acp-2016-617: Review #1

"Surface ozone in the southern hemisphere: 20 years of data from a site with a unique setting in El Tololo, Chile" by J. G. Anet et al.

We would like to thank the anonymous referee #1 for his/her great review of our publication. Below, we provide the answers to his/her comments.

Section 1 - Introduction: For the discussion about the seasonal cycle of tropospheric ozone with maximum in spring or in summer Cooper et al., 2014 (section 4) showing seasonal cycle by hemisphere with a maximum in spring for the south hemisphere using OMI/MLS tropospheric column ozone should be cited.

We added following sentence in the "introduction", Page 3, L20: "*These findings were more broadly confirmed by Cooper et al. (2014), who, using satellite-measured total column ozone datasets, classified the onsets of the Total Column Ozone (TCO) maxima globally. In general, spring TCO maxima are found rather on the SH, while summer TCO maxima are prominent in the NH.*"

Section 3.3 - Methods: Is it possible to further justify the 4 ppbv threshold used to exclude high changes between one hour and the next?

The 4 ppb limit has been defined after thorough trial-and-error experiments. Those 4 ppb created least "false negatives" or "false positives" during automatic filtering, which otherwise would have to be corrected manually. This value changes from station to station, as it depends from the natural ozone variability. We added the following to the paper: "*This value of 4 ppb has been defined as such to avoid too many false positives or negatives during the automatic filtering process, in order to minimize the workload during the manual dataset review process.*"

Section 3.3 - Methods: Would the authors confirm that 8.9% represents data influenced by local pollution and missing data? Does it give a first element that ozone at El Tololo is unlikely driven by local pollution?

Thank you for this valuable comment. The reviewer is right that the 8.9% do cover both the excluded data by the filtering process as well as missing data. This number is dominated by the larger data gaps (see grey periods in Fig. 4 and Table S1) and thus, does not give neither a meaningful impression of the rigor of the filter nor of the frequency of rare events of local pollution. The filtering process only excludes 4.9% from the available data. This information will be added to the manuscript. Indeed, a fraction of only 4.9% being identified as influenced by local pollution is a first benchmark indicating the pristine setting of the station. It is worth to mention that it is generally difficult to unambiguously

classify data points as “local pollution events” or “regional pollution events” as periods with steadily elevated mole fractions would not be excluded by the filter. However, the absence of frequent local emissions (e.g. due to the operation of the back-up diesel generator or heavy traffic on the Cerro Tololo premises) is also in line with the observations of the station operators. Therefore, we rephrased the respective sentence that it reads: "*The filtering of the data excludes approximately 4.9% of the available data indicating the pristine setting of the sampling site with hardly any influence from local pollution sources from the premises' infrastructure.*"

Section 3.3 - Methods: Make clear that the trajectories data set are provided by Skerlak et al.

In order to avoid any misunderstanding, following sentence was reformulated: "*Driven by the wind field of ERAI, Škerlak et al. (2014) calculated kinematic trajectories using an 3-steps iterative Eulerian integration scheme (Sprenger and Wernli, 2015).*"

Section 3.3 - Methods: Would the authors write the definition of the max flux more explicitly as a mathematical formulae? That would be easier for the reader.

Thanks for this good suggestion. We adapted the manuscript accordingly: $\Delta MF_{O_3} \approx n * t * \Delta m_{O_3}$

Section 4.1 - Trends: the authors are using Figure 10 […] to argue in favor of the role of ozone STE max flux in tropospheric ozone seasonal changes and changes in time. This paragraph needs to be clarified. The time periods used to assess the changes of the seasonal cycle of tropospheric ozone over time are 1996-2000 and 2011-2015, whereas the time period in Figure 10 ends in 2013. The shift of ozone maximum from October to August seems to be seen on Figure 10 but not on Figure 5, why?

I would suggest to add the specific humidity in the study. It can be added on Figure 10 and its seasonal cycle could be shown as well. It will give more evidence of the impact of stratospheric ozone on tropospheric ozone changes.

The reviewer's interpretation is totally comprehensible and we would like to clarify certain points. First, the period shown in Fig. 10 runs only from 1997 to 2013, i.e. no data are shown for 1995, 1996, 2014 and 2015, due to the way the analysis routine works. The intrinsic mode functions (IMF) was calculated for 4 year sliding windows, therefore “cutting” the first and last two years. Calculating the IMF over yearly data would lead to less meaningful/representative results due to the relatively low signal-to-noise-ratio (interannual variability). The shift of the timing in the maximum of ozone is less visible in Figure 5 also due to the different data treatment. Figure 5 shows a comparison of 3 different percentiles of 5-year-monthly averages, not absolute maximal values, and data are aggregated in monthly bins. We appreciate the suggestion to use specific humidity. Usually, ozone peaks associated with stratospheric intrusions are mostly accompanied with low relative humidity (RH) as it was also

shown by Rondanelli et al. (2002) for El Tololo. We also looked into the RH measurements. Unfortunately, the RH time series at El Tololo suffers from sensor deterioration which jeopardizes a detailed inclusion of the RH data into the analysis (see Fig. S7 c).

In order to help the reader to interpret the Figures, we changed the manuscript, the caption of Figure 9 and the text (see Section 4.3 of the revised manuscript) as following: "*For calculation, a 4-year sliding window of daily data was defined and run over all data between 1996 and 2015.*"

And: "*Note that the regression is only poorly visible in Fig. 5, in which data are aggregated in monthly bins and a comparison of 3 different percentiles of 5-year-monthly averages, instead of absolute maximal values, is shown.*"

And: "*A 4-year sliding window of daily data was applied. Average values for the years 1996-1999 are shown as the data point in end of December 1997, 1997-2000 in end of December 1998, and so forth until years 2012-2015 which are shown as data point in end of December 2013.*" was added to the caption of Figure 9.

Section 4.1 - Trends: The authors are pointing out a shift of two months between maximum of STE flux (Figure A9) and maximum of ozone mixing ratio (Figure 5). This is true for the time period 2011-2015 but not for 1996-2000. In 1996-2000, the STE max flux shows two maxima. This paragraph needs to be clarified.

Thanks for this remark. It is true that the STE trajectory frequency of the earlier period shows two maxima. We erroneously focused purely on the more recent period. The finding of the two maxima (or rather: the persistent low in August) in the 1996-2000 period is robust and can be found in the absolute maxima, mean, median and 5-95 percentiles. The reason for this finding is not entirely clear – we assume that the state of the QBO and ENSO might have played an important role on STE during this period (relatively strong El Niño, weaker la Niña), as shown in Neu et al. (2014). As can be seen in Fig. 1R1 and Fig. 2R1 (below), the QBO shear index (QBO, red lines) and the multivariate ENSO index (MEI, blue lines) where in-phase during nearly two years between 1996-2000 and relatively out-of-phase during 2011-2015. The effects are known: the tropical upwelling increases, boosting the planetary-scale wave activity and henceforth the stratospheric circulation, finally leading to an increase in STE exchange in the sub- and extratropics (Neu et al. 2014 & references therein). This most likely leads to an overall stronger STE activity especially in JJA 1997 and JJA 1998. Also due to the comments of reviewer #2, we have entirely reformulated section 4.3

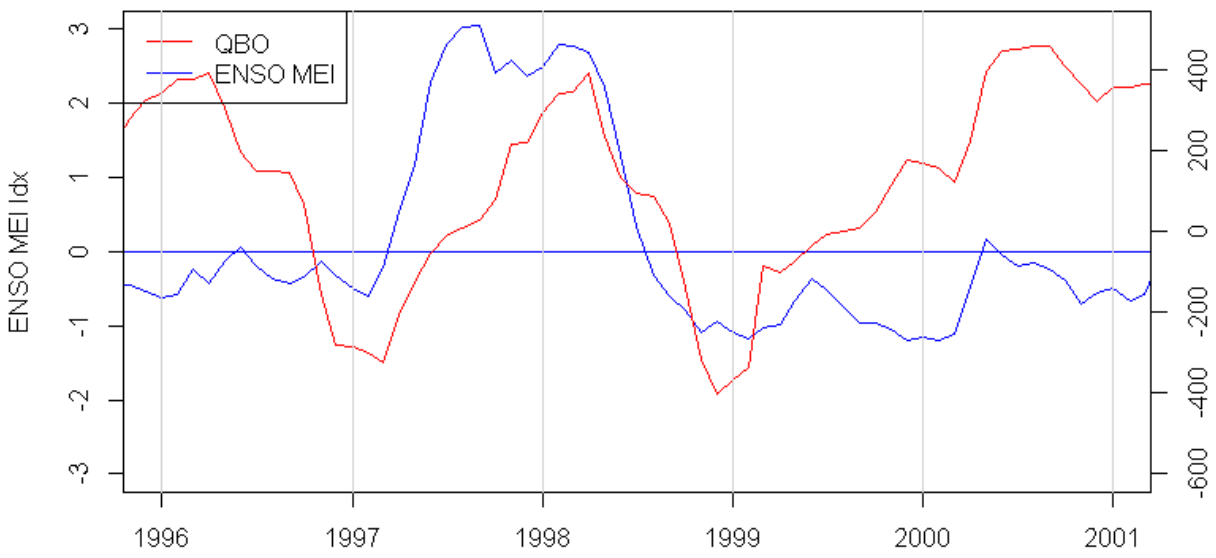

Fig. 1R1: ENSO MEI (blue) and QBO shear index (red) for the 1996-2000 period.

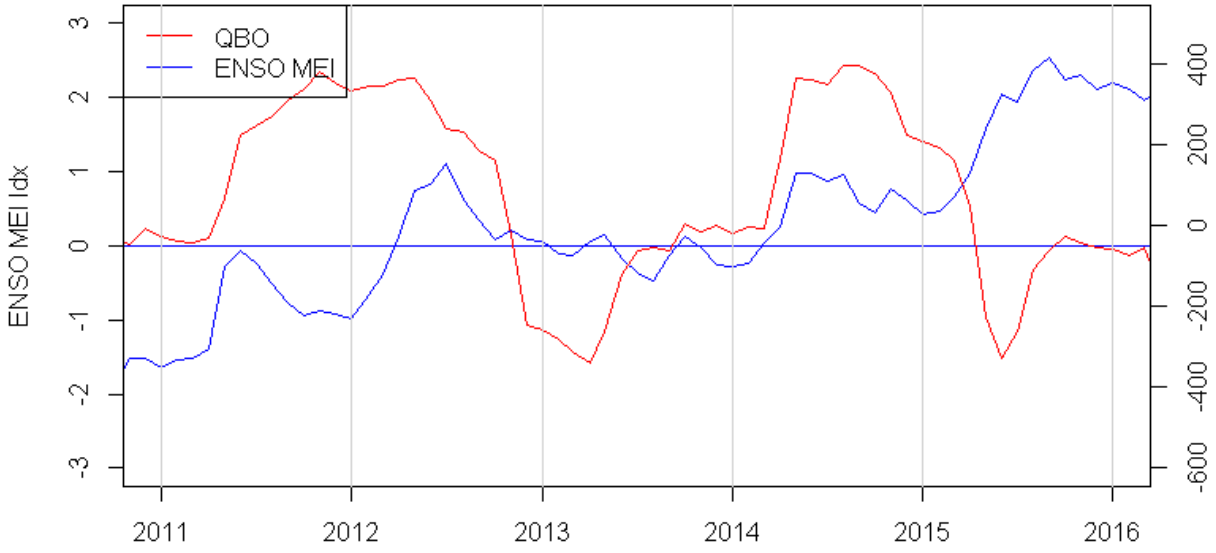

Fig. 2R1: ENSO MEI (blue) and QBO shear index (red) for the 2011-2015 period

Section 4.1 - Trends: Factors influencing the trend in austral fall could be biomass burning in Australia and south of Africa (Cooper et al., 2014), more than Southeast Asia as biomass burning in this region occur mostly in the northern hemisphere.

This is correct. However, by austral fall, the Southeast-Asian branch of the ITCZ has already moved northwards, allowing first pollutants to be transported southbound of the ITCZ into the southern hemisphere. Moreover, biomass burning emissions in Australia and Southern Hemisphere Africa are minimal in austral fall (van der Werf et al., 2006). For clarity, we rewrote the following in Section 4.3 of the revised manuscript: "*An increase of biomass burning in Southeast Asia (e.g. Shi and Yamaguchi, 2014;Verma et al., 2015) and Australia (Cooper et al., 2014) with subsequent eastward transport of ozone precursors, could also explain the positive anomaly in MAM in the 2011-2015 period, as the Northward*

migration of the ITCZ during this time of the year starts to allow effects of NH emissions to be seen in the SH and prevailing westerly conditions (see Fig. 2) exclude any sensitivity of ozone mole fractions at TLL to emissions on the South American continent."

Section 4.3 - Large-Scale influences at TLL: As said above, Figure 10 shows time series from 1996 to 2013 and not 2015. Would it be possible to extent the time period to 2015?

As replied above, this is technically not possible.

Section 4.3 - Large-Scale influences at TLL: The shift in the seasonal cycle has been discussed in section 4.1 - Trends. I would suggest to move the paragraph of the section 4.1 to the section 4.3, otherwise it is confusing for the reader.

This is a very good idea. We moved paragraph "seasonal cycles" in 4.1. to 4.3 and, together with the remarks of reviewer #2, restructured and reformulated the section extensively.

Section 4.3 - Large-Scale influences at TLL: Instead of discuss the relative humidity, I would suggest to discuss the specific humidity which is the absolute value of humidity in the air.

As said above, we could only base ourselves on some very short RH measurements done at TLL, as the sensors mostly were reporting wrong values. Since 2014, the station has been equipped with new sensors, and we totally agree that such a study, relating tropospheric ozone with specific humidity, should absolutely be carried out in near future.

Figures 8 and 10: on the y or x-axis, I would suggest to give the month associated to the given week or day of the year. It will help the reader to follow the analysis which is often based on season or month in the text.

We agree that the reader could be helped by completing the y-axis of Figure 10 with months: We have therefore adapted Figure 10 as the reviewer suggested:

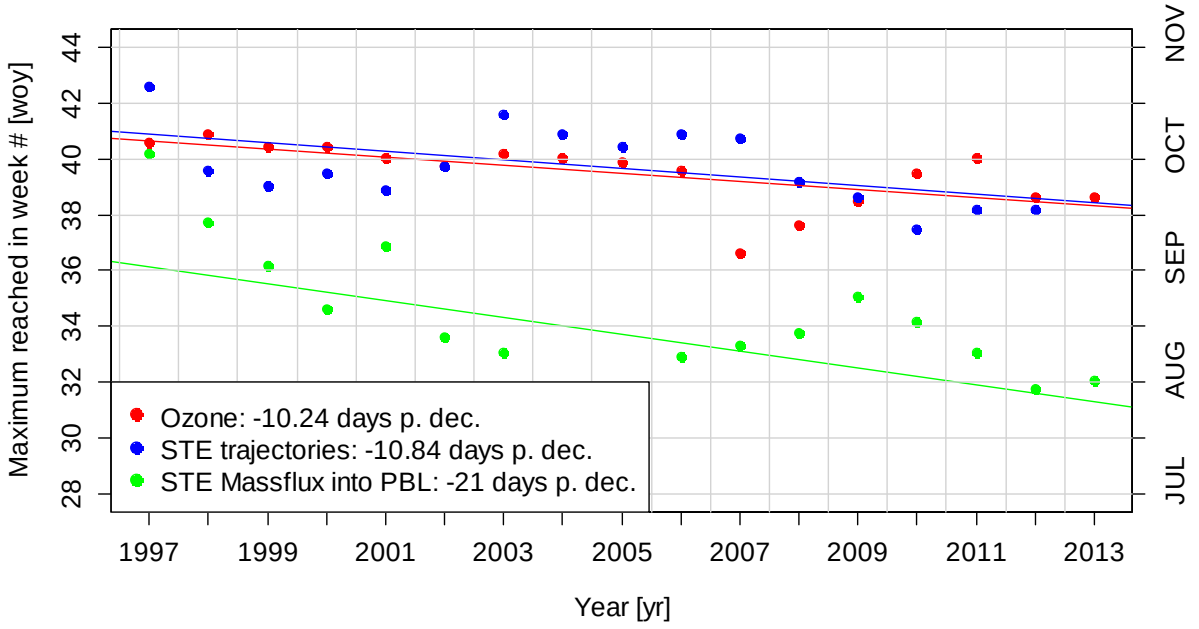

Fig. 3R1: Revised Fig. 10 of original manuscript

Table 1: I would suggest to add the 95% confidence limit and the p-value

We added the 95% confidence limit and the p-value in Table 1 as suggested. On the same time, we slightly revised the numbers as we updated the analysis routine. The latter finding nevertheless does not influence the overall trend discussion.

Typos/Technical corrections

I would suggest to add the longitude and latitude of El Tololo in the title

We agree this would be a good idea. We therefore revised the title: *Surface ozone in the southern hemisphere: 20 years of data from a site with a unique setting in El Tololo, Chile, 30°N, 71°W, 2200 m asl*

L.5 p.3: Change "Unites" to "United"

L.8 p.7: Change "Stratosphere-Troposphere-Transport" to "Stratosphere Troposphere-Exchange"

L.19-20 p.9: I would link both paragraphs (no enter)

L.22 p.9: Change "positive deviations" to "increase"

L.23 p.9: Change "During the remaining of the year" to "For the other months of the year"

We adopted the suggested changes.

L.29 p.9: I would suggest to remove "The attentive reader may have realized that" and start the sentence directly by "The maximum of STE…"

We agree that this makes the text more readable and adapted the manuscript.

L.30 p.9: Change "ration" to "ratio" - L.13 p.10: "biomass burning in …" a word is missing?

We corrected the typos

L.29 to 30 p.: I would suggest to remove this paragraph as everything is already written in the caption of the figure

We assumed that the reviewer meant L29-30 p11, and we have removed the double information from the text about Fig. 8.

Figure 3: I would suggest to change "(a&b)" to "(a) DJF, b) JJA)", same for "(c & d)" and "(e & f)"

Good idea, we changed the figure caption accordingly.

Figure 4: I would suggest to add minor ticks to show all the years - Figure 5: I would suggest to add minor ticks to show all the months

We modified the figures, adding the minor ticks:

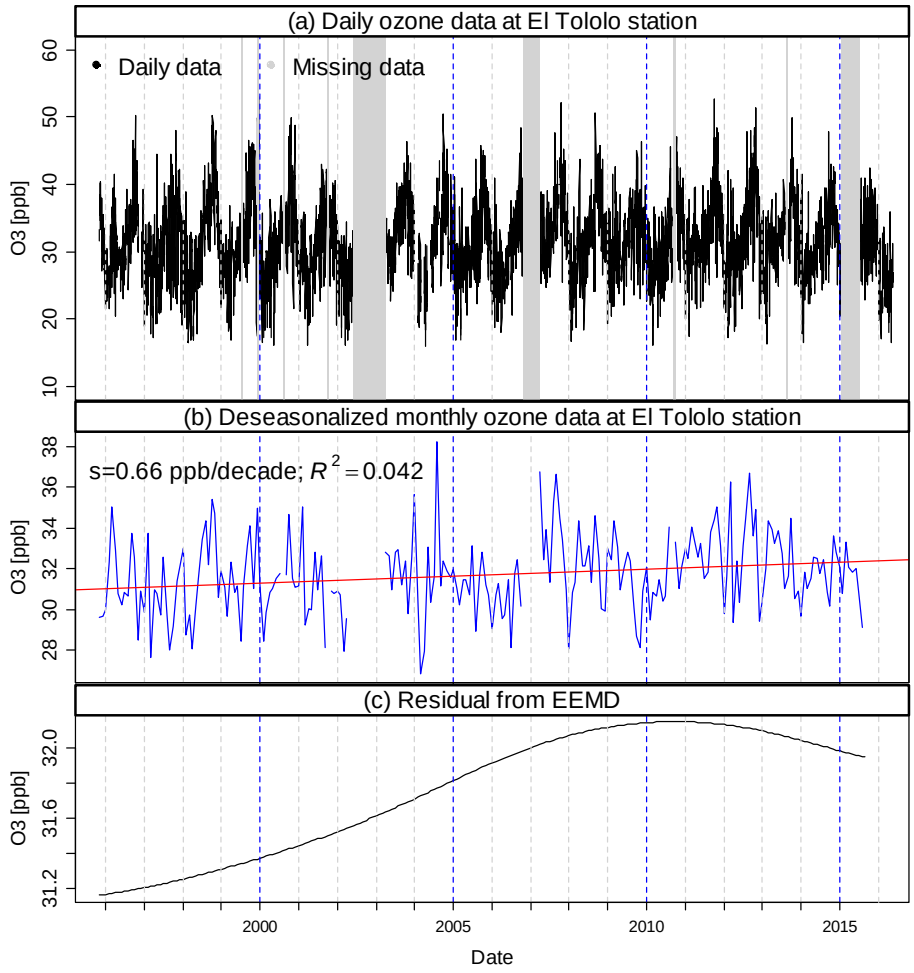

Fig. 4R1: Revised Fig. 4 of original manuscript

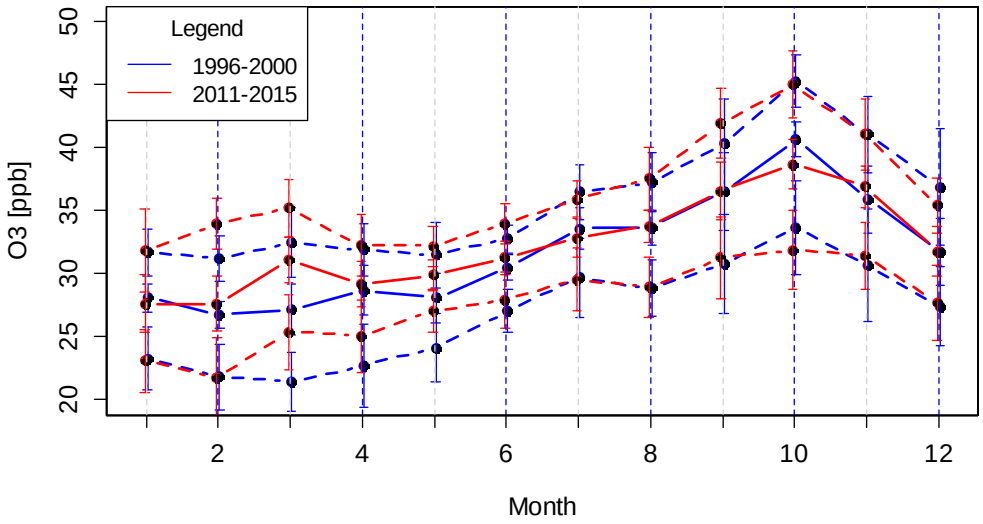

Fig. 5R1: Revised Fig. 5 of original manuscript

Figure A4: I would suggest to change the caption of the figures in order to have the consistent name of the season related to the month: Fall (MAM). It is quite common to order the season as followed: winter, spring, summer, and fall

We modified the figure caption accordingly, as well as the order:

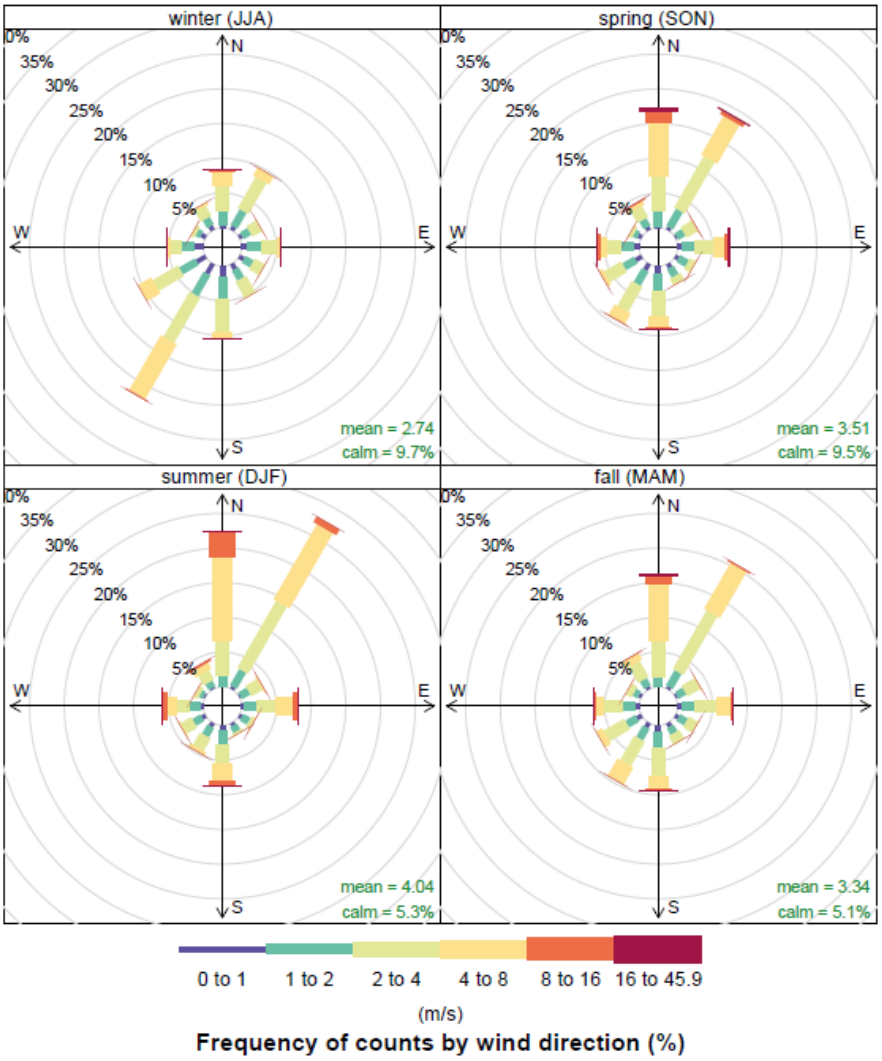

Fig. 6R1: Revised Fig. A4 of original manuscript

Figure A6: I would suggest to follow the common order: winter (JJA), spring (SON), summer (DJF), and fall (MAM)

We reordered the seasons following the suggestion of the reviewer.

Figure A7: I would suggest to add dash lines for the three panels as in Figure 4 and add minor ticks to show all the years

We adapted the minor ticks on the x-axis as suggested:

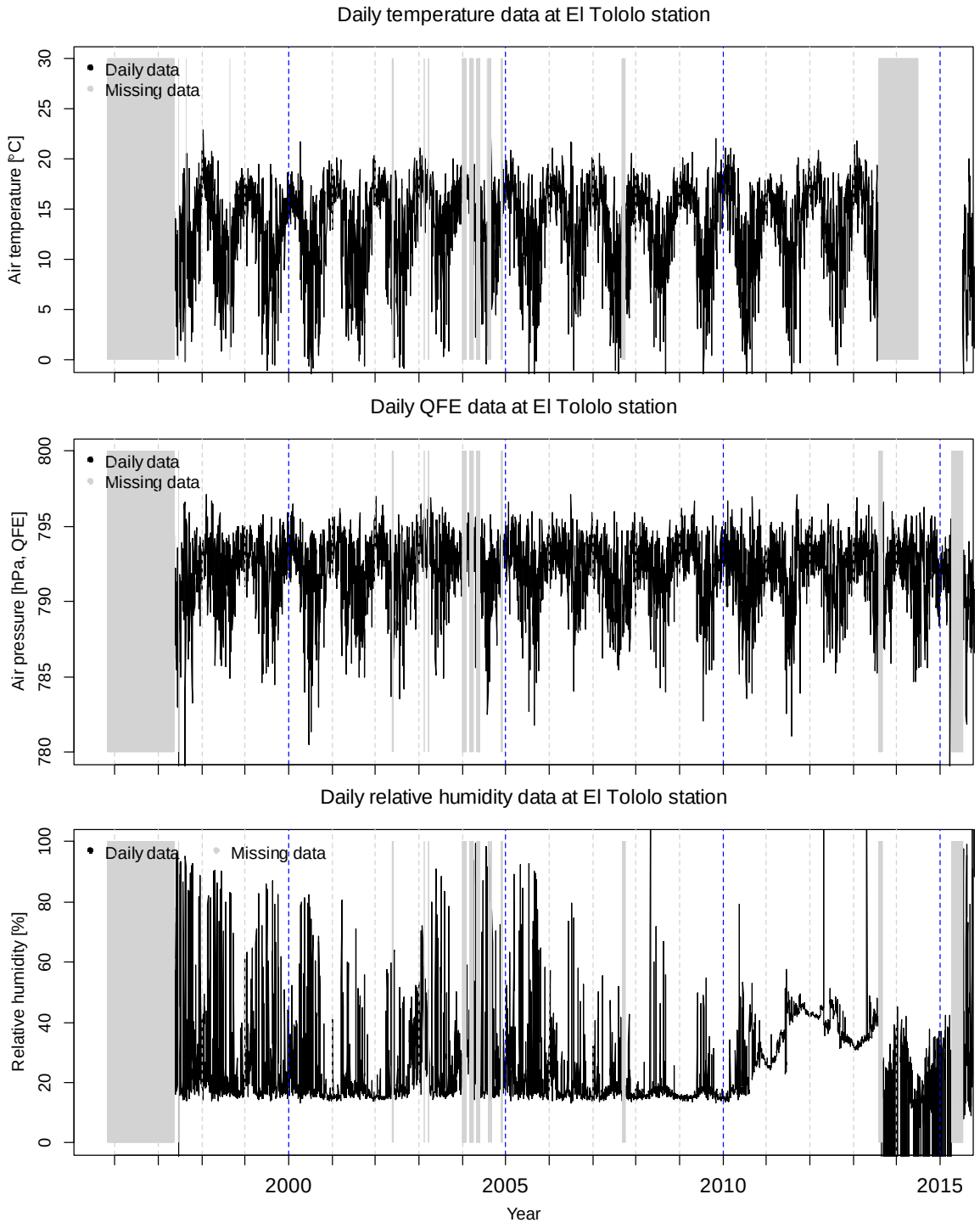

Fig. 7R1: Revised Fig. A7 of original manuscript

Figure A9: I would suggest to add minor ticks to show all the month - In general be careful with the use of "STE" and "STT". STE = stratosphere - troposphere exchange, STT = stratosphere to troposphere transport

Thanks for pointing out the wrong naming. We modified the figure caption and added minor ticks:

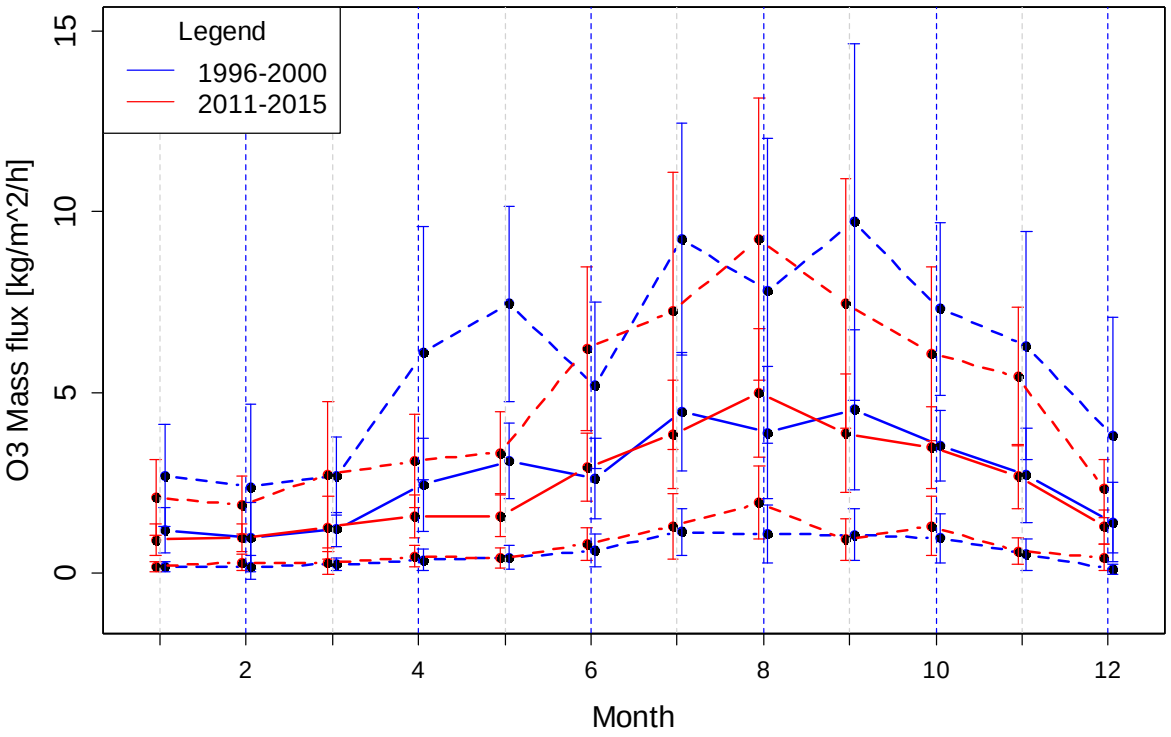

Fig. 8R1: Revised Fig. A9 of original manuscript; Caption modified to: *Mean annual ozone STE mass flux cycle (1995-2000 and 2010-2015) showing mean, upper 95th percentile and lower 5th percentile*

References

Neu, J. L., Flury, T., Manney, G. L., Santee, M. L., Livesey, N. J., and Worden, J.: Tropospheric ozone variations governed by changes in stratospheric circulation, Nature Geosci, 7, 340-344, 10.1038/ngeo2138

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van der Werf, G.R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano, A. F. Jr, Interannual variability in global biomass burning emissions from 1997 to 2004, Atmos. Chem. Phys., 6, 3423–3441, 2006