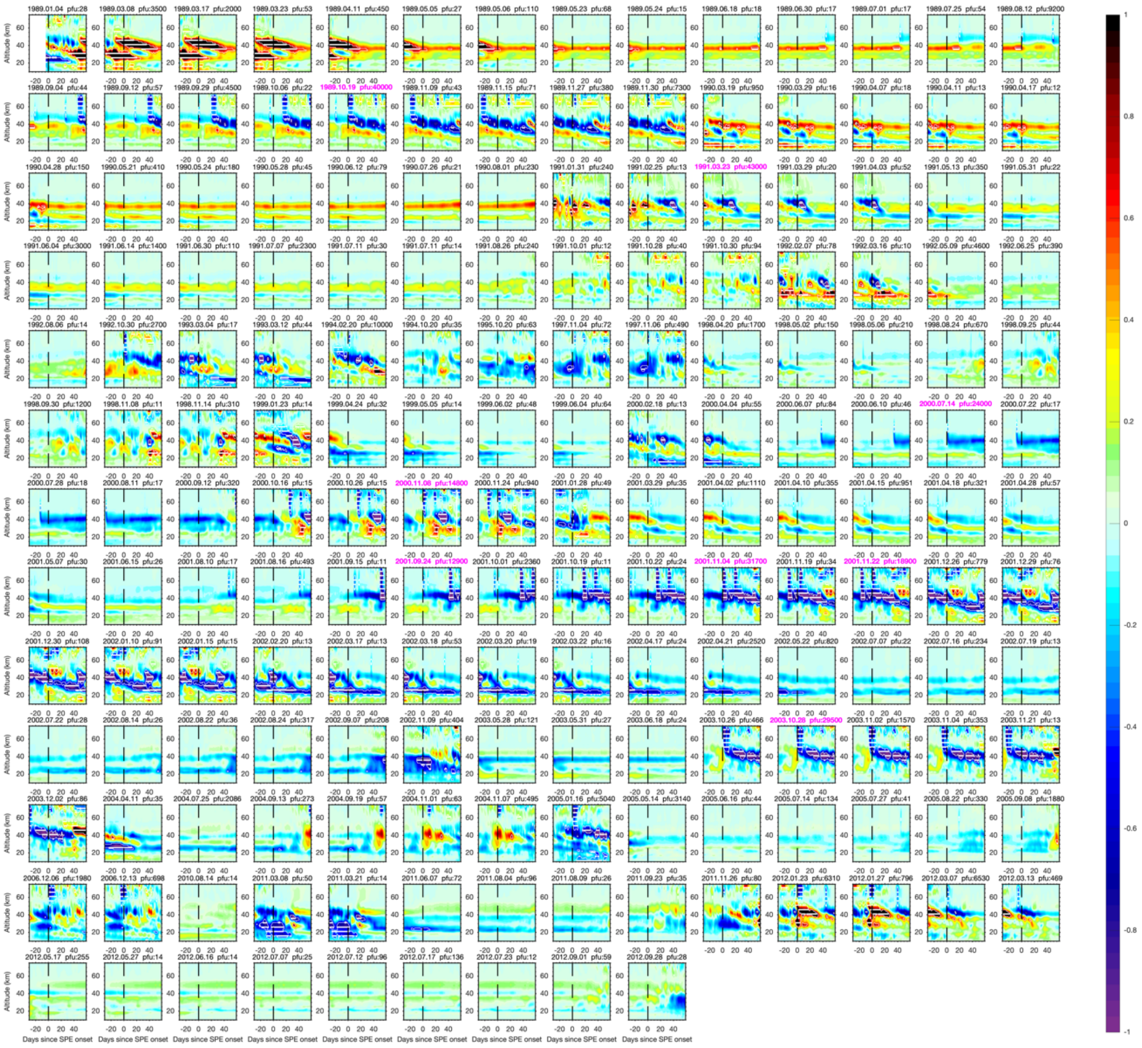

Figure A3. Same as Fig. 4 but after all individual SPEs since 1989.

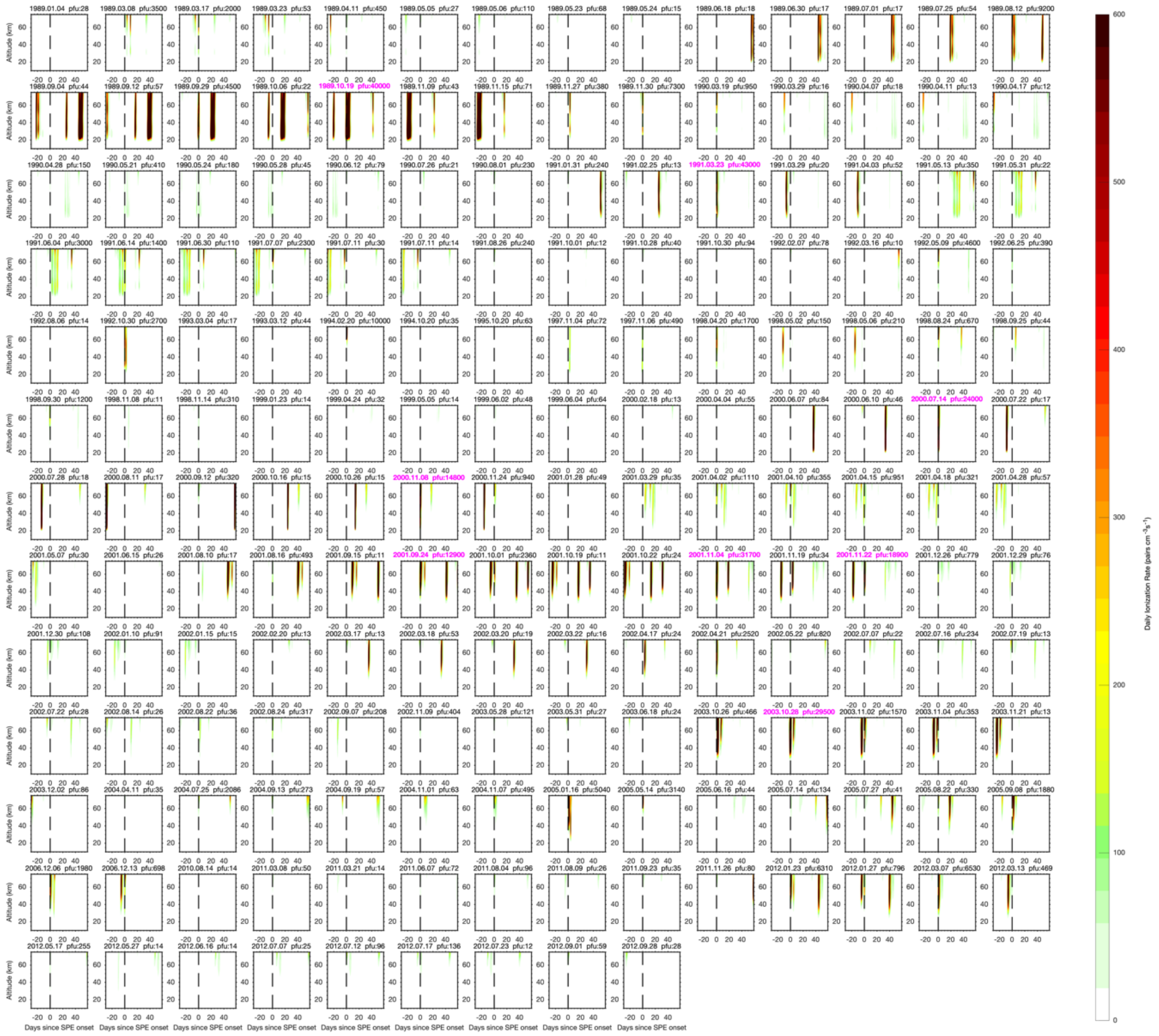

Figure A4. Daily averaged ionisation rate along with altitude at 30 days before and 60 days after individual SPEs since 1989.

Data availability

MLS ozone data used in this study is available at https://mls.jpl.nasa.gov/products/o3_product.php. Proton fluxes and solar proton events are available from https://www.ngdc.noaa.gov/stp/satellite/goes/index.html. Daily geo-magnetic activity Ap-index used in Fig. 1 can be found at https://www.ngdc.noaa.gov/stp/GEOMAG/kp_ap.html. SPE induced ionization rate dataset is available at https://solarisheppa.geomar.de/solarprotonfluxes.

Author contribution

JJ and AK formed the idea of the work. JJ performed the analysis and wrote the paper with contributions from NK, PTV and AK. MES provided the WACCM-D model data. All the authors had intensive discussions about the method and results during the research.

Competing interests

The authors declare that they have no conflict of interest.

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