Dear Guy Brasseur Referee:

We are submitting the point-by-point responses to your comments. We thank you for comments and suggestions, and hope you are satisfied with our responses.

The major changes made in the revised version include:

1. Added related discussion about the role of heterogeneous chemistry in the revised manuscript.
2. Added more discussions on the role of the boundary layer and changing meteorology in O_3 trends.
3. Added a discussion on the HOx chemical source in the revised manuscript.

On behalf of the co-authors,

First Author: Xiaodan Ma
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Point-by-point responses to the Comments/Suggestions of Reviewer #3

Overview

The paper in its present form is clearly written and could be published after revisions. At this stage of the review process, I would like to add a few points and suggestions to be carefully addressed by the authors:

General comments

1. Since the focus of the paper is on the response of ozone to possible forcing processes, I wonder why so little is said about the role of heterogeneous chemistry. There is growing evidence that the increase in ozone is related to the increase in HO_2 due to the reduced scavenging of HO_2 by aerosols (PM) more than a change in the J-values. The paper highlights the change in the $J(NO_2)$ due reduced PM concentrations, but does not provide the quantitative response associated with the change in heterogenous processes. It would be important to discuss this aspect, even if no specific simulation of this effect has been done.

Thanks for the reviewer's suggestion. According to recent researches, decrease in $PM_{2.5}$ was considered as one of the important causes leading to such an increase in surface O_3 mainly due to additional O_3 production associated with reduced sink of hydroperoxyl radicals (HO_2) (Li et al., 2019). They pointed out that increase in surface O_3 associated with decrease in $PM_{2.5}$ was more prominent than that with reduction of NOx emissions over the NCP region where O_3 formation was dominated by VOC-limited regime. Liu and Wang (2020a, 2020b) found the reduction of PM emissions increased the O_3 levels by enhancing the photolysis rates and reducing heterogeneous uptake of reactive gases (mainly HO_2 and O_3), of which the latter is more important than the former. However, the MM model does not include aqueous-phase chemistry that has been implemented in the 3D meteorology/chemistry models (e.g., Li et al., 2019; Liu and Wang, 2020a, 2020b). Thus, inclusion of detailed aerosol chemistry and observation-based uptake coefficients in a box model like MM is necessary to provide more accurate assessment of impact of aerosol radiative effect on surface O_3 change in the future.

We have included the related discussion about the role of heterogeneous chemistry both in the Introduction and Discussion part (See L92-98 and L407-413).

2. A more convincing discussion must be provided regarding the role of the boundary layer, the changing meteorology (e.g., average cloudiness, precipitation, etc.,) and the surface deposition processes. Some of these may not be explicitly treated in a box model, but should be discussed with appropriate references.

Thanks for the comment. We agree with the reviewer that the boundary layer and the changes in other meteorological factors such as cloudiness, precipitation etc. played an important role in increase in surface O_3. In fact, we have conducted two cases (cases F and G in Table 1) to assess the impact of the planetary boundary layer height (PBLH) on the change in surface O_3. In addition, we have included two cases (i.e., E and G in Table 1) to investigate the impact of surface maximum air temperature on increase in surface O_3. To highlight the reviewer's concern with the role of changing meteorology, we cited a relevant reference on this topic, "The influence of changing meteorological factors on the change trend in surface O_3 may vary greatly with regions and time. In addition to air temperature and the boundary layer conditions, other meteorological factors such as cloud cover, precipitation, wind fields played an important role in driving the changes in surface O_3 observed in many places of China (e.g., Liu and Wang, 2020a)" (see L416-420).

3. There should be a discussion about the processes that have changed HO_x chemistry (which affects the ozone production and loss) and this includes, for example, the HONO and formaldehyde photolysis.

We agree with the reviewer that HO_x chemistry is an important factor affecting the production and loss of O_3. Now a statement with "The HONO photolysis as the primary production of OH radicals and the formaldehyde (HCHO) photolysis as the net radical source of HO_2 can lead to major changes in the HOx and NOx budget that may have an important effect on O_3 production and loss (e.g., Aumont et al., 2003; Brasseur et al., 2006; Lin et al., 2012)" is added on the revised version (see L174-179).

References:

Aumont, B., Chervier, F., and Laval, S.: Contribution of HONO sources to the NOx/HOx/O3 chemistry in the polluted boundary layer, Atmos. Environ., 37, 487-498, https://doi.org/10.1016/S1352-2310(02)00920-2, 2003.

Brasseur, G. P., and Solomon, S.: Aeronomy of the middle atmosphere: Chemistry and physics of the stratosphere and mesosphere, Springer Science & Business Media, 2006.

Li, K., Jacob, D. J., Liao, H., Shen, L., and Bates, K. H.: Anthropogenic drivers of 2013-2017 trends in summer surface ozone in China, National. Acad. Sciences., 116, 422-427, https://doi.org/10.1073/pnas.1812168116, 2019.

Lin, Y. C., Schwab, J., Demerjian, K., Bae, M.-S., Chen, W.-N., Sun, Y., Zhang, q., Hung, H.-M., and Perry, J.: Summertime formaldehyde observations in New York City: Ambient levels, sources and its contribution to HOx radicals, J. Geophys. Res., 117, D08305, https://doi.org/10.1029/2011JD016504, 2012.

Liu, Y., and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 – Part 2: The effects of emission changes and implications for multi-pollutant control, Atmos. Chem. Phys. Discuss., 2020, 1-27, 10.5194/acp-2020-53, 2020a.

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., 20, 6305-6321, https://doi.org/10.5194/acp-20-6305-2020, 2020b.