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 #1

In this model study, He et al. examine the effects of changing emissions, climate change, and long-range transport on summertime surface ozone levels in the 2050s. They apply two scenarios, A1B and A1Fi, to two different models, the global NCAR Community Atmospheric Model with Chemistry (CAM-Chem) and the regional chemical transport model CMAQ. The scenarios differ in their magnitude of anthropogenic emissions, with A1Fi showing increasing NOx emissions over the US, and A1B showing a strong decrease. To investigate the role of long-range transport, they perform CMAQ model simulations with some kind of fixed chemical boundary conditions and with boundary conditions derived from the CAM-Chem simulations. To characterize what is called "model deficiency," the authors compared the ozone values simulated by CAM-Chem to those simulated by CMAQ.

Scientific significance.
The paper reveals little new science, and is not suitable for publishing in its current form. The effect of changing intercontinental transport on surface ozone in receptor regions has already been studied extensively (e.g., Wu et al., 2008, 2009; Fiore et al., 2009; Wild et al., 2012; Doherty et al., 2013). Lam et al. (2011), not mentioned here, has previously used CMAQ to examine the effects of climate change and emissions on U.S. surface ozone, as has Penrod et al. (2014). The current study does not, in my view, add significantly to the existing literature. I recommend that the authors review carefully past literature and think how their work advances this topic.

Response:
We agree with the reviewer that we should have explained the uniqueness of this study much more fully and have done so in the revised version. Almost all of section 1 was rewritten so that we can provide more background and better clarify the value of this study. We disagree with the reviewer about there being little new science (if that were the case, the same could be said for most of the papers cited by the reviewer as they basically reexamine and/or incrementally improve on issues discussed in prior studies, including studies we authored earlier that were not cited by the reviewer or even in our original paper – we have remedied this in the new version by adding a number of new references and discussion about those studies). We also should have included further discussion of

past studies and their findings, including the additional references raised by the reviewer. We have revised the text to include a better discussion of the history of the science in a short summary.

This study builds on and extends the earlier studies by investigating the effects of uncertainties, using sensitivity analyses, affecting the air quality findings produced in such a modeling system. This includes examining the uncertainties associated with factors like model structure, the chemical mechanism, and the climate conditions used in the global and regional models. We believe this provides new information and sufficient uniqueness for the paper to be worthy of publication.

Scientific quality.

The description of the model simulations is scant, and the model analysis is insufficient. What methane concentrations are applied? Do the models include lightning NOx, soil NOx, or stratosphere-troposphere exchange? How are the meteorological fields downscaled for use in the regional model? How do emissions of anthropogenic and biogenic VOCs change in the US? (Just the sum is given). How do the model results compare to CASTNET observations? How do anthropogenic emissions change in the source regions (Asia and Mexico) for the two scenarios? How much ozone is transported into the domain as opposed to ozone precursors? Why are there regional differences in the contribution of climate change to the change in ozone (Figure 7)? Why does the contribution of "model deficiency" to model ozone appear so large in the Southeast US (Figure 7)? Isn't "model deficiency" just the difference between two models, both of which could be deficient? The reader is skeptical about the meaningfulness of such a metric, in part because models use the same chemical scheme, which could be flawed, especially in regard to isoprene oxidation (Mao et al., 2013).

Response:

As suggested by the reviewer, we have added to the description of the models and their capabilities. Most of the questions raised by the reviewer were also addressed in our two related earlier papers (Lei et al., 2012, 2013). Nonetheless, we agree that some of the issues needed clarification for this paper (e.g., the treatment of lightning NO_x) and we have done so in the revised paper.

We briefly address some of the reviewer's questions below:

- Methane concentrations are fixed in CMAQ in the background atmosphere for both the current and projected atmospheres; we applied the IPCC A1B and A1Fi projected methane levels for the future global and regional background atmospheres.
- CMAQ is designed for air quality studies, and thus focuses on tropospheric chemistry; it has limited capability for simulating stratospheric chemistry (CMAS, 2007; Yarwood et al., 2005), so we cannot investigate the SSE with that current modeling system. However, the global CAM-Chem runs do include a complete

representation of stratospheric processes.

- The meteorological fields were downscaled using the CMM5 model; we have previously published several papers describing the downscaling technique and its performance for both current and future climate (e.g., Liang et al., 2008, 2006, 2004, 2001). This results in a much better representation of atmospheric dynamics than using global model results directly as has been done in some prior studies.
- The U.S. emissions were processed using SMOKE with NEI and scaled IPCC emissions; the approach is discussed in earlier papers such as Tao et al. (2007) and we also added one paragraph in Section 2 to explain the emissions processing such as the soil NO_x treatment.
- CASTNet observations are included in the EPA AQS database. We did do an analysis of the model relative to all of the EPA AQS sites. CASTNet sites are normally located in suburban or rural locations; the relatively sparse CASTNet network can provide information about the background ozone, while with high resolution CMAQ simulations our goal is investigating the small scale climate signal of future ozone changes. Please see details in our case study about ozone observations and simulations in California.
- The emission changes outside the modeling domain are reflected in dynamic LBCs from CAM-Chem simulations; we published two papers discussing these changes such as in Asia (Lei et al., 2012; Lei et al., 2013). These LBC conditions are concentration profiles of air pollutants (e.g., Figure 3); note that the standard CMAQ model by itself has no ability to estimate the transport of chemical species through the model boundaries so it is reliant on these boundary conditions being far enough from the region of interest that the effects of the transport into the contiguous U.S. get represented properly in the study.
- We agree with the reviewer that model deficiency was not at all the right terminology. The term ‘model deficiency’ has been eliminated and is now referred to as the effect or the differences due to “modeling design”. This term is defined as the bulk difference between the global and regional modeling systems in representing all aspects related to US air quality including basic design, emissions, chemical mechanism, climate conditions, and their interactions. This approach does not allow separate of individual model factors, but represents a rough estimate of how the model system forwarded error may affect the result. Past studies generally lack a quantitative estimate of how much model forwarded errors may account for relative contribution identified among the regional emissions, long-range transport, and climate.
- Through a number of model experiments with both the global and regional models, this study examines a range of uncertainties associated with common modeling assumptions. However, this study is not aimed at evaluating all uncertainties such as the chemistry mechanism example given by the reviewer.

Presentation quality.

Tables 3 and 4 contain such a density of information that they are incomprehensible. Authors would be wise to choose what information is most important for the reader to know and to just present that. It would probably be best to present NOx and VOC emissions for the entire domain and for selected source regions – e.g., Asia and Mexico. Temperature changes would best be viewed as a map. Model validation would also best be viewed on maps such as in Figure 5. Most captions lack information on what exactly is being shown – summertime average surface ozone? Paper has problems in written English, about 1-3 per paragraph.

Response:

We thank the reviewer for these valuable suggestions. We have revised the manuscript to improve the quality of the presentation of the results, including revisions to the original Tables 3 and 4. We have also added figures showing the changes of temperature and emissions in the supplementary materials. However the emissions and climate changes outside the CONUS such as those in Asia are not simulated directly by CMAQ, so they are represented in CMAQ through the LBCs coming from the global CAM-Chem that does include those emissions. Details about changes in these areas can be found in our earlier studies with CAM-Chem (Lei et al., 2012, 2013).