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 reviewers for providing extremely helpful comments and suggestions, leading to what we hope are a greatly improved manuscript. Below we list the detailed responses to the reviews (note that the italicized text is the reviewer's comments, followed by our response as normal text in blue).

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 useful suggestions. We have extensively rewritten the paper to provide better clarity on why this paper does extend the existing literature. We agree with the reviewer that much of the material needs to be better explained and we have rewritten the paper to do so. One of the authors involved in the research but not in preparation of the submitted paper went through it and made significant input to the rewrite. Sections 1 and 2 have almost been entirely rewritten. Section 3 has also been extensively rewritten to improve the description of the results and findings, and include more detailed discussion about the future ozone trend and analysis about relative contribution.

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 have rewritten sections 2 and 3 to better clarify the range of experiments and the overall discussion of the findings. The CMAQ experiments 1-5 use dynamic LBCs from CAM-Chem experiments 11-15, respectively, i.e., CMAQ and CAM-Chem simulations were conducted under the same climate and 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 emphasize that these are chemical LBCs that provide chemical concentration profiles at the CMAQ modeling 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 agree 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, which limited what could be done here. Our past studies have shown

that five-year integrations are adequate for representing the primary signals in air quality effects -- this is a typical practice in regional air quality modeling studies related to climate change. For example, a synthesis study of future US ozone used 2048-2052 results to represent the future cases due to short simulations of regional CTM studies (Weaver et al., 2009) and a WRF-CMAQ study used 2001-2004 and 2057-2059 to represent the present-day and future ozone pollution (Gao et al., 2013). The model results are climatological so only roughly correspond to the 1995-1999 period and are considered representative of the present climatological conditions, and not the specific period where there might be specific major weather events that would need to also be represented.
The regional model used in the downscaling, CMM5, has been developed and tested in a number of previous studies (Liang et al., 2001; Liang et al., 2004). From past studies we know that the climatological findings are not significantly different from using the 5 year period to the first order approximation.

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 the level of methane plays an important role in determining background ozone. In our study, global methane emission projection in CAM-Chem use the recommended concentration changes fron the IPCC Special Report for Emissions Scenarios (IPCC-SRES) as a function of time based on the scenarios analyzed. We added further discussion to the paper to explain how the boundary concentration of methane and other long lived gases were treated.
Because of limited computing resources, we did not conduct sensitivity tests to further examine the methane impacts on future ozone or to do special calculations using methane emissions that would require a much longer time period needed for the calculations if emission boundary conditions were used.

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 can be important for specific events affecting the surface ozone, especially at high elevation areas in the western US. However, there is no indication that such events greatly affect the climatology of surface ozone across the U.S. Because of the limitations of the models, the coarse resolution of the global model and the lack of stratospheric processes in the regional model, we were unable to address these issues in this paper. Admittedly this would be an interesting issue to address in future studies using either a very high resolution global model that includes stratospheric processes (which CAM-Chem does if it were run at higher resolution and/or if stratospheric processes were included in a regional model. We added discussion about STT in Section 4 of the revised manuscript.

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:

In the revised manuscript, we have added additional discussion on the ozone and CO concentrations for the both fixed and dynamic LBCs cases, and have revised Figures 2 and 3 as suggested. The altitude profiles of long-lived species show 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 chemical LBCs and atmospheric circulation.

Regarding the ozonesonde data, while we could compare our results to the in-situ measurements of Trinidad Head, the value of doing this would be very limited because our runs were driven by the CCSM3 climate simulations and not reanalysis data.

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. The Table caption has been modified to make this clearer. In Figure

5, 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. For space limitations, we have not included similar plots for other regions and other seasons in the main article, but we did add US ozone maps to the supplementary material

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:

We agree with the reviewer and have totally rewritten Section 1 to provide a better discussion of past studies and their findings.

About the seasonal variations of future ozone, summer is by far the most important season and while the ozone will increase in the spring season it still is unlikely to be as important as the JJA period. Unfortunately, only the JJA outputs for case 2, 3, 4, and 5 were saved on the server, so we are not able to analyze future ozone changes during other seasons within this study. We do agree it would be interesting to see if the length of the ozone season is lengthened by the changing climate.