Anonymous Referee #2

This manuscript presented a comprehensive modeling analysis on the surface ozone trends over China during 2013-2017. Significant ozone increases have been observed in China over this period in spite of the strong emission control actions implemented. Better understanding the drivers of these trends is of great scientific importance. The authors have conducted an ensemble of numerical simulations using the WRF-CMAQ air quality model to interpret these surface ozone trends, in particular, quantifying the role of meteorology in this manuscript. The results showed that the model had some success in reproducing the Chinese ozone increase trends, supporting the use of it to assess contributions from changes in anthropogenic emissions vs. changes in meteorology. The results further emphasized the importance of interannual variations in meteorology affecting the recent surface ozone trends in China.

This is an important study, representing a great step to understand the drivers of interannual changes in summertime surface ozone pollution in China. The manuscript is well organized and written, and the methodology and results sound solid. I recommend publish after the following comments been addressed.

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.

Specific comments:

[Comment]: 1. Page 5, Line 120-125:

Some recent studies have suggested that the ozone increases in China since 2013 were largely driven by the concurrent decreases in PM2.5 levels and the resulting changes in heterogenous HO_2 uptake by aerosol surfaces (Li et al., 2019a, 2019b). Since the model applied in this study reproduced the observed ozone increases, did the results support the important role of heterogenous reactions? Although the authors may discuss this issue in the second paper, I suggest put some sentences in this paper in the context of these recent findings.

Reference:

Li, K., et al., Anthropogenic drivers of 2013-2017 trends in summer surface ozone in China, P Natl Acad Sci USA, 116, 422-427, 2019.

Li, K. et al., A two-pollutant strategy for improving ozone and particulate air quality in China, Nature Geoscience, 12, 906-910, 2019.

Response: Thanks for pointing out this and suggesting the two recent papers. We have discussed the impact of heterogeneous reactions on the ozone changes in the second paper (Liu and Wang, 2020), which also supported the significant role of heterogeneous reactions in the increasing urban ozone concentration across China. Following the referee's suggestion, we have revised the manuscript and placed the incorporation of comprehensive heterogeneous chemistry into the CMAQ model within the context of these recent findings.

Revision in the main text:

1) Line 121-124:

"The original CMAQ model includes the heterogeneous reactions of only NO_2, NO_3, and N_2O_5 on aerosol surfaces. Recent studies (Li et al., 2019a; Li et al., 2019b) have suggested that the heterogeneous reactions on aerosol surfaces, mainly the uptake of HO_2, played a significant role in the increasing O_3 concentrations in China from 2013 to 2017. To better simulate the effects of aerosol on ozone via heterogeneous reactions, …"

Reference:

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, 2020.

[Comment]: 2. Page 8, Figure 4:

The year 2013 seems to be a special year with particularly low ozone values, for example, as can be seen from Figure 4a) over Beijing (the BTH region). If the 2013 data point was removed from the linear trend calculation, then no trend was observed for Beijing. This is also the case for Guangzhou (Figure 4c) and the long-range transport ozone influences (Figure 7). Can you comment on this?

Response: Thanks for this comment. For some regions, such as Beijing and Guangzhou, the year 2013 does seem to be a special year with particularly low ozone concentration. But this is not the case in other regions such as Shanghai and Chengdu, in which the ozone concentrations in 2013 were higher than those in 2014. Figure 3a-d also shows that the changes in MDA8 O_3 during 2014-2017 relative to 2013 were increases or decreases, depending on regions and years. Such characteristics are attributed to the complex and varying roles of meteorology in ozone changes, which are highlighted in this paper.

As for the impact of long-range transport on ozone changes, we think the year 2013 is also not a special year with the lowest influence from the long-range ozone transport compared with other years. We remove the 2013 data and plot Figure R1 below to show the changes in the simulated MDA8 O_3 due to variations in long-range transport in 2015, 2016, and 2017 relative to 2014. Similar increases in ozone are still found, which supports the increasing contribution from long-range transport to ozone in these years. We have added this statement to the revised manuscript.

Revision in the main text:

1) Line 231-232:
   "As shown in Fig. 4, the changes in observed MDA8 O_3 varied in cities and years, which were generally captured by the model"
2) Line 346-348:
   "Increases in ozone levels were also found if we compared the changes in MDA8 O_3 due to variations in chemical boundary conditions relative to 2014."

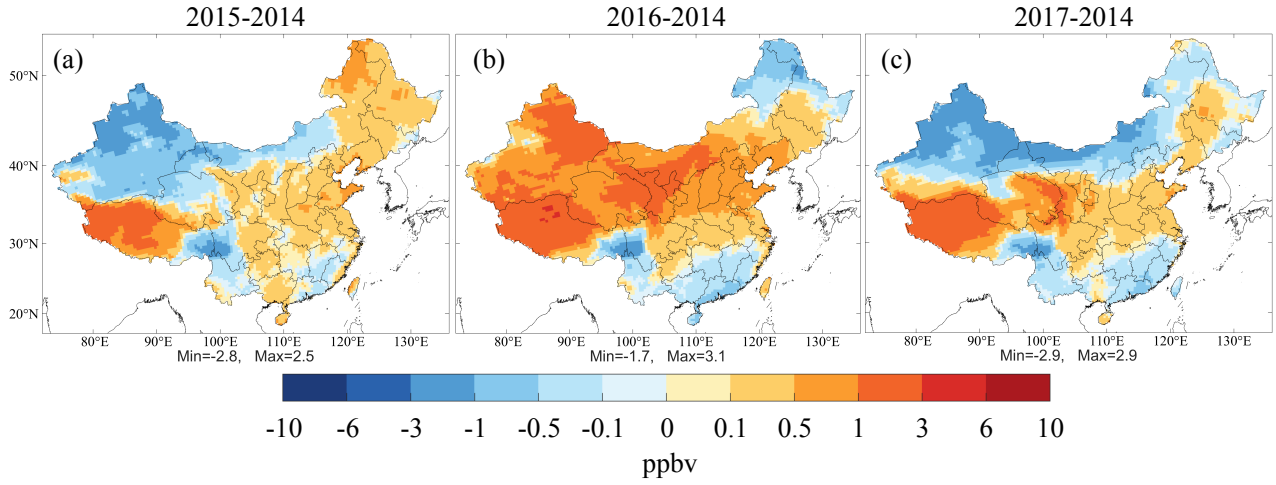

Figure R1 Changes in the simulated summer surface MDA8 O_3 mixing ratios due to variations in long-range transport over China in 2015, 2016, and 2017 relative to 2014.

[Comment]: 3. Page 10, first paragraph of section 3.5:

Figure 6 showed that the 2013-2017 changes in wind significantly increased surface ozone over most regions of China, and the authors attributed the ozone increases to enhanced transport from the lower stratosphere. This is not clear to me. It may explain some of the surface ozone increases in the northern and central China as argued by the PV changes, but how about the southern China where I think stratospheric ozone influences would be low at surface? It is not clear that enhanced ozone transport from the lower stratosphere could lead to 6-10 ppbv surface ozone increases in the southern China. I wonder whether changes in horizontal winds still contribute there, e.g., changes in the wind speed and the summer Asian monsoon. Please clarify.

Response: Thanks for this comment. Referee #1 raised a similar concern. We think that the PV analysis works well for Qinghai-Tibetan Plateau of western China with terrain heights > 3km. But for other regions (eastern and southern), surface ozone increases are due to a decrease in wind speeds, which would help the accumulation of ozone and ozone precursors in these regions and increase ozone concentrations. By examining the wind directions in the two years, we did not find evidence for change in vertical transport from the free troposphere to the surface or change in horizontal transport within the model domain. We have revised this section and clarified the possible reasons for the increasing ozone concentrations in terms of the changes in wind speed, horizontal transport, and vertical transport. In Figure 6, we replaced the panels about PV with those about wind speed.

Revision in the main text:

1) Line 22-23:
   "The results show that the wind field change made a significant contribution to the increase in surface ozone over many parts of China."
2) Line 291-299:
   "Notable increases in MDA8 O_3 in western and eastern China due to the change in wind fields were identified, which contributed significantly to the meteorology-induced increasing ozone (Fig. 3h). In the Qinghai-Tibetan Plateau of western China whose terrain heights are greater than 3 km, the significant increase in the MDA8 O_3 mixing ratio (3 to 9 ppbv) due to wind change from 2013 to 2017 can be attributed in part to the enhanced downward transport from the upper troposphere as indicated by the increase in the potential vorticity (PV) (Fig. S6). In eastern China, the increase in O_3 level can be explained by the decrease in the wind speeds (Fig. 6h), which helps the accumulation of O_3 and its precursors and then increases ozone concentrations. There is no strong evidence for the change in the vertical transport from the free troposphere to the surface in eastern China and the horizontal transport from other regions within the modeling domain between these two years, according to the wind data (Fig. S7)."
3) Line 360-361:
   "The results show that the changes in the wind fields made a significant contribution to the increase in surface ozone levels over many parts of China."
4) Line 382-384:
   "The increase in MDA8 O_3 in Qinghai-Tibetan Plateau from 2013 to 2017 was ascribed to enhanced downward transport from the upper troposphere."
5) Line 646-647 (the caption of Figure 6):
   "wind speed at a height of 10 m (Wind)"

[Comment]: 4. Page 10, Line 301-302:

The statement "we found that the impact of temperature via the change in the chemical reaction rate was more

significant than that via the change in biogenic emissions from 2013 to 2017" need to be more quantitative. It is difficult to read from Figure 5 and Figure 6 (the color bars are too small). It might depend on regions. I suggest compare their values averaged over the key regions and over China.

Response: Thanks for this suggestion. In the revised manuscript, we have enlarged the panels in Figures 5 and 6 and provided high-resolution figures, which can help to compare the impacts of changes in temperature and biogenic emissions on MDA8 O_3. Figure 5o and 6c are put together in Figure R2 below. We also compared their values averaged in four typical cities, namely Beijing, Shanghai, Guangzhou, and Chengdu, in Figure R3 (also being provided in the supplementary material as Figure S8). We noted that the impact of temperature and biogenic emissions on ozone could be different in some regions (e.g., opposite impacts in Beijing), which can be explained by the transport of emitted pollutants. However, the increases (decreases) in MDA8 O_3 due to changes in temperature were generally higher than those due to changes in biogenic emissions, which supported the statement in our manuscript.

Revision in the main text:

1) Line 304-306:

"However, comparing Fig. 5o to Fig. 6c, we found that the impact of temperature via the change in the chemical reaction rates was generally more significant than that via the change in biogenic emissions from 2013 to 2017 (also see Fig. S8 for the quantitative comparisons in different cities)."

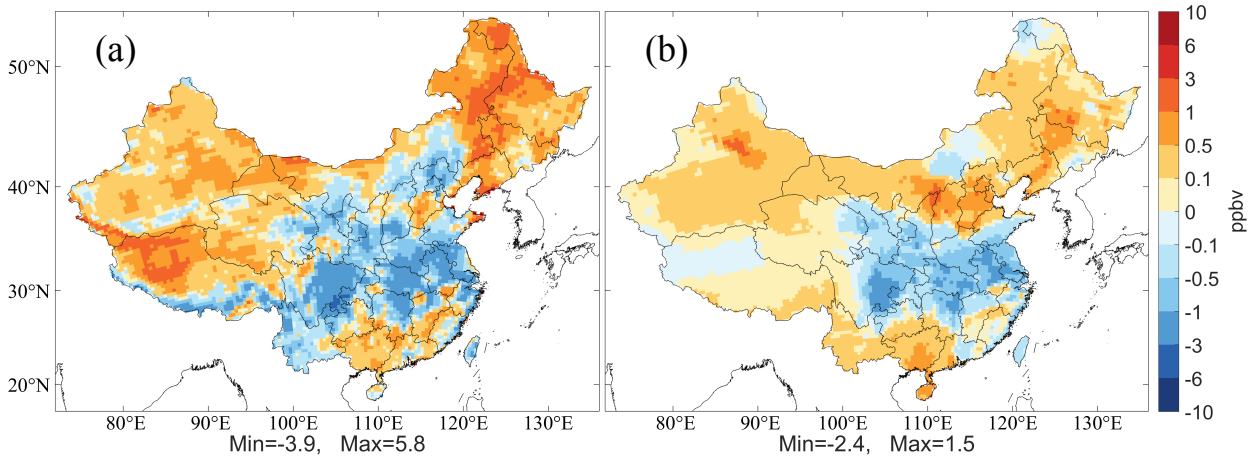

Figure R2 Changes in the simulated summer surface MDA8 O_3 mixing ratios due to the changes in (a) temperature and (b) biogenic emissions in 2017 relative to 2013.

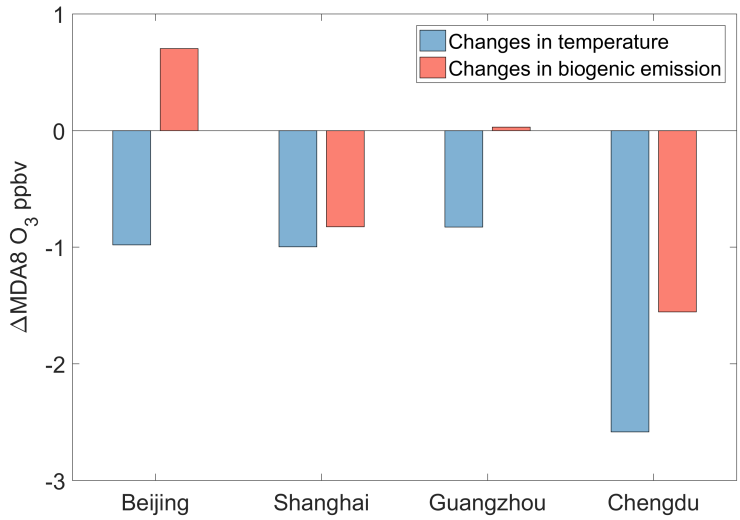

Figure R3 (Figure S8) Changes in the simulated summer surface MDA8 O_3 mixing ratios due to the changes in temperature and biogenic emissions in 2017 relative to 2013 in Beijing, Shanghai, Guangzhou, and Chengdu.

[Comment]: 5. Page 12, Line 342-345:

The statements here seem to imply that transport of PAN led to the long-range transport of ozone influences. How about transport of ozone itself? Which one is the main pathway? One way to quantify and to separate the influences is to conduct a simulation fixing PAN in the 2013 chemical boundary conditions, yet I do not want to push the authors to do more model simulations. Can you explain the issue with present analyses and results?

Response: Thanks for pointing out this issue. The original statement does seem to indicate that the transport of PAN is the only pathway affecting the long-range transport of ozone, which needs to be corrected. Previous observation and modeling studies (West et al., 2009; Wild et al., 2004) have suggested that ozone and its precursors, namely NO_x (or its carrier PAN), VOCs and CO, can be transported a long distance and then affect the ozone concentration in remote regions. We have corrected this statement in the revised manuscript.

We also think that investigating the pathway of long-range O_3 transport is an interesting and important topic and agree that one way to address this issue is to conduct many simulations fixing different pollutants individually in the chemical boundary conditions and compare the simulation results with each other. As the present study focuses on the impact of meteorological parameters, we prefer to address which chemical contributed most during long-range transport in a future study.

Revision in the main text:

1) Line 347-348:

"Ozone and its precursors can be transported a long distance and then affect surface O_3 in remote regions (West et al., 2009; Wild et al., 2004)."

Reference:

West, J. J., Naik, V., Horowitz, L. W., and Fiore, A. M.: Effect of regional precursor emission controls on long-range ozone transport - Part 1: Short-term changes in ozone air quality, Atmos Chem Phys, 9, 6077-6093, 10.5194/acp-9-6077-2009, 2009.

Wild, O., Pochanart, P., and Akimoto, H.: Trans-Eurasian transport of ozone and its precursors, 109, 10.1029/2003jd004501, 2004.