We thank the reviewers for the thorough revisions and for providing constructive comments on our manuscript, "Trends of ground-level O_3 in Mexico during 1993-2014: Comparison of Monterrey with Mexico City and Guadalajara". We are pleased that the editor and reviewer's perspective on addressing O_3 long-term trends in Mexican urban areas is in agreement with our own views on the issue. We have addressed the concerns and recommendations received, and we believe that these helped to improve significantly the quality of our manuscript. Please find below our detailed response to the comments received, which are also highlighted in red in the revised version of the manuscript, submitted along with this response.

Reviewer #2:

This paper by Hernandez Paniagua deals with a very important topic of ground level ozone (O_3) and its trend in Monterrey, Mexico (MMA) from 1993-2014. It also presents comparison of O_3 trend with other big metropolitan areas in Mexico namely Mexico City (MCMA) and Guadalajara (GMA). While the topic is very important for both air quality and human health, the paper needs some revisions before it is suitable for publication in ACP.

Major Comments:

The paper as it stands is not fully digested. This is one of the major weakness of the paper. It could be significantly concise and coherent without losing any of the messages. A lot of figures could be moved to the supplemental section. For example Figures 3, 4, 5, and to some extent 6 do not really add much to the paper. Similarly, sections describing measurements at MCMA and GMA could be moved to the supplemental section as these measurements are not really different from MMA. On the other hand, the paper will benefit by expanding the analysis section.

Response: As requested by the reviewer, Figures 3 and 5 were moved to Supplementary information, now Fig. S3 and S5, respectively. We consider that Figure 4 must be included in the main body of the manuscript, because it shows the wind direction occurrence discussed along the whole manuscript. Similarly, Figure 3 shows the continuous measurements of O_3 discussed in section 3.2, instead of moving it to Supplementary information, lines showing the 5^{th}, median and 95^{th} percentile as requested below, were included for better interpretation, and the trends of such metrics are discussed in Section 3.4. The Sections 2.2 and 2.3 concerning the monitoring of O_3 at the MCMA and the GMA were moved to Supplementary information as requested. See sections: S1.2 and S1.3.

There are also comparisons with places around the world that is not very relevant for MMA. While it is important to compare the results with measurements made at other places around the world, these locations need to be very carefully selected. O_3 levels depend not only on the emissions of precursors but also the availability of the sunlight. The locations selected in the paper are from everywhere in the world from Canada to Japan, Cyprus to Saudi Arabia. These places do not have similar climate and very likely different emission scenarios and as a result not suitable candidate sites for comparison. Surprisingly there were no comparison being made with any locations in United States which likely has places with similar climate and emission sources (at least in terms of vehicular fleet make up). I suggest the other make a more selective comparison.

Response: The reviewer makes a good point. We have modified the introduction to present only relevant studies carried in North America and Europe. See lines: 58-75, 77-92. Additionally, reports of trends in O_3 at US urban areas are cited and discussed along the results section to explain the O_3 trends reported in the current study. See lines: 114-119, 387-400, 496-502, 544-550 and 590-597.

Despite the paper claiming it as a study of “impact of emissions of VOCs and NO_x on trends of ground level O_3”, there is very little analysis of emissions of VOCs and NO_x in the paper. The only analysis presented is the trend in VOC and NOx emission inventory in MMA, GMA and MCMA (Fig. 1). And, Fig. 1 is mainly used in the introduction and not interpreting the observed trends. It is misleading to claim it as the study of “impact of emissions” as it stands. I suggest the authors, present analysis of NO_x and VOCs (CO) measurements and attempt to make connections between VOCs, NO_x and O_3. A more suitable title for the current paper would be “Trends of ground level O_3 in Monterrey, Mexico during 1993-2014: Comparison with Mexico City and Guadalajara”. The lack of NO_x and VOCs trend makes most of the current conclusion seem more like speculations.

Response: We agree with the reviewer, data of O_3 precursors emissions is now presented in the introduction section. See lines: 109-122, 124-129. Additionally, data of NO_x and CO recorded within the MMA were used to explain the observed trends in O_3. See lines: 311-319, 410-416, 457-465 and 566-574. We have also modified the title of our manuscript as suggested to: "Trends of ground-level O_3 in Monterrey, Mexico during 1993-2014: Comparison with Mexico City and Guadalajara".

There are too many different O_3 metrics being used in the paper and the authors constantly switch between them. There is no rationale being presented for why a certain metric is being used. I suggest the authors minimize the number of metric being used if possible or justify the use of different metrics in terms of what it reveals about O_3 in MMA.

Response: The reviewer is right, the importance of each metric discussed was included. See lines: 135, 307-309, 367-369 and 402-403.

<u>Specific Comments:</u>

Line 23: Why is larger AV_d observed at polluted sites close to industrial areas? O_3 being a secondary pollutant should show larger AV_d at downwind sites? In fact, the largest AV_d is observed at STA which is furthest away from the industrial areas.

Response: The reviewer is right, there was a mistake in the description of the AV_d in the abstract. Indeed, Fig. 6 clearly shows that the largest AV_d occur at downwind sites, i.e. SNB and STA. Text modified: "… the largest AV_d are observed at sites downwind of industrial areas.". See lines: 23-24.

Line 30: GPE is described as highly populated area downwind of an industrial area in Table 2. So, GPE qualifies as site with largest and smallest seasonal cycle (AV_s).

Response: The reviewer is right. There was mistake in the GPE site description in Table 2. Indeed, GPE is located within a densely-populated area, with few industries nearby, as shown in Fig. 1. Text modified in Table 2: "Urban background site in the La Pastora park, surrounded by a highly populated area, 450 m from Pablo Rivas Rd". The sentence concerning the AV_s description in the abstract was also corrected, in accordance with the results presented in Fig. 6. Text modified: " The largest amplitudes of the seasonal cycles (AV_s) are typically recorded downwind of urban areas, whereas the lowest values are recorded in highly populated areas and close to industrial areas. ". See lines: 28-30.

Line 70: Introduction: The introduction switches between O_3 trend in rural background and urban areas. I suggest the authors focus solely on the urban areas as this is the focus of this particular paper.

Response: As suggested by reviewer, the introduction was modified to focus on studies of O_3 trends in urban areas. See lines: 70-75, 78-92, 96-102.

Line 133-135: Based on Figure 14, O_3 has gone up in MMA by only around 20-25% and decreased in GMA.

Response: We have rephrased the sentence to clarify that the exceedances mentioned are punctual. Text modified: "For instance, official reports indicate that since 2000, ground-level O_3 at the Guadalajara metropolitan area (GMA, the second most populated city) and the Monterrey metropolitan area (MMA, the third most populated city), has breached the 1-h average standard of 110 ppb O_3 by up to 80 %, and the 8-h running average standard of 80 ppb O_3 by up to 50 % (INE, 2011; SEMARNAT, 2015).". See lines: 98-102.

Line 172-186: This section belongs to the results and discussion section. These could be used to explain some of the observations rather than keeping it in the methodology section. Further, I suggest figures 3 and 4 be moved to a supplemental section to make the paper more concise.
Response: As suggested by the reviewer, the description of Air mass back-trajectories calculation was moved to the results section 3.1. Text modified:
"**3.1 Wind occurrence at the MMA**

The MMA is highly influenced by anti-cyclonic, easterly air masses that arrive from the Gulf of Mexico, especially during summer (Fig. S5). Figure 2 shows the frequency count of 1-h averages of wind direction by site and season within the MMA during 1993-2014. At all sites, apart from OBI, the predominant wind direction is clearly E, which occurs between 35-58 % of the time depending on season. These air masses are augmented by emissions from the industrial area E of the MMA, which are transported across the urban core and prevented from dispersing by the mountains located S-SW of the MMA. On average, the highest wind speeds are observed during summer. By contrast, calm winds of ≤ 0.36 km h^{-1} (0.1 m s^{-1}) occurred less than 2 % of the time at all sites, most frequently in winter, and least frequently in summer.".

Figure 3 was moved to Supplementary information, now Fig. S5. However, we decided to maintain Fig. 4 (now Fig. 2) within the main body of the manuscript, in order to provide information of the wind occurrence at each monitoring site, which is useful for interpreting the results presented along the manuscript.

Line 188: What is the time resolution of these measurements?
Response: The resolution of the O_3 measurements was added. Text modified: " Tropospheric O_3, 6 additional air pollutants (CO, NO, NO_2, SO_2, PM_{10}, and $PM_{2.5}$) and 7 meteorological parameters (wind speed (WS), wind direction (WD), temperature (Temp), rainfall, solar radiation (SR), relative humidity (RH) and pressure) have been monitored continuously, with data summarised as hourly averages, since November 1992 at 5 stations that form part of the Integral Environmental Monitoring System (SIMA) of the Nuevo Leon State Government (Table 2; SDS, 2016). ". See lines: 159-163.

Line 204-235: Section 2.2 and 2.3: I suggest moving these sections to the supplemental as well. Most of the analysis focuses on the MMA and these two sections only describe the locations and measurements at MCMA and GMA. These measurements are not different from MMA. A reference sentence at the end of MMA measurement section would be sufficient.
Response: As suggested by the reviewer, sections 2.2 and 2.3 were moved to Supplementary information S1.2-3. A reference of O_3 monitoring within the MCMA and the GMA was placed at the end of the paragraph. Text added: "The monitoring of O_3 and other air pollutants at the MCMA and the GMA is detailed in the Supplementary Information S1.2-3.". See lines: 177-178.

Line 237: Section 2.4: I suggest the author expand this section to describe all the methods used in the analysis of the data. Please add a brief description for (i) openair package for R, (ii) MAKESENS 1.0 macro, (iii) Seasonal Trend Decomposition technique. Please include how do they work or what is being done in each of these program and what are the advantages of such an analysis to reveal changes in O_3?

Response: As requested, brief descriptions of the *openair* software tools, the MAKESENS 1.0 macro and the STL technique were included in section 2.3. Analysis of data. Additionally, the pertinence of each function and test used in the current study was stated when required along the results section. See lines: 217-230, 232-242, 244-256.

Line 238: I suggest that figure 5 be moved to the supplemental section.
Response: As suggested, Figure 5 was moved to Supplementary information. See Fig. S6.

Line 265: It would be better to show 5, 50 and 95th percentile line for the data in Figure 6 than all the data.
Response: As requested, lines showing the 5^{th}, 50^{th} and 95^{th} percentiles were included in Fig. 3 (before Fig. 6). However, we maintained the points representing the 1-h O_3 averages in order to show the variations in the magnitude of O_3 peaks during the studied period.

Line 270: Add "(see Figure 3)" behind "(winter)".
Response: The text was added: "The highest O_3 mixing ratios (1-h averages) are typically observed in April (spring), with lowest values usually recorded in December and January (winter) (Fig. 3).". See line: 287.

Line 276: Reaction of O_3 with NO to form NO_2 is only a part of the full Ox cycle. Please include the full Ox cycle.
Response: We modified the sentence as requested. See lines: 294-295.

Line 278: Ox cannot have a minimum value of 0. This would require both NO_2 and O_3 to be 0. It is very likely a measurement error or lack of measurements. I suggest the authors employ some kind of data filtering. There are also instances where Ox is lower than O3 which is again not possible.
Response: The reviewer is right, some O_x values were calculated when O_3 or NO_x 1-h averages were missing. Table S2 was corrected.

Line 282-285: It would be very helpful to include trends in measured NOx and CO to interpret the observed trends in O3 and Ox. This would also be in line with the title of the paper.
Response: As requested by the reviewer, the long-term trends in maximum daily 1-h averages for NO_x were included in Figure 4. These were used to discuss the reported results when required. Text modified: "The largest annual increase observed at SNN is likely influenced by the significant (p<0.05) annual growth of 1.90 ppb y^{-1} in NO_x in levels as shown in Fig. 4, which can be ascribed to localised industrial emissions and constant urban growth W of the MMA (ProAire-AMM, 2008; SDS, 2015). By contrast, the non-significant (p>0.05) trend of -0.01 ppb O_3 yr^{-1} observed at STA is may be masked by local import of O_3, combined with air masses stagnation, since NO_x does exhibit a significant (p<0.05) annual increase of 1.59 ppb yr^{-1}. However, long-term monitoring of VOCs trends and sources is needed to determine the origin of the no trend current status at STA.". See lines: 313-319.

Line 287: Please add description of how the data is de-seasonalised.
Response: As requested by the reviewer, the STL filtering procedure is described in section 2.3 Analysis of data. the seasonal component. Text added: "The O_3 and other air pollutants time-series were decomposed into trend, seasonal and residual components using the Seasonal-Trend Decomposition technique (STL; Cleveland et al., 1990). STL consists of two recursive procedures: an inner loop nested inside an outer loop, assuming measurements of x_i (independent) and y_i (dependent) for i = 1 to n. The seasonal and trend components are updated once in each pass through the inner loop; each complete run of the inner loop consists of $n_{(i)}$ such passes. Each pass of the outer loop consists of the inner loop followed by a computation of the robustness weights, which are used in the following run of the inner

loop to minimise the influence of transient and aberrant behaviour on the trend and seasonal components. The initial pass of the outer loop is performed with all robustness weights equal to 1, followed by $n_{(0)}$ passes of the outer loop.".

Additionally, it was noted that STL was used to filter the O_3 data when required. See lines: 309-311, 361-362, 379-383, 403-405.

Line 289: A NO_x trend would also help justify this statement regarding increased localized industrial emissions.

Response: As requested, we have included in Fig. 4 the long-term trend for maximum daily 1-h NO_x averages for all monitoring sites, and the trends determined have been used to explain the increases in O_3 levels. Text modified: "The largest annual increase observed at SNN is likely influenced by the significant (p<0.05) annual growth of 1.90 ppb y^{-1} in NO_x in levels as shown in Fig. 4, which can be ascribed to localised industrial emissions and constant urban growth W of the MMA (ProAire-AMM, 2008; SDS, 2015). By contrast, the non-significant (p>0.05) trend of -0.01 ppb O_3 yr^{-1} observed at STA is may be masked by local import of O_3, combined with air masses stagnation, since NO_x does exhibit a significant (p<0.05) annual increase of 1.59 ppb yr^{-1}.". See lines: 313-318.

Line 290-292: Isn't this the further study to connect emissions with O3?

Response: We discussed in the manuscript that the changes in NO_x levels are likely influencing the changes observed in O_3, however, we also stated that a further study of VOCs long-term trends would help to clarify the origin of the no trend status at STA. See lines: 313-318.

Line 298: It is surprising that new vehicles are only limited to the city center or the impact of new vehicles are only seen there. Is there some kind of restrictions on the age limit of vehicle that can enter the city center? Else you should see the benefit over the whole MMA unless the industrial emissions are offsetting the vehicular emissions.

Response: There is any restriction of automobile age circulation within the city centre of the MMA. We have clarified that because OBI is representative of mobile NO_x sources, the growth reported in the vehicular fleet corresponds mostly to new vehicles equipped with efficient exhaust catalyst technology, and hence the decline in NO_x seen at OBI (Fig. 4). Additionally, we show that the largest increase in maximum daily 1-h NO_x averages is observed at the SNN industrial site. Finally, we also discussed in the manuscript that the increases in NO_x at other sites are related with the constant growth in urbanisation, as can be seen in Fig. 4a for OBI. See lines: 321-330.

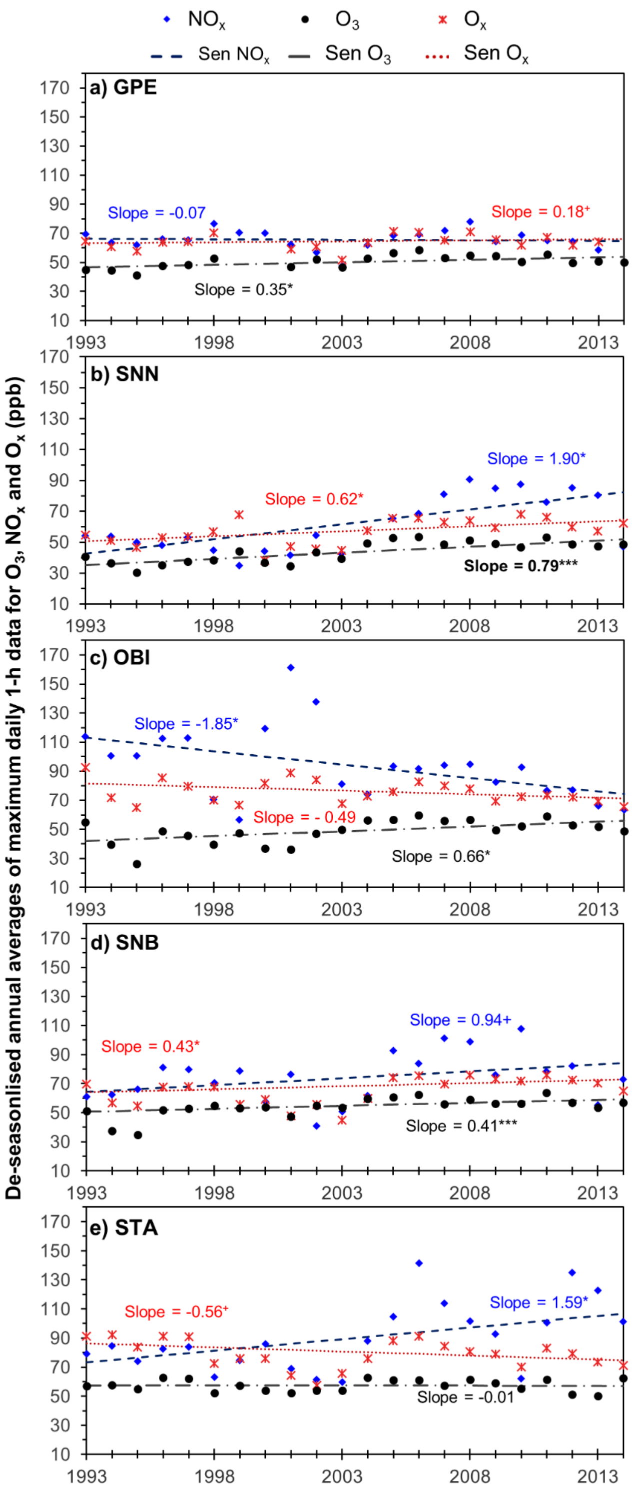

Fig. 4. Long-term trends of daily maximum 1-h values for NO_x, O_3 and O_x observed at the 5 monitoring sites during 1993- 2014 within the MMA. The slopes show annual rates of change expressed in units of ppb yr^{-1}. The dashed lines represent the Sen slopes. Statistical significance is expressed as p<0.1 =$^+$, p<0.05 = *, p<0.01 = ** and p<0.001 = ***.

Line 305: Is there seasonality in rush hour or is the observed shift in O3 dip due to change in time i.e. day light saving?

Response: The 1-h variation in the O_3 daily cycle arises from the change to daylight saving time during spring and summer, which was clarified in the text. Text modified: " Figure 5 shows daily profiles of O_3, O_x, NO, NO_2, NO_x, and SR averaged over the 5 sites within the MMA. O_3 generally dips during rush hour by reaction with NO, which occurs around 07:00 in spring and summer and 08:00 in autumn and winter;

the 1-h difference in the dip derives from the change to daylight saving time during spring and summer. ". See lines: 334-336.

Line 315: How is the normalization performed? Do you subtract the mean O3?

Response: We have stated how the normalisation was made. Additionally, this procedure was included also in the Figure 6 caption. Text modified: "To compare the O_3 diurnal cycles by season, normalised daily profiles were constructed by subtracting daily averages from hourly averages in order to remove the impact of the long-term trends (Fig. 6; Hernández-Paniagua et al., 2015), with daily amplitude values (AV_d; calculated by subtracting the lowest normalised values from the highest normalised values) used to assess diurnal variations in O_3 among seasons.". See lines: 345-349.

Line 318: All the sites show lowest AVd in winter not just SNN.

Response: The reviewer is right. The lowest AV_d are observed at all sites during winter, whereas the lowest ones occur in summer. Text modified: "The lowest AV_d values occur in winter at all sites in response to reduced SR, whereas the largest values observed during summer result from enhanced photochemistry under high SR. ". See lines: 349-350.

Line 318-320: Please justify this statement or add a reference. This statement seems to be misplaced. The lowest AVd during winter is likely due to availability of less sunlight and subsequent slower photochemistry as shown in Section 3.3 and not due to inflow of VOC and NOx laden air masses. These should instead enhance O3 production resulting in larger AVd.

Response: We have clarified that the lowest annual average AV_ds at SNN result from the arrival of NE and E air masses laden with fresh emissions of O_3 precursors, which contrasts with the AV_ds determined at downwind sites receptors of photochemically processed air masses, particularly STA, where the largest average AV_ds were observed. Text modified: "The lowest AV_d values occur in winter at all sites in response to reduced SR, whereas the largest values observed during summer result from enhanced photochemistry under high SR. The lowest AV_d observed at SNN is associated with the inflow of NE and E air masses laden with fresh emissions of O_3 precursors, which are transported to downwind sites (SNB and STA), and become stagnated by the surrounding mountains. This would explain that the largest AV_ds within the MMA are observed at sites receptor of photochemically processed air masses, particularly STA (Fig. 6).". See lines: 349-354.

Line 340: It is hard to see what is going on in Figure 10. Please consider adding a second panel focusing only on one of the years.

Response: As requested, Fig. 8 (before Fig. 10) was modified. The annual cycle of meteorological variables reported in the literature as drivers of the O_3 cycles, was summarised in Fig. 8a. The statistical analysis of O_3 mixing ratios dependence to Temp., rain RH and SR, showed the strongest correlation for O_3 and SR (r=0.76, p<0.05). Figure 8b shows seasonal cycles for O_3 and SR derived from monthly averages filtered with the STL technique. Overall, the relationship between both variables is clear, with peaks for O_3 in spring and early autumn, and for SR in early summer. Figure 8c shows the trends in the O_3 AV_s for all sites within the MMA, with a clear decline in O_3 AV_s before and during the economic crisis between 1994-1996 as result of decreased emissions of O_3 precursors, and an increasing trend since 1998 in response to the recovery of the economy. Finally, Figure 8d shows the annual rates of change in AV_s for the 5 monitoring sites within the MMA from 1993 to 2014.

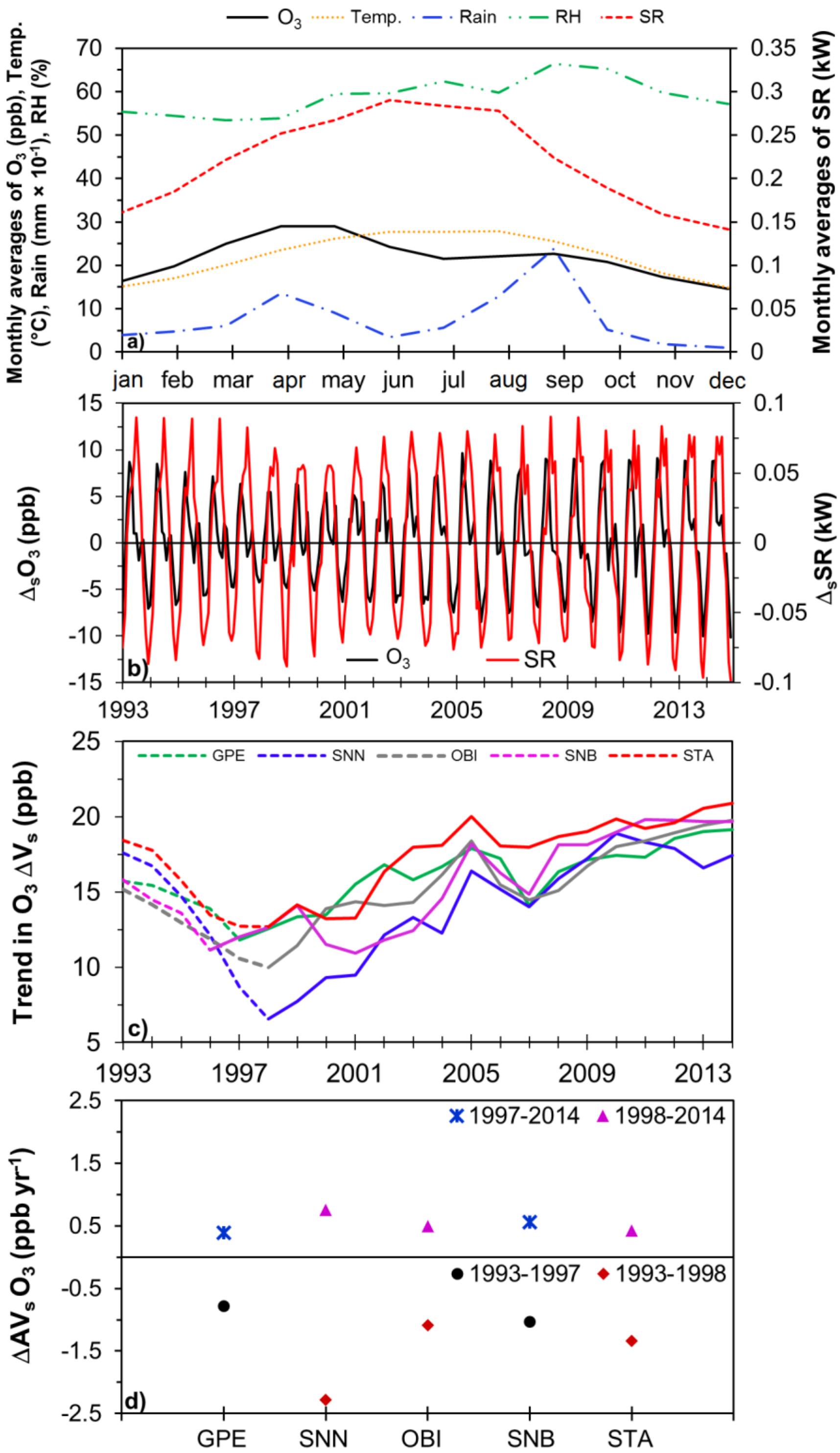

Fig. 8a). Annual cycles of O_3, temp., rain, RH and SR constructed by averaging records from 1993 to 2014 for a 1-year period. **b).** Average seasonal cycles in O_3 and SR within the MMA, constructed from monthly averages filtered with the STL technique developed by Cleveland et al. (1990). **c).** Trends in AV_s of O_3 recorded at the 5 monitoring sites within the MMA from 1993 to 2014. The decline in AV_s observed is due to the economic crisis experienced in Mexico during 1994-1996, followed by persistent increases in AV_s since 1998. **d).** Annual rates of change in O_3 AV_s by site, before and after the 1994-1996 economic crisis.

Line 341: How does reduced rainfall decrease O3 levels? In line 345-346, it is mentioned that frequent rain storms suppresses O3 levels in late summer and early autumn. These two statements are contradictory.

Response: The reviewer is right, there was a mistake in the sentence. Lee et al. (2014) reported increases in O_3 levels during the rainy season due to increased concentrations of the hydroxyl radical. We have corrected the sentence and included such study. Text modified: "By contrast, downward spikes in the seasonal cycles of O_3 within the MMA are observed recurrently between July-August (Fig. 8b), which likely result from high wind speeds (>6 km h^{-1} in average) that disperse O_3 precursors and increase the boundary layer height (ProAire-AMM, 2008), and high day-time temperatures (>40° C) that could suppress the O_3 formation. Steiner et al. (2010) reported that within VOC-limited areas, temperatures >38° C may lead to decreases in O_3 formation, in response to a decrease in the peroxyacetyl nitrate lifetime (NO_x sink). The peak in O_3 observed in September is characteristic of humid regions, and can be ascribed to an increase in OH radicals derived from the increment in RH during the rainy season (Lee et al., 2014). Zheng et al. (2007) reported that this O_3 secondary peak became less noticeable since 2000 over the mid-western and eastern US regions. Indeed, the O_3 secondary peak is characteristic of the Asian summer monsoon, which transports maritime clean air to land with constant rainfall, thereby increasing RH (Xu et al., 2008).". See lines 390-400.

Lee, Y. C., Shindell, D. T., Faluvegi, G., Wenig, M., Lam, Y. F., Ning, Z., Hao, S., and Lai, C. S.: Increase of ozone concentrations, its temperature sensitivity and the precursor factor in South China, Tellus B. Chem. Phys. Meteorol., 66, doi:10.3402/tellusb.v66.23455, 2014.

Steiner, A. L., Davis, A. J., Sillman, S., Owen, R. C., Michalak, A. M., and Fiore, A. M: Observed suppression of ozone formation at extremely high temperatures due to chemical and biophysical feedbacks, Proc. Natl. Acad. Sci. U.S.A., 107, 19685-19690, doi:10.1073/pnas.1008336107, 2010.

Line 359: What is the benefit of calculating AVs. It is currently not clear. One could do a similar trend analysis without calculating AVs.

Response: We have stated the relevance of analysing AV_s. Since the O_3 seasonal cycle is related with the periodic component of the O_3 time series (Cleveland et al., 1990), an increase in the AV_s may imply an increase in the mixing ratios related to seasonality. Moreover, the changes in AV_s determined for the MMA, may reflect the changes in ambient levels of O_3 precursors between 1993-2014. The analysis of AV_s presented in the current study confirms the dominant role of emissions from the industrial region over tropospheric O_3, since the greater changes were observed at the industrial site SNN compared to the urban site GPE. Text modified: "The seasonal amplitude value (AV_s) may provide insights regarding the response in O_3 production to year-to-year variations in the emissions of O_3 precursors and climate.". See lines: 402-405.

Line 364: How did NOx and CO change during the economic crisis? A large decrease in NOx was observed in US and Europe during the last recession (Castellanos et al., 2012, Russell et al., 2012). Do you see similar decrease in NOx as well?

Response: As suggested by the reviewer, we conducted an analysis of NO_x and CO long-term trends of data recorded within the MMA and cited and discussed the provided references. Overall, significant decreases are observed for GPE, SNN and OBI of ca. 5-20 % between 1994-1996, although such decreases are discussed in detail in section 3.8. Additionally, we included Fig. S8, which shows changes in the national Gross Domestic Product (GDP) from 1993 to 2014. Overall, the GDP decreased significantly in 1995 by 7.8 % relative to 1994, during the 1994-1996 economic crisis in Mexico, and in 2009, by 6.4 % in relation to 2008 in response to the worldwide economic recession. This is in agreement with the decreases observed in the US and in some European urban areas during 2007-2009. Text added: "It is very likely that the observed decline in O_3 AV_s is ascribed to the economic crisis experienced in Mexico during 1994-1996 (Tiwari et al., 2014; INEGI, 2016), which caused a reduction in VOCs and NO_x emissions from the industrial activity as reflected in the gross domestic product in 1995 (Fig. S8). Moreover, the reported recovery of the economy since 1997 may have driven the increases in precursor emissions leading to the observed increases in O_3 AV_s. During the global economic recession of 2008-2009, Castellanos and Boersma (2012) observed a reduction of 10-30 % in the tropospheric levels of NO_2 over large European urban areas, which is consistent with a faster decline of 8 ±5 % yr^{-1} in the NO_2 column density during the same period detected by Russell et al. (2012) at US urban regions.". See lines: 425-435.

Line 376: Please see previous comments regarding NOx trend.

Response: As requested, long-term trends of NO_x daily maximum 1-h averages and annual averages were included and discussed in the corresponding sections, as evidence of the changes in emissions with the MMA from 1993-2014.

Line 385: Please see previous comments regarding new vehicle being limited to city centers.

Response: The statement was re-phrased, clarifying that the observed decrease in NO_x observed at OBI is likely due to decreased NO_x emissions from mobile sources, which is offset at other sites by industrial emissions and urban growth. See lines 454-465.

Line 399: Why would accumulation or stagnation of air mass not result in an increasing trend? If more O3 and precursors are coming in from nearby places, then O3 should go up?

Response: We have stated that the lack of a trend at STA is likely due to the occurrence and stagnation of photo-chemically processed air masses with high loading of O_3 and NO_x, and VOCs depleted, which is line with the increasing NO_x trend observed at STA. Text modified: "The large growth rates both in O_3 and NO_x identified at SNN are likely the result of increased emissions from a growing number of industries and sub-urban development E of the MMA. However, at OBI, the increasing positive trend in O_3 contrasts with the NO_x decreasing trend of 0.40 ppb NO_x yr^{-1}, which may arise from the O_3 production non-linear response in the VOC-sensitive MMA airshed, to increasing emissions of VOCs and decreasing NO_x emissions (Sierra et al., 2013; Menchaca-Torre et al. 2015).". See lines: 457-462.

Line 400: Figure 4 does not show stagnation at STA.

Response: We do not agree with this comment. Fig. 4 shows that STA is dominated by the arrival of E and SE air masses in all seasons. Moreover, with the exception of a significant increase of NW air masses occurrence in winter, low occurrence is observed from other wind directions, which is due to the influence of mountains surrounding STA, which act as natural barriers to the N and S-SW-W. Hence, this would confirm that air masses towards the S-SW-W and N would likely stagnate over STA.

Line 423: SNB has growth rate of -0.06 ppb not SNN.

Response: The reviewer is right, there was a mistake. The sentence was corrected. Text modified: " Table 3 shows that significant (p<0.05) annual O_3 growth ranged from -0.05 ppb O_3 yr^{-1} for STA and W, to 0.66 ppb O_3 yr^{-1} for OBI and SE.". See lines: 508-509.

Line 427: Please add p value.

Response: The p-value was added. Text modified: " By contrast, significant (p<0.05) decreasing trends of 0.48 ppb O_x yr^{-1} and 1.52 ppb NO_x yr^{-1} were calculated for the SW sector at OBI, whereas non-significant (p>0.05) trends were apparent at STA.". See lines: 513-514.

Line 431: Are there any sites upwind of the MMA industrial area? This would help interpretation of the data.

Response: Although, there is a monitoring site E of SNN, it was set in 2011 and has experienced instrumentation problems since then. Therefore, the data recorded there were not used in the current study.

Line 442: Figure 1 shows largest emissions for GMA in recent years. This contradicts the statement being made about Figure 1 describing the magnitude of AVds in three cities.

Response: Since the main comment was the consistency of the methodology used to obtain the emission estimates in each NEI release, we included these in section 2.2 and modified Fig. 1 accordingly (now Fig. S1). Additionally, we discussed in the introduction the uncertainties reported for the NEI data, although, Fig. 13 clearly shows that largest mixing ratios of O_3 are observed at the MCMA and the lowest ones at the MMA, which is in agreement with the AV_d reported. See section 2.2 and lines 534-542.

Line 445-448: The statement regarding weekend effect is not clear. Figure 13 does not differentiate between weekday and weekend. It is not clear whether NOx or VOCs decrease during the weekend. So, the statement regarding why no differences in O3 is observed between weekday and weekend is not appropriate.

Response: As requested, the discussion of the weekly cycles was re-written. Text modified: "No significant differences (p>0.05) were observed at any of the metropolitan areas between O_3 AV_d during weekends and weekdays. This lack of a weekend effect in O_3 was reported previously at the MCMA for 1987-2007 by Stephens et al. (2008), who attributed it to weekday O_3 production being limited by VOCs and inhibited by NO_x; this was also observed by Song et al. (2010). By contrast, simultaneous decreases in emissions of VOCs and NO_x mostly from vehicle sources during weekends could have counteracting effects on the O_3 production rates, leading to similar levels of O_3 during weekdays at the 3 metropolitan areas. This behaviour was reported previously by Wolff et al. (2013) for US urban areas of the Northeast, Midwest and Coastal California regions, which exhibited similar or even higher (±5 %) O_3 levels during weekdays than at weekends, despite lower O_3 precursor emissions during weekends. Moreover, Wolff et al. reported that from 1997-1999 to 2008-2010 the sites studied exhibiting a weekend effect decreased from ca. 35 % to less than 5 %, which was attributed to an increase in the VOC/NO_x emission ratio derived from a greater decline in NO_x than in VOCs emissions (Pusede et al., 2014).

It is likely that the O_3 weekly patterns observed at the metropolitan areas arise from reduced traffic activity during weekends, leading to increases in ratios of VOCs/NO_x. Within the MMA, this would be confirmed by lower NO_x mixing ratios (on average 5 %) during weekends, changing to a transition O_3 production between VOC- and NO_x-limited during weekends. Moreover, a change to a NO_x-limited O_3 production derived from the reduction in NO_x seems unlikely since this would result in lower O_3 levels during weekends, not observed at any of the studied urban areas (Torres-Jardon et al., 2004).". See lines 531-549.

Line 450: Section 3.7: I suggest the authors consolidate the trend in O3 in the three metropolitan areas to the observation based only on the trend line. A statement regarding why the trend line is more appropriate than randomly choosing the start and end data to get the reduction/increase in O3 is justified.

Response: As suggested by the reviewer, section 3.7 was modified. Text modified: "Figure 13 shows long-term trends for these pollutants determined with the Mann-Kendall and Sen's estimate. Within the MMA, a significant (p<0.05) increasing trend of 0.20 ppb O_3 yr^{-1} is observed during 1993-2014, within the MCMA a significant (p<0.05) decreasing trend of 0.71 ppb O_3 yr^{-1} occurred during the same period, while within the GMA, a non-significant (p>0.05) trend of -0.09 ppb O_3 yr^{-1} is evident during 1996-2014. The observed trends in O_3 during the studied period, reflect the response to decreasing NO_x (1.24 ppb yr^{-1}; p<0.05) within the MCMA (Fig. 13a), and increasing NO_x (0.28 ppb yr^{-1}; p<0.05) within the MMA (Fig. 13c). Such changes in tropospheric NO_x of 1.0 % yr^{-1} within the MMA and of -1.24 % yr^{-1} within the MCMA, agree with those reported by Duncan et al. (2016), in the NO_2 column during 2005-2014 over the MMA (0.8 % yr^{-1}) and MCMA (-0.1 % yr^{-1}). The status of no trend in O_3 within the GMA contrasts with the significant decrease in NO_x levels (1.47 ppb yr^{-1}; p<0.05) observed both at ground-level (-2.0 % yr^{-1}) and in the NO_2 column (-0.2 % yr^{-1}).". See lines: 554-564.

Line 462: Why is there such a large variance in the annual averages?

Response: We have discussed along the manuscript that changes in O_3 precursor emissions during the economic crisis in Mexico between 1994-1996, and the global recession in 2008-2009 may have led to decreases in O_3. See lines: 472-483, 617-625.

Line 471: Figure 1 shows both VOCs and NOx are going up for MCMA not going down. So, why is O_3 going down with both the precursors going up?

Response: Fig. 1 (now Fig. S1) was modified, and now shows that NO_x emissions decreased during 1999-2008 and VOCs emissions remained constant during 2005-2008. This is in agreement with the observed trends in O_3, despite the uncertainties reported. Additionally, data of NO_x measurements were included and discussed, which show a decreasing trend during 1993-2014. Text modified: "Within the MMA, a significant (p<0.05) increasing trend of 0.20 ppb O_3 yr^{-1} is observed during 1993-2014, within the MCMA a significant (p<0.05) decreasing trend of 0.71 ppb O_3 yr^{-1} occurred during the same period, while within the GMA, a non-significant (p>0.05) trend of -0.09 ppb O_3 yr^{-1} is evident during 1996-2014. The observed trends in O_3 during the studied period, reflect the response to decreasing NO_x (1.24 ppb yr^{-1}; p<0.05) within the MCMA (Fig. 13a), and increasing NO_x (0.28 ppb yr^{-1}; p<0.05) within the MMA (Fig. 13c). Such changes in tropospheric NO_x of 1.0 % yr^{-1} within the MMA and of -1.24 % yr^{-1} within the MCMA, agree with those reported by Duncan et al. (2016), in the NO_2 column during 2005-2014 over the MMA (0.8 % yr^{-1}) and MCMA (-0.1 % yr^{-1}). The status of no trend in O_3 within the GMA contrasts with the significant decrease in NO_x levels (1.47 ppb yr^{-1}; p<0.05) observed both at ground-level (-2.0 % yr^{-1}) and in the NO_2 column (-0.2 % yr^{-1})." See lines: 555-564, 566-576.

Line 487: Which standard is used for Table 5, new or the old one? If it is a mixed of two then, the data is not directly comparable. I suggest using the new O3 standard for all years.

Response: We clarified in the text how the number of annual exceedances was calculated. Text modified: " Such standards are applicable for whole calendar years and were not used in this study to determine the annual exceedances.". See lines 607-608.

Line 493: It is hard to evaluate the statement without not knowing what is represented in table 5. 2012 and 2013 showed a significant reduction in number of days exceeding the standard. Then, there is a big jump in 2014. If the big jump in 2014 due to the change in the standard, then it is kind of misleading to say "recommended that more stringent emission controls are introduced in order to improve air quality within the MMA".

Response: We clarified in the text that the decrease in the number of annual exceedances between 2012-2013 could be due to decreases in NO_x emissions observed particularly at SNN, and possibly ascribed to decreased primary emissions from industries upwind the MMA. Therefore, a decrease of industrial emissions upwind would have a positive impact as observed in the MMA airshed. Text modified: "Between 2012-2013, the number of annual exceedances decreased at all sites, possibly ascribed to an acute deacceleration of the Mexican economy reflected in declines in ground-level NO_x, which is observed particularly at SNN (Fig. 10). Such decrease in primary emissions from the industries upwind the urban area may impact positively the MMA airshed, leading to the observed decreases in annual exceedances.". See lines: 617-625.

Table 3: I suggest moving it to the supplemental section.

Response: As suggested by the reviewer, Table 3 was moved to Supplementary information, now Table S1.

Table 5: Which standard is being used to calculate these exceedances?

Response: The annual exceedances of the O_3 1-h and 8-h running averages were calculated using the old NOM-020-SSA1-1993, which set the maximum permitted O_3 levels in 110 ppb (1-h), and 80 ppb (8-

h). Additionally, in order to permit a better interpretation of the annual exceedances of the O_3 NOM, the results from Table 5 were depicted in Fig. 14.

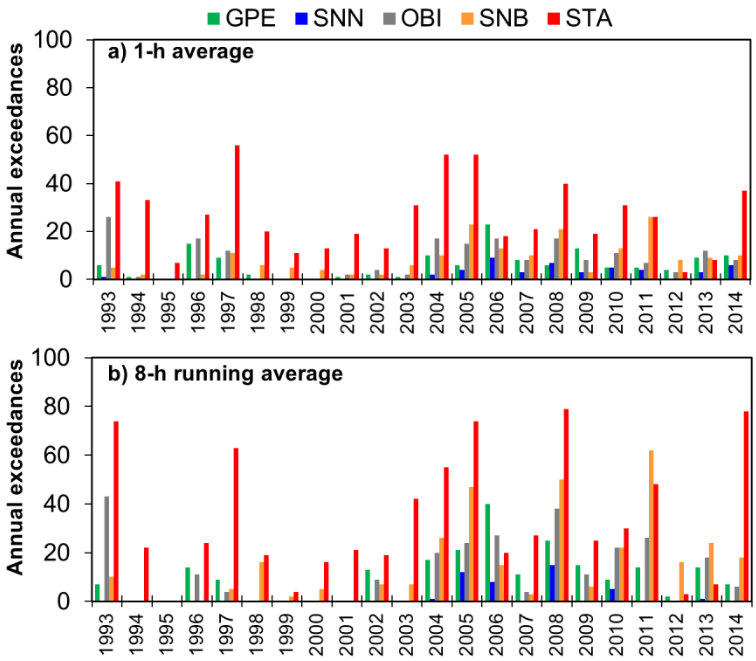

Fig. 14. Annual exceedances of the O_3 NOM for 1-h averages (110 ppb) and 8-h running averages (80 ppb) at the 5 monitoring sites within the MMA from 1993 to 2014.

Figure 2: Please remove the wind rose plot from OBI and add predominant wind direction for each site as a single arrow for each site.

Response: As requested by the reviewer, Figure 2 (now Fig. 1) was modified.

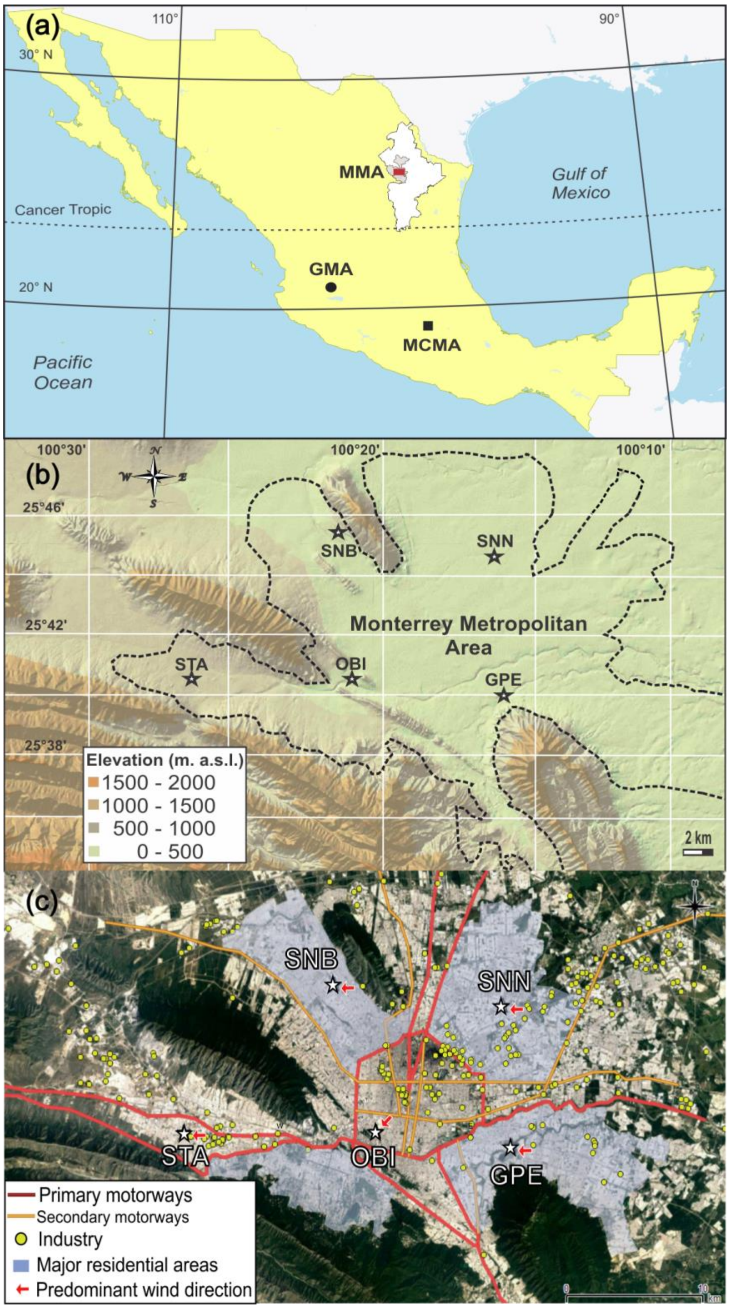

Fig. 1(a). The MMA, MCMA and GMA in the national context. **(b).** Topography of the MMA and distribution of the 5 monitoring sites over the area. **(c).** The 5 monitoring sites in relation to primary and secondary motorways, industries and major residential areas. The red arrows show the predominant wind direction at each site from 1993 to 2014.

Figure 9: Legend is missing STA. There are two GPE s instead.

Response: The legend in Fig. 9 (now Fig. 6) was corrected.

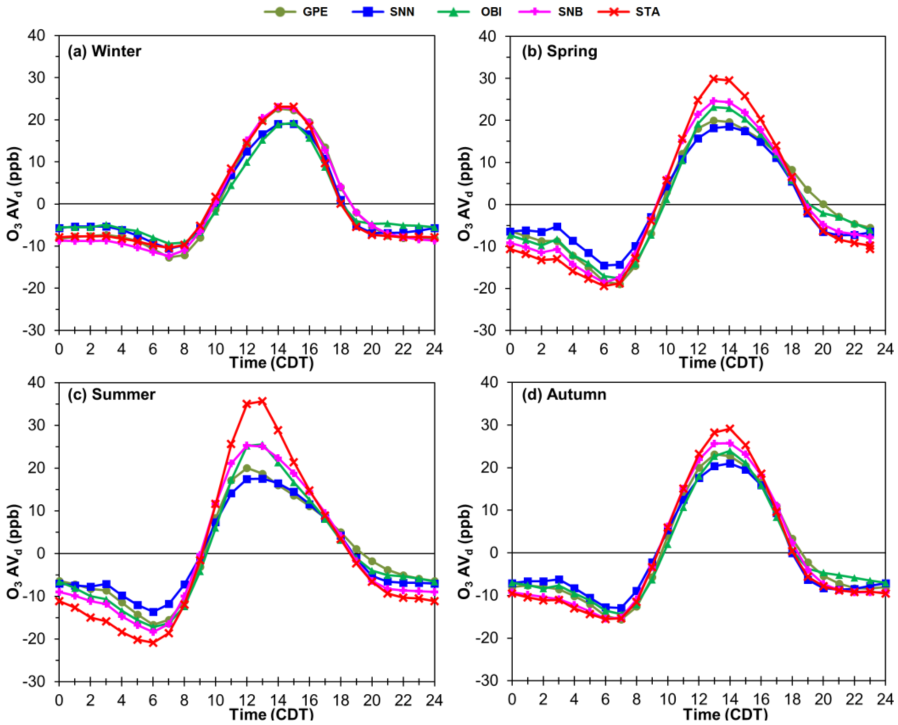

Fig. 6. O_3 de-trended daily profiles by season observed within the MMA during 1993-2014. De-trended O_3 daily cycles were constructed by subtracting daily averages from hourly averages to remove the impact of the long-term trends.

Figure 10: Please add a zoom into one of the years.

Response: Figure 10 was included as a panel of Fig. 8. Additionally, instead of including a zoom in a given year, the average annual cycle for O_3 and meteorological variables reported to be associated with O_3 seasonal variations was included in Fig. 8.

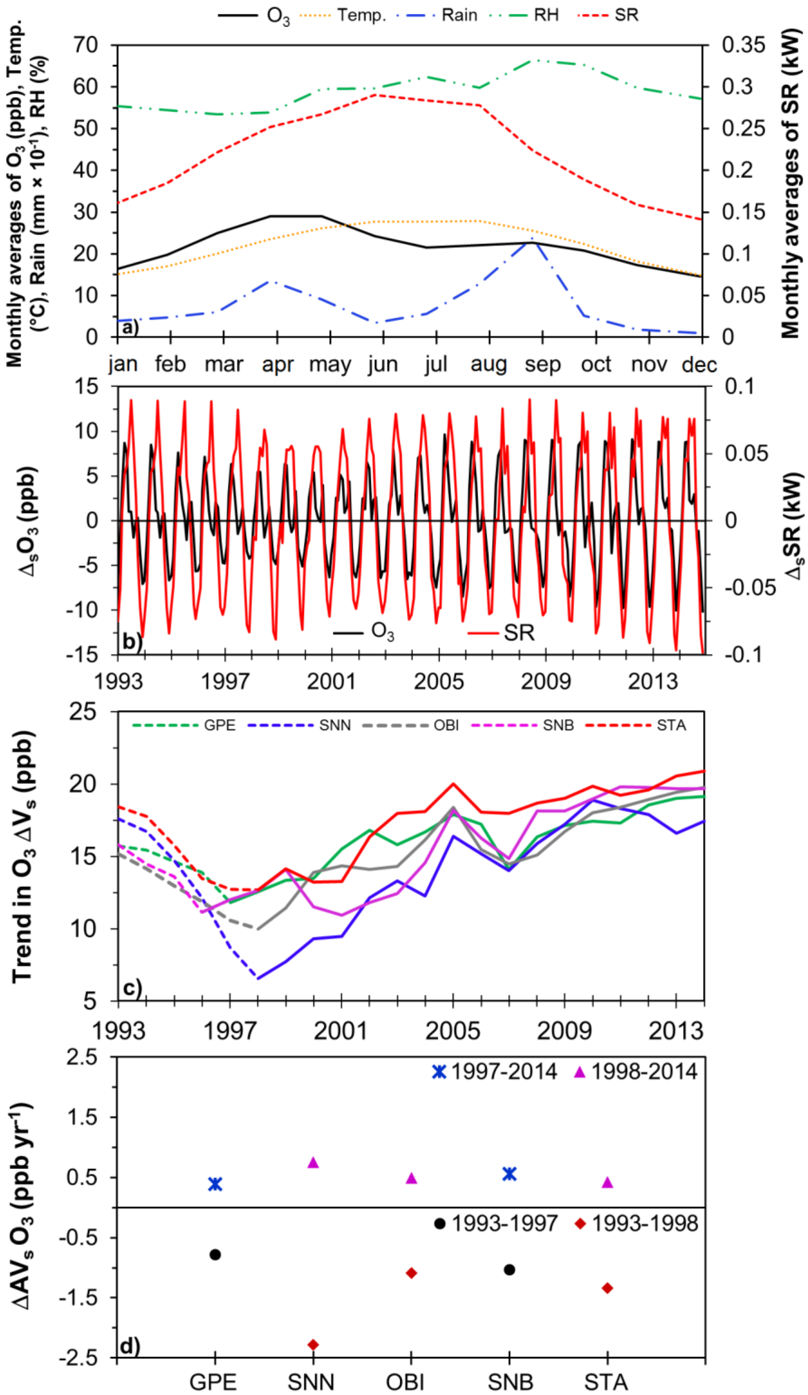

Fig. 8a). Annual cycles of O_3, temp., rain, RH and SR constructed by averaging records from 1993 to 2014 for a 1-year period. **b).** Average seasonal cycles in O_3 and SR within the MMA, constructed from monthly averages filtered with the STL technique developed by Cleveland et al. (1990). **c).** Trends in AV_s of O_3 recorded at the 5 monitoring sites within the MMA from 1993 to 2014. The decline in AV_s observed is due to the economic crisis experienced at the country during 1994-1996, followed by persistent increases in AV_s since 1998. **d).** Annual rates of change in O_3 AV_s by site, before and after the economic crisis within the country.

References:

Castellanos, P. and Boersma, K. F.: Reductions in nitrogen oxides over Europe driven by environmental policy and economic recession, Scientific Reports, 2, 265, 1–7, doi:10.1038/srep00265, 2012.
Russell, A. R., Valin, L. C., and Cohen, R. C.: Trends in OMI NO2 observations over the United States: effects of emission control technology and the economic recession, Atmos. Chem. Phys., 12, 12197-12209, doi:10.5194/acp-2-12197-2012, 2012.