Response to Reviewer 1

The manuscript titled “Impacts of meteorology and emissions on surface ozone increases over Central Eastern China between 2003 and 2015” present a very interesting and useful model study showing an increase in surface ozone over Central Eastern China (CEC) between July 2003 and July 2015 which is in agreement with recent studies (Xu et al., 2016, 2018; Lu et al., 2018; Gaudel et al., 2018).

According to this present study, emission changes have a higher impact on the Maximum Daily 8-h Average of ozone (MDA8 ozone) than the meteorological changes. However the meteorological changes would better explain larger ozone increases (delta MDA8 ozone >=10 ppbv between July 2003 and July 2015) than the emission changes. By this latter result, the authors would like to point out that, the long-range transport of ozone and its precursors from neighboring areas should be taken into account, for air pollution control.

The manuscript is well written and the quality of the text and its structure is very much appreciated. However I suggest major revisions, especially regarding one main conclusion of the manuscript that would need more evidence. Indeed, the impact of the transboundary transport on the surface O3 above CEC that would explain an increase of surface O3 between July 2003 and July 2015 is not very much convincing as it is written now.

You can find below general comments followed by more specific comments.

Response: we thank the reviewer for the helpful comments and suggestions, which are very helpful for improving our original manuscript. Below we address all of these comments and have revised the manuscript accordingly. For clarity, the reviewer’s comments are listed below in black italics, whilst our responses and changes in the manuscript are shown in blue and red, respectively.

General comments:

1) I would suggest the authors to cite the following recent studies to put even better in perspective the increase of surface ozone between July 2003 and July 2015 using long-term time series:  Xu et al. (2016, 2018), Lu et al. (2018) and Gaudel et al. (2018)


station, China-Part 2: The roles of anthropogenic emissions and climate variability. Atmospheric Chemistry & Physics, 18(2).


Response: these recent studies have been cited in the revised manuscript.

2) I find hard to understand why the authors have chosen the two periods July 2003 and July 2015. Could the authors explain more this choice? According to the Tropospheric Ozone Assessment Report database (https://join.fz-juelich.de) and seasonal cycles studies (e.g. Sun et al., 2016), the month with higher ozone for most of the sites above CEC would be June. Why would the authors choose July? In addition, according to Figure 1 of the present manuscript, the observations of surface ozone are available mostly in 2004, why would the authors choose 2003 instead of 2004? It should be clarified in the text.

Response: we are sorry that the original discussion is unclear. We chose 2003 and 2015 for simulation mainly because some recent studies (especially our previous study of Sun et al., 2016) have reported the significant increase of summertime ozone over the CEC region. And the modelling results indeed indicated the significant differences in either meteorology or anthropogenic emissions between these two years.

Yes, June is usually the month with the highest surface ozone concentrations in North China. In the present study, June was not selected for simulation because of the varying crop residue burning activities. The crop residue burning usually lasts from late May to late June in CEC, and these emissions had varied greatly over the past decade, which would introduce large uncertainty to the evaluation of impacts from anthropogenic emissions. Thus, we didn’t focus on the O₃ change simulations in June.

To further confirm the conclusions drawn from the comparison between 2003 and 2015, we have conducted additional simulations for July 2004 and July 2014. There are little difference in the modelled regional-mean and spatial distributions of MDA8 O₃ between 2003 and 2004 as well as between 2014 and 2015. Overall, the modelling results in 2004 and 2014 supported the major conclusions derived from 2003 and 2015.

The following discussions have been added in the revised manuscript and the supporting information to elaborate this issue.
The last Paragraph in Section 1:

“This is a follow-up study of Sun et al. (2016) that found a significant increase of summertime O₃ at a regional site in North China from 2003 and 2015. We integrate the global GEOS-Chem model and its Asian nested model to investigate the spatial distributions of surface O₃ over the whole CEC region, and to quantify the relative contributions from changes in meteorological and anthropogenic emission between 2003 and 2015.”

The first graph in Section 2.1:

“The models are run with the full standard NOx-Ox-hydrocarbon-aerosol tropospheric chemistry (Mao et al., 2013) for January to August of 2003 and 2015, including the spin-up time of six months (January to June) for each simulation, but only the results for July are discussed in this paper. The results of August 2003 and 2015 are discussed in supplementary document to confirm the result of this study. Since the crop residue burning usually lasts from late May to late June over CEC and the emissions had varied greatly over the past decade, which introduces large uncertainty to the evaluation of impacts from anthropogenic emissions (Chen et al., 2017; Wu et al., 2018), we don’t focus on the O₃ change simulations in June. For comparison, we also conducted model simulations for July 2004 and July 2014, and the results supported the major findings obtained from 2003 and 2015 (see results in the supplement).”

Page 7, Line 15:

“The simulated surface O₃ in 2004 was also compared against these observations in Figure S2.”

Page 9, Line 17:

“The spatial distributions of MDA8 O₃ in July 2004 and 2014 in Figure S8 present similar patterns to July 2003 and 2015. The regional mean MDA8 O₃ increased from 67.8±6.2 ppbv in July 2004 to 74.8±9.8 ppbv in July 2014. In addition, the regional mean MDA8 O₃ increased from 63.4±4.9 ppbv in August 2003 to 73.8±5.0 ppbv in August 2015 (Figure S9). These results are comparable to those derived from the comparison between July 2003 and July 2015. The detail description is provided in the supplement.”

Supplementary document:

1. The model simulated monthly-mean MDA8 O₃ in July 2004 and July 2014

   To further confirm the conclusions drawn from the comparison between 2003 and 2015, we also conducted model simulations for July 2004 and July 2014. Figure S2 shows the comparison of observed versus simulated monthly-mean surface O₃ levels at six rural sites in July 2004. The model captures well the observed O₃ concentrations at Mt Tai, SDZ, and Mt Hua, with only minor bias (-1.6–4.0 ppbv). In comparison, the model tends to overestimate
the O$_3$ levels at Mt Huang, Lin’an and Hok Tsui. Figure S8 shows similar spatial distributions of MDA8 O$_3$ over CEC in July 2003, 2004, 2014 and 2015. In July 2004, the regions with MDA8 O$_3$ >75 ppbv moved to the south of North China Plain compared to July 2003, mostly due to the different atmospheric circulation patterns. The regional mean MDA8 O$_3$ in July 2004 is 67.8±6.2 ppbv, a little higher than that in July 2003 (65.5±7.9). The regional mean MDA8 O$_3$ in July 2014 is 74.8±9.8 ppbv, which is comparable to that in July 2015 (74.4±8.7 ppbv). We can find the significant increases of MDA8 O$_3$ from 2004 to 2014 as well as from 2003 to 2015. The different concentrations and spatial distributions of O$_3$ between 2003 and 2004 (as well as between 2014 and 2015) should be mostly due to the inter-annual variability in meteorological conditions. Overall, the modelling results in 2004 and 2014 supported the major conclusions derived from 2003 and 2015.

2. The model simulated monthly-mean MDA8 O$_3$ in August 2003 and August 2015

Figure S9 shows the spatial distributions of MDA8 O$_3$ in August 2003 and August 2015. In August 2003, there is no region with monthly-mean MDA8 O$_3$ >75 ppbv. In August 2015, the region with monthly-mean MDA8 O$_3$ >75 ppbv is comparable to that in July 2015, but the O$_3$ levels are generally smaller than those in July 2015. The regional mean MDA8 O$_3$ in August 2003 and 2015 are 63.4±4.9 and 73.8±5.0 ppbv, yielding an increase of 10.4 ppbv. The regional mean MDA8 O$_3$ during July-August of 2003 and 2015 are 64.5±6.4 and 74.1±6.8 ppbv, giving a comparable increase of 9.6 ppbv from 2003 to 2015. The difference in the regional mean MDA8 O$_3$ between July and August of 2003 is 2.1 ppbv, a little higher than that between July and August in 2015 (0.6 ppbv). Such levels are much lower than the difference between 2003 and 2015. Overall, the modelling results in August of 2003 and 2015 supported the major conclusions derived from July 2003 and July 2015.
Figure S8. Monthly-mean spatial distributions of surface MDA8 O₃ in July over East China. (a) 03E03M: 2003 standard simulation; (b) 04E04M: 2004 standard simulation; (c) 14E14M: 2014 standard simulation and (d) 15E15M: 2015 standard simulation. Black contours indicate the regions with MDA8 O₃ > 75 ppbv. Filled circles in (d) show the observed MDA8 O₃ at 115 sites of the network of Chinese National Environmental Monitoring Center in July 2015. The red rectangle represents the Central Eastern China region (CEC: 103°E-120°E, 28°N-40°N).

Figure S9. Monthly-mean spatial distributions of surface MDA8 O₃ in August over East China. (a) 03E03M: 2003 standard simulation and (b) 15E15M: 2015 standard simulation. Black contours indicate the regions with MDA8 O₃ > 75 ppbv. The red rectangle represents the Central Eastern China region (CEC: 103°E-120°E, 28°N-40°N).

3) I didn’t find a strong argument supporting the impact of transboundary transport on the surface O₃ above CEC that would explain the increase of surface O₃ between July 2003 and July 2015. The authors should add more evidence or be clearer in their analysis.

Response: we agree with the reviewer. The phrase of “transboundary transport” may be not appropriate, and “transport” should be better here. As indicated by the budget analysis, the transport pattern in July 2015 tended to export less O₃-laden air masses than in 2003, which means that more O₃ were accumulated within CEC. Since the O₃ levels in July 2015 is higher than that in July 2003, if the transport pattern in 2015 was the same as 2003, it would transport much more O₃ air masses from the CEC. As well, the difference in transport pattern also led to the different spatial distributions of surface O₃ over the CEC region between 2003 and 2015. The following discussions have been added in the revised manuscript to clarify this issue.

“We found that in July 2015, the wind speeds over southern and eastern boundaries of CEC were much lower than that in July 2003 (Figure S13), leading to much lower O₃ flux across these two boundaries. The low O₃ over southern CEC in July 2003 was mainly due to the strong south-westerly wind, decreasing O₃ levels in this area.”
“In July 2003, the air flows into CEC through the south boundary, and then out across the other three boundaries. In contrast, the air masses flow into this area across the east boundary in July 2015, and then out across the left three boundaries. The larger O$_3$ flux from each boundary in July 2003 is due to stronger winds. Compared to the 03E03M simulation (-897 Gg mon$^{-1}$; negative value means export of O$_3$ from this region), 03E15M shows a much lower O$_3$ flux (-401 Gg mon$^{-1}$), indicating that weather conditions in 2015 play a more important role in pollutant accumulation, which is consistent with our analysis in Section 4.”

**Specific comments:**

1. **Title:**

   I would suggest to add “July” in the title.
   
   **Response:** The title has been revised as follows. Note that the analysis in the present study covers July and August, and June was not considered due to the variation in biomass burning.

   “Impacts of meteorology and emissions on summertime surface ozone increases over Central Eastern China between 2003 and 2015”

   2. **Abstract:**

   Line 23 p.1: Change “The increase in regional averaged O$_3$ resulting from…” to “The increase in averaged O$_3$ in the CEC region resulting from…”
   
   **Response:** Changed

3. **Sections 2 to 6:**

   Line 19 p.4: The authors are using the global chemical transport model GEOS-Chem v11-01 but the current version reported on the website cited in the manuscript is v11-02. Would the use of v11-02 instead of v11-01 change the results? Could the authors add a word about it in the text?
   
   **Response:** We used the version of v11-01 in this study. The web link has been modified as follows.


4. **Line 28 p.4:** Could the authors say how long they estimate the spin-up? How many month?

   **Response:** The following statements have been added in the revise manuscript to clarify this.

   “The models are run with the full standard NOx-Ox-hydrocarbon-aerosol tropospheric chemistry (Mao et al., 2013) for January to August of 2003 and 2015, including the spin-up
time of six months (January to June) for each simulation, but only the results for July are discussed in this paper.”

5. Line 17-20 p.6: I would suggest to change “The contribution of anthropogenic NOx (NMVOCs) emission changes can be calculated by the difference between the 2015 standard simulation and 03N15M (03V15M), defined as 15E15M-03N15M (15E15M -03V15M). ” to “The contribution of anthropogenic NOx and NMVOCs emission changes separately can be calculated by the difference between 15E15M (the 2015 standard simulation) and 03N15M (the 2003 NOx emission simulation), and between 15E15M and the 2003 NMVOCs emission simulation (03V15M).”

Response: changed.

6. Line 25 p.6: It is very useful to cite the website link of the Chinese Data Center but unfortunately, there is no English version. Do the authors know whether it is planned to implement the English version? If yes, could the authors say a word about it?

Response: we found the English version but it does not provide the data now. It may be used in the future. We have added the website of the English version in the revised manuscript.

http://datacenter.mee.gov.cn/aqiweb2/getAirQualityDailyEn (in English)


7. Line 27 p.6: The authors used the word “background sites”. If the authors refer to observed ozone at sites which are not influenced by recent, locally emitted or produced anthropogenic pollution, they should use the word “baseline” instead for consistency purposes (Cooper et al., 2014; Dentener et al., 2011)


Response: the “background sites” have been changed to “baseline sites” in the revised manuscript.

8. Line 4 p.7: Why the authors didn’t choose the year 2004 for all the sites? Is July 2003 comparable with July 2004?

Response: the data we have for Mt. Tai and Hok Tsui were in 2003, and the data for the
other sites were only available in 2004 and were taken from literature. We have simulated the surface O₃ in July 2004. The comparison between simulated and observed O₃ data in July 2004 is shown in Figure S2. We found the simulated O₃ concentrations in July 2003 are comparable to those in July 2004. The original statement has been changed as follows in the revised manuscript.

“We compare the simulated surface O₃ concentrations with the 2003 observations for Mt Tai and Hok Tsui but with the 2004 observations for the other four sites (Figure 1(a)). The simulated surface O₃ in 2004 was also compared against these observations in Figure S2.”


Response: yes, the modeling set-up was the same between these simulations. It started from January 2004.

10. Line 25-30 p.7: Could the authors explain how they chose the “nine representative sites”? Would it be possible to show the diurnal cycle from observations and the simulations for the July month for each site with the standard deviation? The comparison observations/model, day/night would be more straightforward.

Response: we chose non-urban sites to represent the O₃ concentrations in major cities over CEC. In general, the selected non-urban sites are sub-urban or rural sites which are far away from the urban and industrialized areas. We have compared the diurnal cycles of O₃ (and CO and NOx) from observations and simulations. The following discussions have been added in the revised manuscript and supplements.

“To avoid the influence of local emission, photochemical and deposition processes in small-scales of urban area, we selected one non-urban site to represent the O₃ concentrations of each city over CEC. In general, the selected non-urban sites are sub-urban or rural sites which are far away from the urban and industrialized areas. For cities where no non-urban sites are available, we chose the stations that are least affected by local pollution (i.e., sites relatively far away from roads, factories, power plants, etc.).”

“Time series and diurnal variations of hourly O₃ concentrations from the model and observations at Mt Tai in 2003 and nine representative sites in 2015 are compared in Figures S3, S4 and S5, respectively. The nine observation sites are carefully selected to be far away from urban areas in the capital cities of nine provinces and municipalities, including Beijing, Tianjin, Ji’nan, Taiyuan, Zhengzhou, Wuhan, Chongqing, Changsha, and Nanjing. The model reproduces the time series of O₃ with a normalized mean bias of 4% at Mt Tai. The overestimation of O₃ concentrations in the afternoon is likely to be due to the overestimated precursor emissions in the model. For the nine sites, the model captures most day-to-day
variability and diurnal variations (Figures S4 and S5).”

11. Line 3-4 p.8: Does the trend of observed O$_3$ at Mt Tai come from Sun et al. (2016)? The paper should be explicitly cited. Does the simulated increase of about 1.3 ppbv yr$^{-1}$ refer to the same model: nested version of Geos-Chem or GFDL-AM3? I would suggest showing the time series observations/model in a figure in the supplement material. Could the authors report the 95% confidence intervals with the trends?

Response: the original statement is confusing. Yes, the trend of observed O$_3$ at Mt. Tai was taken from Sun et al. (2016), and the simulated increase of ~1.3 ppbv yr$^{-1}$ was calculated from the Geos-Chem model in the present study. The use of “trend” should be not appropriate as we only performed simulations for two years. The original statement has been modified as follows in the revised manuscript.

“In addition, the observed results of Sun et al. (2016) reported the MDA8 O$_3$ at Mt Tai increased from 75.9±15.9 to 102.1±28.1 ppbv in July-August from 2003 to 2015, which is higher than the simulated result in this study (i.e., from 71.1±10.0 ppbv in July 2003 to 90.4±18.5 ppbv in July 2015). Nonetheless, the model captures the significant increase in surface O$_3$ levels over CEC between July 2003 and July 2015.”

12. Line 15 p.8: Is the increasing rate calculated from a delta between 2 years looking at one month comparable to the increasing rate calculated from a full time series? Are the authors sure of the rate of 0.74 ppbv yr$^{-1}$: 74.4 ppbv compare to 65.5 ppbv, 11 years apart would give around 0.8 ppbv yr$^{-1}$, wouldn’t it?

Response: we agree with the reviewer, and the original descriptions have been modified as follows.

“Table 2 shows the monthly mean MDA8 O$_3$ over CEC. The regional mean MDA8 O$_3$ increased from 65.5±7.9 ppbv in July 2003 to 74.4±8.7 ppbv in July 2015, showing an increase of about 8.9±3.9 ppbv in twelve years. According to the limited reports of observed long-term (>10 years) changes of O$_3$ concentrations, we find significant increases of summertime O$_3$ (1–3 ppbv yr$^{-1}$) in the north part (Beijing), east part (Mt Tai) and south part (Lin’an) of CEC over the past two decades (Ding et al., 2008; Ma et al., 2016; Sun et al., 2016; Xu et al., 2008; Zhang et al., 2014b). Our result shows that both daily mean O$_3$ concentration and MDA8 O$_3$ were significantly higher in July 2015 than in July 2003 over most areas of CEC (Figure 3).”

13. Line 29-30 p.8: I am not sure to understand the reason of the choice of focusing on MDA8 O$_3$ and not the daily mean. Could the authors clarify this point?

Response: MDA8 O$_3$ has been widely used as an indicator to present the O$_3$ pollution levels. And due to the NO titration at nighttime, the O$_3$ concentrations at night may not reflect the actual O$_3$ pollution conditions. So we choose the MDA8 O$_3$ for analyses, rather than the daily
mean. The following statement has been added in the revised manuscript to clarify this.

“Considering that the nighttime O\textsubscript{3} is easily titrated by NO and the MDA8 O\textsubscript{3} is a good indicator for the overall O\textsubscript{3} pollution condition, we focus on the MDA8 O\textsubscript{3} changes over CEC between July 2003 and July 2015 instead of daily-mean O\textsubscript{3}.”

14. Line 9 p.9: Please clarify “domain-averaged”?

Response: it has been changed to “regional mean”.

15. Line 13 p.10: I would suggest to explain Table 2 earlier in the text when the authors first refer to the Table in section 3.2.

Response: we adopt this suggestion and move it to section 3.2.

16. Line 15 p.11: “even if” would suggest that the authors would expect another result for \(\Delta\text{MDA8 O}_3\) greater than 10 ppbv. If it is the case, could the authors say a word about what result they would expect?

Response: the original statements may be unclear and have been clarified as follows in the revised manuscript.

“It is worth noting that in the polluted regions where MDA8 O\textsubscript{3} > 75 ppbv, the contribution of emission change increases from \(5.0\pm1.8\) ppbv for \(\Delta\text{MDA8 O}_3 \geq 0\) ppbv case to \(5.2\pm1.7\) ppbv for \(\Delta\text{MDA8 O}_3 \geq 10\) ppbv case, whilst the contribution of meteorology change increases from \(3.7\pm3.2\) ppbv to \(5.0\pm2.5\) ppbv. Even if the \(\Delta\text{MDA8 O}_3\) is greater than 10 ppbv, the O\textsubscript{3} increase caused by emission change is still higher than that caused by meteorological change, indicating the dominant effect of emissions on O\textsubscript{3} pollution in the highly polluted regions.”

17. Line 4 p.12: Do the authors still mean MDA8 O\textsubscript{3} using the words “O\textsubscript{3} concentrations”? Please be consistent.

Response: we have changed the “O\textsubscript{3} concentration” to “MDA8 O\textsubscript{3}”.

18. Line 22 p.12: Remove “in”.

Response: removed

19. Line 7-9 p.13: According to Table 4, the larger O\textsubscript{3} flux in 15E03M would be 1906 Gg mon\(^{-1}\) and not -1232 Gg mon\(^{-1}\), is it correct? Could the author explain where -1232 Gg mon\(^{-1}\) come from?

Response: it should be -1100 Gg mon\(^{-1}\) and has been corrected in the revised manuscript.

20. Line 14 p.13: Change “excessive O\textsubscript{3} production” to “excessive net O\textsubscript{3} production”

Response: changed.

formed O$_3$ in the CEC region in July 2003 cannot be removed by transport in July 2015 as it is not the same air masses 11 years later.

**Response:** agree. This statement has been rephrased as follows.

“In July 2003, about half of the net photochemically formed O$_3$ in the CEC region was removed by transport (897 out of 1629 Gg mon$^{-1}$). In comparison, only 1/4 of the net photochemically produced O$_3$ (502 out of 2158 Gg mon$^{-1}$) was transported out of CEC in July 2015.”

22. **Line 27-28 p.13:** The sentence “[...] the absolute value of O$_3$ transport flux increased by 395 Gg mon$^{-1}$” is confusing. According to Table 4 all fluxes for both horizontal and vertical transport are actually decreasing when comparing simulation 03E03M with 15E15M. This is in agreement with stronger winds in July 2003 than in July 2015 shown in Figure S6. The sentence needs to be rephrased.

**Response:** the original sentence has been rephrased as follows.

“Comparing the results of the 2003 and 2015 standard simulations (15E15M-03E03M), we find less O$_3$ export from CEC in 2015, which means about 395 Gg mon$^{-1}$ (2015-2003) O$_3$ was accumulated in this region.”

23. **Line 30-31 p.13:**

“As a result, the increase in O$_3$ concentrations from July 2003 to July 2015 is mainly due to transboundary horizontal transport, vertical transport and photochemical reactions.”

Regarding the transboundary horizontal transport, what would be the source regions of O$_3$ that would affect O$_3$ above CEC? Regarding the vertical transport, do the results imply there is an increase in stratospheric intrusion?

According to Figure S6 that shows weaker winds in July 2015 than in July 2003, the transboundary horizontal transport doesn’t seem to be a major process that would explain an increase in surface O$_3$ above CEC. Indeed, from Table 4, the local photochemical processes rather than the transport processes seem to be leading the increase in O$_3$. The slow changes in time of the dry deposition process could also explain the increase in surface O$_3$ because it cannot compensate the increase in net photochemical production of O$_3$.

**Response:** the original statement is somewhat misleading. The increase in O$_3$ concentrations from July 2003 to July 2015 should be due to the enhanced photochemical production (mainly due to the increased emissions) and the weakened export (due to the meteorological conditions). This statement has been revised as follows in the revised manuscript.

“As a result, the increase in O$_3$ concentrations from July 2003 to July 2015 should be due to the enhanced photochemical production (mainly due to the increased emissions) and the
weakened export (due to the meteorological conditions).”

Conclusions:

24. Line 4 p.14: Would it be possible to put the numbers in perspective with long-term time series study above individual sites such as Mount Waliguan, Mount Tai, Shangdiazi?

Response: the results from these long-term time series study have been discussed in the main context, and were not listed again in the conclusion section.

25. Line 14-15 p.14: The terms “transport” can refer to the winds, which are part of the “meteorological” condition. Could the authors explicitly use the words “winds”, “humidity” and “temperature”? That would help the reader.

Response: the original statement has been modified as follows.

“The meteorological conditions (mostly due to wind patterns) in July 2015 tended to accumulate pollution and reduced O₃ export over the central part of CEC and thus enhanced O₃ levels there. Air temperature and relative humidity does not promote the O₃ production in July 2015.”

Line 24-25 p.14: To my opinion, the manuscript does not show a strong argument to support the theory of the impact of transboundary transport on surface O₃ above CEC. Could the authors be clearer and bring more evidence?

Response: the phrase of “transboundary transport” may be misleading. From the budget analysis, we found that large-scale regional transport is an important contributor to the spatial distributions and inter-annual variations of surface O₃ over the CEC region. The original statement has been modified as follows.

“Transport issues in local O₃ control strategies should go beyond transport from neighbouring areas (e.g., cities) and account for the long-distance transport (e.g., across provinces).”