acp-2016-617: Review #2

"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 #2 for his/her critical review of our publication. Below, we provide the answers to his/her comments.

Major comments of the reviewer

1) According to Figure 5, ozone measured observed at El Tololo increases substantially in the austral autumn (March-April) but shows some decreases in October. The largest ozone differences between El Nino and La Nina years also appear in March-April and September-October (Figure 11). However, the meteorological fields in Figure 3 are shown for DJF and JJA, which are not relevant to the key seasonal features shown in Figures 5 and 11.

Thanks for pointing this out. It is true that the climatology, as it is shown now, does not create any benefit for the reader. We therefore decided to show MAM and SON instead of DJF and MAM in Fig. 3 of the revised manuscript:

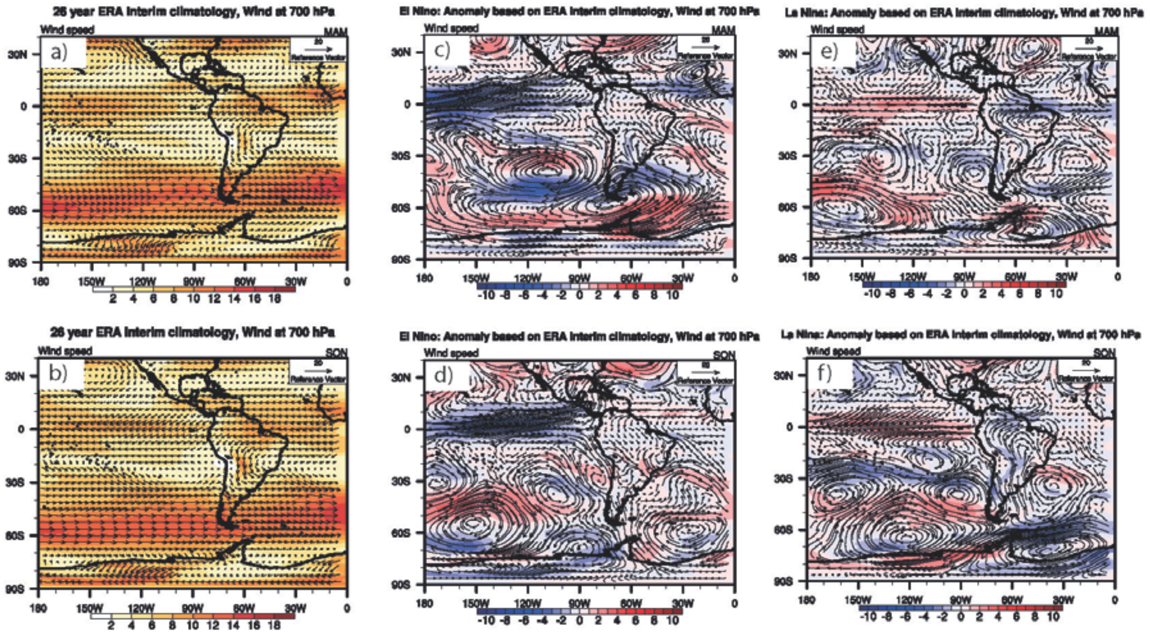


Fig. 1R2: ERA Interim wind climatology at 700 hPa (a, MAM and b, SON) and wind change in vector and strength during an exemplary El Niño event (1997-1998) (c, MAM and d, SON) and a La Niña event (1988-1989) (e, MAM and f, SON).

2) Figure 4b shows a time series for the deseasonalized monthly ozone data. Based on the plot, the authors noted in the text that it is not clear to see an ENSO signal. However, the influence of ENSO on ozone is known to have a strong seasonality (see also Figure 11). Why not also show a

time series of monthly ozone in March-April and September-October, respectively, and correlate the time series with the ENSO index?

Thanks for this valuable critique. It is true that the entire section 4.3 needs a thoughtful restructuring. Based on your suggestion, we tried to include a more in-depth discussion of the ENSO signal. We also included your idea to correlate the ozone time series with the MEI for the two periods (March-April and September-October) and have modified sect. 4.3 accordingly. We refer to "recommendations of the reviewer" for a more complete reply.

Should the slope shown on the top of Fig.4b be ppb/decade rather than ppb/y?

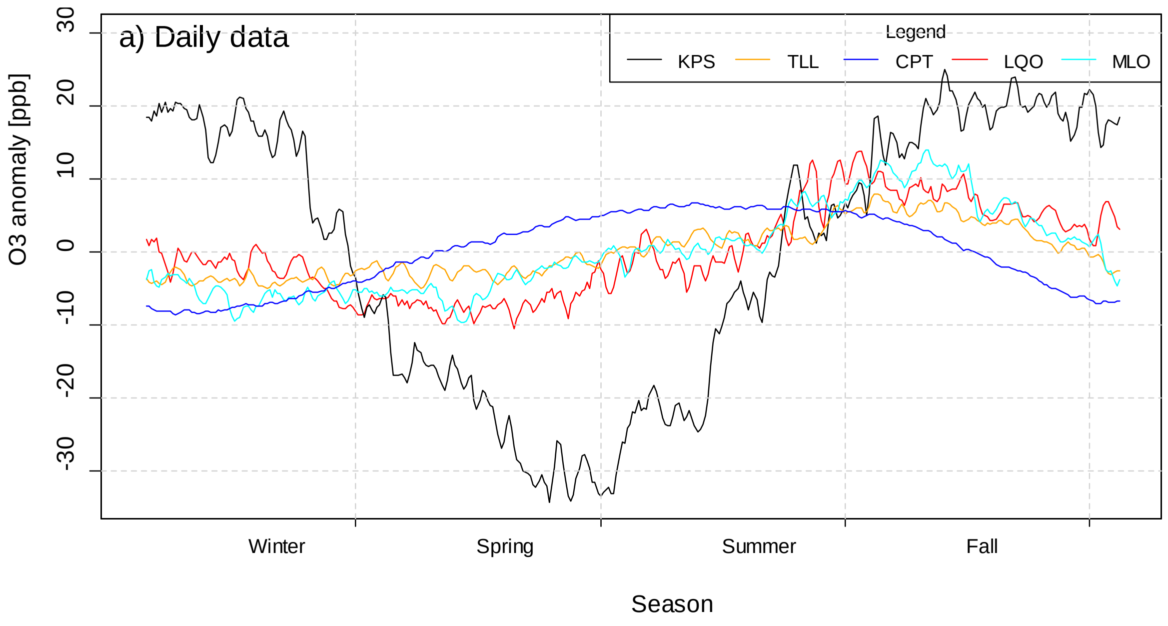
This is correct. We modified the slope legend.

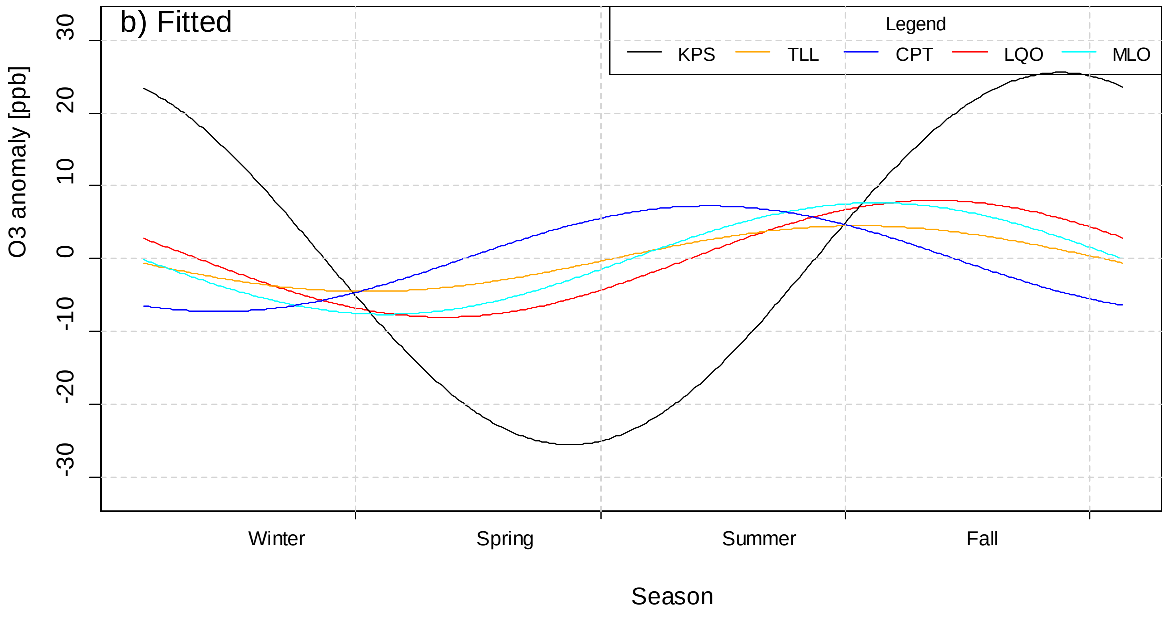
It would be nice to also report the 95% confidence limits of the trends for annual mean and for each season.

We added, as also recommended by reviewer #1, the p-value and the confidence interval. Significant trends with 90% and 95% confidence are now labelled with * and **, respectively.

3) Figure 7 is very confusing. The figure caption notes that data at some sites are shifted by 182 days. Can you just separate the plot for the sites in the Northern Hemisphere versus the Southern Hemisphere without shifting the days and clearly label the latitude and longitude of each site?

Thanks for this comment. We agree that the shifting by half a year may be surprising to the reader, but makes the comparison of stations on the Northern hemisphere and the Southern hemisphere easier, as the seasons on both hemispheres will be aligned over another. In order to increase the readability, we modified the x-axis labels to name the season, and no more the day of year. We prefer this version rather than a separation of the plots, as this would result in a total of 4 plots, without significantly increasing the clarity. Moreover, we decided to add Mauna Loa (MLO) to the plot in order to include the station in the entire discussion.





Another possible factor contributing to the seasonal cycle of ozone in the southern Hemisphere is biomass burning emissions, which the authors did not discuss at all. Please check the seasonal cycle of biomass burning activity in this region as reported in the published literature.

We are thankful that the reviewer pointed out this topic. It is however not entirely true that we did not discuss biomass burning at all. Several times, biomass burning is shortly discussed as a possible factor. Different studies have been cited concerning this topic. We can only repeat that regional biomass burning in the El Tololo region as well as over most of the Southern American continent is not a factor considering the footprint of air parcels travelling to El Tololo, and that influences from Africa or Australia are rather improbable (van der Werf et al., 2006). We nevertheless considered this point when editing section 3.4.

Recommendations of the reviewer

Similar to Mauna Loa Observatory in the Northern Hemisphere (Lin et al., 2014,Nature Geoscience), El Tololo (30S) is located in the subsiding branch of the Hadley Cell in the Southern Hemisphere. Ozone measured at Mauna Loa increases during boreal autumn (Sep-Oct) but shows no significant trend during boreal spring (March-April). Interestingly, ozone measured at El Tololo shows an increase during austral autumn (March-April) but no trend in austral spring (Sep-Oct). While the mechanisms controlling ozone trends at these two sites may be different, there are some similarities on their seasonal ozone trends. Thus, the referee strongly encourages the authors to carefully read Lin et al. (2014) and organize the analyses and associated discussions for El Tololo in a similar way to Mauna Loa. (*…following suggestion of restructuring the order of the figures…*)

We are grateful that the reviewer makes the link to Mauna Loa, another remote, high-altitude station located on the Northern Hemisphere. We did not discuss Mauna Loa due to different reasons:

- This paper was meant to present and discuss 20 years of ozone data from a southern hemispheric station, additionally to put it in context with other stations on both hemispheres, but not to compare it with one similar station from the northern hemisphere
- It is closer to upstream regions with large anthropogenic activities and thus, most likely stronger influenced by ozone precursors emissions
- Its location, more than 1000 km closer to the Equator, makes it difficult to classify it as a non-equatorial station

Yet, we realize that this was possibly a bit short-sighted. As noted above, we restructured section 4.3 entirely, also including a short discussion of similarities and differences to Mauna Loa, including the implicated MEI and QBO shear indexes. However, we tend to disagree that we should organize the analyses similar to the paper of Lin et al. (2014) due to following reasons:

- The manuscript intends to present a new station to the scientific community, first explaining its “special setting” compared to other GAW stations on the world
- We attempt to start from straightforward, rather simple-to-understand connections (like a simple trend analysis) and to later focus into deeper insights, analyzing annual cycles, comparing the station to other stations as well as ozone sonde measurements (in order to get a 3D-picture) and finally trying to link the recognitions with large-scale interactions like ENSO, or STE
- We followed the approaches of recent multi-station or multi-platform (in-situ, but also soundings, airborne observations, satellite products) O3 trend analysis (Logan et al., 2012;Parrish et al., 2013, 2012;Cooper et al., 2014) where changes in emissions and ozone photochemistry, changes in transport and pollution transport pathways, and potential of influence of climate change are investigated side-by-side

Please see also Fig.4 in the following manuscript. Their model also shows that free tropospheric ozone near El Tololo increases during austral autumn (MAM) but there is no significant trend in austral winter (JJA). I wonder if the observed ozone increase at El Tololo has something to do with the poleward shift of the subtropical jet stream in the Southern Hemisphere or changes in geopotential height patterns. The analysis suggested above will help in interpreting the cause of the seasonal trends.

We have attentively read the manuscript of Lin et al. (2016) and included a short discussion about the subtropical jet stream into section 4.3.

Reading the following articles might be useful for understanding the connections between ENSO and ozone variability. Introduction in the current manuscript focuses too much on precursor emission changes that are most relevant for polluted regions in the Northern Hemisphere. I think citing and discussing the findings from the following papers are more relevant to your analysis.

Thanks for these recommendations. They were of great help when restructuring section 4.3. We also included some ENSO-related introductions, citing the suggested publications and others:

Section 1: *Wang et al. (1998) and, more recently, Lin et al. (2015) state that at least some NH spring peaks originate from a combination of ozone-rich stratospheric influx (February-April) and formation by local ozone chemistry (April-June).*

Section 1: *At low latitudes, tropospheric ozone levels at remote sites are known to be sensitive to teleconnections like the El Niño/Southern Oscillation (ENSO). For example, Lin et al. (2014) analyzed the seasonal cycle of a long-term ozone dataset from Mauna Loa (Hawaii). There, long-term springtime ozone observations only marginally increased in the 2000s whereas fall ozone was observed to significantly augment in the 1990s. Lin et al. (2015) found the cause to be the ENSO, which, by altering SST, therefore convection and also large-scale atmospheric patterns, reduced (augmented) advection of air masses from Asia in spring (fall) during La Niña (El Niño) events. The ENSO-sensitive pattern does not limit itself to tropospheric ozone, but also to TCO, as has been shown in Ziemke et al. (2010), who retrieved an independent ENSO index based on TCO anomalies in the pacific region (Ozone ENSO index, OEI).*

Section 3.4: *There are several large-scale processes which potentially impact either short-term or long-term ozone variations at TLL. These factors include large-scale advection of air masses either via the subtropical jet or via potential vorticity cutoffs from the polar jet region, stratosphere-troposphere exchange of ozone-rich air from the stratosphere, as well as pattern changes in tropical up- or downwelling. The impact of these different elements vary not only over the course of a day (storm-scale) or of a year (seasonal cycles), but may also oscillate over timescales of two to seven years, following teleconnections and large-scale oscillations from features like the quasi biennial oscillation (QBO) or ENSO.*

All these influences are visible in the different ozone data products from TLL. We initially focus on the changes in the mean annual cycle of ozone over time and, based on this, elaborate further on the timing of the maximum and on the shape of the cycle. We conclude this section with a short overview of the observed short-term variations (week-scale) including a possible interpretation for those findings.

First, two mean seasonal cycles for two 5-year periods, one at the beginning of the measurements (1996-2000), and the other one in the recent past (2011-2015), were analyzed for potential differences. In Figure 5, the monthly means, with the upper 95th and the lower 5th percentiles including associated uncertainties are presented. Table 1 summarizes the findings in numbers. The two periods show a very similar annual cycle. However, there are subtle differences: Especially in austral fall (February-March), the 5th percentile, mean, and the 95th percentile increased remarkably from the first to the second period. Among the three curves, the 5th percentile shows the most persistent increase from February up to June in the more recent period. For the other months of the year, changes are minor except for October, where 2011-15 shows slightly lower values of O_3*. The annual cycle and some of the differences between the two periods are mainly driven by the annual cycle of ozone STE mass flux (see Fig. 5 and Fig. S9 in the supplementary material). However, the increase of ozone mole fractions from March to May cannot be explained by STE only, as at that time, ozone STE mass flux shows negative anomalies (Fig. S9). Here, three other factors like QBO or ENSO, change in large-scale dynamics and precursor species have to be taken into account. i): As Neu et al. (2014) showed, positive QBO shear and the multivariate ENSO index (MEI) lead to increased stratospheric air circulation, negative upper troposphere ozone anomalies (due to upwelling of relatively ozone-depleted air) and therefore to potentially decreased ozone-STE activity in the sub- and extratropics. Doherty et al. (2006) and Sekiya and Sudo (2012)) explained the decrease in total column ozone found in the eastern Pacific region during El Niño conditions with a decrease in NOx-production due to a decrease in lightning activity. The ozone anomalies at TLL and the MEI show significant correlations especially in September-October (cor=-0.78), and the annual cycles of ozone during El Niño years and La Niña years indeed show significantly different values especially in austral fall and spring (Fig. 11). During La Niña events, ozone levels reach higher values especially from September to November than during El Niño events. Fig. S10 in the supplementary material illustrates that the 1996-2000 period not only had a weak positive correlation (cor=0.28) of both (QBO shear/MEI) indexes, but also had one single strong El Niño event lasting nearly two years, possibly leading to a decreased ozone-STE burden in 1997/1998. The second, later period showed nearly no in-phase correlation of MEI and QBO shear index (cor=-0.04) and no significant El Niño event. ii): The subtropical jet has to be considered as contributing factor to the MAM anomaly in the 2011-2015 period. We assume that with the broadening of the Hadley Cell (HC, Choi et al., 2014;Nguyen et al., 2013), the extratropical jet, moving to higher latitudes, increasingly advected more polluted air from Southeast Asia, e.g. in form of Peroxyacetylenitrate (PAN, see also Jiang et al., 2016), during this period of the year, when biomass burning prevails (e.g. Streets et al., 2003). More work has to be done to confirm this theory, e.g. using satellite measurements, as this would go beyond the scope of this work. iii): 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 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. 3) exclude any sensitivity of ozone mole fractions at TLL to emissions on the South American continent. At most, the ozone increase may originate from regional pollution from the La Serena region, which – in fall – may get transported upwards due to the PBL height and occasional support by frontal systems. This latter assumption remains, however, hypothetical. A confirmation would require high-resolution numerical simulations to resolve the transport in the mountainous terrain of TLL. In summary, we conclude that the annual cycle is mostly STE-driven from June to January. From February to April however, the broadening of the HC with subsequent transport of pollutants from Southeast Asia, the contrast to El Niño dominated (1996-2000) versus average years (2011-2015) and the increase in precursor species in Southeast Asia are the best explanations for the positive ozone anomaly in the more recent period. From Table 1, we conclude furthermore that in general, differences between nighttime and daytime trends are very low which indicates that TLL is a very good background station with similar ozone levels under free tropospheric conditions and under planetary boundary layer (PBL) influence. Mean ozone mole fractions at TLL only vary between 32.5 and 31.0 ppb during day and night, while other stations located near greater cities (e.g. eastern US, Bloomer et al., 2010) report up to 50 ppb peak differences between nighttime and daytime. This finding is most probably attributed not only to the remote location, far away from pollutant sources, but also to the high altitude located above the PBL.

Regarding the 2-month lag between the recent maximum of STE ozone mass flux and the recent maximum of ozone mixing ratio in Figs. 7 vs. S9, we explain this delayed response of ozone to STE by following mechanisms: i) a certain amount of time is needed to equally distribute ozone stemming from STE in the lower troposphere (e.g. titration of NOx & HOx residing in the atmosphere) in order to reach chemical equilibrium and ii) deep convection underestimation as well as seasonal cycle uncertainties within the ERA Interim dataset (Škerlak et al., 2014) lead to doubts concerning the exact onset of ozone STE mass flux maximum around the cordillera.

What concerns the timing of the maximum, Fig. 8 shows a strong correlation (cor=0.89) between the mean annual cycle of STE trajectories (Škerlak, 2014) and of the ozone concentration. The two parameters show a strikingly similar pattern, indicating that STE may be a strong driver for O_3. Note that the mass flux illustrated in Fig. S9 shows a slightly earlier peak occurrence than the number of STE trajectories. Another indication for the coupling of O_3 concentration and STE is a coherent shift in the maximum of these quantities over the observation period towards an earlier occurrence in the year. This is illustrated in Fig. 10. For calculation, a 4-year sliding window of daily data was defined and run over all data between 1996 and 2015. Then, an empirical mode decomposition was done (Huang and Wu, 2008;Wu and Huang, 2009). Out of the Hilbert periodogram, the IMF resembling the most to an annual cycle is selected and the IMF-datapoints are extracted. The latter are averaged to get an average of IMF over the 4-year window. Finally, the day-of-year matching the maximum value of the IMF is extracted. For the ozone time series, a regression of -10 days per decade was calculated. For STT, an even larger trend of -11 or -21 days per decade was obtained for the maximum number of trajectories of stratospheric origin and for the mass flux into the PBL, respectively. Note that the regression is only poorly visible in Fig. 5, where 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.

This shift in the seasonal cycle to earlier times in the year has already been presented in other studies for other locations (Parrish et al., 2013;Lin et al., 2014). For instance, spring peaks are observed in the NH to *regress with a rate of 3 to 14 days per decade (Parrish et al., 2013). Parrish et al. (2013) also suggest that the relative contribution from the stratosphere may at least partly explain the shift in the annual cycle at high-altitude stations in the NH like Jungfraujoch, being located at 3580 meters asl. Yet, a conclusive explanation for this shift of the seasonal cycle remains missing. Schnell et al. (2016) recently suggested that future climate change will shift the maximum of the ozone seasonal cycle to earlier in the year, but they did not provide any clear explanation for this phenomenon.*

Considering the short-term variations, it is known from previous studies that a (anti)correlation between ozone and relative humidity exists at TLL, but only in very specific cases. Gallardo et al. (2000) analyzing the first years of data collected at Tololo, found such an anticorrelation between ozone and water vapor in summer in connection with upslope transport of boundary layer air associated with a thermally driven circulation. Rondanelli et al. (2002) investigated the effect of troughs associated with a frontal zone passing over TLL, and could classify their observations in two categories: wet and dry events. During wet events, relatively humid air from the PBL is advected to TLL, and shortly after regression of relative humidity, ozone is rising rapidly. During dry events, ozone is rising, but relative humidity stays at normal, dry levels or drops even further. Carbon monoxide, a good PBL pollutant and hence an optimal tracer, has been measured in TLL since April 2013. Therefore, the dependence of CO and ozone was investigated. This analysis revealed a significant correlation (not shown) in rare, specific episodes, during which less pristine air from the PBL – originating from the La Serena, Valparaiso and Santiago regions – is reaching TLL. Those events were not always associated with low potential vorticity values (PV streamer, reconstructed from ERA Interim data, not shown) or frontal zones, but some of them were. This confirms the finding of Rutllant et al. (2013), who, during the VOCALS-REx-campaign, found a persistent, regular South-Westerly advection via thermals, being able to transport air masses in the afternoon from the marine region into the Andes which would allow inbound transport of slightly more polluted air masses to TLL. An in-depth analysis of CO-O_3-correlations over several years may be promising to provide more robust conclusions. However, the CO time series is still limited in time and an extended study would go beyond the scope of the paper.

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