Dear Dr. West:

We truly appreciate the reviewers for carefully reading the manuscript and providing helpful suggestions. Below we include a point-by-point response (in bold blue) to the reviews, responding to *their comments (in italic)* and explaining the changes made to the manuscript (in light blue).

Best regards,
Meiyun Lin (on behalf of the authors)
May 5, 2020

Reviewer #1

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 areas 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.

RE: Thank you for the comments. We have revised the manuscript following your suggestions.

Major comments:

2. *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).*

RE: The relevant discussion in the Introduction has been rephrased (see Lines 80-85):

"Large inter-model differences not only exist in seasonal means but also in day-to-day variability (e.g., Fiore et al., 2014; Dolwick et al., 2015; Jaffe et al., 2018). An event-oriented multi-model comparison,

tied closely to intensive field measurements, is needed to provide process insights into the model discrepancy. ”

We have also cited Dolwick et al (2015) and Emery et al (2012) in Section 5 (see Lines 1040-1050).

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.

RE: We have moved the time series analysis of MDA8 O3 to Section 3 and added the statistics for model performance at all CASTNet sites in Table S1. The purpose of Section 3 is to provide an overall evaluation of the models. The event-specific evaluation of the models is discussed in Section 4.

-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).

RE: We agree with the reviewer that the 50-km resolution of AM4 may be insufficient to represent the local-scale photochemical production. We have added some related discussions in Section 4.4 – Regional and local anthropogenic pollution events.

-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?

RE: This is now clarified in the revised manuscript (Line 197):

"The high-resolution BASE and sensitivity simulations for January – June 2017 are initialized from the corresponding nudged C96 (~100x100 km^2) simulations spanning from 2009 to 2016 (8 years)."

-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.

RE: Thank you for the comment. This is clarified in the revised manuscript (Lines 178-182; Section 2.2).

"We implement a stratospheric O_3 tracer (O3Strat) in GFDL-AM4 to track O3 originating from the stratosphere. The O3Strat is defined relative to a dynamically varying e90 tropopause (Prather et al., 2011) and is subject to chemical loss in the same manner as odd oxygen of tropospheric origin and deposition to the surface (Lin et al., 2012a; Lin et al., 2015a)."

-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.

RE: The GEOS-Chem simulations period and the spin-up process are now clarified in the revised manuscript (Line 223).

"We conduct two nested high-resolution simulations with GEOS-Chem for February-June 2017: BASE and a USB simulation with anthropogenic emissions zeroed out in the U.S. (Table S3). Initial and boundary conditions for chemical fields in the nested-grid simulations were provided by the corresponding BASE and USB GEOS-Chem global simulations at 2° × 2.5° resolution for January-June 2017. Only April-June results from the nested simulations are analysed in this study. The three-month spin-up period (January-March) used for GEOS-Chem is relatively short compared to the multi-year GFDL-AM4 simulations, although it should be sufficient given that the lifetime of ozone in the free troposphere is approximately three weeks (e.g., Young et al., 2016)."

-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 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.

RE: We only apply the NOx emissions adjustments to the Eastern US. To our knowledge, the findings of Travis et al. are not just limited to the NEI 2013 used in their study. They imply that NOx emissions over the eastern U.S. in all global emission inventories may be overestimated since almost all of the current global models using a varies of emission inventories in different years overestimate surface ozone in the southeast US.

Comparison of regional NO_x, CO, and NMVOCs emissions from AM4 and GC is shown in Table S1.

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.

RE: We have added H_2O mixing ratios in Table 1 and revised the event classification to reflect uncertainties. We discussed in the text how the attribution in Table 1 is done (Lines 325-400):

"The attribution is based on a combination of observational and modeling analyses. First, we examine the O_3/CO/H_2O relationships and collocated meteorological measurements from the NOAA/ESRL mobile lab deployed at Angel Peak to provide a first guess on the possible sources of the observed high-O_3 events (Section 4.1). Then, we analyze large-scale meteorological fields (e.g., potential vorticity), satellite images (e.g., AIRS CO), and lidar and ozonesonde observations to examine if the transport patterns, the high-O_3 layers and related tracers are consistent with the key characteristics of a particular source (Section 4.2-4.5). Available aerosol backscatter measurements and multi-tracer aircraft profiles are also used to support the attribution (Sections 4.3 and 4.6). Finally, for each event we examine the spatiotemporal correlations

of model simulations of total O_3, background O_3, and its components (e.g., stratospheric ozone tracer), both in the free troposphere and at the surface. For a source to be classified as the dominant driver of an event, O_3 from that source must be elevated sufficiently from its mean baseline value. ".

3.1. *Even characterization for stratospheric intrusions:*

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

RE: These two events occurred before the FAST-LVOS campaign began. However, the stratospheric influence on these two events is evident in the time series analysis of MDA8 O_3 now shown in Fig.4, as we discussed in Section 3.2. More detailed analyses are provided in the Supplemental Material. We have clarified these in Table 1.

-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.

RE: Thanks. We have added anomalies in AM4 stratospheric ozone tracer for June 11-14 in Fig.9 and discussed this in the main article (Section 4.2, Lines 530-535).

"Over the areas where observed MDA8 O_3 levels are 60–75 ppbv, GFDL-AM4 estimates 50–65 ppbv USB O_3 with simulated O_3Strat 20–40 ppbv higher than its mean baseline level in June."

-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?

RE: The June 14 event likely resulted from mixing of regional anthropogenic pollution and transported stratospheric O_3 residual from the previous day, as we clarified in Table 1 and discussed in the revised manuscript:

"*Mixing of stratospheric ozone with regional pollution on June 14:*

Stratospheric air masses that penetrate deep into the troposphere can mix with regional anthropogenic pollution and gradually lose their typical stratospheric characteristics (cold and dry air containing low levels of CO), challenging diagnosis of stratospheric impacts based directly on observations (Cooper et al., 2004; Lin et al., 2012b; Trickl et al., 2016). On June 14, O_3 measured at Angel Peak is positively correlated with CO ($\Delta O_3/\Delta CO = 0.75$; Fig. 6b), similar to conditions of anthropogenic pollution on June 16 (Fig. 6c–d). TOPAZ lidar shows elevated O_3 of 70–80 ppbv concentrated within the boundary layer below 3 km altitude (Fig. 8b). These observational data do not provide compelling evidence for stratospheric influence. However, GFDL-AM4 simulates elevated O_3Strat coinciding with the observed and modeled total O_3 enhancements within the PBL, indicating that O_3 from the deep stratospheric intrusion on the previous day may have been mixed with regional anthropogenic pollution to elevate O_3 in the PBL. At the surface (the bottom panel in Fig.9), AM4 simulates high USB O_3 and elevated O_3Strat (20-40 ppb above its mean baseline) over Arizona and New Mexico where MDA8 O_3 greater than 70 ppb were observed. The fact that GEOS-Chem is unable to simulate the ozone enhancements in lidar measurements and at the surface further supports the possible stratospheric influence. This case study demonstrates the value of integrating observational and modeling analysis for the attribution of high-O_3 events over a region with complex O_3 sources. **"**

We agree that the discussion on O3/NOz ratios is not relevant here and thus have deleted it from the revised manuscript.

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 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?*

RE: We agree with the reviewer that the increased aerosol backscatter aloft is a more compelling evidence of the wildfire influence, in contrast to the O3/CO/H2O relationship, during this event. Thus, we have moved the aerosol backscatter analysis (previously Fig.14) to Section 4.3 (now Fig.10) to support the discussions. Indeed, the models have difficulty accurately simulating the observed ozone enhancements during this event. We have stated

the uncertainties both in Section 4.3 and in the Conclusions as the reviewer suggested. Particularly, in the Conclusions, we stated:

"The two models also differ substantially in total and background O_3 simulations during the June 22 wildfire event. GEOS-Chem captures the broad O_3 enhancement in lidar observations but overestimates surface MDA8 O_3 at some sites during this event. It remains unclear whether higher USB O_3 simulated by GEOS-Chem during this event is from greater O_3 produced from wildfire emissions or excessive lightning NO_x emissions in the model. Although GFDL-AM3 captures the observed interannual variability in O_3 enhancements from large-scale wildfires over the WUS (Lin et al., 2017), GFDL-AM4 has difficulty simulating the observed O_3 enhancements during the relatively small-scale wildfire event on June 22. Sensitivity simulations with fire emissions constrained at the surface or with part of fire NO_x emissions emitted as PAN do not substantially improve simulated O_3 on June 22. Wildfires typically occur under hot and dry conditions which also enable the buildup of O_3 produced from regional anthropogenic emissions, complicating an unambiguous attribution of the high-O_3 events solely based on observations. Screening of exceptional events due to wildfire emissions remains a serious challenge."

Regarding the poor correlation between CO and O_3 measured at Angel Peak, we added the following discussions in Section 4.1:

"In particular, exceptionally high CO levels (~100–440 ppbv) on June 22 (Fig. 6e) suggest influences from wildfires. Ozone enhancements were measured by the TOPAZ ozone lidar on June 22 (Section 4.3) although the correlation between CO and O_3 at Angel Peak is not strong. The net production of O_3 by wildfires is highly variable, with many contradictory observations reported in the literature (Jaffe and Wigder 2012). The amount of O_3 within a given smoke plume varies with distance from the fire and depends on the plume injection height, smoke density, and cloud cover (Faloona et al., 2020)."

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.

RE: We have revised discussions on the regional pollution events: June 16, June 2, and June 29-30. Please see tracked changes in Section 4.4.

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.

RE: We agree completely that elevated ozone at 6-8 km altitude reflects the Asian influence while elevated O3 at 1-4 km altitude appears to be influenced by local/regional pollution. We have revised the discussions accordingly. Particularly, we discuss whether the Asian contribution is sufficiently larger than its average value, as suggested by Reviewer #2:

"Both GFDL-AM4 and GEOS-Chem capture the observed O_3-rich plumes at surface–4 km and 6–8 km altitude above Clark County during this event. Elevated O_3 at 6-8 km altitude reflects the long-range transport from Asia, as supported by concurrent enhancements in total and USB O_3 in both models and by the large difference in the AM4 BASE simulation and the sensitivity simulation with Asian anthropogenic emissions zeroed out. Elevated O_3 at 1-4 km altitude appeared to be influenced by a residual pollution layer from the previous day; this plume was later mixed into the growing PBL (up to 4 km altitude), elevating MDA8 O_3 in surface air on May 24. Further supporting the impact from regional or local pollution below 4 km altitude, both models simulate much larger enhancements in total O_3 (70-90 ppbv) than in USB O_3 (~50 ppbv).

On May 24th, MDA8 O_3 approached or exceeded the 70-ppbv NAAQS at multiple sites in California, Idaho, Wyoming, and Nevada (Fig. 15a), likely reflecting the combined influence from regional pollution and the long-range transport of Asian pollution. MDA8 O_3 at four surface sites in Clark County was above 65 ppbv. More exceedances would have occurred if the level for the NAAQS were lowered to 65 ppbv. In parts of Idaho, Wyoming, California where observed MDA8 O_3 were higher than 60 ppbv, the contribution of Asian anthropogenic emissions as estimated by GFDL-AM4 were 8–15 ppbv (Fig. 15a), much higher than the springtime average contribution of ~5 pppv estimated by previous studies (e.g., Lin et al., 2012), supporting the episodic influence from Asian pollution during this event. At several high-elevation sites in California such as Arden Peak (72 ppbv) and Yosemite National Park (70 ppbv) where observed MDA8 O_3 exceeds the NAAQS level, the contribution of Asian pollution is approximately 9 ppbv. Ozone produced from regional and local anthropogenic emissions dominates the observed MDA8 O_3 above 70 ppbv in the Central Valley of California. "

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?*

RE: Indeed, discussions for this event rely heavily on limited observational evidence. We have revised the discussions and provided information you asked for when possible. Please see tracked changes in Section 4.6.

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.*

RE: Thank you for the suggestion. We have revised the Conclusions to reflect uncertainties. Please see tracked changes in Section 6. Specifically, we follow the suggestions by Reviewer #2 and discuss whether the models are useful for screening of exceptional events due to STT and wildfires.

Other specific comments:

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

RE: Revised.

2. *Figure 6: suggest marking location of O3 sonde in panel a)*

RE: Done.

3. *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.*

RE: Considering that Fig 7 is large and busy, a separate color palette will make the figure difficult to read. We've stated in the figure caption that the stratospheric ozone tracer was scaled by 2.5.

4. *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.*

RE: Revised. Now all sites are shown in Figure 1 and Table S1.

5. *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.*

RE: Done.

6. Figure 17: Note that the upper tail of O3 concentrations simulated by AM4 is overpredicted. 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.

RE: Thank you. We have added the discussions in the revised manuscript.

7. 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.

RE: Good suggestion. There are only a few low-elevation CASTNET sites in the SWUS. We aid some analyses for the AQS sites in the Clark County. Due to coarse model resolution (particularly AM4), most of the sites reside in the same model grid, making the comparison not meaningful. We agree completely with the reviewer that it is important to show the differences between high-elevation and low-elevation sites. We plan to do this in a forthcoming study using a higher resolution version of the models.

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

RE: Anomalies in AM4 O_3Strat for the June 11-14 event are now shown in Fig.9 in the main article.

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

RE: Done and the results are briefly discussed in Section 3.

10. *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.*

RE: This is now clarified in Section 2.3.

11. *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.*

RE: Thank you for your insight. We have avoided using the term such as "standard-exceeding" as it has a specific policy meaning.

Response to Reviewer #2

Reviewer #2

This paper leverages intensive field measurements from the Fires, Asian, and Stratospheric Transport-Las Vegas Ozone Study (FAST-LVOS) and two global chemistry models to attribute high ozone events in the Las Vegas area to various causes: wildfires, stratosphere to troposphere ozone intrusions and transport from Asia. For many of the events the disagreement between the two models is notable. Overall, I found this to be an interesting, well-written paper with well-crafted figures. After the authors address a few points I would recommend publication.

RE: Thank you for your supporting comments. We have revised the manuscript following your suggestions.

Major Comments:

I. *It is unclear how the different events are attributed to different sources. A more clear discussion of attribution is necessary.*

(i) *As the author's state: "Identifying the primary source of the high-O3 events solely based on observations is challenging; additional insights from models thus needed as we demonstrate below." Thus, how are the events attributed in Table 1? Is the measured data in section 4.1 used to support the modeled attribution? Or is it the primary attribution mechanism? Are both models used to attribute a particular event or just one? Is the event attribution through the preponderance of evidence?*

(ii) *On a more philosophical note it seems the authors often make good qualitative arguments for a particular type of event but is this sufficient to attribute an event to a particular cause? In particular, as air from various sources is mixes together how does one determine the cause of an ozone exceedance? Quantitatively, doesn't one need to know the average ozone contribution from a particular source and if ozone from that source is elevated sufficiently from its average one can attribute the ozone exceedance to that source (even if it is not the dominant source)? Or does one require a particular source to be the dominant source to attribute an event to it? At any rate more detail should be given as to what it means to attribute an event to a source.*

RE: Thank you for the suggestion. In Lines 325-400, we have clarified how the attribution in Table 1 is done. Please see also our Reponses to Reviewer 2 - Source characterization for specific O_3 events. For a source to be classified as the dominant driver of an event, O_3 from that source must be elevated sufficiently from its mean baseline value. We have followed this guideline throughout the manuscript for discussions of each event. For example, for the STT events, we show that on days and locations when observed MDA8 O3 exceeded 65 ppbv, AM4 O_3Strat is 20-40 ppbv above its mean baseline level.

II. *The difference between the models in Figure 6 is deeply disturbing to me. While the authors concentrate on the differences in the stratospheric ozone intrusion, the figures overall are very different, not only in their ozone but in the isentropes. Is this a resolution problem, an interpolation problem, a meteorological analysis problem or possibly a result of differences in the advection algorithm? This seems quite important to determine as one of the main differences between the models seem to be in their handling of stratospheric intrusions. It seems as a minimum the authors could look at: (i) potential vorticity and potential temperature surfaces in the native resolution of the two meteorological datasets. Are these the same or different? (ii) Then examine potential vorticity and potential temperature in the resolution of the two model grids. Does changing the model grid do something to the fields? (iii) Finally they could examine the ozone differences between the figures. Is the ozone similar in the stratosphere in general in the two models which would point to greater numerical diffusion in GEOS-CHEM diluting the stratospheric intrusion? Or perhaps GEOS-CHEM has less ozone in the stratosphere. The paper seems to imply that the simplified stratospheric chemistry and dynamics in GEOS-CHEM is the reason for the discrepancy between the two models. What evidence do the authors have for this assertion?*

RE: Thank you for your insight. We have revised Fig.6 and the isentropes between the two models are similar. We have also conducted the additional analysis suggested by the reviewers. As shown in Supplemental Fig.S6, the synoptic-scale patterns of potential vorticity at 250 hPa and ozone levels in the UTLS are similar between the two models. We added the following statement in the revised manuscript:

"For comparison, GEOS-Chem simulates a much weaker and shallower intrusion (Fig. 7b), despite the similar synoptic-scale patterns of potential vorticity at 250 hPa and ozone levels in the UTLS (Fig.S6), suggesting possibly greater numerical diffusion in GEOS-CHEM diluting the stratospheric intrusion"

III. *When comparing differences between the AM4 and GEOS-CHEM model it would be useful to know the extent to which these differences might be due to differences in emissions. At a minimum*

the authors should discuss some of the emission differences, and the extent to which these might contribute to the difference in the USB ozone. Ideally these type of studies should be done with the same emissions. However, the authors should take any emission differences into account in their analysis, or at the least discuss that these may be a source of uncertainty in analyzing the differences between the models.

RE: Good point! We have summarized regional anthropogenic and fire emissions of NOx, CO, and NMVOCs in the U.S, China, and Europe in Table 1. Overall, the differences in anthropogenic and fire emissions used in the models are very small. The largest emission discrepancies are likely from lightening NOx emissions as we discussed in the paper. Unfortunately, lightening NOx calculated by the models are not archived in these simulations.

IV. *One of the important aspects of this study seems to be in the attribution of exceedances. I think the authors should quantitatively assess the skill of the models in diagnosing exceedances (perhaps over a longer period of time than the FAST-LVOS timeframe itself, if possible). There are many measures of this skill in the literature. What percent of time when an exceedance is measured do each of the models predict the exceedance? And how often do the models get a false positive (predict an exceedance when one doesn't occur)?*

RE: We now report the percentage of site-days with MDA8 O3 above 70 or 65 ppbv at Clark County and CASTNet sites in Table S5. The results are briefly discussed in Section 5.

" Tables S4 and S5 report year-to-year variability in the percentage of site-days with springtime MDA8 O_3 above 70 ppbv (or 65 ppbv) and simulated USB levels during 2010–2017. The percentage of site-days with MDA8 O_3 above 70 ppbv during April–June 2017 is 0.9% from observations at CASTNet sites, 2.0% from GFDL-AM4, and 0.1% from GEOS-Chem. GFDL-AM4 captures some aspects of the observed year-to-year variability despite mean-state biases. For example, the observed percentage of site-days with MDA8 O_3 above 70 ppbv at CASTNet sites is highest (9.4%) in April-June 2012, compared to 3.1±3.2% for the 2010–2017 average. That statistics from GFDL-AM4 are 7.7% for 2012 and 4.0±2.9% for the 2010–2017 average. May–June mean USB MDA8 O_3 at Clark County sites are 50.9 ppbv in 2017, 55.3 ppbv in 2012, and 52.3±2.0 ppbv for the 2010–2017 average. Supporting the conclusions of Lin et al. (2015a), these results indicate that background O_3, particularly the stratospheric influence, is an important source of the observed year-to-year variability in high-O_3 events over the WUS during spring."

V. *I feel the conclusions could be made stronger. The authors give for the most part a detailed comparison between the two models. I think the paper would be strengthened if the authors stepped back a little.*

(i) First, it would be interesting to discuss model skill in simulating the exceptional events as discussed in the first paragraph in the paper. Giving the difference between the two model

simulations to what extent are we confident that they can screen exceptional events? What is the skill of the models in assessing extreme events (see comment IV).

(ii) Second, in the case of an exceptional event, to what extent can these models be used to attribute the event to a particular cause?

(iii) The authors claim that much of the pattern of USB in AM4 is due to STT and the ability to simulate STT is an important difference between the models. This seems like an important conclusion, but how strong is the evidence? It would be good if the authors would summarize the reasons they conclude this. Do the differences in USB coincide with locations where the stratospheric tracer is high? Some more analysis might be beneficial to really make this point.

(iv) While the authors point out some differences in USB it would be nice to quantify the uncertainty here. I think a difference map in the USB between the two models would be very helpful. Do we know USB within 10% or 20% (at least based on these two models) and how important is this for policy considerations?

RE: Thank you for the suggestion. We have revised the Conclusions substantially to include discussions on uncertainties and whether the models can be used for screening of exceptional events due to STT or wildfires. Please see tracked changes in Section 6.

Minor Comments:

1. *P2, l47 "contribute". Should this be contribute episodically?*

RE: Yes. Revised.

2. *P2, l58 "independent". This seems a bit strong. As has been shown in climate models (e.g., Knutti et al., 2013) models are really not independent from each other due to the sharing of information and algorithms across groups. For example the two models in this paper share the MEGAN scheme, but probably also share other aspects. Thus, I would delete the word "independent" here and in other locations.*

RE: Good point. We have deleted "independent" in the revised manuscript.

3. *Fig. 6, Please make the vertical and horizontal scales identical so these figures are easier to compare.*

RE: Revised as suggested.

4. *P6, l46 Please give the frequency of measurements used here and elsewhere.*

RE: Done.

5. *P7, l188 What is GEOS-FP meteorology?*

RE: Clarified in the revised manuscript:

"… using the Goddard Earth Observing System – Forward Processing (GEOS-FP) assimilated meteorological data."

6. *P8, l229 What is the standard representation of lightning?*

RE: Rephrased to:

"The model calculates lightning NO_X emissions using monthly climatology of satellite lightning observations coupled to model deep convection (Murray et al., 2012). The calculation of lightning NO_X in this study differs from that in Zhang et al. (2014), who used the U.S. National Lightning Detection Network (NLDN) data to constrain model flash rates. "

7. *P9, l235 Are the emissions the same in AM4 and its predecessor? If not to what extent is the comparison between them simply a matter of the different emissions. At the minimum the authors should mention these comparisons use different emissions and the extent to which these differences can explain the differences between the models.*

RE: We noted in the revised manuscript:

"These improvements are mainly credited to the changes in dynamics/convection schemes in AM4 (Zhao et al., 2018a), according to our sensitivity simulations (not shown). The difference in emissions inventories contribute to some of the O_3 differences but is not the major cause because the largest differences between the two models in simulated free tropospheric O_3 occur during the cold months (November-April) when photochemistry is weak (Fig.2b). "

8. *P12, l319. PVU is not a unit. Please give the mks units for a PVU.*

RE: Done.

9. *P14, l394. The authors suggest excessive lightning NOX in GEOS-chem causes excessive ozone. They cite a number of older papers to make their point. What is the evidence in this study for excessive lightning NOX? For example, P18 l496 the authors state the overestimate is likely due to lightning NOX. On P20 l569 the author categorically state it is the abundance of lightning NOx that results in higher background ozone in GEOS-chem. Without more analysis it seems lightning NOX is a possible explanation. However, if they authors claim this is the likely explanation they need to give some more evidence.*

RE: The influence of excessive lightning NOx emissions on the overestimates of background ozone has been discussed in detail by Zhang et al. (ACP, 2014). We have cited Zhang et al. (ACP, 2014) to support our discussions.

10. *P14, section 4.3: The authors made a number of sensitivity simulations with respect to the simulation of fire plumes. I did not get a sense as to which of these sensitivities improved or degraded the simulation. Please give some overall conclusions.*

RE: Done. Please see Line 783 and the Conclusion section.

11. *P15, l14 "dominant source". Could you clarify? If the local emissions are 20-30 ppb and simulated emissions are over 60 ppb, why are local emissions the dominant source?*

RE: Rephrased to

"Both models show boundary layer O_3 enhancements in total O_3 simulations but not in USB simulations (Fig. 11b), indicating that regional or local anthropogenic emissions are the primary source of observed O_3 enhancements."

12. *P17 l481 How were the STT events diagnosed?*

RE: That sentence has been rephrased and moved to Section 3. We now say:

"The two models show large differences in simulated total and USB O_3 on days when AM4 O_3Strat indicates a stratospheric influence (highlighted in blue shading). AM4 O_3Strat indicates frequent STT events during April–June with MDA8 O_3 exceeding or approaching the current NAAQS of 70 ppbv."

13. *P17 l486 "underestimates the magnitude of STT". The authors show that GEOSCHEM underestimates the ozone concentrations in stratospheric folds, at least the ones they examined. First this sentence needs to be qualified as to where and when. Secondly while GEOS-CHEM may underestimate the ozone in the stratospheric intrusions this does not mean it underestimates STT (the model might simply be excessively diffusive while still simulating the same exchange).*

RE: That sentence has been rephrased and moved to Section 3.2. Lines 310-320:

"The two models show large differences in simulated total and USB O_3 on days when AM4 O_3Strat indicates a stratospheric influence (highlighted in blue shading)."

14. *P18 l519, and p19 l20: "Many of the standard O3 events. . ." Could you quantify this? From the figure it appears to be less than half the events?*

RE: Rephrased.

15. *P19 l549 "contributing to ~30ppbv to surface ozone". I believe this is a modeling result. Please state this.*

RE: Rephrased.

16. *P20 l555 "wildfire event". This would be a good place to summarize whether any of the sensitivity tests resulted in a better capture of the wildfire event.*

RE: Agreed. Please see below in the Conclusion section:

"Although GFDL-AM3 captures the observed interannual variability in O_3 enhancements from large-scale wildfires over the WUS (Lin et al., 2017), GFDL-AM4 has difficulty simulating the observed O_3 enhancements during the relatively small-scale wildfire event on June 22. Sensitivity simulations with fire emissions constrained at the surface or with part of fire NOx emissions emitted as PAN do not substantially improve simulated O3 on June 22. Wildfires typically occur under hot and dry conditions which also enable the buildup of O3 produced from regional anthropogenic emissions, complicating an unambiguous attribution of the high-O3 events solely based on observations. Screening of exceptional events due to wildfire emissions remains a serious challenge."

17. *P18, l500 "likely reflect": what are the arguments for this? It might be interesting to show a difference map for USB, getting at the uncertainty in USB between two state-of-the-art models.*

RE: The difference map for USA is now shown in Fig.S12.

Characterizing sources of high surface ozone events in the southwestern U.S. with intensive field measurements and two global models

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Abstract

The detection and attribution of high background ozone (O_3) events in the southwestern U.S. is challenging but relevant to the effective implementation of the lowered National Ambient Air Quality Standard (NAAQS; 70 ppbv). Here we leverage intensive field measurements from the Fires, Asian, and Stratospheric Transport–Las Vegas Ozone Study (FAST-LVOS) in May–June 2017, alongside high-resolution simulations with two global models (GFDL-AM4 and GEOS-Chem), to pinpoint the sources of O_3 during high-O_3 events. We show stratospheric influence on four out of the ten events with daily maximum 8-hour average (MDA8) surface O_3 above 65 ppbv in the greater Las Vegas region. While O_3 produced from regional anthropogenic emissions dominates pollution in the Las Vegas Valley, stratospheric intrusions can mix with regional pollution to push surface O_3 above 70 ppbv. GFDL-AM4

captures the key characteristics of deep stratospheric intrusions consistent with ozonesondes, lidar profiles, and co-located measurements of O_3, CO, and water vapor at Angel Peak, whereas GEOS-Chem has difficulty simulating the observed features and underestimates observed O_3 by ~20 ppbv at the surface. On days when observed MDA8 O_3 exceeds 65 ppbv and AM4 stratospheric ozone tracer shows 20-40 ppbv enhancements, GEOS-Chem simulates ~15 ppbv lower U.S. background O_3 than GFDL-AM4. The two models also differ substantially during a wildfire event, with GEOS-Chem estimating ~15 ppbv greater O_3, in better agreement with lidar observations. At the surface, the two models bracket the observed MDA8 O_3 values during the wildfire event. Both models capture the large-scale transport of Asian pollution, but neither resolves some fine-scale pollution plumes, as evidenced from aerosol backscatter, aircraft, and satellite measurements. U.S. background O_3 estimates from the two models differ by 5 ppbv on average (greater in GFDL-AM4) and up to 15 ppbv episodically. Our multi-model approach tied closely to observational analysis yields process insights, suggesting that elevated background O_3 may pose challenges to achieving a potentially lower NAAQS level (e.g., 65 ppbv) in the southwestern U.S.

Keywords: background ozone, stratospheric intrusions, wildfires, Asian pollution

1 Introduction

Surface ozone (O_3) typically peaks over the high-elevation southwestern U.S. (SWUS) in late spring, in contrast to the summer maximum produced from regional anthropogenic emissions in the low-elevation eastern U.S. (EUS). The springtime O_3 peak in the SWUS partly reflects the substantial influence of background O_3 from natural sources (e.g., stratospheric intrusions) and intercontinental pollution (Zhang et al., 2008; Fiore et al., 2014; Jaffe et al., 2018). These "non-controllable" O_3 sources can episodically push surface daily maximum 8-hour average (MDA8) O_3 to exceed the NAAQS (Lin et al., 2012a; Lin et al., 2012b; Langford et al., 2017). Identifying and quantifying the sources of springtime high-O_3 events in the SWUS has been extremely challenging owing to limited measurements, complex topography, and various O_3 sources (Langford et al., 2015). As the O_3 NAAQS becomes more stringent (lowered from 75 ppbv to 70 ppbv since 2015), quantitative understanding of background O_3 sources is of great importance for screening exceptional events, i.e. "…unusual or naturally occurring events that can affect air quality but are not reasonably controllable using techniques that tribal, state or local air agencies may implement…" (U.S. Environmental Protection Agency, 2016). Here we leverage intensive measurements

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from the 2017 Fires, Asian, and Stratospheric Transport-Las Vegas Ozone Study (FAST-LVOS; Langford et al., manuscript in preparation), alongside high-resolution simulations with two global atmospheri chemistry models (GFDL-AM4 and GEOS-Chem), to characterize the sources of high-O_3 events in the region. Through a process-oriented analysis, we aim to understand the similarities and disparities between these two widely-used global models in simulating O_3 in the SWUS.

Mounting evidence shows that a variety of sources contribute to the high surface O_3 found in the SWUS during spring. For example, observational and modelling studies show that deep stratospheric intrusions can episodically increase springtime MDA8 O_3 levels at high-elevation SWUS sites by 20–40 ppbv (Langford et al., 2009; Lin et al., 2012a). Large-scale transport of Asian pollution across the North Pacific also peaks in spring due to active mid-latitude cyclones and westerly winds, contributing to high-O_3 events and raising mean background O_3 levels over the SWUS (Jacob et al., 1999; Lin et al., 2012b; Lin et al., 2015b; Langford et al., 2017; Lin et al., 2017). Moreover, frequent wildfires add complexity to the study of O_3 in the SWUS (Jaffe et al., 2013; Baylon et al., 2016; Lin et al., 2017; Jaffe et al., 2018). In the late spring and early summer, increased photochemical activity from U.S. domestic anthropogenic emissions can further complicate the unambiguous attribution of observed high-O_3 events in this region to background influence.

Quantifying the contributions of different O_3 sources relies heavily on numerical models. Previous studies, however, have shown large model discrepancies in the estimates of North American background O (NAB), defined as O_3 that would exist in the absence of North American anthropogenic emissions. Zhang et al. (2011) applied GEOS-Chem to quantify NAB O_3 during March–August of 2006–2008 and estimate a mean NAB O_3 of 40±7 ppbv at SWUS high-elevation sites, while Lin et al. (2012a) estimated an averag of 50±11 ppbv for the late spring to early summer of 2010 with GFDL-AM3. Emery et al. (2012) estimated mean NAB O_3 to be 20–45 ppbv with CAMx and 25–50 ppbv with a regional model driven b GEOS-Chem boundary conditions, during spring-summer. Large inter-model differences not only exist i seasonal means but also in day-to-day variability (e.g., Fiore et al., 2014; Dolwick et al., 2015; Jaffe et al 2018). An event-oriented multi-model comparison, tied closely to intensive field measurements, is needed to provide process insights into the model discrepancy.

Deploying targeted measurements and conducting robust model source attribution are crucial t characterize and quantify the sources of elevated springtime O_3 in the SWUS (Langford et al., 2009,

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Langford et al., 2012; Lin et al., 2012a; Lin et al., 2012b). This is particularly true for inland areas of the SWUS, such as greater Las Vegas, where air quality monitoring sites are sparse, making it difficult to assess the robustness of model source attribution (Langford et al., 2015; Langford et al., 2017). Using field measurements from the Las Vegas Ozone Study (LVOS) in May–June 2013 and model simulations, Langford et al. (2017) provided an unprecedented view of the influences of stratosphere-to-troposphere transport (STT) and Asian pollution on the exceedances of surface O_3 in Clark County, Nevada. This study suggests that O_3 descending from the stratosphere and sometimes mingled with Asian pollution can be entrained into the convective boundary layer and episodically brought down to the ground in the Las Vegas area in spring, adding 20–40 ppbv to surface O_3 and pushing MDA8 O_3 above the NAAQS. However, uncertainties remain in previous analyses due to the use of relatively coarse-resolution simulations and limited measurements to connect surface O_3 exceedances at high-elevation baseline sites and low-elevation regulatory sites. High-resolution simulations and more extensive observations are thus needed to further advance our understanding of springtime peak O_3 episodes in the region.

In May–June 2017, the NOAA Earth System Research Laboratory Chemical Sciences Division (NOAA/ESRL CSD) carried out the FAST-LVOS follow up study in Clark County, NV. During this campaign, a broad suite of near-continuous observations was collected by in situ chemistry sensors deployed at a mountain-top site and by state-of-the-art ozone and Doppler lidars located in the Las Vegas Valley. These daily measurements were supplemented by ozonesondes and scientific aircraft flights during four 2 to 4 day long intensive operating periods (IOPs) triggered by the appearance of upper-level troughs above the U.S. West Coast. These extensive measurements, together with high-resolution simulations from two global models (GFDL-AM4 and GEOS-Chem), provide us with a rare opportunity to pinpoint the sources of elevated springtime O_3 in the SWUS. We briefly describe the FAST-LVOS field campaign and model configurations in Sect. 2. Following an overall model evaluation (Sect. 3), we present process-oriented analyses of the high-O_3 events from deep stratospheric intrusions, wildfires, regional anthropogenic pollution, and the long-range transport of Asian pollution (Sect. 4). Sect. 5 summarizes differences between the simulated total and background O_3 determined by the two models during FAST-LVOS. Finally, in Sect. 6, the implications of the study are discussed.

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2 Measurements and Models

2.1 FAST-LVOS measurement campaign

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The FAST-LVOS experiment was designed to further our understanding of the impacts of STT, wildfires, long-range transport from Asia, and regional pollution on air quality in the Las Vegas Valley. The field campaign was carried out between May 17 and June 30, 2017 in Clark County (NV) which includes the greater Las Vegas area (Fig. 1). The measurement campaign consisted of daily lidar and in situ measurements supplemented by aircraft and ozonesonde profiling during the four IOPs (May 23–25, May 31–June 2, June 10–14, and June 28–30). The daily measurements included chemical composition (e.g., CO and O_3) and meteorological parameters (e.g., air temperature and water vapor) recorded with high temporal resolution by instruments installed in a mobile laboratory (Wild et al., 2017) parked on the summit of Angel Peak (36.32°N, 115.57°W, 2682 m above sea level, a.s.l.), the site of the 2013 LVOS field campaign. This mountain-top site, located ~45 km northwest of the Las Vegas City (see Fig. 1), is far from anthropogenic emission sources and mostly receives free tropospheric air during nights, but is frequently influenced during the day by air transported from the Las Vegas Valley through upslope flow in late spring and summer (Langford et al., 2015). The Tunable Optical Profiler for Aerosols and oZone (TOPAZ) 3-wavelength mobile differential absorption lidar (DIAL) system, which was previously deployed to Angel Peak during LVOS, was relocated to the North Las Vegas Airport (NLVA, Fig. 1) where it measured 8-minute averaged vertical profiles of O_3 and aerosol backscatter from 27.5 m to ~8 km above ground level (a.g.l.) with an effective vertical resolution (for O_3) ranging from ~10 m near the surface to ~150 m at 500 m a.g.l. and ~900 m at 6 km a.g.l. The aerosol backscatter profiles were retrieved at 7.5 m resolution. TOPAZ was operated daily, but not continuously, throughout the campaign. NOAA also deployed a continuously operating micro-Doppler lidar at NLVA to measure vertical velocities and relative aerosol backscatter throughout the campaign. Boundary layer heights were inferred from the micro-Doppler measurements following the method in Bonin et al. (2018).

The routine in situ and lidar measurements described above were augmented during the four IOPs by ozonesondes launched up to four times per day (30 launches total during the entire campaign) from the Clark County Department of Air Quality Joe Neal monitoring site located ~8 km north-northwest of the NLVA. Aircraft measurements were also conducted by Scientific Aviation to sample O_3, methane (CH_4), water vapor (H_2O), and nitrogen dioxide (NO_2) between NLVA and Big Bear, CA during the IOPs. Readers can refer to our previous studies (Langford et al., 2010; Alvarez II et al., 2011; Langford et al., 2015; Langford et al., 2017; Langford et al., 2019) for detailed descriptions and configurations of the

TOPAZ and the other measurement instruments. The FAST-LVOS field campaign is also described in more detail elsewhere (Langford et al., manuscript in preparation).

The FAST-LVOS measurements were augmented by hourly surface O_3 measurements from Joe Neal and other regulatory air quality monitoring sites operated by the Clark County Department of Air Quality (Table S1). Surface observations of O_3 from these and other mostly urban sites were obtained from the U.S. Environmental Protection Agency (EPA) Air Quality System (AQS; https//www.epa.gov/aqs). We average the AQS measurements into 0.5° × 0.625° grids for a direct comparison with model results (as in Lin et al., 2012a, b). Surface observations from rural sites and more representative of background air were obtained from the EPA Clean Air Status and Trends Network (CASTNet; https//www.epa.gov/castnet).

2.2 GFDL-AM4 and GEOS-Chem

Comparisons of key model configurations are shown in Table S2. AM4 is the new generation of th Geophysical Fluid Dynamics Laboratory chemistry-climate model contributing to the Coupled Mode Intercomparison Project, Phase 6 (CMIP6). The model employed in this study, a prototype version o AM4.1 (Horowitz et al., 2020), differs from the AM4 configuration described in Zhao et al. (2018a, 2018b by including 49 vertical levels extending up to 1 Pa (~80 km) and interactive stratosphere-troposphere chemistry and aerosols. Major physical improvements to GFDL-AM4, compared to its predecessor GFDL-AM3 (Donner et al., 2011), include a new double-plume convection scheme with improved representation of convective scavenging of soluble tracers, new mountain drag parametrization, and the updated hydrostatic FV^3 cubed-sphere dynamical core (Zhao et al., 2016; Zhao et al., 2018a, b). For tropospheric chemistry, GFDL-AM4 includes improved treatments of biogenic VOCs photo-oxidation, photolysis rates, heterogeneous chemistry, and sulfate and nitrate chemistry and deposition processes (Mao et al., 2013a; Mao et al., 2013b; Paulot et al., 2016; Li et al., 2016; Paulot et al., 2017), as described in more details in Schnell et al. (2018). We implement a stratospheric O_3 tracer (O_3Strat) in GFDL-AM4 to track O_3 originating from the stratosphere. The O_3Strat is defined relative to a dynamically varying e90 tropopause (Prather et al., 2011) and is subject to chemical loss in the same manner as odd oxygen of tropospheric origin and deposition to the surface (Lin et al., 2012a; Lin et al., 2015a). The model is nudge to NCEP reanalysis winds using a height-dependent nudging technique (Lin et al., 2012b). The nudging minimizes the influences of chemistry-climate feedbacks and ensures that the large-scale meteorological conditions are similar across the sensitivity simulations. We conduct a suite of AM4 simulations at C192 (~50×50 km^2) horizontal resolution for January–June 2017: (1) a BASE simulation with all emission

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included; (2) a sensitivity simulation with anthropogenic emissions zeroed out over North America (15°–90°N, 165°–50°W; NAB); (3) a sensitivity simulation with anthropogenic emissions zeroed out in the U.S. (USB); (4) a sensitivity simulation with Asian anthropogenic emissions shut off, and (5) a sensitivity simulation with wildfire emissions zeroed out (see Table S3). The high-resolution BASE and sensitivity simulations for January–June 2017 are initialized from the corresponding nudged C96 (~100×100 km^2) simulations spanning from 2009 to 2016 (8 years). Compared to the NAB simulation, the USB simulation includes additional contributions from Canadian and Mexican anthropogenic emissions. The USB estimates are now generically defined as "background O_3" and used by the U.S. EPA. Over the WUS, the vertical model resolution ranges from ~50–200 m near the surface to ~1–1.5 km nea the tropopause and ~2–3 km in much of the stratosphere.

Goddard Earth Observing System coupled with Chemistry (GEOS-Chem; http://geos-chem.org) is a widely-used global chemical transport model (CTM) for simulating atmospheric composition and air quality (Bey et al., 2001; Zhang et al., 2011), driven by assimilated meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO). We used a nested-grid version of GEOS-Chem (v11.01) (Wang et al., 2004; Chen et al., 2009) and conducted high-resolution simulations over North America (10°–70°N, 140°–40°W) at a 0.25° (latitude) × 0.3125° (longitude) horizontal resolution using the Goddard Earth Observing System – Forward Processing (GEOS-FP) assimilated meteorological data. The model uses a fully coupled NOx-Ox-hydrocarbon-aerosol-bromine chemistry mechanism in th troposphere ("Tropchem"), whereas a simplified linearized chemistry mechanism (Linoz) is used in th stratosphere to simulate stratospheric ozone and cross-tropopause ozone fluxes (McLinden et al., 2000). Although GEOS-Chem is also equipped with the Universal tropospheric-stratospheric Chemistry eXtension (UCX) mechanism that simulates interactive stratosphere-troposphere chemistry and aerosols (Eastham et al., 2014), this option was not used in the GEOS-Chem simulations presented in this study due to computational constraints. To further save computational resources, we used a reduced vertical resolution of 47 hybrid eta levels, by combining vertical layers above ~80 hPa from the native 72 level of GEOS-FP. The thickness of model vertical layer over the WUS ranges from ~15–100 m near the surface to ~1 km near the tropopause and in the lower stratosphere. Similar GEOS-Chem simulations with simplified treatments of stratospheric chemistry and dynamics have been previously used to estimate background O_3 for U.S. EPA policy assessments (Zhang et al., 2011; Zhang et al., 2014; Fiore et al., 2014; Guo et al., 2018). Thus, it is important to assess the limitation of this model in representing high

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background O_3 events from stratospheric intrusions. We conduct two nested high-resolution simulations with GEOS-Chem for February–June 2017: BASE and a USB simulation with anthropogenic emission zeroed out in the U.S. (Table S3). Initial and boundary conditions for chemical fields in the nested-grid simulations were provided by the corresponding BASE and USB GEOS-Chem global simulations at 2° × 2.5° resolution for January–June 2017. Only simulations during April–June are analyzed in this study. The three-month spin-up period (January–March) used for GEOS-Chem is relatively short compared to the multi-year GFDL-AM4 simulations, although it should be sufficient given that the lifetime of ozone in the free troposphere is approximately three weeks (e.g., Young et al., 2016)(e.g., Young et al., 2018).

2.3 Emissions

Anthropogenic emissions used in GFDL-AM4 are modified from the CMIP6 historical emission inventory (Hoesly et al., 2018). The CMIP6 emission inventory does not capture the decreasing trend in anthropogenic NO_X emissions over China after 2011 as inferred from satellite-measured tropospheric NO_2 columns (Liu et al., 2016; Fig. S1). We thus scale CMIP6 NO_X emissions over China after 2011 based on a regional emission inventory developed by Tsinghua University (personal communications with Qiang Zhang at Tsinghua University; Fig. S1). The adjusted NO_X emission trend over China agrees well with the NO_2 trend derived from satellite retrievals. We also reduce NO_X emissions over the EUS (25°–50° N, 94.5°–75° W) by 50% following Travis et al. (2016), who suggested that excessive NO_X emissions may be responsible for the common model biases in simulating O_3 over the southeastern U.S. These emission adjustments reduce mean MDA8 O_3 biases in GFDL-AM4 by ~5 ppbv in spring and ~10 ppbv in summer over the EUS (Fig. S2). The model applies the latest daily-resolving global fire emission inventory from NCAR (FINN) (Wiedinmyer et al., 2011), vertically distributed over six ecosystem-dependent altitude layers from the ground surface to 6 km (Dentener et al., 2006; Lin et al., 2012b). Biogenic isoprene emissions (based on MEGAN; Guenther et al., 2006), lightning NO_X emissions, dimethyl sulfide, and sea salt emissions are tied to model meteorological fields (Donner et al., 2011; Naik et al., 2013).

For GEOS-Chem, anthropogenic emissions over the United States are scaled from the 2011 U.S. NEI to reflect the conditions in 2017 (https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data). Similar to AM4, we reduce EUS anthropogenic NO_X emissions in GEOS-Chem by 50% to improve simulated O_3 distributions. Anthropogenic emissions over China are based on the 2010 MIX emission inventory (Li et al., 2017), with NO_X emissions scaled after 2010 using the same trend as in

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GFDL-AM4. Biogenic VOC emissions are calculated online with MEGAN (Guenther et al., 2006). Biomass burning emissions are from the FINN inventory but implemented in the lowest model layer. The model calculates lightning NO_X emissions using monthly climatology of satellite lightning observation coupled to model deep convection (Murray et al., 2012). The calculation of lightning NO_X in this study differs from that in Zhang et al. (2014), who used the U.S. National Lightning Detection Network (NLDN) data to constrain model flash rates.

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3 Overall model evaluation

3.1 GFDL-AM4 versus GFDL-AM3

[Figure 2 about here]

We first compare O_3 simulations in AM4 with its predecessor, AM3, which has been extensively used to estimate background O_3 in previous studies (Lin et al., 2012a; Lin et al., 2012b; Fiore et al., 2014; Lin et al., 2015a). Figure 2 shows the comparisons of simulated and observed March mean O_3 vertical profiles and mid-tropospheric O_3 seasonal cycles at the Trinidad Head and Boulder ozonesonde sites. Free tropospheric O_3 measured at both sites in March is representative of background conditions with little influence from U.S. anthropogenic emissions. Thus, we also show O_3 from the NAB simulations with North American anthropogenic emissions zeroed out. As constrained by available AM3 simulations from previous studies, we focus on the 2010–2014 period and compare the NAB estimates as opposed to th USB estimates used in the rest of the paper. Compared with AM3, simulations of free tropospheric O_3 are much improved in AM4. Mean O_3 biases are reduced by 10–25 ppbv in the middle troposphere and 20–65 ppbv in the upper troposphere in AM4, reflecting mostly an improved simulation of background O_3 (Fig. 2a). These improvements are mainly credited to the changes in dynamics/convection schemes in AM4 (Zhao et al., 2018a), according to our sensitivity simulations (not shown). The difference in emissions inventories contribute to some of the O_3 differences but is not the major cause because the largest differences between the two models in simulated free tropospheric O_3 occur during the cold months (November-April) when photochemistry is weak (Fig.2b).

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3.2 GFDL-AM4 versus GEOS-Chem

[Figure 3 about here]

Next, we examine how GFDL-AM4 compares with GEOS-Chem in simulating mean distribution and day-to-day variability of total and USB O_3 in the free troposphere (Fig. 3) and at the surface (Fig. 4 and

Fig. S3) during FAST-LVOS. Comparisons with ozonesondes at Joe Neal show that total O_3 below 70 hPa simulated by the two models often bracket the observed values (Fig. 3a). Between 700–300 hPa GFDL-AM4 better captures the observed mean and day-to-day variability of O_3, as evaluated with standard deviation. Further comparison with lidar measurements averaged over 3–6 km altitude above La Vegas shows that total and USB O_3 in GFDL-AM4 exhibits larger day-to-day variability than in GEOS-Chem (σ = 8.1 ppbv in observations, 8.1 ppbv in AM4, and 6.7 ppbv in GEOS-Chem; Fig. 3c). For mean O_3 levels in the free troposphere, AM4 estimates a 7 ppbv contribution from U.S. anthropogenic emission (total minus USB), while GEOS-Chem suggests only 3.5 ppbv. The largest discrepancies between the tw models occurred on June 11–13 (the blue shaded period in Fig. 3c), which we later attribute to a stratospheric intrusion event (Sect. 4). During this period, AM4 simulates elevated O_3 (70–75 ppbv broadly consistent with the lidar and sonde measurements, while GEOS-Chem considerabl underestimates the observations by 20 ppbv. Consistent with total O_3, USB O_3 in GFDL-AM4 is much higher than GEOS-Chem on June 11–13.

[Figure 4 about here: Surface MDA8 O_3 time series]

Figure 4 shows the times series of observed and simulated surface MDA8 O_3 at four high-elevation site and one low-elevation site in the region during the study period. Statistics for the comparison of all site are shown in Table S1. The two models show large differences in simulated total and USB O_3 on day when AM4 O_3Strat indicates a stratospheric influence (highlighted in blue shading). AM4 O_3Stra indicates frequent STT events during April–June with MDA8 O_3 exceeding or approaching the curren NAAQS of 70 ppbv. Compared with observations, GFDL-AM4 captures the spikes of MDA8 O_3 an elevated USB O_3 during these STT events (e.g., April 23, May 13, and June 11). On these days, GEOS Chem underestimates observed O_3 by 10–25 ppbv and simulates much lower USB O_3 levels than GFDL AM4. The two models also differ substantially in total and USB O_3 (14–18 ppbv) on June 22 (yellow shading), with GEOS-Chem overestimating observations at high-elevation sites while GFDL-AM4 underestimating observations at both high- and low-elevation sites. We will provide more in-dept analysis of these events in Sect. 4 and identify the possible causes of the model biases.

4 Process-oriented analysis of high-ozone events during FAST-LVOS

[Table 1 about here]

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Deleted: It is also worth noting that more exceedances would have occurred at these sites if a standard of 65 ppbv were implemented (dotted lines in Fig. 15). The O_3 standard-exceeding rate in Clark County would have increased by 3–4 times during late spring to early summer given a 65-ppbv O_3 NAAQS (e.g., increased from 2.4% to 10.8% in 2017; Fig. S12).¶
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We identify ten events with observed MDA8 O_3 exceeding 65 ppbv at multiple sites in the greater La Vegas area during April–June 2017. Table 1 provides an overview of the events, the dominant source fo each event, the surface sites impacted, and associated analysis figures presented in this article. Th attribution is based on a combination of observational and modeling analyses. First, we examine th O_3/CO/H_2O relationships and collocated meteorological measurements from the NOAA/ESRL mobile la deployed at Angel Peak to provide a first guess on the possible sources of the observed high-O_3 event (Sect. 4.1). Then, we analyze large-scale meteorological fields (e.g., potential vorticity), satellite image (e.g., AIRS CO), and lidar and ozonesonde observations to examine if the transport patterns, the high-O layers and related tracers are consistent with the key characteristics of a particular source (Sect. 4.2–4.5). Available aerosol backscatter measurements and multi-tracer aircraft profiles are also used to support th attribution (Sect. 4.3 and 4.6). Finally, for each event we examine the spatiotemporal correlations of mode simulations of total O_3, background O_3, and its components (e.g., stratospheric ozone tracer), both in the free troposphere and at the surface. For a source to be classified as the dominant driver of an event, O from that source must be elevated sufficiently from its mean baseline value.

4.1 Observed O_3/CO/H_2O relationships

[Figures 5-6 about here]

Relationships between concurrently measured O_3 and CO are useful to identify the possible origins o elevated surface O_3 (Parrish et al., 1998; Herman et al., 1999; Langford et al., 2015). During FAST-LVOS in-situ 1-min measurements at Angel Peak show differences in $\Delta O_3/\Delta CO$ and water vapor content betwee air plumes during a variety of events (Figs. 5, 6 and S4). Notably, on June 11, O_3 was negatively correlate with CO ($\Delta O_3/\Delta CO$ = -3.79). This anti-correlation is distinctly different from the O_3/CO relationship during other periods (e.g., $\Delta O_3/\Delta CO$ = 0.68–0.70 on June 16 or $\Delta O_3/\Delta CO$ = 1.08 on June 2). The negativ correlation (high O_3 together with low CO) serves as a strong evidence of a stratospheric origin of the ai masses on June 11, since O_3 is much more abundant in the stratosphere than in the troposphere wherea CO is mostly concentrated within the troposphere where it is directly emitted or chemically forme (Langford et al., 2015). On the contrary, simultaneously elevated O_3 and CO suggests influences by wildfires (e.g., June 22) or anthropogenic (e.g., June 16) pollution (Figs. 6b–d and S4). In particular exceptionally high CO levels (~100–440 ppbv) on June 22 (Fig. 6e) suggest influences from wildfires Ozone enhancements were measured by the TOPAZ ozone lidar on June 22 (Sect. 4.3) although th correlation between CO and O_3 at Angel Peak is not strong. The net production of O_3 by wildfires is highl

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variable, with many contradictory observations reported in the literature (Jaffe and Wigder, 2012). The amount of O_3 within a given smoke plume varies with distance from the fire and depends on the plume injection height, smoke density, and cloud cover (Faloona et al., 2020).

We gain further insights by examining water vapor concurrently measured at Angel Peak. Air masses from the lower stratosphere are generally dry, whereas wildfire/urban plumes from the boundary layer are relatively moist (Langford et al., 2015). Thus, the dry conditions of the air masses on June 11 support our conclusion that the plume was from the lower stratosphere and transported downward to Angel Peak (Fig. 6a). These conditions are in contrast to those of the urban/wildfire plumes transported from the Las Vegas Valley (Fig. 6c–6d). Additionally, we separate the anthropogenic plumes on June 16 into daytime and nighttime conditions because of a diurnal variation of air conditions (relatively dry at night versus wet during daytime; Figs. 6c–d). This analysis further demonstrates that the anthropogenic pollution plume during nighttime is wetter than the stratospheric air on June 11. On June 14 (Fig. 6b), measured O_3 was positively correlated with CO, indicating regional/local pollution influence, but the lower levels of water vapor than those in regional pollution and wildfire plumes suggest that the stratospheric air which reached Angel Peak earlier may have been mixed with local pollution. On June 28 (Fig. 6f), O_3 was positively correlated with CO and the air masses were relatively dry, indicating that the plume was likely from aged pollution transported from Asia or Southern California as opposed to from fresh pollution from the Las Vegas Valley. Identifying the primary source of the high-O_3 events solely based on observations is challenging; additional insights from models are thus needed as we demonstrate below.

4.2 Characteristics of stratospheric intrusion during June 11–14

[Figures 7-8 about here]

Analysis of the 250 hPa potential vorticity and the AM4 model stratospheric O_3 tracers shows significant stratospheric influence on surface O_3 in the SWUS on April 22–23 (Fig. S5), May 13–14 (Fig. S5), and June 11–14 (Figs. 7–8). During these events, surface MDA8 O_3Strat in AM4 was 20-40 ppbv higher than the mean baseline level (15–20 ppbv; see dashed purple lines Fig. 4). Below, we focus on the June 11–14 event, which was the subject of a 4-day FAST-LVOS IOP with 60 hours of continuous O_3 lidar profiling and 13 ozonesonde launches, in addition to continuous in situ measurements at Angel Peak.

Deep stratospheric intrusion on June 11–13

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Synoptic-scale patterns of potential vorticity (PV) indicate a strong upper-level trough over the northwes
U.S. on June 12 (PV = 4–5 PVU in Fig. 7a). The PV pattern displays a "hook-shaped" streamer of ai
extending from the northern U.S. to the Intermountain West, a typical feature for a STT event (Lin et al
2012a; Akritidis et al., 2018). This upper-level trough penetrated southeastwardly towards the SWUS
facilitating the descent of stratospheric air masses into the lower troposphere. Ozonesondes launched a
Joe Neal on June 12 recorded elevated O_3 levels of 150–270 ppbv at 5–8 km altitude (color-coded circle
in Fig. 7b). Consistent with the ozonesonde measurements, GFDL-AM4 shows that O_3-rich stratospheri
air masses descended isentropically towards the study region, with simulated O_3 reaching 90 ppbv at ~
km altitude. For comparison, GEOS-Chem simulates a much weaker and shallower intrusion (Fig. 7b)
despite a similar synoptic-scale pattern of potential vorticity at 250 hPa and ozone levels in the UTLS (Fig
S6), suggesting possibly greater numerical diffusion in GEOS-Chem diluting the stratospheric intrusion.

TOPAZ lidar measurements at NLVA vividly characterize the strength and vertical depth of intruding O
tongues evolving with time (Fig. 8a). A tongue of high O_3 exceeding100 ppbv descended to as low as 2–
km altitude on June 12. GFDL-AM4 captures both the timing and structure of the observed high-O_3 laye
and attributes it to a stratospheric origin as supported by the O_3Strat. In contrast, GEOS-Chem
substantially underestimates the depth and magnitude of the observed high-O_3 layers in the fre
troposphere. Zhang et al. (2014) also showed that GEOS-Chem captures the timing of stratospheri
intrusions but underestimates their magnitude by a factor of 3.

[Figure 9 about here]

Surface observations show that high MDA8 O_3 exceeding 60 ppbv first emerged on June 11 over Southern
Nevada (Fig. 9), consistent with the arrival of stratospheric air masses as inferred from the negativ
correlation between O_3 and CO measured at Angel Peak (Fig. 6a). Over the next few days, the areas wit
observed MDA8 O_3 approaching 70 ppbv gradually shifted southward from Nevada and Colorado t
Arizona and New Mexico. By June 13, observed surface MDA8 O_3 exceeded 70 ppbv over a large
proportion of the SWUS, including Arizona and New Mexico. GFDL-AM4 captures well the observe
day-to-day variability of high-O_3 spots over the WUS, although the model overall has high biases. Ove
the areas where observed MDA8 O_3 levels are 60–75 ppbv, GFDL-AM4 estimates 50–65 ppbv USB O
with simulated O_3Strat 20–40 ppbv higher than its mean baseline level in June. GEOS-Chem
underestimates observed surface MDA8 O_3 by 10–20 ppbv during this event and estimates 15 ppbv lowe
USB than AM4 (Fig. 9). These results are consistent with the fact that GEOS-Chem does not capture th

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Stratospheric air masses that penetrate deep into the troposphere can mix with regional anthropogenic pollution an gradually lose their typical stratospheric characteristics (cold and dry air containing low levels of CO), challenging diagnosi of stratospheric impacts based directly on observations (Coop et al., 2004; Lin et al., 2012b; Trickl et al., 2016). On June 14, O_3 measured at Angel Peak is positively correlated with CO ($\Delta O_3/\Delta CO$ = 0.75; Fig. 5b), similar to conditions of anthropogenic pollution on June 26 (Fig. 5c–d). However, observed $\Delta O_3/\Delta NOz$ in the plume (11.4) was much larger thar those (~1–7) of typical urban plumes (Kleinman et al., 2002). TOPAZ lidar shows elevated O_3 of 70–80 ppbv concentrated within the boundary layer below 3 km altitude (Fig. 7b). GFDL-AM4 captures the observed O_3 enhancements within th PBL and estimates a stratospheric contribution of 20–30 ppbv (30% of the total), suggesting that O_3 from the deep stratospheric intrusion on the previous days had been mixed with regional anthropogenic pollution to elevate O_3 in the PBL on June 14. GEOS-Chem is unable to simulate the observed features. This case study demonstrates the value of integrating ... [4]
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structure and magnitude of deep stratospheric intrusions during the period (Figs. 3, 7, and 8) possibly du
to the simplified treatments of stratospheric chemistry and dynamics (Sect. 2.3).

Mixing of stratospheric ozone with regional pollution on June 14

Stratospheric air masses that penetrate deep into the troposphere can mix with regional anthropogenic pollution and gradually lose their typical stratospheric characteristics (cold and dry air containing low levels of CO), challenging diagnosis of stratospheric impacts based directly on observations (Cooper et al., 2004; Lin et al., 2012b; Trickl et al., 2016). On June 14, O_3 measured at Angel Peak is positively correlated with CO ($\Delta O_3/\Delta CO = 0.75$; Fig. 6b), similar to conditions of anthropogenic pollution on June 16 (Fig. 6c–d). TOPAZ lidar shows elevated O_3 of 70–80 ppbv concentrated within the boundary layer below 3 km altitude (Fig. 8b). These observational data do not provide compelling evidence for stratospheric influence. However, GFDL-AM4 simulates elevated O_3Strat coinciding with the observe
and modeled total O_3 enhancements within the PBL, indicating that O_3 from the deep stratospheric intrusion on the previous day may have been mixed with regional anthropogenic pollution to elevate O_3 in the PBL. At the surface (the bottom panel in Fig. 9), AM4 simulates high USB O_3 and elevated O_3Stra
(20–40 ppbv above its mean baseline) over Arizona and New Mexico where MDA8 O_3 greater than 70
ppbv were observed. The fact that GEOS-Chem is unable to simulate the ozone enhancements in lida
measurements and at the surface further supports the possible stratospheric influence. This case study demonstrates the value of integrating observational and modeling analysis for the attribution of high-O_3 events over a region with complex O_3 sources.

The extent to which stratospheric intrusions contribute to surface O_3 at low-elevation sites over the WUS is poorly characterized in previous studies. Notably, surface O_3 at three low-elevation (~700–800 m a.s.l.) air quality monitoring sites in Clark County exceeded the current NAAQS level of 70 ppbv on June 14: 74 ppbv at Joe Neal, 73 ppbv at North Las Vegas Airport, and 71 ppbv at Walter Johnson. The number of monitoring sites with O_3 exceedances would have increased to eleven in Clark County if the NAAQS had been lowered to 65 ppbv. While O_3 produced from regional anthropogenic emissions still dominates pollution in the Las Vegas Valley (Fig. S7), our analysis shows that stratospheric intrusions can mix with regional pollution to push surface O_3 above the NAAQS.

4.3 Wildfires on June 22

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[Figure 10 about here; Aerosol backscatter]

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Significant enhancements in aerosol backscatter were observed at 3–6 km altitude above NLVA on Jun 21–22, indicating the presence of wildfire smoke (Fig. 10a). Under the impact of the wildfire plume mobile lab measurements at Angel Peak (~3 km altitude) detected elevated CO as high as 440 ppbv in warm and moist air masses (Fig. 6e). The lidar measurements at NLVA on June 22 show broad O enhancements (80–100 ppb) from the surface to 4 km altitude (Fig. 11a). After 12:00 PDT (19:00 UTC) a deep PBL (3–4 km) developed and O_3 within the PBL was substantially enhanced (> 80 ppbv), likel due to strong O_3 production through reactions between abundant VOCs in the wildfire plumes and NO in urban environments (Singh et al., 2012; Gong et al., 2017). Surface MDA8 O_3 exceeded 70 ppbv a multiple sites in the Las Vegas Valley during the event (Table 1). Unfortunately, the synoptic condition did not trigger an IOP, so there was no aircraft or ozonesonde measurement during this event.

GFDL-AM4 has difficulty simulating the O_3-rich plumes above Clark County on June 22 (Fig. 11a) GEOS-Chem captures the observed high-O_3 layers within the PBL but overestimates O_3 at 3–6 km altitud (see also Figs. 3b and 11a), likely due to excessive O_3 produced from lightning NO_x over the southern U.S. (Zhang et al., 2011; Zhang et al., 2014). At the surface, total MDA8 O_3 simulated by the two model bracket the observed values at sites in the Las Vegas area (see yellow shading in Fig. 4) and across th Intermountain West (Fig. 12a). AM4 does not simulate elevated O_3 during this event, while GEOS-Chem simulates elevated total and USB O_3 levels across the entire Southwest region. GEOS-Chem simulations during this wildfire event agree better with the observed MDA8 O_3 enhancements (> 70 ppbv) at Joe Nea (Fig. 4). At the high-elevation sites Angel Peak and Spring Mountain Youth Camp, however, GEOS-Chem overestimates the observed MDA8 O_3 by 10–15 ppbv. Overall, GEOS-Chem seems to be more consistent with observations than GFDL-AM4 during this wildfire event. However, we cannot rule ou the possibility that the better agreement between observations and GEOS-Chem simulations during thi event may reflect excessive O_3 from lightning NO_X in the model (Zhang et al., 2014).

Meteorological conditions (e.g., temperature and wind fields) on June 22 in the reanalysis data used by GFDL-AM4 and GEOS-Chem are similar over the WUS (not shown). The two models use the sam wildfire emissions (FINN) but with different vertical distributions. Fire emissions are distributed between

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the surface and 6 km altitude in GFDL-AM4 but are placed at the surface level in GEOS-Chem. We conduct several sensitivity simulations with GFDL-AM4 to investigate the causes of the model biases. Placing all fire emissions at the surface in GFDL-AM4 results in ±5 ppbv differences in modeled MDA8 O_3 on June 22 (Fig. S8). Observations suggested that 40% of NO_X can be converted rapidly to PAN and 20% to HNO_3 in fresh boreal fire plumes over North America (Alvarado et al., 2010). Both models currently treat 100% of wildfire NO_X emissions as NO. We conduct an additional AM4 sensitivity simulation, in which 40% of the wildfire NO_X emissions are released as PAN and 20% as HNO_3. This treatment results in ±2 ppbv differences in simulated monthly mean MDA8 O_3 during an active wildfire season (August 2012; Fig. S9). Overall, these changes do not substantially improve simulated O_3 on June 22. Future efforts are needed to investigate the ability of current models to simulate O_3 formations in fire plumes (Jaffe et al., 2018).

4.4 Regional and local anthropogenic pollution events

[Figure 12 about here]

Regional and local anthropogenic emissions were important sources of elevated O_3 in Clark County during FAST-LVOS, contributing to three out of ten observed high-O_3 events above 65 ppbv during April–June 2017 (Table 1). Below, we focus on the June 16 event when severe O_3 pollution with MDA8 O_3 exceeding 70 ppbv occurred over California, Arizona, parts of Nevada, and New Mexico. Analysis for the June 1 and June 29–30 pollution events are shown in the supplemental material (Figs. S4, S10, and S11). The TOPAZ lidar measurements on June 16 show elevated O_3 of 55–90 ppbv in the 4-km–deep PBL (Fig. 11b). However, this event did not trigger an IOP, so ozonesonde and aircraft measurements are unavailable. Both GFDL-AM4 and GEOS-Chem capture the buildup of O_3 pollution in the PBL on June 16 (Fig. 11b). Both models show boundary layer O_3 enhancements in total O_3 simulations but not in USB simulations (Fig. 11b), indicating that regional or local anthropogenic emissions are the primary source of observed O_3 enhancements. Similar to June 16, GEOS-Chem clearly show enhancements in total O_3 in the PBL but not in USB O_3 on June 2 and June 29–30 (Fig. S10). The model attribution to U.S. anthropogenic emissions is consistent with the positive correlation between O_3 and CO measured at Angel Peak on June 16 (Fig. 6c–6d), June 2, and June 29–30 (Fig. S4). It is noteworthy that with a higher horizontal resolution GEOS-Chem better resolves the structure of the O_3 plumes as observed by TOPAZ lidar for all of the three pollution events. At the surface, both models capture the large-scale MDA8 O_3 enhancements across

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the SWUS on June 16 (Fig. 12b). The surface O_3 enhancements on June 2 and June 29–30 are relatively localized in Southern California and the Las Vegas area (Fig. S11), and both models have difficulty simulating the observed peak MDA8 values (Fig. 4).

4.5 Long-range transport of Asian pollution on May 20–24

[Figures 13-15 about here]

During May 20–24, long-range transport of Asian pollution toward the WUS was observed via large-scale CO column observations with Atmospheric Infrared Sounder (AIRS) on NASA's Aqua satellite (Fig. 13a). These Asian plumes spent a few days travelling eastward across the Pacific and reached the west coast of the U.S. on May 23 during the first FAST-LVOS IOP (May 23–25). The lidar measurements at NLVA on May 24 clearly showed high-O_3 plumes (> 70 ppbv) concentrated within the layers of 1–4 km and 6–8 km altitude above the Las Vegas Valley throughout the day (Fig. 14a). Both GFDL-AM4 and GEOS-Chem capture the observed O_3-rich plumes at surface–4 km and 6–8 km altitude above Clark County during this event. Elevated O_3 at 6–8 km altitude reflects the long-range transport from Asia, as supported by concurrent enhancements in total and USB O_3 in both models and by the large difference in the AM4 BASE simulation and the sensitivity simulation with Asian anthropogenic emissions zeroed out. Elevated O_3 at 1–4 km altitude appeared to be influenced by a residual pollution layer from the previous day; this plume was later mixed into the growing PBL (up to 4 km altitude), elevating MDA8 O_3 in surface air on May 24. Further supporting the impact from regional or local pollution below 4 km altitude, both models simulate much larger enhancements in total O_3 (70–90 ppbv) than in USB O_3 (~50 ppbv).

On May 24, MDA8 O_3 approached or exceeded the 70-ppbv NAAQS at multiple sites in California, Idaho, Wyoming, and Nevada (Fig. 15a), likely reflecting the combined influence from regional pollution and the long-range transport of Asian pollution. MDA8 O_3 at four surface sites in Clark County was above 65 ppbv. More exceedances would have occurred if the level for the NAAQS were lowered to 65 ppbv. In parts of Idaho, Wyoming, California where observed MDA8 O_3 were higher than 60 ppbv, the contribution of Asian anthropogenic emissions as estimated by GFDL-AM4 were 8–15 ppbv (Fig. 15a), much higher than the springtime average contribution of ~5 ppbv estimated by previous studies (e.g., Lin et al., 2012b) supporting the episodic influence from Asian pollution during this event. At several high-elevation sites in California such as Arden Peak (72 ppbv) and Yosemite National Park (70 ppbv), where observed MDA8 O_3 exceeds the NAAQS level, the contribution of Asian pollution is approximately 9 ppbv. Ozone

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produced from regional and local anthropogenic emissions dominates the observed MDA8 O_3 above 7(
ppbv in the Central Valley of California.

4.6 An unattributed event: June 28

The lidar measurements from June 28 show a fine-scale structure with a narrow O_3 layer exceeding 100 ppbv at 3–4 km altitude during 08:00–14:00 PDT (15:00–21:00 UTC shown in Fig. 14b). An ozonesond launched at 12:00 PDT also detected a high-O_3 layer (~115 ppbv) between 3.5 and 4 km altitude (no shown). This high-O_3 filament appears to descend and mix into the PBL after 14:00 PDT (21:00 UTC), contributing to elevated O_3 within the PBL in the afternoon. (Fig. 14b). Both models are unable t represent this fine-scale transport event, possibly due to diffusive mixing of the narrow layer. We therefore, focus on available airborne and in situ measurements to investigate the origin of this fine-scal O_3 filament.

Our examinations of large-scale satellite CO column measurements reveal a migration of high-CO plume during June 23–27 from Asia that arrived at the west coast of the U.S. on June 27 (Fig. 13b). GFDL-AM estimates 5–6 ppbv contributions from Asian pollution over the WUS on June 28 (Figs. 15b), which d not represent a significant enhancement above the mean Asian contribution. Aircraft measurements above the Las Vegas Valley showed collocated enhancements in CH_4 and O_3 coincident with low, fre tropospheric water vapor values at 3–4 km altitude (Fig. 10b). In-situ measurements at Angel Peak sho concurrent increases in CO and O_3 coincident with relatively dry conditions (Fig. 6f). These observations indicate that the O_3-rich plume appears to be unrelated to stratospheric intrusions. Aerosol backscatte measurements at NLVA show only a very slight enhancement in backscatter within the elevated O_3 laye on June 28, in contrast to the thick smoke observed on June 22 influenced by fresh wildfires in the Las Vegas Valley (Fig. 10). HYSPLIT and FLEXPART analyses presented in Langford et al., (in preparation suggest a possible connection to the Schaeffer Fire (https://en.wikipedia.org/wiki/Schaeffer_Fire) in th Sequoia National Forest in California. Another possible source is the fine-scale lofting of pollution fro Southern California followed by transport into the free troposphere over Las Vegas (Langford et al., 2010) This event further demonstrates the complexity of O_3 sources in the SWUS. We recommend measurements of atmospheric compounds like acetonitrile (CH_3CN, abundant in fire plumes) and methyl chloride (CH_3Cl, abundant in Asian pollution) (Holzinger et al., 1999; Barletta et al., 2009) via aircra

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Deleted: Overall, our analyses presented here suggest that the most likely sources for this high-O_3 filament on June 28 are aged fire plumes or fine-scale Asian pollution plume, although the lofting of pollution above Southern California followed by transport into the free troposphere over Las Vegas cannot be ruled out (Langford et al., 2010).

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and in situ platforms in future field campaigns in the region to help identifying the sources of such high O_3 filaments.

5 Comparison of background ozone simulated with GFDL-AM4 and GEOS-Chem

[Figure 16 about here]

Here, we summarize the differences in total and background O_3 between the two models over the WUS. GFDL-AM4 and GEOS-Chem differ in the spatial distributions and magnitudes of April–June mean USB O_3 at the surface and in the free troposphere over the U.S. (Fig. 16). USB O_3 in GFDL-AM4 peaks over the high-elevation Intermountain West at the surface (45–55 ppbv) and over the northern U.S. in the free troposphere (3–6 km altitude; 50–65 ppbv) due to the influence of STT. In comparison, GEOS-Chem simulates higher USB O_3 levels in southwestern states (e.g., Texas), both at the surface (45–50 ppbv) and at 3–6 km altitude (55–65 ppbv), likely due to excessive lightning NO_X during early summer (Zhang et al., 2011; Zhang et al., 2014). These discrepancies in USB between the two models likely reflect that GFDL-AM4 simulates stronger STT influences over the WUS while it produces less O_3 from weaker lightning NO_X emissions in the free troposphere over the southern U.S. than GEOS-Chem (Fiore et al., 2014). Despite a quantitative disparity, both models simulate higher USB O_3 levels over the WUS (45–55 ppbv in GFDL-AM4 and 35–45 ppbv in GEOS-Chem) than over the EUS at the surface (Fig. 16a). Our USB O_3 estimates with GEOS-Chem are generally consistent with the estimates in previous studies using GEOS-Chem or regional models driven by GEOS-Chem boundary conditions (Zhang et al., 2011; Emery et al., 2012; Dolwick et al., 2015; Guo et al., 2018). In contrast to NAB O_3 estimates in earlier studies by zeroing out North American anthropogenic emissions (Zhang et al., 2011; Lin et al., 2012a; Fiore et al., 2014; Zhang et al., 2014), USB O_3 estimates in our study include the additional contribution from Canadian and Mexican emissions. USB O_3 at Clark County sites is ~4 ppbv greater than NAB O_3 estimated by the same GFDL-AM4 model (Table S4). We also find that NAB O_3 estimated with the new GFDL-AM4 model is ~5 ppbv lower than the NAB estimates by its predecessor GFDL-AM3 (Lin et al., 2012a) for the WUS during March–April (Fig. S13), consistent with an improved simulation of free tropospheric ozone in AM4 during spring (Fig. 2). During early summer, the NAB O_3 levels estimated by AM3 and AM4 are similar (Fig. S13).

[Figure 17 about here]

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Here, we summarize the differences in total and background C between the two models over the WUS. Figure 15 shows the times series of observed and simulated O_3 at four high-elevation sites and one low-elevation site in the region during the study period. Notably, STT events (highlighted in blue shading) occurred frequently during April–June with MDA8 C exceeding or approaching the current NAAQS of 70 ppbv. Compared with observations, GFDL-AM4 captures the spikes of MDA8 O_3 during STT events associated with elevated USE O_3 and stratospheric O_3 (e.g., April 23, May 13, and June 11). During these events, GEOS-Chem significantly underestimate observed O_3 by 10–25 ppbv and simulates much lower USB C levels than GFDL-AM4, since the model underestimates the magnitude of STT (Sect. 4.2). The two models also differ substantially in total and USB O_3 (14–18 ppbv) during the Jun 22 wildfire event (yellow shading), with GEOS-Chem overestimating observations at high-elevation sites while GFDL-AM4 underestimating observations at both high- and [21]

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1127 We further compare simulated surface MDA8 O_3 against observations at 12 high-elevation sites (> 150
1128 m altitude; including 11 CASTNet sites and Angel Peak; see Table S1 and black circles in Fig. 1) in th
1129 WUS (Fig. 17). The observed high MDA8 O_3 events above 65 ppbv at these high-elevation sites ar
1130 generally associated with enhanced background O_3 in both models (USB O_3 = 50–60 ppbv in GFDL-AM
1131 and 45–55 ppbv in GEOS-Chem; Fig. 17a). Stratospheric intrusions are an important source of th
1132 observed events above 70 ppbv (Fig. S14), as suggested by GFDL-AM4, which better captures these high-
1133 O_3 events influenced by elevated background O_3 contributions, whereas GEOS-Chem underestimate
1134 these extreme events. For mean MDA8 O_3 at these sites, GFDL-AM4 is biased high by 3 ppbv whil
1135 GEOS-Chem is biased low by 5 ppbv. A recent study by Lin et al. (2019) found that an improved treatmen
1136 of ozone dry deposition can reduce mean springtime ozone biases in GFDL-AM4 by 5 ppbv. Mean USE
1137 O_3 simulated with GFDL-AM4 is 51.4±7.8 ppbv at WUS sites, higher than that in GEOS-Chem (45.7±5.
1138 ppbv; Fig. 17b). Probability distributions show that GFDL-AM4 simulates a wider range of total and USE
1139 O_3 than GEOS-Chem, reflecting relative skill in capturing the day-to-day variability of O_3.

1140 Tables S4 and S5 report year-to-year variability in the percentage of site-days with springtime MDA8 O
1141 above 70 ppbv (or 65 ppbv) and simulated USB levels during 2010–2017. The percentage of site-days
1142 with MDA8 O_3 above 70 ppbv during April–June 2017 is 0.9% from observations, 2.0% from GFDL-
1143 AM4, and 0.1% from GEOS-Chem. GFDL-AM4 captures some aspects of the observed year-to-yea
1144 variability despite mean-state biases. For example, the observed percentage of site-days with MDA8 O
1145 above 70 ppbv is highest (9.4%) in April-June 2012, compared to 3.1% for the 2010–2017 average. Tha
1146 statistics from GFDL-AM4 are 7.7% for 2012 and 4.0% for the 2010–2017 average. May–June mea
1147 USB MDA8 O_3 at Clark County sites are 50.9 ppbv in 2017, 55.3 ppbv in 2012, and 52.3±2.0 ppbv for
1148 the 2010–2017 average. Supporting the conclusions of Lin et al. (2015a), these results indicate tha
1149 background O_3, particularly the stratospheric influence, is an important source of the observed year-to-
1150 year variability in high-O_3 events over the WUS during spring.

1151 **6 Discussion and Conclusions**

1152 Through a process-oriented analysis of intensive measurements from the 2017 FAST-LVOS field
1153 campaign and high-resolution simulations with two global models (GFDL-AM4 and GEOS-Chem), we
1154 study the sources of observed MDA8 O_3 above 65 ppbv in the SWUS. Attribution of each event to a
1155 specific source is sometimes challenging, despite an integrated analysis of multi-tracer, multi-platform

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observations and model simulations as we demonstrated in this study. We identify the high-O_3 events associated with stratospheric intrusions (April 22–23, May 13–14, and June 11–13), mixing of local pollution and transported stratospheric O_3 (June 14), regional anthropogenic pollution (June 2, June 16 and June 29–30), wildfires (June 22), and mixing of Asian pollution with regional pollution (May 24). We also discuss an event (June 28) likely resulting from the fine-scale transport of fire plumes or pollution from Southern California, although a solid attribution for this event is challenging based on available data.

During the June 11–13 deep intrusion event, the NOAA mobile lab measurements at Angel Peak show a sharp increase in O_3 coinciding with a decrease in CO and water vapor, a marker for air of stratospheric origin. These characteristics are in contrast to the concurrent increases in O_3 and CO in humid and warm urban plumes and wildfires plumes transported from the Las Vegas Valley. The observed O_3/CO/H_2O relationships can serve as a useful first guess for the high-O_3 events influenced directly by a deep intrusion. However, once transported stratospheric O_3 is mixed into regional pollution, model diagnostic tracers are needed to quantify the stratospheric impact. For instance, on June 14, observations at Angel Peak show positive O_3/CO correlations while O_3Strat in GFDL-AM4 shows 20–30 ppbv enhancements above its mean level at Angel Peak and surface sites across the SWUS where the observed and simulated total MDA8 O_3 were above 70 ppbv.

GFDL-AM4 and GEOS-Chem differ significantly in simulating STT events, affecting their ability to simulate USB mean levels and extreme events. During the June 11–14 STT event, GFDL-AM4 captures the key characteristics of deep stratospheric intrusions consistent with lidar profiles and ozonesondes, whereas GEOS-Chem with simplified stratospheric chemistry and dynamics has difficulty simulating the observed features. At the surface on days when observed MDA8 O_3 exceeds 65 ppbv and AM4 O_3Strat is 20–40 ppbv above its mean baseline level, AM4 simulates 15–20 ppbv greater USB O_3 than GEOS-Chem (Figs. 4 and 9). During these STT events, total MDA8 O_3 simulated by the two models often brackets the observed values, as noted previously by Fiore et al. (2014). The FAST-LVOS analysis, combined with our earlier multi-year studies (Lin et al. 2012a; Lin et al., 2015a) indicates that GFDL AM3/AM4 capture the timing and locations of the observed O_3 enhancements in surface air and aloft during STT events, and are thus useful for screening of exceptional events due to STT. Considering the high biases in AM3/AM4 simulated total MDA8 O_3 during some STT events, we recommend bias-correction to simulated USB O_3 such as the approach used by Lin et al. (2012a). For the future application of GEOS-Chem for USB estimates, we recommend the version with the Universal tropospheric-stratospheric Chemistry eXtension

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(UCX) mechanism (Eastham et al., 2014) and process-oriented evaluation using daily ozonesondes and lidar profiles.

The two models also differ substantially in total and background O_3 simulations during the June 22 wildfire event. GEOS-Chem captures the broad O_3 enhancement in lidar observations but overestimates surface MDA8 O_3 at some sites during this event. It remains unclear whether higher USB O_3 simulated by GEOS-Chem during this event is from greater O_3 produced from wildfire emissions or excessive lightning NO_x emissions in the model. Although GFDL-AM3 captures the observed interannual variability in O_3 enhancements from large-scale wildfires over the WUS (Lin et al., 2017), GFDL-AM4 has difficulty simulating the observed O_3 enhancements during the relatively small-scale wildfire event on June 22. Sensitivity simulations with fire emissions constrained at the surface or with part of fire NO_x emissions emitted as PAN do not substantially improve simulated O_3 on June 22. Wildfires typically occur under hot and dry conditions which also enable the buildup of O_3 produced from regional anthropogenic emissions, complicating an unambiguous attribution of the high-O_3 events solely based on observations. Screening of exceptional events due to wildfire emissions remains a serious challenge.

The multi-model approach tied closely to intensive measurements provides insights into the capability and uncertainty of models in background O_3 estimates and harnesses the strengths of individual models to characterize the sources of high-O_3 events. Stratospheric intrusions, Asian pollution, and wildfires are important sources of the observed high-O_3 events above 65 ppbv in the SWUS, although uncertainties remain in the quantitative attribution. Surface ozone in China continues to increase despite regional NO_x emission controls in recent years (Liu et al., 2016; Li et al., 2019; Sun et al., 2016). Furthermore, the increasing frequency of wildfires under a warming climate (e.g., Westerling et al., 2006; Dennison et al., 2014) and growing global methane levels (e.g., West et al., 2006; Morgenstern et al., 2013) may foster higher background O_3 levels in the coming decades (Lin et al., 2017). These potentially increasing background O_3 sources, together with year-to-year variability in stratospheric influence (Lin et al., 2015a) will leave little margin for O_3 produced from local and regional emissions, posing challenges to achieving a potentially tightened O_3 NAAQS in the SWUS.

Data availability. Model simulations presented in this manuscript are available upon request to the corresponding author (Meiyun.Lin@noaa.gov). Field measurements during FAST-LVOS are available at https://www.esrl.noaa.gov/csd/projects/fastlvos.

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Author contributions. MYL conceived this study and designed the model experiments; LZ performed the GFDL-AM4 simulations and all analysis under the supervision of MYL; EK and YXW conducted the GEOS-Chem simulations; LWH and YXW assisted in the interpretation of model results; AOL, CJS, RJA, IP, PC, JP, TBR, SSB, ZCJD, GK, and SC carried out field measurements. LZ and MYL wrote the article with inputs from all coauthors.

Competing interests. The authors declare that they have no conflict of interest.

Disclaimer. The statements, findings, and conclusions are those of the author(s) and should not be construed as the views of the agencies.

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Table 1. List of high-O_3 events above 65 ppbv in the greater Las Vegas region during April-June 2017 (unit: ppbv). ¶

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