Interactive comment on "Tropospheric Ozone Seasonal and Long-term Variability as seen by lidar and surface measurements at the JPL-Table Mountain Facility, California" by M. J. Granados-Muñoz and T. Leblanc

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General Comments:

This paper is a very useful contribution to the rather limited literature of the vertical distribution of ozone over the USA west coast. The analytical techniques employing trajectory analysis, stratospheric-tropospheric folding dynamical structures, time series and variability analysis, and attribution analysis all contribute to the value of this work. The major shortcomings concern the choices for trajectory/attribution parameters (primarily time scales) and the absence of a concise conclusion section. Minor shortcomings concern the details of the trend analysis and some inconsistencies in the attribution. This paper should be accepted after these issues are successfully addressed.

Response:

First of all, we would like to thank the reviewer for the comments and suggestions which have helped to improve the quality of the manuscript. Especially, section 3.4 is now much improved and more significant results were found. Comments and suggestions have been really helpful to improve the section and the quality of the manuscript.

The trajectories attribution section has been modified considering the suggestions and comments from both reviewers. An in-depth sensitivity analysis to optimize the residence times over each region and the duration of the trajectories has been performed and new criteria have been established. Additional details on the selection of the parameters and the duration of the trajectories are now included in the manuscript. More information is provided next.

For the sensitivity test we varied the residence time over each region from 6 to 288 hours (when appropriate) and the duration of the trajectories between 5 and 15 days. Resulting composite ozone profiles, the number of trajectories and the air masses paths associated to each category were analyzed in detail to optimize the criteria for the classification.

Regarding the duration of the trajectories, the sensitivity test reveals that that the composite profiles are not statistically different when using different duration of the trajectories. The main conclusions regarding the ozone mixing ratio values associated to each region are still valid, independently of the trajectories duration (see figure below). However, the number of trajectories associated to each region (included in table 4 in the manuscript) do vary significantly, especially for the Asian and Pacific air masses. For trajectories duration below 10 days, the number of trajectories from Asia is slightly underestimated. A detailed analysis of the trajectories indicates that a value of 12 days for the trajectories significantly improves the results and most of the Asian air masses are correctly identified. Therefore, the trajectories duration has been established

at 12 days for the new version of the manuscript. The residence times above each region has been optimized already considering these 12-day backward trajectories.



Figure: Composite profiles of the ozone mixing ratio associated with the different categories for each season and for different duration of the trajectories: 8 days (solid line), 10 days (dashed line), 12 days (dot-dash line) and 15 days (dotted line). Results are shown only when the number of samples for a given category was larger than 5% of the total number of samples.

In the case of the stratosphere, a residence time of only six hours already shows the influence of the stratosphere on the profiles, which clearly show increased ozone values. Results are very similar for residence times between 6 and 48 hours. Longer residence time leads to an underestimation of the stratospheric cases, since a large fraction of the trajectories descend to the troposphere after this time. The attached figure shows the average composite profiles obtained varying the residence time for the stratosphere between 6 and 48 hours and the associated standard deviation. As indicated by the low standard deviation, results are not highly dependent on the chosen residence time when selected within this range (6-48 hours). The selection of the residence time in the stratosphere affects the profiles associated to the other regions, as observed in the figure. The largest standard deviations are associated to the Asian air masses, but variability is still quite low for most of the profiles.



Figure: Mean composite profiles of the ozone mixing ratio associated with the different categories for each season. Error bars are the standard deviation obtained when varying the residence time for the stratosphere between 6 and 48 hours. Residence times for the other categories are fixed (Central America: 96 h, ABL and AFT: 6 h, and Pacific Ocean: 276 h) and the trajectories duration is 12 days. Results are shown only when the number of samples for a given category was larger than 5% of the total number of samples.

In the case of Central America, the residence time has been increased to 96 hours to avoid the influence of additional sources. When lower values were used, air masses coming from Asia were included within the Central America category leading to an overestimation of the number of cases and influencing the ozone values. Larger values of the residence time, on the other hand, lead to an underestimation of the number of cases. Additionally, the Central America region has been slightly extended further north to 40°N. Based on the analysis of the obtained trajectories, this extended region is more adequate to group the trajectories associated to the North American monsoon circulation.

For the Asian air masses, 6 hours residence time is enough to observe the influence of the Asian air masses on the ozone composite profiles. With the new criteria established for the previous regions and duration of the trajectories of 12 days, almost no variation is observed in the ozone values for residence times above Asia between 6 and 48 h hours. However, residence times longer than 6 hours are more restrictive leading to an underestimation of the Asian number of cases. Residence times of 48 hours for this region with a total duration of the trajectories of 8 days were too restrictive and the number of Asian cases was strongly underestimated. The manuscript has been modified according to the new results.

The Pacific region includes only those air masses with a residence time larger than 276 hours in the region. That way, influence from additional sources is avoided and this region can be considered as our background region.



Figure: Mean composite profiles of the ozone mixing ratio associated with the different categories (except for the stratosphere) for each season. Error bars are the standard deviation obtained when varying the residence time for the ABL, AFT, Central America and Pacific regions between 6 and 288 hours. Residence times for the stratosphere are fixed to 12 hours and the trajectories duration is 12 days. Results are shown only when the number of samples for a given category was larger than 5% of the total number of samples.

A conclusion sections was already added to the manuscript published in ACPD. Please note that the version published and the version attached by the reviewer as a supplement are different. The conclusions section has now been modified in the new version of the manuscript according to the new results obtained in Section 3.4.

Specific Comments: L36: 'No outstanding influence from Asia was identified'. This absence of Asian influence is strongly dependent on the somewhat arbitrary selection of trajectory time-scale parameters. This conclusion is also somewhat inconsistent with the early spring maximum in figure 4. Consider additional analyses to resolve this discrepancy by providing compelling evidence to support your finding.

Response:

We agree with the reviewers that the chosen parameters in Section 3.4 were not the most appropriate and the sensitivity test performed was not exhaustive enough, obtaining misleading results. As previously explained, a new analysis has been performed based on a more exhaustive sensitivity analysis to optimize the residence times over each region and the duration of the trajectories. The new criteria used in the back-trajectories analysis section reveal now the influence of air masses coming from Asia on the ozone profiles measured at JPL-TMF and results are in agreement with previous studies. The manuscript has been modified accordingly. L44: 'Tropospheric ozone can be directly emitted to the troposphere, ': Direct emissions (separate from STE injections) are a very small fraction of tropospheric ozone sources. Suggest you omit this sentence.

Response: Sentence has been removed.

L273: Removing data +/- 1 sd for a correlation calculation is not a legitimate approach. That process will remove approximately 1/3 of the data and will certainly enhance the correlation between the remaining data, but one cannot justify removing that many data and one would certainly not call all those data 'outliers'.

Response:

This paragraph has been omitted from the manuscript considering both reviewers' concerns.

L296: Suggest you use p-values of 0.05 to be consistent with the 95% statistics used elsewhere.

Response:

A discussion using p-Values of 0.05 is also included to be consistent and ease comparison with previous studies. Nonetheless, the criterion of p-Values lower than 0.1 is also used in atmospheric sciences (e. g. Xia et al., 2008; Wilson et al., 2012) and we consider it is worthy to maintain it in the study. A confidence level larger than 90% is still quite significant and these trends should not be neglected. Tables and text has been modified accordingly.

Xia, X., T. F. Eck, B. N. Holben, G. Phillippe, and H. Chen (2008), Analysis of the weekly cycle of aerosol optical depth using AERONET and MODIS data, J. Geophys. Res., 113, D14217, doi:10.1029/2007JD009604.

Wilson, R. C., Fleming, Z. L., Monks, P. S., Clain, G., Henne, S., Konovalov, I. B., Szopa, S., and Menut, L.: Have primary emission reduction measures reduced ozone across Europe? An analysis of European rural background ozone trends 1996–2005, Atmos. Chem. Phys., 12, 437-454, doi:10.5194/acp-12-437-2012, 2012.

Section 4: The summary should be expressed in the Abstract. No need for another summary here. The more discussions should be moved to the section under discussion or a new section heading inserted. The paper needs a short 'Conclusions' section (not summary or discussion). The conclusions should be succinct and describe the main take-home points derived from the paper.

Response:

The discussions have been inserted in Section 3 with the corresponding results as suggested by Reviewer 1. Therefore the discussion section is no longer included in the manuscript.

Technical corrections: See attached .docx for suggested tracked changes. Please also note the supplement to this comment:

http://www.atmos-chem-phys-discuss.net/acp-2016-70/acp-2016-70-RC2-supplement.pdf

Response:

Technical corrections suggested by the reviewer have been addressed in the new version of the manuscript.

Regarding the comment on Line 500 (The wintertime negative results require an explanation), additional details has been added to explain the discrepancies with previous studies. A possible cause for the differences between JPL-TMF and other stations such as those included in Cooper et al., (2014, 2012) might be related to the different sampling or the differences in the analyzed period. Values in the period 2011-2015, not included in the study by Cooper et al., (2012) seem to strongly contribute to the decreasing trend (see Figure 6). Excluding this years from the analysis and analyzing the same period as in Cooper et al., (2012), we still observe a negative trend at TMF (-0. 07 ppbv.year⁻¹), but it is not statistically significant for this shorter period (p=0.83). These discrepancies between the different studies highlight the strong influence of sampling on the obtained results, as already suggested in Lin et al., (2015b). Possible causes for the decreasing trend in winter have been investigated, but no significant results that can satisfactorily explain this trend were obtained. According to sections 3.4 and 3.5, an important source of ozone during winter would be the stratosphere, but no significant trends for the ozone values associated to the stratospheric air masses were found in winter. No anomalous behavior or significant decrease in the number of stratospheric cases was observed either during the analyzed period. The same results were obtained for the Asian air masses and the tropopause folds data during winter. It is worthy to note that the number of data is significantly reduced when they are subdivided in seasons and in the different categories associated to the five regions considered in the trajectories analysis, affecting the significance of the results. A combination of the lidar database with additional data (e.g. model, satellite, additional stations...) would be necessary to reach significant conclusions in this respect.