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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterizing sources of high surface ozone events in the southwestern U.S. with intensive field measurements and two global models" *by* Li Zhang et al.

Anonymous Referee #1

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In this article, Zhang et al evaluate ambient and modeled data in the Las Vegas area during the 2017 FAST-LVOS study. They aim to determine dominant source categories for high observed ozone events. They leverage both enhanced monitoring data and sensitivity simulations from multiple global models to understand O3 events at this time and location. The evaluations shown here highlight the challenges with determining O3 sources. Even with these detailed datasets the evidence for categorizing O3 events on many of the event days is not definitive. I think this analysis is a valuable addition to the literature, but I think the uncertainties in the analysis need to be more clearly communicated. I believe that the authors have overstated the confidence in their ability to categorize ozone events from the data provided. In addition, there are certain ar-

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eas where the article needs additional details and background, specifically: 1) include references to additional relevant articles on background ozone, 2) include more comprehensive model evaluation information especially as it relates to the specific times and locations of the ozone events of interest, 3) provide more systematic information with which to compare indicator values in each these events through Table 1 or additional tables/figures.

Major comments:

1. Introduction: Please reference the recent comprehensive review of background ozone by Jaffe et al (2018). For the paragraph summarizing past modeling to predict USB, please also include references to Dolwick et al (2015), Emery et al (2012) and additional references cited in Jaffe et al (2018). In lines 80-85, please note that the Dolwick et al (2015) modeling included analysis of daily O3 and plotted USB against total O3 showing the range of daily values. Jaffe et al (2018) also included daily quantification of background ozone. Similarly, when discussing past literature on USB estimates in section 5, please also cite and compare to Dolwick et al (2015) and Emery et al (2012).

2. Modeling description and evaluation:

- Given the heavy reliance on AM4 results to categorize events in this paper, the authors should provide more detailed information on model performance. Model performance is only shown for aloft measurements. I suggest adding model performance of ground-level O3 based on measurements at CASTNET sites. Also, it would be useful to report mean bias of ground-level O3 (based on nearby CASTNET and FAST-LVOS measurement) for each specific episode day (i.e. what is the model performance at the times and locations of interest and how does it change on different days examined). This will provide important context for interpreting modeling results used to classify the different O3 episodes. This could be done as part of Table 1 or as a separate table in the paper. In addition, performance information that is already available in Figures 15

and 17 could be brought forward and expanded upon in section 3.

-Given 50 km resolution of AM4, the model may be better suited to quantify O3 from some sources than from others. The coarse resolution may not matter as much for stratospheric intrusions and for transport from Asia but may be insufficient for capturing photochemical production from sources that may have gradients in precursor emissions (fires and local/regional US anthropogenic sources).

-The AM4 model simulations were conducted for January-June 2017 and the first episode evaluated was in late April. That corresponds to ∼4-month spin-up period which seems short for global simulations that are tracking impact from long-range sources. Please address this short spin-up period? Why is this length of spin-up appropriate for the simulations conducted here?

-A stratospheric tracer is implemented in AM4. Please provide details on whether this tracer is inert or reactive (i.e. can be degraded by chemistry and deposition). If the tracer is inert, then it should not be used to quantify stratospheric impacts (e.g. line 313) because the lack of degredation processes will lead to an overestimate of stratospheric O3 influence. In this case it could still be used qualitatively to identify times and locations of stratospheric influence.

-GEOS-Chem description did not specify the simulation period. Was this also Jan-June 2017? If so, please also address the relatively short spin-up on the GEOS-Chem simulations.

-I have several questions/comments on the US emissions adjustment. You state that NOx emissions were cut by 50% in the Eastern US, does this mean the NOx emissions were left unchanged in the Wester US? The Travis et al paper was based on an analysis for 2013 and NOx emissions and vehicle fleet characteristics (age, vehicle emissions control systems etc) have been continually changing in the US. Is it appropriate to apply scaling factors based on a 2013 analysis to this 2017 time period? In addition, GEOS-Chem emissions already account for decreasing NOx emissions in more recent

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years based on EPA trends information. Since the EPA trends include improvements to inputs for mobile source emissions calculations in more recent years, this 50% adjustment may be double-correcting for adjustments that are already included in the more recent EPA data. In addition, recent papers coming out of the 2017 WINTER campaign (Salmon et al., 2018; Jaegle et al. 2018) suggested that EPA NOx emissions were unbiased in winter, so should same 50% NOx cut be applied in winter months as in summer? If this adjustment was only made in the Eastern US, perhaps the impact of these adjustment are limited for this analysis which focuses on the Las Vegas area. Also, why do you use monthly climatology for lightning NO rather than a method based on NLDN? Perhaps the impact of lightning NO representation on model performance noted on Line 387 would be less if actual emissions rather than monthly climatology were used. It would be useful to add a table to the supplemental information that included emissions levels (tons of NOx, CO, VOC) by region (Eastern US, Western US, China, EU, Fires etc) used for the 2 models.

3. Source characterization for specific O3 events:

For many of the episodes listed in table 1, the evidence is suggestive but not compelling for the classifications given.

I suggest adding to Table 1 a characterization of how confident you are in the classification. Based on evidence presented in the paper, the only episode that I would rate a "high confidence" is the June 11th stratospheric intrusion.

I suggest adding fields to Table 1 so that episodes can be more easily compared: H2O mixing ratio (Avg +/- SD), O3:CO slope, O3/NOz. Several of these indicators are mentioned for one of the episode days in the text but not provided for all days so that the reader can compare what they look like during different types of events.

It would be useful if Table 1 provided a more detailed accounting of which lines of evidence were used to classify each of the episodes examined. It seems that in most cases, the ambient data can be indicative of influence from different types of sources

but can't provide quantitative estimates of how much O3 is from US sources versus fires/stratosphere/Asia etc. The model zero out simulations are often the basis for determining that O3 during an episode is primarily from one source. Given this dependence on the model, more detail on model methods and model performance compared to ground-based measurement on each of the episode days is warranted.

3.1. Even characterization for stratospheric intrusions:

-Apr 22-23 and May 13-14 - no details are provided to evaluate these events

-June 11-13 - anti-correlated O3:CO and very dry conditions are compelling for predominant stratospheric impact. I suggest adding this episode to figures S5 and S6 to show what the stratospheric tracer looks like for these days.

-June 14 - Evidence not compelling. O3:CO is positively correlated and slope is similar to slope from "US anthropogenic events". O3/NOz evidence not provided for any other events for comparison and reference from 1998 analysis may not be relevant to current conditions. Note that a recent paper by Henneman et al (2017) found that O3/NOz values have increased over time as US NOx levels have decrease leading to more efficient O3 production. They reported that for Atlanta, the O3/NOz increased from 5.4 in 2001 to 9.3 in 2011. Similarly, in urban Gulfport, FL the ratio increased from 11.1 in 2001 to 20.5 in 2011. So, the range of 1-7 based on Kleinman et al (2002) may not be relevant for current urban conditions in the US. H2O mixing ratio and O3:CO slope appear to be in same range as other events classified as from US anthropogenic influence. Vertical profiles and modeling impacts are not compelling; vertical profiles look similar to what I would expect for local formation with low overnight PBL concentrations due to titration, some mixing down of residual layer in the morning accompanied by increasing boundary layer O3 during the day. What does the stratospheric tracer look like on this day?

3.2. Event characterization for wildfires (June 22): The increased aerosol backscatter aloft on the previous day (Figure 14) is suggestive that wildfires may be advected to

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this general location. The O3:CO and H2O mixing ratio is not convincing. Why are O3 and CO not correlated if they are both originating from the fire event? Also, the H2O mixing ratio looks like it is in the range of what was observed on June 16th, a "US anthropogenic emissions" event. What are typical H2O mixing ratios (average +/- SD) for Las Vegas during June? Is 3.5 g/kg really outside what you might get from normal meteorological variation? In fig 9 it is clear that the models cannot accurately capture O3 on this day, so it does not appear that modeling evidence should be used here leaving us with no way to quantify the impact of wildfires on the ground-level ozone. Do the fire sensitivities described on lines 394-406 impact the vertical profiles of O3? If you were to recreate Figure 9 using those sensitivities would the models be more able to capture O3 vertical profile?

3.3. Event characterization for regional/local pollution events:

-June 16th: O3 and CO are positively correlated suggesting that O3 formed from an emissions source that also emitted CO. This appears to be based primarily on modeled predictions of total O3 and USB. How does this compare to other events? What is the mean bias for ground-level O3 in Las Vegas on this day? Please state the level of elevated ozone aloft measured by TOPAZ (55-70 ppb?)

-June 2 and June 29-30: Not much information provided. Event characterization for Asian transport event (May 24): AIRS CO images from the days leading up to May 24 are suggestive of transport from Asia. No information is provided on O3:CO or H2O mixing ratio. Modeling from AM4 predicts 5-10 ppb influence from Asia. Is 5-10 ppb of O3 from Asia enough to classify this as "Asian transport" alone since O3 as > 70 ppb? Doesn't this suggest that there was at least one other major source? What is the MB for O3 at ground-level in Las Vegas on this day? Also note that the elevated O3 at 1-4 km altitude could also be local/regional O3 in the residual layer from previous days. The elevated O3 at 6-8 km is more definitively long-range transport. For this episode, I would say it is inconclusive. The models predict an Asian contribution, but the observations are not conclusive as they could also be showing local/regional

photochemical production over several days. The evidence from AIRS is suggestive but cannot provide any quantification of how much of the O3 comes from Asia.

3.4. Event characterization for unattributed event (June 28): Much of the evidence here seems to rule out rather than support any particular source. The elevated filament seems uncharacteristic of longer range transport over which time the filament is likely to disperse. Yet the satellite CO suggests possible transport form Asia. The back trajectories suggest possible influence from a California wildfire but the relatively dry air and lack of elevated aerosol backscatter makes it seem unlikely that a narrow filament of O3 could be from a wildfire without substantially elevated PM. The narrow filament of O3 could be from the stratosphere but the elevated CO and CH4 suggest photochemical formation. It should also be noted that Sequoia NP often receives pollution from California's San Joaquin Valley and Los Angeles, so the back trajectories make it seem equally likely that the source is US emissions vs a wildfire. How do the deltaO3/deltaCO and H2O mixing ratio compare to other events. Is the O3:CO slope statistically different from slopes on other days (using 95 % CO from the regression)? Could you use a T-test on the mixing ratio to determine whether it is statistically different from H2O levels on other episode days? One line 307, you indicate that the dryer air during this episode indicates the plume was from Asia or Los Angeles, which is a different conclusion than you present on lines 470-472 that states it is likely from a wildfire plume. What is the model mean bias (in ppb) for ground-level O3 on this day?

4. Conclusions: I think the language in the conclusion is too strong since there appears to be a lot of uncertainty in the characterization of O3 sources for most of the events evaluated in this paper. For instance, I do not believe that this analysis has been successful in "pinpointing sources of observed MDA8 O3" (line 536) and the language in general should reflect the uncertain nature of the conclusions that can be drawn for the analysis so far.

Other specific comments:

-Line 300: did you mean to indicate specific panels of Figure 5?

-Figure 6: suggest marking location of O3 sonde in panel a)

-Figure 7: Suggest using a separate color palette for the rightmost panel of AM4 rather than scaling by 2.5 to avoid confusion by the reader.

-Figure 15: Some but not all of these monitors are shown in the map in Figure 1. I suggest you provide a map that has locations of all monitors used in Figure 15.

-Figure 16: Why do you use 2 different scales in panels a) and b)? It looks like the O3 covers the same range of values in both. It is confusing to the reader when trying to compare the results from the various panels. I suggest just using the color range from panel a) for all panels in this figure.

-Figure 17: Note that the upper tail of O3 concentrations simulated by AM4 is over-predicted. Since this upper tail is important for characterizing O3 events above the NAAQS, this should be discussed in the enhanced model performance section recommended above.

-Figure 17 and S14: I suggest you add analogous figures that shows this same information but for CASTNET sites in the SW US at low elevation. It is important to be able to compare the results you are finding for high-elevation sites to what is predicted at lower elevation sites.

-Figure S6: Suggest showing the June 11-14 event in this figure as well.

-Table S1: suggest adding AM4 and GEOS-Chem model statistics of Apr-June 2016 O3 simulations to this table (mean bias, r etc).

-Table S2: Table states that MEGAN was used in AM4 simulations, but this is not clear in description in section 2.3. If MEGAN emissions were used for AM4, then add this to 1st paragraph of section 2.3. Also, Table refers to “section 2.4” which does not appear to exist in the paper.

-Note that the US O3 NAAQS is only exceeded when O3 is > than the level of the standard, not >= the level and that ozone values are truncated to the nearest ppb. Therefore, a measured concentration of 70.9 ppb is meeting the current NAAQS. When showing levels that violate the standard either in figures or tables, a line at 71 ppb would be more appropriate than a line at 70 ppb. For instance, in Figure S12, the cutoff for the blue bars should be >= to 71ppb.

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