We thank Referee # 1 for the positive remarks about the significance of our study and easiness of reading, and for the helpful suggestions to improve our manuscript. Our answers to the referee's comments are listed below.

Specific comments

Referee comment:

P3, L16. The authors should point out that one of the key findings of the first Tex-AQs study was the disproportionate role of highly reactive VOCs (HRVOCs), primarily alkenes, released from petroleum refineries in the rapid production of ozone in the Houston area. These "upset" emissions were greatly reduced before the second study took place, greatly reducing the local ozone contributions.

Author response:

We agree with the statement made by the referee, which is consistent with previous studies, which were not included in our paper (Ryerson et al., 2003; Daum et al., 2004). We added the following on P3, L16-18:

> "The O_3 pollution in this region was likely a result of abundant precursors emitted locally from urban and industrial sources (particularly, the highly reactive VOCs (HRVOCs) from the petroleum refineries) and the local chemistry sustained by the high summer temperature and land-sea breeze effects. However, the emissions of HRVOCs have been considerably reduced after the first campaign, resulting in lower local contributions to O_3."

We also included two more studies in the cited references on P3, L16-17:

> "Two intensive air quality campaigns investigated peak O_3 in the HGB region during 2000 and 2006, respectively (Ryerson et al., 2003; Daum et al., 2004; Banta et al., 2005; Rappenglück et al., 2008; Parish et al., 2009; Pierce et al., 2009; Langford et al., 2010).

Referee comment:

P3, L25-29. The authors should consider including the study of Darby et al. in their Introduction

Author response:

The short term cluster analysis on hourly wind and ozone maxima in the Houston area (Darby et al., 2005) is a good suggestion, not only for this line. It could also be added on P3, L14 as it points out that the transition from offshore to onshore flow causes high O_3 concentrations (>140 ppb) on a 1-h basis, which is in the line with the land-sea breeze effects described in that paragraph. Therefore we included it among other studies on P3, L14:

> "The land-sea breeze effect complicates this picture through recirculation of local pollution and formation above the coast of the Gulf of Mexico (GOM) of stagnant air masses that entrain local precursors and favor local chemistry and formation of O_3 (Banta et al., 2005; Darby et al, 2005; Nielsen-Gammon et al., 2005; Rappenglück et al., 2008; Langford et al., 2009)."

Recently, we also found that Souri et al. (2016) did cluster analysis on 900 hPa winds and surface O_3 and reported long-term temporal trends in MDA8 O_3 by wind cluster. Darby et al. (2005) did something similar in terms of describing 1-h O_3 maxima by wind patterns but on a much shorter term. Therefore we made the following revision on P3, L22-28:

> "Regional background O_3 in the HGB region has been quantified by many studies but results vary, depending on the temporal scale, spatial scale and the altitude of observations used in data analysis (Banta et al., 2005; Darby et al., 2005; Nielsen-Gammon 2005; Rappenglück et al., 2008; Kemball-Cook et al., 2009; Langford et al., 2009; Zhang et al., 2011; Banta et al., 2011; Berlin et al., 2013; Liu et al, 2015; Souri et al., 2016). Most of the above studies used the MDA8 O_3 to quantify background O_3. Overall, regional (continental) background O_3 ranges from 16 to 107 ppb, while marine background has values between 18 and 40 ppb. Local O_3 contributions are between 25 and 80 ppb. Observations from 1-h average O_3 data and using wind patterns resulted in higher O_3 mixing ratios, particularly during stagnation in the afternoon (>140 ppb) (Darby et al., 2005). Meteorological variables, such as wind patterns, were used separately to characterize the transport regime and its diurnal

transition in the HGB region and interpret their findings from data analysis; their covariance with O_3 and NO_x was not considered."

Referee comment:

P6, L8. Did the Varimax rotation make any difference in the interpretation compared to the unrotated PCs?

Author response:

The rotation gave different loadings for each PC. Because the primary interpretation of the PCs was based on the loading values, yes it made a difference. For instance, in approach A (independent PCA on MDA8 O_3 and 8-h average NO_x at 5 sites), the absolute values of the loadings for PC1 were all greater than 0.9, without rotation. However, when using the Varimax rotation, only 2 out of 5 sites had significant loadings (absolute values nearly or greater than 5). The situation is similar for NO_x except that the loadings in PC1 were all greater than 0.6 (no rotation) and only 1 out of 5 sites had a significant loading value after rotation (0.95).

Referee comment:

P6, L18. A logical extension of this work would be to apply the PCA techniques to the diurnal 1-h median values of Ox (=O3+NOx), which is more conservative. Indeed, an analysis of the nighttime Ox concentrations when there is no photochemical activity might provide the best estimate of background ozone.

Author response:

Yes, it would be interesting to apply the PCA method to 1-h median O_x, which is defined by Daum et al. (2004) as being the sum of O_3 and NO_2, to estimate regional background O_3. We did not use it because we wanted to assess the relationship between regional NO_x and regional O_3; this required independent analyses of 1-h median O_3 and NO_x. Moreover, due to the limitation of the measurement method, NO_2 might include other oxidation products (PAN, HNO_3, etc.). The nighttime background could also be the recirculated local pollution from the previous day and might be different than the "regional" background in the following day. Our focus was on daytime regional background because of its important contribution to peak O_3.

Referee comment:

P7, L26. As noted later in the paper, PC1 equally represents the marine and continental backgrounds depending on the sign.

Author response:

We agree that on the scale of the high O_3 season (May-Oct), both marine and continental influences are described by PC1 based on the sign of its loadings. However, the statement in this line is related to the proximity to the GOM of the high PC1 loadings. We rephrased the text on P7, L26 to read: "The proximity to the GOM emphasizes that PC1 is largely influenced by marine background during summer."

Referee comment:

P8, L6. Figure 2 suggests that the primary NOx PC loadings are associated with the W.A. Parish and other power plants; is this the case?

Author response:

The primary NO_x PC loadings are the results of interpolating between the monitoring sites. Some monitoring sites are in the proximity of power plants. The W.A. Parish power plant is in the northwest of the PC1 pattern.

Referee comment:

P8, L29. Another explanation for the difference is that much of the NOx responsible for the background O3 production has been converted to NOy (e.g. HNO3 and PAN). This would include most lightning generated NOx. Also, depending on the season, a significant amount of the background ozone may also have originated from the stratosphere.

Author response:

We agree with the referee regarding NO_x conversion to NO_y but we don't think that lightning NO_x and stratospheric O3 are important contributions in the HGB region based on available studies. Therefore, we added the following on P8, L31:

> "It is also possible that a fraction of background NO_x (including lightning NO_x) was converted to PAN and HNO_3, which was accounted for in the total NO_x by the measurement method, reducing the potential of background NO_x to explain background O_3. Stratospheric O_3 also may explain some of the background O_3 in the HGB. However, stratospheric O_3 contributions are either overestimated at mid-latitudes by the global cross-tropopause transport models (Liu et al., 2016) or the relationship between the cosmogenic beryllium-7 associated with particulate matter and surface O_3 observed in the HGB region is not conclusive enough (Gaffney et al., 2005). Modeling based estimates of lightning NO_x in the Gulf of Mexico suggest that this source is negligible near the surface, ranging from near zero to 50 ppt during two summer months (Pickering et al. 2016)."

Referee comment:

P9, L30. See comment above about VOCs and Daum et al. (for example)

Daum, P. H., L. I. Kleinman, S. R. Springston, L. J. Nunnermacker, Y.-N. Lee, J. Weinstein-Lloyd, J. Zheng, and C. M. Berkowitz (2004), Origin and properties of plumes of high ozone observed during the Texas 2000 Air Quality Study (TexAQS 2000), J. Geophys. Res, 109, doi:10.1029/2003JD004311.

Author response:

The referee points out the work of Daum et al. (2004), which provides support to our statement "The unexplained portion for the 1-h level (70%) is quite significant. We believe it is related to rapid VOC chemistry in this area of the HGB region."

We thank the referee for this suggestion. Therefore, we added the following on P9, L31:

> "Daum et al. (2004) measured various plumes for almost two weeks in late summer of 2000 and showed that six of them were different from typical urban plumes: they were rich in formaldehyde and peroxides, attributable to hydrocarbon oxidation and photochemistry, respectively. They also found that O_3 formation in these plumes was very efficient (6.4-11 ppbv O_3/ppbv of NO_x). These plumes were tracked back to sources of NO_x and hydrocarbons in the proximity of the Houston Ship Channel. Using zero-dimensional model predictions, they found that O_3 formed very fast (140 ppbv/h). Compared to urban plumes, the authors found that the formation of O_3 in plumes from the Ship Cannel was more NO_x-limited, but uncertainties remain whether the production of O_3 in this area is NO_x- or VOCs-limited."

Referee comment:

P11, L12. PC5 is not significant for O3.

Author response:

Indeed, PC5 is not significant for O_3 (eigenvalue less than 1).

We made the following change on P11, L12:

> "However, we retained all five components because they were not significantly different in explaining the variance in the original variables, particularly for NO_x (Table 3). PC5 was not significant for O_3."

Referee comment:

P13, L25+. The variations in Fig. 4 suggest that the 1-h median approach is (not surprisingly) more strongly influenced by the persistent onshore flow during July than the 8-h MDA8 approach.

Author response:

We added the following on P13, L25-28:

> "In Fig. 4, the hourly median approach also reveals a stronger onshore effect than the MDA8 O_3 approach. This could be because of the smaller time scale of observations, which allows the median to capture better the influence of the onshore flow in terms of O_3.

We also added the following on P13, L30:

> "Regardless of the approach, background O_3 drops in July, which is consistent with the bimodal variation of the annual 8-h average background O_3 (Nielsen-Gammon et al., 2005) and with the less intense and a more easterly Bermuda High during July (Wang et al., 2016)."

Referee comment:

P16, L13. The slopes all agree within the margins of error and are not significantly different.

Author response:

The slopes from different approaches in this study and other studies are not different given the error bars (Fig.1, at the end of this author response), but those from this study appear to have lower uncertainties, regardless of the approach.

Referee comment:

P16, L15. The background ozone trend estimates derived from the current analysis may be twice as precise as those in Berlin et al. (2013), but they are not necessarily more accurate. Indeed, the large interannual variations in the method B data plotted in Figures 6 and 7 suggest that a linear model is not really appropriate. Some discussion of this is in order.

Author response:

We propose the following change on P16, L11-15:

> "Overall, the slope we report in our study (-0.68 ± 0.27 ppb y^{-1}) is larger but more certain compared to the slopes reported by Berlin et al. (2013), which were quantified regardless of the WD (-0.33 ± 0.39 ppb y^{-1} and -0.21 ± 0.39 ppb y^{-1}). Compared to the values reported by Berlin et al. (2013), which represent the trend associated with SE winds only (-0.92 ± 0.74 ppb y-1 or -0.75 ± 0.55 ppb y^{-1}), our slope derived from Approach B is smaller but twice as certain (-0.68 ± 0.27 ppb y^{-1}) and compares better with that reported by Souri et al. (2016) in terms of absolute error (-1.0 ± 0.55 ppb y^{-1})."

The linear model is appropriate despite the larger interrannual variation in the early years, particularly for approach B. These variations are probably due to the fact that we only used five sites or to the fact that local chemistry was much more important in earlier years due to high emissions of O_3 precursors from petrochemical facilities, making it difficult to extract the regional background from surface data during those years. We think that the spatial scale also has an effect on the interannual variability and we see it in the slightly smaller error bars in Approach C, when meteorology and chemistry are covaried between twice as many sites as used in Approach B. Statistically, we cannot reject the linear model to quantify the temporal trends in background O_3 and NO_x because the model parameters are significant. We also used the linear model because previous studies used it, and we wanted to be able to compare our trends. Physically, we agree that a linear model might not be appropriate because there are many confounding factors that influence background O_3 and NO_x on the long term. However, we could not account for all these factors in our study to test for non-linearity.

Technical corrections

Referee comment:

P1, L8 (Abstract). Suggest omitting the "the" to give: "…photochemistry is most active…"

Author response:

We made the correction on P1, L8:

> "Ozone (O_3) in the lower troposphere is harmful to people and plants, particularly during summer, when photochemistry is most active and higher temperatures favor local chemistry."

Referee comment:

P1, L24 (Abstract). Suggest replacing the "the" to give: "…since 2007 and an increase in…"

Author response:

We changed the following on P1, L24:

> "This decline is likely caused by a combination of state of Texas controls on precursor emissions since 2007 and an increase in frequency of flow from the Gulf of Mexico over the same time period.

Referee comment:

P3, L16. "Parrish" is misspelled in the reference.

Author response:

We corrected it on P3, L16:

> "Two intensive air quality campaigns investigated peak O_3 in the HGB region during 2000 and 2006, respectively (Banta et al., 2005; Rappenglück et al., 2008; Parrish et al., 2009; Pierce et al., 2009; Langford et al., 2010)."

Referee comment:

P9, L24. What does VOCs mean? Is this a typo?

Author response:

Yes, it is a typo. We made the correction on this line:

> "However, the high scores in July and August might be related to NO_x and VOCs chemistry, rather than vertical mixing due to a higher boundary layer."

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Figures

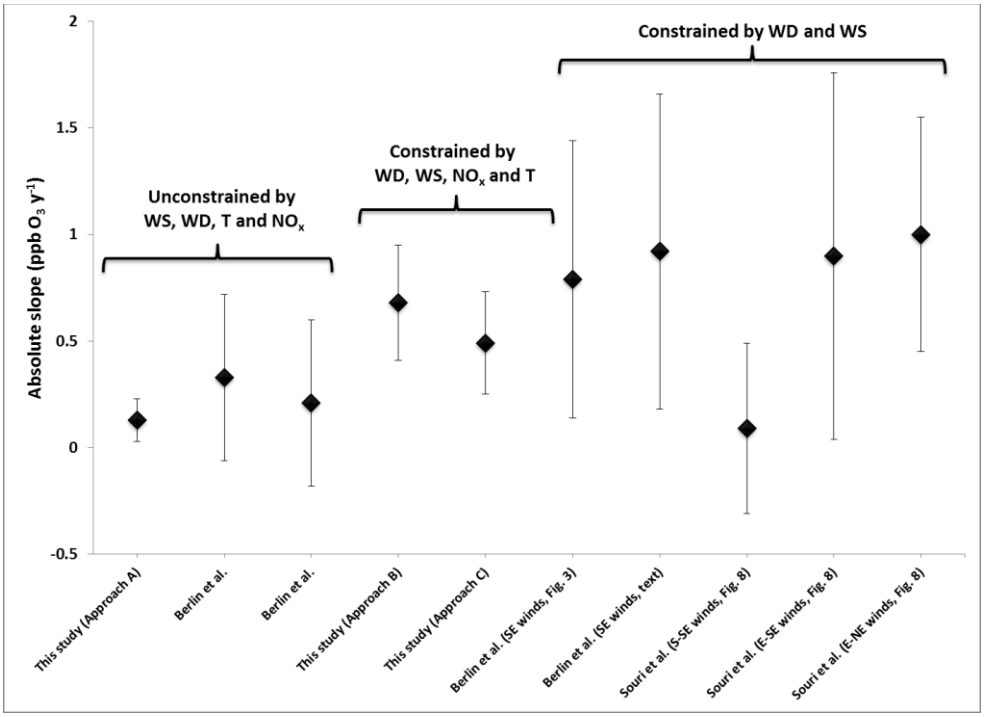

Figure 1: Comparison between the slopes of temporal trends in regional background O_3 in the HGB