

Reviewer #1

Comment#1-1: The authors examined source contributions of anthropogenic, background, and individual natural sources to surface ozone over China, as well as their differences between 2016 and 2017, in a chemical transport model GEOS-Chem. They found that natural background accounted 70-80% of surface ozone in China. Domestic anthropogenic sources contributed 30% during May-August and up to 69% during polluted ozone days. Ozone increases in 2017 relative to 2016 are due to hotter and dryer weather conditions. This topic is of interest and the manuscript is well written. I would suggest publishing after addressing my comments below.

Response#1-1: We thank the reviewer for the valuable comments. All of them have been implemented in the revised manuscript. Please see our itemized responses below.

Comment#1-2: My main concern is about the method for calculating source contribution. The authors considered the contribution from one source as the differences in ozone between BASE and sensitivity simulations with individual source emissions turned off. They did not consider the nonlinearity of source contribution. As the authors mentioned in the discussion that zero-out and 20% off emission would produce different contribution value. Ozone chemistry is complex. The nonlinearity exists non only in the amount of emission perturbed, but also in the species of emissions (VOCs or NO_x) and the location of emission perturbed. Directly comparing the source contribution derived from the differences of two simulation could be biased. The authors needs to quantify how large the nonlinearity would perturb the results with more simulations or, at least, discuss the potential biases of results related to the nonlinearity.

Response#1-2: To addressed the concern, we have further conducted an additional high-resolution simulation for July 2017 by decreasing Chinese anthropogenic emissions by 20% in the model, and presented the results in Figure S9. We state in the Section 5 (Discussions and Conclusion) “To further estimate the non-linear response of ozone to changes of sources, we conduct an additional sensitivity simulation with Chinese domestic anthropogenic emissions reduced by 20% in the model for July 2017. As shown in Figure S9, compared to the zero-out method, the 20% perturbation method estimates much lower domestic anthropogenic contributions (6.8 ppbv vs. 11.8 ppbv averaged over China in July 2017) with similar spatial distributions, which is consistent with Ni et al. (2018). Such strong non-linear responses of ozone to precursor emissions in China reflects that more stringent anthropogenic emission control measures are required to mitigate ozone pollutions.”.

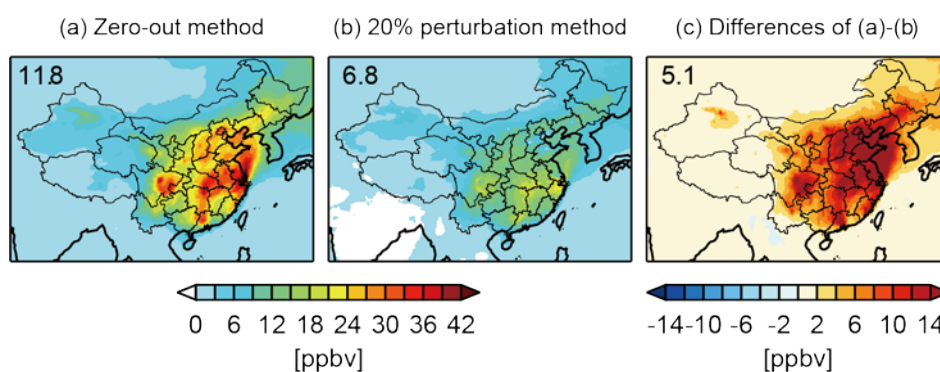


Figure S9. Spatial distributions of Chinese domestic anthropogenic contributions to surface MDA8

ozone in 2017 estimated from (a) zero-out methods (difference between the BASE simulation and the noCH simulation), (b) 20% perturbation method (five times of difference between the BASE simulation and the CH20off simulation). (c) shows the difference between (a) and (b).

Comment#1-3: The ozone contribution from each source was estimated as the ozone difference between the BASE simulation and each sensitivity simulation for BVOC, lightning NO_x, soil NO_x and biomass burning emissions. But the stratospheric contribution was quantified using an ozone tagging method. Is it appropriate to compare contributions derived from two different methods? How large the uncertainty it has?

Response#1-3: Thanks for pointing it out. Ozone in the lower stratosphere has much a longer lifetime (several years). Using the zero-out method to estimate stratospheric influences on tropospheric ozone in the model requires a very long spin-up time (more than ten years) and it is computationally unaffordable. Therefore, the tagged ozone simulation is typically used to quantify stratospheric influences on tropospheric ozone in chemical transport models. We now explain in the text: “Due to the long lifetime of ozone in the lower stratosphere (~years, Wang et al., 1998), the stratospheric contribution to tropospheric ozone is typically quantified using the tagged ozone simulation (TagO3) (Wang et al., 1998; Zhang et al., 2014), instead of perturbing the stratospheric ozone chemistry that requires a long spin-up time.”

Reference in the text:

Wang, Y., Jacob, D. J., and Logan, J. A.: Global simulation of tropospheric O₃-NO_x-hydrocarbon chemistry: 3. Origin of tropospheric ozone and effects of nonmethane hydrocarbons, *J. Geophys. Res.*, 103, 10757-10767, <http://doi.org/10.1029/98jd00156>, 1998.

Comment#1-4: How does the model treat the emission injection height? In the recent study, Yang et al. (2019) found that uncertainty in anthropogenic emission height strongly affects surface SO₂ concentration by about 80%. The inaccurate emission injection height can also lead to the bias of simulated surface ozone concentration.

Response#1-4: Thanks for pointing it out. We now state in the Section 2.2: “All anthropogenic emissions are emitted at the lowest layer in the model with a thickness of 120 m. A recent study found that uncertainties in industrial emission injection height could affect surface SO₂ concentrations by about 80% (Yang et al., 2019), and this may further affect surface ozone concentrations that requires further study.”

Reference in text:

Yang, Y., Smith, S. J., Wang, H., Lou, S., and Rasch, P. J.: Impact of Anthropogenic Emission Injection Height Uncertainty on Global Sulfur Dioxide and Aerosol Distribution, *J. Geophys. Res.*, <http://doi.org/10.1029/2018jd030001>, 2019.

Comment#1-5: Line 320: What are the ‘interactional’ and ‘pure’ contributions?

Response#1-5: We now state in the text to further elaborate the ‘interactional’ and ‘pure’ contributions: “These values are considered as the actual contributions of BVOCs on ozone, which include the pure contributions, i.e., contributions of BVOCs on ozone in the absence of

all other sources, and the interactional effects of BVOCs with other sources.”

Comment#1-6: Table: I suggest adding values for percentage change.

Response#1-6: Changed as suggested.