

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 #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 NO_x 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: Our project included two major parts, the global CAM-Chem simulations and the regional CMAQ simulations which focus on the US with more detailed emissions and improved climate conditions at regional scales. The effects of changing intercontinental transport of ozone and its precursors on the receptor regions such as Europe and the United States are discussed in our previous publications using results from the global CAM-Chem model (Lei et al., 2012; Lei et al., 2013), which is not the goal of this study. The results of regional CTMs such as CMAQ depend on regional meteorology downscaled (in this study by CMM5) from GCMs (or reanalysis for validation purpose) and lateral boundary conditions (LBCs) for providing cross-boundary transport of chemical species including ozone and its precursors, as well as the projected changes of future emissions within the computational domain. Previous regional CTM studies normally focus on two of these three factors, such as Penrod et al. (2014) and

other papers cited in our manuscript. In our study, we synthesize robust regional climate downscaling from CMM5, dynamic intercontinental transport from CAM-Chem, and future IPCC emission predictions into the regional CMAQ model. Thus our study is one of very few studies investigating all of these factors together.

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. Accordingly we have 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). ...”

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 NO_x, soil NO_x, 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 discussed above, this study is part of our project which involves lots of our previous efforts such as global CTM simulations and regional climate downscaling. We appreciate the valuable suggestion to include more details about our modeling system, and will add them into the revised manuscript. We briefly answer some questions here. Methane concentrations are fixed in CMAQ as background and we applied the IPCC A1B and A1Fi projected future methane levels as global background; lightning NO_x and soil NO_x were not included in the CMAQ version 4.6 used in this study; CMAQ only focuses on tropospheric chemistry and has no stratospheric chemistry (CMAS, 2007; Yarwood et al., 2005), so we cannot investigate the SSE with the current modeling system. The meteorological fields were downscaled using the CMM5 model developed by our group; we published several papers describing the downscaling technique and its performance for both current and future climate (Liang et al., 2008; 2001; 2004; 2006). The US emissions were processed using SMOKE with NEI and scaled IPCC emissions; the results were discussed before (Tao et al., 2007). CASTNet observations are included in the EPA AQS database. 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), and the standard CMAQ has no ability to estimate the transport of chemical species through the modeling boundaries. The ‘model deficiency’ is defined to describe the bulk difference between the global and regional modeling systems, representing all aspects related to US air quality including emissions, chemical mechanism, climate conditions, and their interactions. This approach does not allow separation of individual model deficiency factors, but represent a rough estimate of how the overall model system formulation errors may affect the result. The past studies generally lack a quantitative estimate of how much model formulation errors may account for relative contribution identified among the regional emissions, long-range transport, and climate. Our study attempts to provide a rough account of the uncertainty that may be included in the factor analysis. We will add the above description about the ‘model deficiency’ into the revised manuscript. The isoprene oxidation chemistry was not explicit described even in the latest CMAQ, so in this project we have no ability to identify the effects from isoprene oxidation.

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 NO_x 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 will add figures showing the changes of temperature and emissions in the auxiliary materials. However the emissions and climate changes outside the CONUS such as Asia are not simulated by CMAQ, so they are out of the scope of this study. Details about changes in these areas could be found in our CAM-Chem studies (Lei et al., 2012; Lei et al., 2013). Lastly, we will carefully revise this manuscript to improve the presentation quality.

References:

- CMAS: Community Modeling and Analysis System: CMAQ v4.6 operational guidance document, report, Univ. of North Carolina, Chapel Hill, 2007.
- Fiore, A. M., Dentener, F. J., Wild, O., Cuvelier, C., Schultz, M. G., Hess, P., Textor, C., Schulz, M., Doherty, R. M., Horowitz, L. W., MacKenzie, I. A., Sanderson, M. G., Shindell, D. T., Stevenson, D. S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I., Carmichael, G., Collins, W. J., Duncan, B. N., Faluvegi, G., Folberth, G., Gauss, M., Gong, S., Hauglustaine, D., Holloway, T., Isaksen, I. S. A., Jacob, D. J., Jonson, J. E., Kaminski, J. W., Keating, T. J., Lupu, A., Marmer, E., Montanaro, V., Park, R. J., Pitari, G., Pringle, K. J., Pyle, J. A., Schroeder, S., Vivanco, M. G., Wind, P., Wojcik, G., Wu, S., and Zuber, A.: Multimodel estimates of intercontinental source-receptor relationships for ozone pollution, *Journal of Geophysical Research-Atmospheres*, 114, 21, D04301:10.1029/2008jd010816, 2009.
- Langford, A. O., Aikin, K. C., Eubank, C. S., and Williams, E. J.: Stratospheric contribution to high surface ozone in Colorado during springtime, *Geophys. Res. Lett.*, 36, L12801, doi:10.1029/2009gl038367, 2009.
- Lei, H., Wuebbles, D., and Liang, X. Z.: Projected risk of high ozone episodes in 2050, *Atmos. Environ.*, 59, 567-577, 2012.
- Lei, H., Wuebbles, D. J., Liang, X. Z., and Olsen, S.: Domestic versus international contributions on 2050 ozone air quality: How much is convertible by regional control?, *Atmos. Environ.*, 68, 315-325, 10.1016/j.atmosenv.2012.12.002, 2013.
- Leibensperger, E. M., Mickley, L. J., and Jacob, D. J.: Sensitivity of US air quality to mid-latitude cyclone frequency and implications of 1980-2006 climate change, *Atmos. Chem. Phys.*, 8, 7075-7086, 2008.
- Liang, X. Z., Kunkel, K. E., and Samel, A. N.: Development of a regional climate model for US midwest applications. Part I: Sensitivity to buffer zone treatment, *J. Clim.*, 14, 4363-4378, 10.1175/1520-0442(2001)014<4363:doarc>2.0.co;2, 2001.
- Liang, X. Z., Li, L., Dai, A., and Kunkel, K. E.: Regional climate model simulation of

- summer precipitation diurnal cycle over the United States, *Geophys. Res. Lett.*, 31, 4, L24208, doi:10.1029/2004gl021054, 2004.
- Liang, X. Z., Pan, J. P., Zhu, J. H., Kunkel, K. E., Wang, J. X. L., and Dai, A.: Regional climate model downscaling of the U.S. summer climate and future change, *Journal of Geophysical Research-Atmospheres*, 111, D10108, doi:10.1029/2005jd006685, 2006.
- Liang, X. Z., Kunkel, K. E., Meehl, G. A., Jones, R. G., and Wang, J. X. L.: Regional climate models downscaling analysis of general circulation models present climate biases propagation into future change projections, *Geophys. Res. Lett.*, 35, L08709 10.1029/2007gl032849, 2008.
- Lin, J. T., Wuebbles, D. J., and Liang, X. Z.: Effects of intercontinental transport on surface ozone over the United States: Present and future assessment with a global model, *Geophys. Res. Lett.*, 35, 6, L02805, doi:10.1029/2007gl031415, 2008.
- Lin, M., Horowitz, L. W., Oltmans, S. J., Fiore, A. M., and Fan, S.: Tropospheric ozone trends at Mauna Loa Observatory tied to decadal climate variability, *Nature Geosci*, 7, 136-143, 10.1038/ngeo2066 <http://www.nature.com/ngeo/journal/v7/n2/abs/ngeo2066.html#supplementary-information>, 2014.
- Tao, Z. N., Williams, A., Huang, H. C., Caughey, M., and Liang, X. Z.: Sensitivity of U.S. surface ozone to future emissions and climate changes, *Geophys. Res. Lett.*, 34, 5, L08811, doi:10.1029/2007gl029455, 2007.
- Turner, A. J., Fiore, A. M., Horowitz, L. W., and Bauer, M.: Summertime cyclones over the Great Lakes Storm Track from 1860–2100: variability, trends, and association with ozone pollution, *Atmos. Chem. Phys.*, 13, 565-578, 10.5194/acp-13-565-2013, 2013.
- Wu, S. L., Duncan, B. N., Jacob, D. J., Fiore, A. M., and Wild, O.: Chemical nonlinearities in relating intercontinental ozone pollution to anthropogenic emissions, *Geophys. Res. Lett.*, 36, L05806, 10.1029/2008gl036607, 2009.
- Yarwood, G., Rao, S., Yocke, M., and Whitten, G. Z.: Updates to the Carbon Bond chemical mechanism: CB05., Final Report to the US EPA, RT-0400675, December 8, 2005, http://www.camx.com/publ/pdfs/CB05_Final_Report_120805.pdf, 2005.