Anonymous Referee #1

This is a very well written paper, which provides a thorough analysis of the impacts of gas and PM emission controls on ozone formation across China. The paper is appropriate for ACP and it works well with the companion paper that is also under review with ACPD. As described below, there are a few items that need to be addressed, after which the paper would be suitable for publication in ACP.

Response: We thank the referee for providing a thoughtful and detailed review of our paper. 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.

Major Comments:

[Comment]: 1. Lines 230-232 Here some context needs to be provided for these trends, and some evaluation against observations is warranted. Figure S1 shows that observed ozone increased by 18% across all urban areas with ozone monitors. However, the model indicates that urban ozone increased from 55 to 57 ppbv, which is just a 3.6 % increase, a rate that is five times less than the observed rate. Why are the modeled trends so low compared to the observed trends, and which processes are being missed by the models? To help the readier understand the discrepancy between the model and observations the authors need to directly compare the model to observations. For example they can compare the modeled trend in the grid cell (or cells) above Beijing to all of the monitors with data from 2013-2017. They can make similar plots for the other urban areas of YRD, PRD and SCB

Across all of China the model predicts a very small ozone decrease of 0.6 ppbv, or just 1%. It's difficult to believe that this tiny decrease has any real meaning. How is the p-value (0.006) so low? What kind of statistical test was used? To have such a tiny decrease with such a low p-value indicates that the signal-to-noise ratio is very high, which implies that there is very little interannual variability. But Part I of this study shows that meteorology creates substantial interannual variability.

Response: Thanks for this valuable comment. The simulated MDA8 O_3 increase (~2 ppbv) in the nightlight-classified urban areas from 2013 to 2017 is much lower than the average increase observed at 493 sites in 74 cities (~9 ppbv). The discrepancy can be explained as follows. The urban areas determined using the nightlight data are not exactly the same as those 493 sites and cover some rural areas (with decreasing ozone) and additional small townships (see Fig R1 below). If we match the model output with the observation sites, then the model can capture 57% of the average rate of increase at those sites, as shown in Figure R2 below. We had compared the simulated and observed MDA8 O_3 changes in Beijing (BTH), Shanghai (YRD), Guangzhou (PRD), and Chengdu (SCB) in Section 3.3 of Part 1 (Liu and Wang, 2020). The result showed that the model could also generally capture the changes in observed MDA8 O_3 in different cities. We have added some texts in the revised manuscript to clarify this discrepancy.

The p value was calculated using the F-test statistical method. As shown in Fig. 1, the MDA8 O_3 mixing ratio across all of China did present a small decreasing trend with a high confidence level ($p=0.006$). Part 1 of this study showed the large variability of meteorological impacts on O_3 in regions and years, but this regional variability can be 'averaged out' over the whole China, leading to a clearer ozone trend. The very small ozone decreases in China indicated that the ozone concentration has leveled off in recent years, attributable to the decrease in large rural areas due to the NO_x emission reduction. The recently published studies also supported our model predicted ozone decreases in rural areas of eastern China. Wang et al. (2019) revealed no significant change in O_3 levels observed at a coastal site (Hok Tsui) in South China in the outflow of air mass from eastern China during 2007-2018. Xu et al. (2020) reported decreasing O_3 mixing ratios from 2013 to 2016 at two rural sites in BTH (Shangduanzi) and YRD (Linan).

Revision in the main text:

1) Line 227-242:

"The model predicted that the MDA8 O_3 mixing ratio in urban areas increased at a rate of 0.46 ppbv per year (ppbv a^{-1}) ($p = 0.001$). This simulated increase (~2 ppbv from 2013 to 2017) in the nightlight-classified urban areas is much lower than the average increase observed at 493 sites in 74 cities (~9 ppbv, Fig. S1d). The discrepancy can be explained as follows. The urban areas determined using the nightlight data are not exactly the same as those 493 sites and cover some rural areas (with decreasing ozone) and additional small townships (Fig. S3). When we matched the modeled locations to the 493 observation sites, the model captured 57% of the rate of increase of MDA8 O_3 averaged at those sites (see Fig. S3 in Part 1 (Liu and Wang, 2020)). Part 1 also showed a large variability of meteorological impacts on O_3 in different regions (e.g., Beijing, Shanghai, Guangzhou, and Chengdu), and the simulated overall urban O_3 trend with a high confidence level ($p = 0.001$) suggests that this regional variability in meteorological impact can be 'averaged out', leading to a clearer urban O_3 trend driven by emission changes.

The simulated MDA8 O_3 mixing ratio in rural areas decreased at a rate of 0.17 ppbv a^{-1} ($p = 0.005$), which is supported by the recently reported rural ozone trends in China. Wang et al. (2019c) revealed no significant change in O_3 levels observed at a coastal site (Hok Tsui) in South China in the outflow of air mass from eastern China during 2007-2018. More recently, Xu et al. (2020) reported decreasing O_3 mixing ratios from 2013 to 2016 at two rural sites in BTH (Shangduanzi) and YRD (Linan). Overall, MDA8 O_3 mixing ratio in China exhibited a slightly decreasing trend (0.15 ppbv a^{-1}, $p = 0.006$) due to the decrease in a large rural area, which suggested that the ozone concentration has leveled off in recent years."

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.

Wang, T., Dai, J., Lam, K. S., Nan Poon, C., and Brasseur, G. P.: Twenty-Five Years of Lower Tropospheric Ozone Observations in Tropical East Asia: The Influence of Emissions and Weather Patterns, 46, 11463-11470, 10.1029/2019gl084459, 2019c.

Xu, X., Lin, W., Xu, W., Jin, J., Wang, Y., Zhang, G., Zhang, X., Ma, Z., Dong, Y., Ma, Q., Yu, D., Li, Z., Wang, D., and Zhao, H.: Long-term changes of regional ozone in China: implications for human health and ecosystem impacts, Elem Sci Anth, 8, 13, 10.1525/elementa.409, 2020.

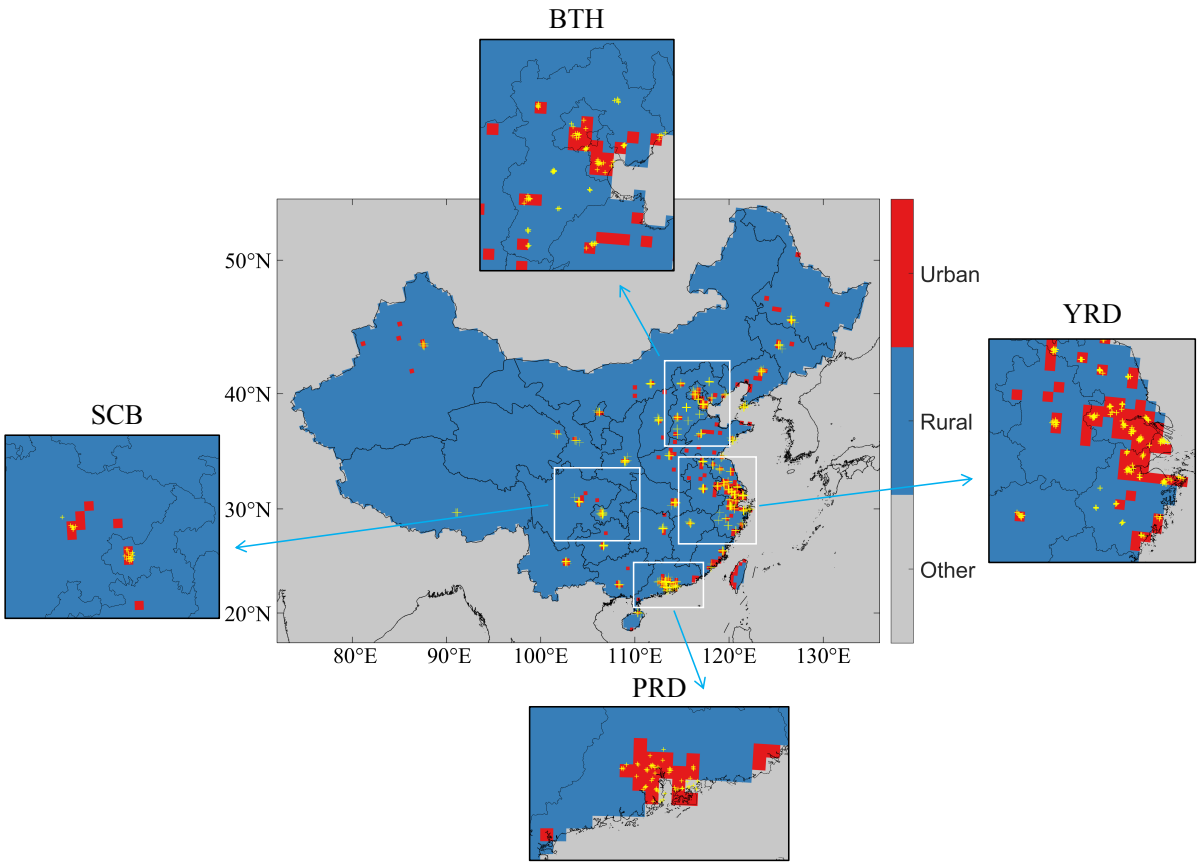

Figure R1 (Figure S3) Spatial distribution of the urban and rural areas in land areas of China identified by using the nighttime light data. The yellow cross "+" represents the locations of 493 environmental monitoring stations in 74 cities since 2013. BTH, YRD, PRD, SCB are the Beijing-Tianjin-Hebei, Yangtze River Delta, Pearl River Delta, and Sichuan Basin regions, respectively.

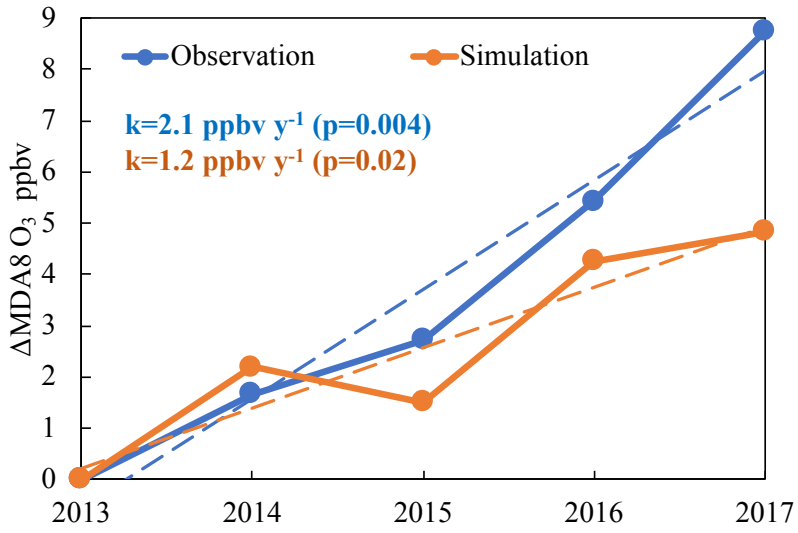

Figure R2 (Fig. S3 in Part 1 (Liu and Wang, 2020)) Changes in observed and simulated summer surface MDA8 O_3 mixing ratios averaged in 493 sites of 74 cities during 2013-2017 relative to those of 2013.

[Comment]: 2. This science paper strays into the realm of policy recommendations, as follows: Line 308-310 "The inter-city variations in the dominant causes of increases in O_3 concentrations mean that the government should adopt additional, localized emission-reduction measures as part of policies aimed to alleviate urban O_3 pollution (see section 3.5)."

Line 343 "3.5 The need for concurrent reduction of anthropogenic VOCs emissions"

Line 370 "Therefore, VOCs emission controls should be implemented together with the PM-targeted measures."

"Line 377-379 We thus conclude that VOCs controls should be implemented in current and future emission-reduction measures to improve the overall air quality."

I understand that the authors want their paper to be beneficial for improving air quality in China, and their results will certainly be useful. However, the recommendations will have to be re-phrased so that this science paper does not sound like a policy document. Fortunately, this is a straightforward editorial process. Instead of saying what the government "should" do, the authors can say something like: "Recent emission controls across China have not reduced ozone and have actually increased ozone in urban areas. If the government wishes to adopt new emissions control policies that will reduce ozone in urban and rural areas we propose the following recommendations for VOC controls. . .." By phrasing it like this, your paper offers very useful options to the government without sounding like a policy paper.

Response: It was our intention to emphasize the policy implications of the results. We understand the referee's viewpoint. In the revised region, we have rephrased these descriptions and made it not reading like a policy paper.

Revision in the main text:

1) Line 316-318:
   "The inter-city variations in the dominant causes of increases in O_3 concentrations suggest that if the government wishes to alleviate urban O_3 pollution, they can adopt additional, localized emission-reduction measures as part of policies (see section 3.5)."
2) Line 350:
   "3.5 The anthropogenic VOCs emission control to reduce O_3"
3) Line 376-377:
   "Therefore, we suggest VOCs emission controls be implemented together with the PM-targeted measures in order to alleviate the urban O_3 pollution."
4) Line 391-392:
   "We thus recommend that VOCs control be implemented in current and future emission-reduction measures to improve the overall air quality."

[Comment]: 3. This study focuses on summer, but did the authors also look at ozone changes during the winter months? TOAR-Climate (Gaudel et al., 2018) compares surface ozone trends at non-urban sites across North America, during 2000-2014, a period of deceasing NO_x emissions. Ozone decreases across much of the continent in summer, but increases in winter (see their Figures 13, 14 and 15). I wonder if a similar pattern has occurred across China in winter.

Gaudel, A., et al. (2018), Tropospheric Ozone Assessment Report: Present- day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, Elem Sci Anth, 6(1):39, DOI: https://doi.org/10.1525/elementa.291

Response: We had also examined the ozone trend in winter. Figure R3 below depicts the observed ozone changes during the winter months (January, February, and December) from 2013-2017 at the same 493 cities. Like summer, the averaged MDA8 O_3 concentration also presented an overall increasing trend in winter. As the present study

focuses on the photochemically active summer season, we do not discuss the winter result.

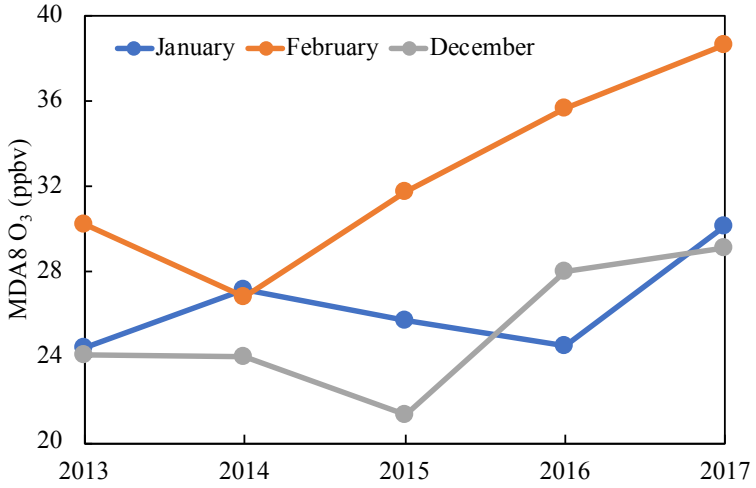

Figure R3 Variation in observed MDA8 O_3 mixing ratios in 493 environmental monitoring stations of 74 cities in January, February, and December during 2013-2017.

Minor Comments

[Comment]: 4. Line 286-288 Here the authors state that, in general, BC has a major impact on photolysis rates. But the overall conclusion from this study is that the impact of PM reductions on ozone production is mainly through the changes in heterogeneous chemistry, with the impact on photolysis rates being secondary. Given the conclusions of the study it would be a good idea to provide some additional context for the impact of BC on photolysis rates and ozone production.

Response: Following the referee's suggestion, we have provided some additional context for the impact of BC in the Conclusion section.

Revision in the main text:

1) Line 385-386:
   "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."

[Comment]: 5. Line 104 Here and elsewhere, there is no such word as "uptakes". To make it plural you can use "uptake rates"

Response: Thanks for the suggestion. We have replaced the word "uptakes" throughout the manuscript.

Revision in the main text:

1) Line 103-104:
   "the absorptions of NO_2, NO_3, and N_2O_5 on aerosol surfaces"
2) Line 106:
   "those of HO_2, O_3, OH, and H_2O_2"
3) Line 199-200:
   "the incorporation of their heterogeneous reactions"
4) Line 335-336:

"the heterogeneous reactions of NO_2, NO_3, and OH"

[Comment]: 6. Line 143 This sentence would sound better as: "The companion paper (Part 1; (Liu and Wang, 2020)) presented validation results. . ."
Response: Thanks for the suggestion. We have changed "comprised validation results" into "presented validation results" in Line 143 in the revised manuscript.

[Comment]: 7. Line 208 Change "observation" to "observations"
Response: Changed in Line 205.

[Comment]: 8. Line 209 Here and throughout the paper, when mentioning a trace gas value in units of ppbv, then the quantity must be referred to as a mixing ratio, and not a concentration, which has units of mass per volume.
Response: Thanks for this suggestion. We have changed "concentration" into "mixing ratio" in Line 206 in the revised manuscript. We also carefully went through the document and made similar changes throughout the revised manuscript.

[Comment]: 9. Line 331 has should be was ". . .where the $PM_{2.5}$ concentration was high and WAS subject. . ."
Response: Thanks for the suggestion. We have changed "has subject to" to "was subject to" in Line 338 in the revised manuscript.