Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-1226-RC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Foreign and domestic contributions to springtime ozone over China" *by* Ruijing Ni et al.

Anonymous Referee #1

Received and published: 13 February 2018

General Comments:

This study examines the domestic and foreign influence of anthropogenic emissions on ozone over China using the GEOS-Chem model and two methods of identifying contributions, a "zero-out" approach and a tagging approach (which seems to be missing from the manuscript). After first validating the model's capabilities against surface and ozonesonde observations, they proceed to characterize the spatial influence (horizontally and vertically) of natural, background, foreign anthropogenic and domestic anthropogenic emissions on ozone over China.

Much of the analysis in this manuscript contains significant insights into the ozone chemistry over China and the impact of foreign and domestic emissions on tropospheric ozone. This manuscript could be a valuable contribution to ACP and to our

understanding of ozone attribution over China, but there are several major items that need to be addressed before I can recommend publication. I discuss two major issues below, and conclude with technical comments.

Specific Comments:

First, this study examines a single 3-month period (spring) in 2008 and draws extensive conclusions based on this period. The nature of emissions, ozone chemistry, meteorology, and atmospheric transport make it difficult to believe in the robustness of results drawn from such a short period without some characterization of the trends, variability, and uniqueness/non-uniqueness of this particular spring in 2008. While the authors point out the reasons for selecting this time period (L102-107), and while they mention some of these issues (e.g. NOx trends in L282-285, differences in emissions and meteorology in L311-312), I do not believe there is a sufficient demonstration of the robustness of their results, and there are many questions that need to be addressed. Are the results drawn throughout Sections 4 and 5 robust for different years, or are they sensitive to chemical and meteorological variability and thereby vary from year-to-year? How much to they vary? Where does the spring of 2008 fit into the bigger ozone/chemistry/meteorology context over China?

I feel that either: (1) additional simulations including at least one additional year are required to demonstrate the simulated variability of ozone over China and the robustness of these results; or (2) the manuscript requires additional literature reviews and a careful description of the ozone variability over China as a demonstration of the robustness of the results. In L247-253 the authors discuss an additional year of simulation, which could certainly provide some of this temporal variability context. Some of the publications below could provide some of this context and reasons why 3-months is not long enough to draw strong conclusions, especially with regards to ozone:

Xu, X., Lin, W., Wang, T., Yan, P., Tang, J., Meng, Z., and Wang, Y.: Long-term trend of surface ozone at a regional background station in eastern China 1991–2006: enhanced variability, Atmos. Chem. Phys., 8, 2595-2607, https://doi.org/10.5194/acp-8-2595-2008, 2008.

Jin, X., and T. Holloway, Spatial and temporal variability of ozone sensitivity over China observed from the Ozone Monitoring Instrument, J. Geophys. Res. Atmos., 120, 7229–7246, doi:10.1002/2015JD023250, 2015

W.N. Wang, T.H. Cheng, X.F. Gu, H. Chen, H. Guo, Y. Wang, F.W. Bao, S.Y. Shi, B.R. Xu, X. Zuo, C. Meng, X.C. Zhang, Assessing spatial and temporal patterns of observed ground-level ozone in China, Sci. Rep., 7 (1), p. 3651, 10.1038/s41598-017-03929-w, 2017.

Garcia-Menendez, F., Monier, E., and Selin, N. E.:The role of natural variability in projections of climate change impacts on U.S. ozone pollution, Geophys. Res. Lett., 44, 2911–2921, 2017.

Brown-Steiner, B., Selin, N. E., Prinn, R. G., Monier, E., Tilmes, S., Emmons, L., and Garcia-Menendez, F.: Maximizing Ozone Signals Among Chemical, Meteorological, and Climatological Variability, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-954, in review, 2017.

Second, the manuscript at times leaves out critical information or does not sufficiently describe methods, definitions, and figures. While this manuscript contains many valuable results, there were several moments where I didn't feel there was enough information provided to understand what was done, or why it was done, and times when I had to search for descriptions and/or infer some explanations on my own. The following list summarizes areas and issues that need to be addressed and revised:

(1) The authors state that they combine zero-out simulations with tagged ozone simulations (and the tagged ozone simulations are mentioned in Table 2 and on L110, L114, and L183-187), but nowhere throughout the rest of the manuscript are the tagged ozone results described or shown. Were they not used? Where are the descriptions of

these results?

(2) A linear weighting method is used to adjust the ozone attribution results, and is described on L188-195, but the description is insufficient. I am not familiar with this method, so I do not fully understand what Equation 1 means, and tracing back to the Li et al. (2016a) citation brings me to a 'normalized marginal method' used for radiative forcing attribution, not ozone attribution. It is not clear to me where the precise formulation of Equation 1 came from, what it does, what impact the adjustment has on the results, or why it was selected.

(3) Many of the comparisons to observations compared the simulated spring of 2008 with other years (e.g. L281-285, L297-298, L311-312), and given the variability in ozone, chemistry, and meteorology (see above), I'm not sure these are wholly valid comparisons, especially without the broader temporal context of ozone over China. Some sort of quantification of measurement-model uncertainty and sensitivity to the time periods compared needs to be included.

(4) The authors define 'natural ozone' on L353, 'background ozone' on L363, 'domestic anthropogenic ozone' on L369, but do not define 'foreign anthropogenic ozone,' leaving it to the reader to infer a definition. Also, I'm not sure that 'natural ozone' is an accurate description of what is described, as humans have influenced atmospheric chemistry beyond just anthropogenic emissions, perhaps 'non-anthropogenic ozone' instead?

(5) Figure 10 should include a plot of the regions where Chinese emissions are the dominant contributor. Figure 10b shows that on average, China contributes $\sim$50% to surface ozone, and it's clear from Figure 8 that Chinese emissions dominate southeastern China's ozone. I'm not sure it's worthwhile then to point out the dominant foreign contribution to surface ozone over regions where Chinese emissions are dominant, especially when the foreign contribution is so low (Figure 8b,c). On its own, Figure 1a is an incomplete representation.

(6) Figure 11 is hard to parse, and given the large spatial heterogeneity shown in the

other Figures, it is not clear to me that a single vertical plot averaging all of China provides valuable information, or if it muddles interesting information through the averaging. This also applies to Figure 10b. Perhaps split these vertical profiles up into regions dominated by domestic and foreign contributions? Or perhaps apply some population weighing? In addition, Figure 11a should also include total ozone and a comparison should be made of total ozone (from the CTL run) and the sum of natural ozone, domestic anthropogenic ozone, and foreign anthropogenic ozone. It's not clear that these will match up, but it would speak to the non-linearity of the ozone simulations and contribution sensitivity simulations. Finally, I had difficulty in understanding Figure 11c as I initially assumed that Figure 11c was just a reformulation of Figure 11b in percentages rather than ppbv. The caption of the figure and the description on L484-485 are not clear, and as there is no description of how they arrived at this calculation, I'm unsure precisely what Figure 11c plots. The analysis summarized in these plots is interesting, but as is I have more questions that could be answered by subdividing these plots.

Technical Corrections:

Throughout the manuscript there are many acronyms that are used but not defined (e.g. MOZART, NAQPMS, PKUCPL).

L91: The ozone itself doesn't differ, but the plumes and chemical regimes, which produce and destroy the ozone, does differ.

L156-171: This paragraph mostly duplicates the information already in Table 1, and I do not feel that this redundancy is necessary.

L198-199: Figure 3 should be Figure 2

L250: These numbers do not match those found in Table 3

L265-266: The authors claim that the biases are due to overestimated free tropospheric and stratospheric transport, but it's not clear to me how this conclusion was reached.

The color scales in Figures 8a,b,d,e need to be consistent, as it requires extra effort to compare the Chinese Anthropogenic and Foreign Anthropogenic contributions. There is a risk that a casual reader would assume that the color scales in Figures 8a,b,d,e are the same, which would lead to incorrect conclusions.

L384: I don't feel that describing the air over the Sichuan Basin as "more isolated" is the correct description; rather the ozone chemistry of the region is controlled and dominated by domestic emissions and chemistry rather than foreign emissions.

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