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Interactive comment on "Ozone trends over the United States at different times of day" *by* Yingying Yan et al.

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Referee #1

The authors analyzed the inter-annual variability and trends of daytime, nighttime and daily mean ozone during 1990-2014 over the United States based on air quality monitoring data at about 1000 stations, and also assessed the impacts of anthropogenic emissions versus climate variability on the ozone trends during 2004-2012 by the GEOS-Chem modeling. This work combines observations and global modeling to evaluate the ozone trends and driving factors in the past two decades over US, both diurnally and spatially, and provide useful information about the ozone trends at the non-peak hours. The manuscript is clearly organized and well written, and the inter-

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pretation of the observational and modeling results is also fairly well. I recommend that this paper can be considered for publication after the following comments being addressed.

We thank the reviewer for comments, which have been incorporated to improve the manuscript.

Specific comments:

1. On the "Control" simulation: the model only accounted for the inter-annual variations of emission inventory for NOx and CO, but ignored that for NMVOCs. The authors argued that the US anthropogenic NMVOC emissions are much smaller than the natural ones and are hence negligible. The effect of the reduction in anthropogenic NMVOC emissions in US on the ozone trends should be evaluated, especially in the urban areas. Furthermore, the changes of the NMVOC emissions in other regions (ca. Europe and Asia) should be also taken into account, as it may influence the subsequent modeling estimation of the Asian contribution to the US ozone trend. At least, sensitivity modeling studies should be done to check if considering the changes of NMVOC emissions in different regions could affect the conclusions of the modeling study.

In our simulations, similar to the anthropogenic emissions for CO and NOx, global anthropogenic emissions of NMVOC use the REanalysis of the TROpospheric chemical composition (RETRO) monthly global inventory for 2000 (Hu et al., 2015). Emissions over China, rest of Asia, the US and Europe are further replaced by the MEIC (base year is 2008; www.meicmodel.org), INTEX-B (base year is 2006 (Zhang et al., 2009)), NEI05 (base year is 2005, ftp://aftp.fsl.noaa.gov/divisions/taq/), and EMEP (base year is 2005 (Auvray and Bey, 2005)) regional inventories, respectively.

To address this comment, we have conducted a sensitivity simulation, as discussed in the end of the revised Sect. 5.1:

"A sensitivity simulation was conducted to test the effect of anthropogenic NMVOC

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emission changes not included in the “Control” simulation. In the sensitivity simulation, we scaled the NMVOC emissions to the years of 2004 and 2012 based on the EDGARv4.3.2 database which provides the emission time series (1970–2012). Emissions of NMVOC were scaled according to emissions over five regions (China, rest of Asia, the US, Europe and rest of world). Other emissions are the same with the “Control” simulation. In this sensitivity simulation, the modeled change in annual mean ozone from 2004 to 2012 is 1.7 ppb (equivalent to 0.21 ppb/yr) averaged over the US, with local ozone changes ranging from -1.9 to 8.1 ppb across the selected 124 grid cells. This magnitude of ozone change is consistent with the “Control” simulation results with the modeled ozone trend at 0.22 ppb/yr during 2004–2012 (Table 5). For the urban sites, the mean ozone change in the sensitivity simulation (2.1 ppb, or 0.26 ppb/yr) is also close to the “Control” simulation (0.28 ppb/yr). These results suggest that changes in anthropogenic NMVOC emissions have not led to a systemic ozone trend across the US on top of the effect of NOx emission changes, consistent with the results in Simon et al. (2015).”.

2. Figure 3: the ozone growth rate during 2004-2012 is much faster than that in 1990-2014, especially during nighttime hours. The authors explain this in Section 5 partly due to the choice of beginning and end years. Is there any other reason for the stronger ozone trend in the recent decade?

We have added another possible reason in the second paragraph of the revised Sect. 5:

“A possible reason for the stronger ozone trend in the recent decade is that anthropogenic emissions of NOx decline much more rapidly (Fig. 4b) over 2004–2012 (at a rate of 4.1%/yr relative to 2004) than over 1990–2014 (2.1%/yr relative to 1990). Also, a heat wave swept much of the US in 2012, partly contributing to the high value in that year.”

3. Section 3: it would be better if the authors can compare the observed ozone trends

in US with those from other regions of the world, such as Europe and Asia.

In the second paragraph of the revised Sect. 3, we have added the following information to compare the annual mean ozone trends across the globe:

“Similar to the enhanced US annual average ozone, increasing trends of ∼1 ppb/yr in the annual mean ozone are observed at mountainous sites (e.g., Tanimoto et al., 2009) and regional background stations (e.g., Wang et al., 2009) in Asia. In contrast, European annual mean ozone levels have on average been decreasing during the last 20 years (e.g., Sicard et al, 2013). Furthermore, annual mean surface ozone at a background station in eastern China has declined (Xu et al., 2008).”

In the third paragraph of the revised Sect. 3, we have also compared the summertime US ozone trends with those in Europe:

“Similarly, a substantial decrease in precursor concentrations over the last decades in Europe, in line with the long-term emission declines (Colette et al., 2011; Wilson et al., 2012), has resulted in a reduction in ozone episodes (Guerreiro et al., 2014). In contrast, the warm season afternoon ozone over eastern China has been growing at rates of 1–3 ppb/yr over the past 20 years (Sun et al., 2016; and references therein) as a result of rapidly growing precursor emissions.”

4. Section 4: the analyses revealed the weaker correlations between climate variability and the nighttime ozone anomaly (compared to the daytime and daily average), and between climate variability and ozone anomalies over western US (compared to eastern US). The authors need comment on the possible reasons for the weak relationships between climate and nighttime ozone, and western US ozone.

In the fourth paragraph of the revised Sect. 4, we have added the possible reasons for the weak correlation between climate and nighttime ozone:

“A possible reason for weaker correlations between climate indices and the nighttime ozone anomaly (compared to the daytime and daily anomalies) is the distinct chemistry

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at night (i.e., titration by nitrogen), compared to the daytime photochemistry. As the vertical mixing weakens at night, the chemical process becomes more localized and may be less sensitive to large-scale climate variability. ”

In the third paragraph of the revised Sect. 4, we have added the possible reasons for the western US:

“This is likely due to compensating effects on different transport and chemical processes. For example, a positive AMONI are associated with enhanced ozone production (due to enhanced temperature and reduced precipitation in the context of a positive AMO), weakened trans-pacific ozone transport from Asia (due to a negative ONI with a La Niña pattern), and strengthened STE.”

5. Page 12, Line 30-31: may the authors comment on the significance, quantitatively, of the Asian contribution in comparison with those from US and climate variability?

We have added the following information to the last paragraph in Sect. 5.2:

“The rising Asian emissions contribute to the US annual mean ozone trends (0.01–0.02 ppb/yr for daily mean, daytime and nighttime ozone) much less than the contributions from the US anthropogenic emission changes (0.08–0.22 ppb/yr) and climate variability (0.05–0.07 ppb/yr).”

This sentence is a summary of the detailed discussion on various contributing factors in the revised Sect. 5.2.

6. Tables 1-3: provide the unit, ppbv yr-1?

Yes, we have added the unit.

7. Table 4: what do the numbers in this table mean? Correlation coefficient (r or r2)?

It means the linear Pearson correlation coefficient (r). We have added this information in the table.

8. Figure 4a: provide p-values for the trends.

We have added the p-values in the revised Figure 4a.

9. Page 6, Line 32: delete one “mean”.

Deleted.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-659/acp-2017-659-AC1-supplement.pdf

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