Anonymous Referee #2

The authors examined the effects of 2013-2017 changes in anthropogenic emissions on summertime ozone pollution over China, and they found that the emission controls for reducing aerosols have worsened urban ozone through the non-linear chemistry of ozone and the complex effects of aerosols. The current increasing trend of ozone in China is of great concern and this topic is well within the scope of ACP journal. The authors here present a very comprehensive study, and the manuscript is well structured. The estimated effects of emissions of individual chemical species on ozone are valuable for air quality planning in China. I would recommend it to be accepted after addressing the following comments.

Response: We thank the referee for providing a thoughtful review of our paper and the recognition of our work. The referee's comments have helped to improve this manuscript. Below, we provide a point-by-point response to the referee's comments and summarize the changes that have been made in the revised manuscript.

[Comment]: 1. Heterogeneous uptake of ozone. The simulated increases in ozone from this pathway (Fig.5h) are high over regions with high $PM_{2.5}$ concentrations other than regions with high levels of mineral dust. I am wondering if you are applying this effect for all the aerosols or just on dust particles. The uptake of ozone by aerosols are only well documented for mineral aerosol. Bauer et al. (2004) also suggested that the lower limit of uptake coefficient (3x10-6) seems to be more appropriate for global modeling.

Response: We applied an uptake coefficient of O_3 (γ_{O_3}) of $1x10^{-5}$ to all aerosols in our model simulations. Previous laboratory and measurement studies indicated that γ_{O_3} varied in a wide range on different aerosols: 10^{-6}-10^{-4} on dust (Michel et al., 2002, 2003; Hanisch and Crowley, 2003), up to 10^{-4} on sodium chloride aerosols (Abbatt and Waschewsky, 1998), and 10^{-5}-10^{-3} on soot particles (Longfellow et al., 2000). Most previous modeling studies adopted $1x10^{-5}$ (Liao et al., 2004; Liao and Seinfeld, 2005; Pozzoli et al., 2008), while one recommended a lower value ($3x10^{-6}$) for dust particles (Bauer et al., 2004). We think our choice of 10^{-5} is reasonable.

Revision in the main text:

1) Line 134-138:

"Previous laboratory and measurement studies of the heterogeneous reaction of O_3 have given a wide range of (γ_{O_3}, from 10^{-6} to 10^{-4} on dust (Michel et al., 2002, 2003; Hanisch and Crowley, 2003), up to 10^{-4} on sodium chloride aerosol (Abbatt and Waschewsky, 1998), from 10^{-5} to10^{-3} on soot particles (Longfellow et al., 2000). Most previous modeling studies adopted $1x10^{-5}$ (Liao et al., 2004; Liao and Seinfeld, 2005; Pozzoli et al., 2008), while one recommended a lower value (3×10^{-6}) for dust particles (Bauer et al., 2004). We applied 10^{-5} to the uptake of O_3 on all the aerosols in our simulation."

Reference:

Abbatt, J. P. D., and Waschewsky, G. C. G.: Heterogeneous Interactions of HOBr, HNO_3, O_3, and NO_2 with Deliquescent NaCl Aerosols at Room Temperature, The Journal of Physical Chemistry A, 102, 3719-3725, 10.1021/jp980932d, 1998.

Bauer, S. E., Balkanski, Y., Schulz, M., Hauglustaine, D. A., and Dentener, F.: Global modeling of heterogeneous chemistry on mineral aerosol surfaces: Influence on tropospheric ozone chemistry and comparison to observations, J Geophys Res-Atmos, 109, 10.1029/2003jd003868, 2004.

Hanisch, F., and Crowley, J. N.: Ozone decomposition on Saharan dust: an experimental investigation, Atmos. Chem. Phys., 3, 119-130, 10.5194/acp-3-119-2003, 2003.

Liao, H., Seinfeld, J. H., Adams, P. J., and Mickley, L. J.: Global radiative forcing of coupled tropospheric ozone and

aerosols in a unified general circulation model, J Geophys Res-Atmos, 109, 2004.
Liao, H., and Seinfeld, J. H.: Global impacts of gas-phase chemistry-aerosol interactions on direct radiative forcing by anthropogenic aerosols and ozone, 110, 10.1029/2005jd005907, 2005.
Longfellow, C. A., Ravishankara, A. R., and Hanson, D. R.: Reactive and nonreactive uptake on hydrocarbon soot: HNO_3, O_3, and N_2O_5, 105, 24345-24350, 10.1029/2000jd900297, 2000.
Michel, A. E., Usher, C. R., and Grassian, V. H.: Heterogeneous and catalytic uptake of ozone on mineral oxides and dusts: A Knudsen cell investigation, 29, 10-11-10-14, 10.1029/2002gl014896, 2002.
Michel, A. E., Usher, C. R., and Grassian, V. H.: Reactive uptake of ozone on mineral oxides and mineral dusts, Atmos. Environ., 37, 3201-3211, https://doi.org/10.1016/S1352-2310(03)00319-4, 2003.
Pozzoli, L., Bey, I., Rast, S., Schultz, M. G., Stier, P., and Feichter, J.: Trace gas and aerosol interactions in the fully coupled model of aerosol-chemistry-climate ECHAM5-HAMMOZ: 1. Model description and insights from the spring 2001 TRACE-P experiment, 113, 10.1029/2007jd009007, 2008.

[Comment]: 2. The updated model will decrease NO_2 concentration, and it compares better with surface NO_2 in summer 2013. But the model also has low biases for summers 2014- 2017 (in Table 2 of the companion paper). Please have more explanations on this.

Response: In Part 1 (Liu and Wang, 2020), we had given one main reason for the low biases of simulated NO_2 for summers 2014-2017, which is related to the fact that the NO_2 concentrations in the national observation network were measured using the catalytic conversion method, which overestimates NO_2, especially during periods of active photochemistry (Xu et al., 2013; Zhang et al., 2017; Fu et al., 2019). Therefore, the updated model has improved simulations for all the years.

Reference:

Fu, X., Wang, T., Zhang, L., Li, Q., Wang, Z., Xia, M., Yun, H., Wang, W., Yu, C., Yue, D., Zhou, Y., Zheng, J., and Han, R.: The significant contribution of HONO to secondary pollutants during a severe winter pollution event in southern China, Atmos. Chem. Phys., 19, 1-14, 10.5194/acp-19-1-2019, 2019.
Liu, Y., and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 – Part 1: The complex and varying roles of meteorology, Atmos. Chem. Phys. Discuss., 2020, 1-28, 10.5194/acp-2019-1120, 2020.
Xu, Z., Wang, T., Xue, L. K., Louie, P. K. K., Luk, C. W. Y., Gao, J., Wang, S. L., Chai, F. H., and Wang, W. X.: Evaluating the uncertainties of thermal catalytic conversion in measuring atmospheric nitrogen dioxide at four differently polluted sites in China, Atmos. Environ., 76, 221-226, 10.1016/j.atmosenv.2012.09.043, 2013.
Zhang, L., Li, Q. Y., Wang, T., Ahmadov, R., Zhang, Q., Li, M., and Lv, M. Y.: Combined impacts of nitrous acid and nitryl chloride on lower-tropospheric ozone: new module development in WRF-Chem and application to China, Atmos Chem Phys, 17, 9733-9750, 10.5194/acp-17-9733-2017, 2017.

[Comment]: 3. The simulated decreases in NO_3 and N_2O_5 (Lines 185-195) could be also induced by decreased ozone in the updated simulation.

Response: Thanks for this comment which we agree. We have modified the statements in the revised manuscript.

Revision in the main text:

1) Line 187:
"The simulated NO_3 mixing ratio decreased slightly (~1 pptv) due to the decrease in NO_2 and O_3 mixing ratios"
2) Line 192-193:

"the decrease in NO_2 and O_3 levels resulted in a decrease in N_2O_5"

[Comment]: 4. The Conclusion section needs to be rewritten. Currently the 9 lines of conclusion are not a good summary of what have been done in the manuscript. Quantitative conclusions should be given in both Abstract and Conclusion section.

Response: We didn't intend to repeat the content of the abstract. Nonetheless, we understand the referee's view and have added more content in the revised conclusion section.

Revision in the main text:

1) Line 379-393:

"This study has quantified the effects of changes in pollutant emissions from anthropogenic activities on the summer surface O_3 concentrations over China from 2013 to 2017. The control measures, while successful in reducing the concentrations of primary pollutants and particulate matter, were found to increase urban O_3 but reduce rural O_3; overall, the NO_x emission reduction has helped to contain total ozone production in China. The reduction in NO_x emission and slight increase in VOC emissions led to ozone increase in urban areas due to the non-linear chemistry of O_3, and the large reductions in PM and SO_2 emissions contributed to urban ozone increase resulting from the complex effects of aerosols on radiation and chemical reactions. Among the primary PM components, the emission decrease in BC increased O_3 more than that for OC despite its smaller reduction compared to OC, resulting from BC being a strong absorber of solar radiation. The dominant causes of the urban ozone increase due to emission change varied among different cities, and they were NO_x and PM in Beijing, NO_x and VOC in Shanghai, NO_x in Guangzhou, and PM and SO_2 in Chengdu. For the aerosol effects, the decrease in heterogeneous uptake of reactive gases was more important than the increase in photolysis rates. Only the CO emission cut helped to decrease urban ozone. Our results show that comparable percentage reductions in anthropogenic VOCs to that achieved for NO_x could have prevented the increases in urban O_3 concentrations. We thus recommend that VOCs control be implemented in current and future emission-reduction measures to improve the overall air quality. In view of the importance and complexity of the uptake of reactive gases on aerosol surfaces, more research should be conducted in this area."

[Comment]: 5. The manuscript is not clear about the impact of boundary conditions of chemical species on simulated O_3 in China. Ideally the chemical boundary conditions are different for 2013 and 2017, considering the differences in anthropogenic emissions and in meteorology outside the model domain. How would these differences at the boundary influence simulated changes in O_3 in China over 2013-2017? Some discussions can be added in Conclusion section.

Response: The impact of boundary conditions has been simulated and discussed in Part 1 (Liu and Wang, 2020), so we will not repeat them in the present paper. Briefly, the chemical boundary conditions for the CMAQ model were derived from the results of the MOZART global model, and they varied in 2013-2017. We found that the impact of the long-range transport from outside the CMAQ modeling domain to China contributed to the increase in MDA8 O_3 in China during 2013-2017, especially on the Qinghai-Tibetan Plateau (with an increase of 1 to 4 ppbv). More discussions are presented in Part 1, Section 3.6 (Liu and Wang, 2020).

Revision in the main text:

1) Line 78-80:

"The role of meteorological variation and total emission changes, the effect of changes in individual meteorological factors, and the impact of changes in long-range transport of O_3 and its precursors from outside

the modeling domain, are discussed in a companion paper, Part 1 (Liu and Wang, 2020).”

Reference:

Liu, Y., and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 – Part 1: The complex and varying roles of meteorology, Atmos. Chem. Phys. Discuss., 2020, 1-28, 10.5194/acp-2019-1120, 2020.