Review of "Attribution of future US ozone pollution to regional emissions, climate change, long-range transport, and model deficiency," by He et al.

We thank the anonymous reviewers for thoroughly reading our manuscript and providing helpful comments and suggestions, which will lead a significant improvement of our manuscript. The detailed responses to major point comments are listed below (text in italic is the reviewer's comments, and the normal text is our response):

Anonymous Referee #2

Overview
This study uses a regional chemical transport model driven by chemical boundary conditions from a global model to examine changes in U.S. surface ozone between present-day and two future emission/climate scenarios (A1B and A1Fi). High resolution modeling is valuable as it might provide more detailed spatial and temporal information on the response of surface ozone to changes in emissions versus climate. However, as I will discuss in detail below, this manuscript does not represent a substantial contribution to process-level understanding of present-day to future changes in U.S. ozone pollution. The discussions presented are often incomplete or scientifically inaccurate. In my view, the paper cannot be published in ACP in its present form.

Response: We appreciate the positive comments and good suggestions about our study. We will add description and analysis according to these comments in the revised manuscript.

Specific Comments:
1. Changes in emissions versus climate?
It is not clear how the role of changes in emission versus climate on the ozone inflow to US is separated in the experiment sets. In Table 1, Figures 4, 6, and 7, the authors stated that the CMAQ experiments Cases 1-5 are driven by dynamic boundary conditions from the CAM-Chem global model, but it is not clear which CAM-Chem experiment among Cases 11-15 is used. If the same CAM-Chem experiment has been used in the CMAQ simulations, the extent to which changes in the ozone inflow to the US are driven by the impact of changes in non-US anthropogenic emissions versus climate change in A1B and A1Fi scenarios? This question cannot be answered by the comparisons between CASES 1-5 and 6-10 experiments! In addition to changes in hemispheric emissions, shifts in atmospheric circulation patterns can also impact decadal variability in the strength of Asian pollution inflow to the U.S., as demonstrated by Lin et al (2014, Nature Geosci). Global and regional circulation patterns are likely to change under future climate scenarios. Thus, it is important to design the model experiments to be able to separate the role of emission vs. climate on global to regional scales.

Response: We thank the reviewer for the useful suggestions. The CMAQ experiments 1-5 used dynamic LBCs from CAM-Chem experiments 11-15, respectively, i.e., CMAQ and CAM-Chem simulations were conducted under the same climate and emissions except for refinements through dynamic downscaling along with more detailed emissions. The ozone inflow into the CMAQ modeling domain is determined by both the lateral boundary conditions (LBCs) and the regional climate, i.e. atmospheric circulation. In our modeling approach, these LBCs are derived from the global CAM-Chem simulations (driven by the global CCSM3 climate) while the regional meteorology is downscaled using CMM5 (also driven by the same global CCSM3 climate). For instance, case 1 and case 6 are driven by the same emissions (NEI) and same meteorology (CMM5 climate downscaling), so these two cases have the same change in atmospheric circulation. In the revised manuscript, we will clarify that these LBCs do not include meteorology fields but only provides chemical LBCs, i.e., concentration profiles at CMAQ modeling domain boundaries. Therefore, when we compare case 1 and case 6, the difference reflects the effects through replacing the fixed LBCs by the dynamic LBCs.

Related to this comment on atmospheric circulation variability, it seems odd to me that the authors define present-day climate as conditions during the five-year period of 1995-1999 but provide no discussion on the extent to which this short, five-year period can represent present-day climate. The frequency of mid-latitude cyclones [e.g., Leibensperger et al., 2008; Turner, et al., 2013] as well as hemispheric pollution transport patterns (Lin et al., 2014) can change significantly from year to year and even from decade to decade: their variability clearly affects ozone in the US. How does their variability during the 1995-1999 period compares with the past 20-30 years?

Response: We understand that long-time integration is necessary for studying the climate effects on future air pollution. However, running regional models, i.e., regional climate model (CMM5), regional CTM (CMAQ), and emissions model (SMOKE), requires substantial amount of computing sources. For instance, CMAQ alone requires about 5k CPU hours and 2k GB disk space for one summer (JJA) simulation. Therefore we can only afford short time period such as 5-yr integration to represent the present-day and future cases. This is a typical practice in regional air quality modeling studies, for instance a synthesis study of future US ozone used 2048-2052 results to represent the future cases from regional CTM simulations (Weaver et al., 2009); a WRF-CMAQ study used 2001-2004 and 2057-2059 to represent the present-day and future ozone pollution (Gao et al., 2013). The downscaling tool, CMM5, has been developed and tested in our previous studies for 10 to 30-yr simulations, which can provide robust climate signals driven by different GCMs (Liang et al., 2001; Liang et al., 2004). Due to the relatively short integration, the effects of decadal climate change such as the variation of hemispheric pollution patterns are not within the scope of this study. We will add this discussion section and cite Lin et al. 2014 paper in the revised manuscript.

2. Changes in methane?

The methane level might play an important role in changes in background ozone in the US (e.g. Wild et al., 2012; Clifton et al, 2014). How does the methane level change in the A1B and A1Fi scenarios? How their changes are represented in the global CAM-Chem simulations? How do they affect the ozone inflow to the US in the CMAQ simulations?

Response: We agree with the reviewer that methane level plays an important role in the background ozone. In our study, global methane emission projection in CAM-Chem simply follow the IPCC Special Report for Emissions Scenarios (IPCC-SERS). The Figure 1. shows the change curves under each scenario. We are using the marker scenarios (A1B and A1Fi). Like other emissions, this projection include the major changes in anthropogneic sources, agriculture sources, and other land use sources.
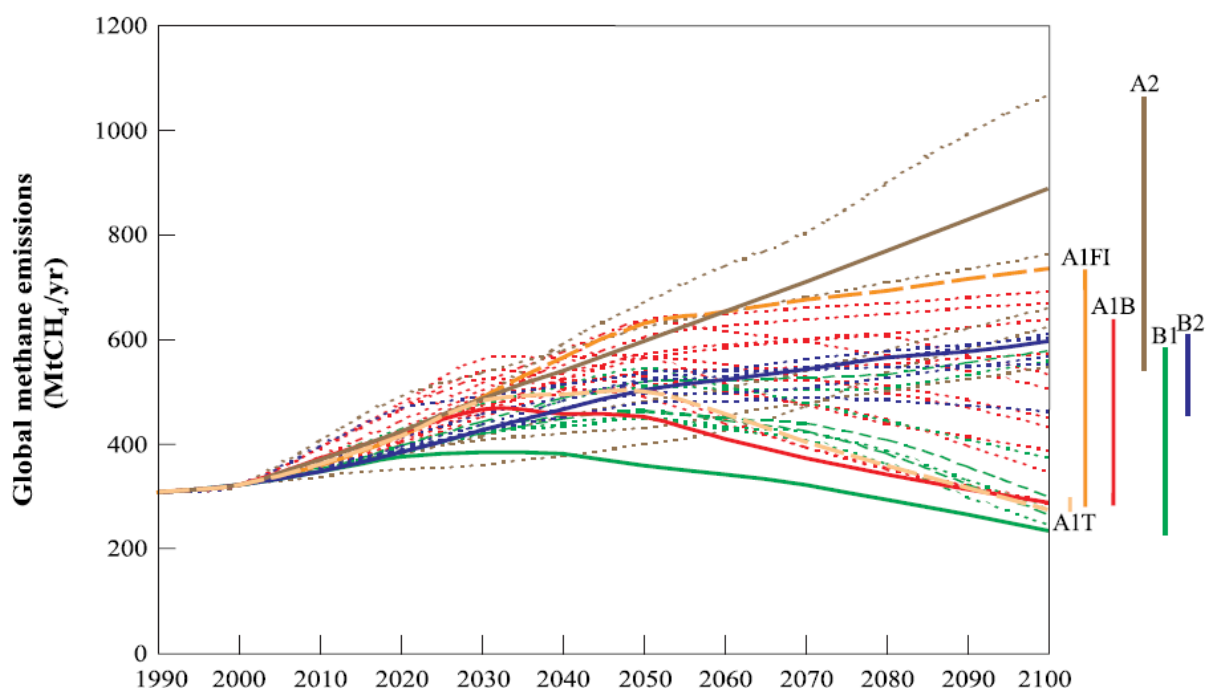

Figure 1. from IPCC SERS Report. Standardized (to common 1990 and 2000 values) global annual methane emissions for the SRES scenarios (in $MtCH_4/yr$). The range of emissions by 2100 for the six scenario groups is indicated on the right. Illustrative (including marker) scenarios are highlighted.

CMAQ used fixed background methane concentrations, and we simply used the IPCC A1B and A1Fi future prediction of methane levels as the global background. Unfortunately, due to the limited computing resources, we did not conduct sensitivity to identify the methane impacts on future ozone. We will add discussion on methane levels in the revised manuscript.

3. Changes in stratosphere-to-troposphere ozone transport?

Recent work has shown that deep stratosphere-to-troposphere transport (STT) of ozone contributes substantially to high-ozone events observed at Western U.S. high

elevation sites (e.g. Langford et al., 2009; Lin et al, 2012). Studies have also shown that the STT ozone flux is likely to increase under a warming climate (e.g. Collin et al., 2003; Hegglin et al., 2009). How does STT change from present-day to future climate in your model simulations? Why not also include the analysis of ozone changes in the intermountain west region (Figure 1)?

Response: We agree with the reviewer that STT is important for the surface ozone especially in high elevation areas in the western US. However CMAQ does not have stratospheric chemistry (CMAS, 2007; Yarwood et al., 2005), so we cannot use it to investigate the effects of STT on the future ozone. That is also the reason why our study does not investigate the changes of future ozone in the intermountain west region, where future ozone is expected to be influenced substantially by STT which is missing in our modeling system.

4. Changes in lateral boundary conditions of long-lived chemical species? (Figure 2)

In Figure 2, it is awkward that the authors examined changes in lateral boundary conditions of short-lived species: NOx and VOCs. These short-lived species are not expected to make a substantial contribution to long-range of ozone. Why not examine changes in relatively long-lived species like ozone, CO, and PAN? It would be even better if the authors can look at how the two models compare to the ozonesonde data at Trinidad Head, California and discuss how the inflow changes.

Please change the y axis in all vertical profiling plots (Figs. 2, 3: : :) to pressure in hPa or altitude in km. A sigma value has no meaning as it depends on the model top!

Response: We appreciate the valuable comments for the long-lived species. In the revised manuscript, we will add discussion about O_3, and CO concentrations in the both fixed and dynamic LBCs (Fig. 2). The altitude profiles of long-lived species shows that the dynamic LBCs transport more CO but less O_3 into the modeling domain. As discussed above, transport into the CMAQ modeling domain is determined by both LBCs and atmospheric circulation. Unfortunately, the CMAQ model used in this study has no ability to calculate the inflow/outflow through the modeling boundaries, so we cannot quantitatively quantify these effects. We will add detailed discussion into the revised manuscript.
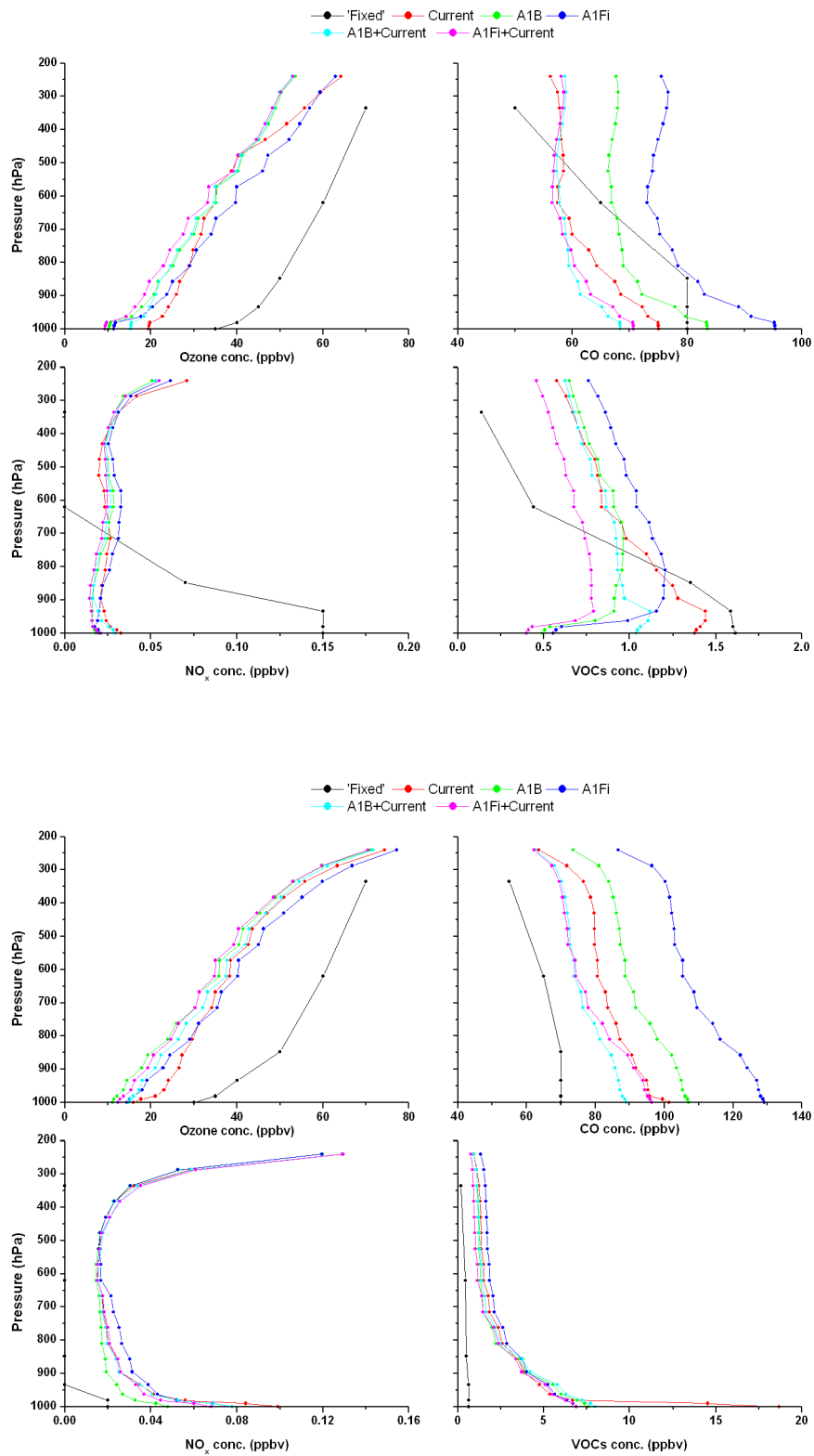
Figure 2.   O$_3$, CO, NO$_x$, and VOCs profile concentrations at a) west boundary and b) east boundary.

About the ozone sonde data, we cannot compare our results to the in-situ measurements of Trinidad Head because our runs were driven by the CCSM3 climate simulations but not reanalysis data. We will also modify the y-axis of Figure 2 to pressure levels in the revised manuscript (Fig. 3).
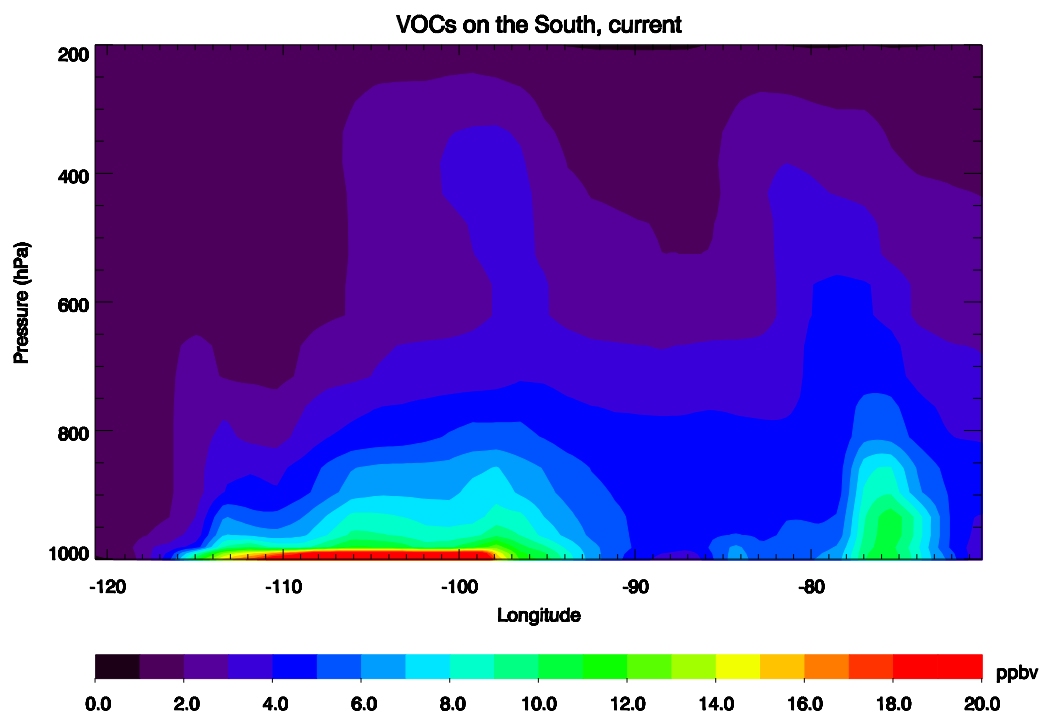

Figure 3. Revised Fig. 2 of the original manuscript through replacing Sigma levels by pressure levels.

5. Model evaluation.

Table 2: It is not clear in the caption whether this is for annual mean or for a specific season. Figure 5: Why not show the maps for each season and for the entire US?

Response: This study focuses only on the summertime ozone, so all ozone values presented are MDA8 means of JJA. In Figure 5 of the manuscript, we show the results of a case study (details in section 3.2) about the present-day ozone simulation in California where the largest discrepancy between model results and observations is found; and in this study we only focus on the summertime ozone. So we will not show similar plots for other regions and other seasons. We will emphasize these in the revised manuscript.

Recommendations:

Revision of the paper will greatly benefit from a thorough literature review on the various drivers of long-term changes in tropospheric ozone and explicitly discuss how their model experiments are designed to address these issues. It would be also useful

to do the analysis for each season, at least separating ozone changes during spring vs summer, as the specific drivers in different seasons can be very different.

Response: In the current manuscript, we only summarize previous studies using regional CTMs, so we sincerely appreciate the suggestion to include the global CTM studies in the literature review. We revised the following sentences:

"… Global CTMs represent atmospheric chemistry and transport processes, including long-range transport (LRT), and resulting changes in atmospheric composition, across the planetary scale. However, lack of detailed emissions and inadequate spatial resolution in existing studies can cause substantial errors (e.g., Lin et al., 2008; Lei et al., 2013). …"

to discussion citing more global CTM studies as:

"… Global CTMs represent atmospheric chemistry and transport processes, including long-range transport (LRT), and resulting changes in atmospheric composition, across the planetary scale. Global CTMs are widely used to investigate the future ozone pollution influenced by intercontinental transport on the receptor regions (Fiore et al., 2009; Langford et al., 2009; Wu et al., 2009), shifts in atmospheric circulation patterns (Lin et al., 2014), and changes of mid-latitude cyclone frequency (Leibensperger et al., 2008; Turner et al., 2013). However, lack of detailed emissions and inadequate spatial resolution in existing studies can cause substantial errors (e.g., Lin et al., 2008; Lei et al., 2013). …"

About the seasonal variations of future ozone, due to the limited computing sources discussed above, we do not have full-year runs for all of our cases. For instance, we only have JJA simulations for case 2, 3, 4, and 5, so discussion about future ozone changes during other seasons is not within scope of this study.

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