Response to Referee 2

This manuscript aims to quantify the contributions of emission changes and meteorological conditions on surface ozone changes over Central Eastern China between July 2003 and 2015. An ensemble of simulations using the GEOS-Chem model were conducted to diagnose the impacts of meteorology, anthropogenic and natural emissions. The results show comparable and spatially different contributions from emissions and meteorology on surface ozone changes between the two months, and further point out the importance of chemical production and pollution transport on surface ozone over Central Eastern China. The manuscript is generally well written and fits the scope of ACP. The results are valuable for better understanding the ozone pollution over Eastern China. I recommend publish on ACP but after the following comments being addressed.

Response: we thank the referee for the positive comments and helpful suggestions. We have addressed all of these comments in the revised manuscript, as detailed below in the responses to individual comments. For clarify, the referee's comments are listed below in black italics, while our responses and changes in the manuscript are shown in blue and red, respectively.

Main Comments

1) This study focused on surface ozone changes between July 2003 and July 2015, yet it is not clear why the two particular years (2003 and 2015) were selected. Why not analyze other years, for example, 2014, as the Chinese anthropogenic emissions in the model were based on 2014? Were meteorological conditions in the two years distinctly different from each other, in order to emphasize the impact of meteorology as analyzed in this study? Please clarify.

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. 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 O_3 between 2003 and 2004 as well as between 2014 and 2015. The difference of MDA8 O_3 between 2003 and 2014 (or 2004 and 2015) is much higher than the difference between 2003 and 2004 (or 2014 and 2015). This means that the impact of inter-annual variability should be smaller than the decadal change in meteorology and emissions. The following discussions have been added in the revised manuscript and supporting information to elaborate this issue.

"This is a follow-up study of Sun et al. (2016) that found a significant increase of summertime O_3 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 O_3 over the whole CEC region, and to quantify the relative contributions from changes in meteorological and anthropogenic emission between 2003 and 2015."

“For comparison, we also conducted additional model simulations for 2004 and 2014, and the results supported the major findings obtained from 2003 and 2015 (see results in the supplement).”

“In July 2004, the regions with MDA8 O_3 > 75 ppb moved to 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). In comparison, 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).”

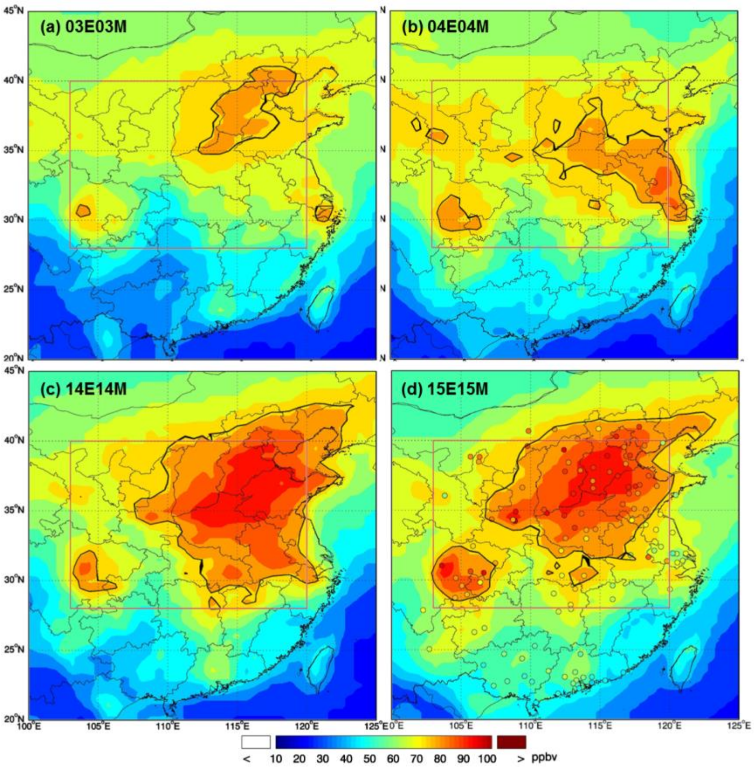

Figure S8. Monthly-mean spatial distributions of surface MDA8 O_3 in July over East China. (a) 03E03M: 2003 standard simulation; (b) 04E04M: 2004 standard simulation; (c) 14E14M: 2014 standard simulation and (d) 15E15M: 2015 standard simulation. Black contours indicate the regions with MDA8 O_3 > 75 ppbv. Filled circles in (d) show the observed MDA8 O_3 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).

2) Since only two months were analyzed in the study, it needs to be careful with the interpretation of the surface ozone changes between the two periods. The manuscript described the ozone changes mainly as an increasing trend and compared it with previous trend observations, e.g., the paragraphs in Sect. 3.1 and Sect. 3.2 (Page 8). It should be well noted here that surface ozone changes between July 2003 and 2015 may largely reflect ozone inter-annual variability, not a trend.

Response: we totally agree with the reviewer that the O_3 changes between two specific years are largely influenced by the inter-annual variability, rather than a trend. We have deleted the phrase of "trend" in the revised manuscript. Nonetheless, the long-term observations have indeed indicated a significant increasing trend of O_3 over CEC in the past decades, and our model simulations in 2003-2004 and 2014-2015 also supported this argument.

Specific Comments

1) Page 7, Line 4:

"but with the 2004 observations for the other four sites". Do you mean there was no observations available for the other four sites in 2003?

Response: yes, the observational data at these four sites were obtained in 2004, not in 2003. We have compared the 2004 modelling results with the observation data in 2004. Generally, the simulation results for the six sites in July 2004 are comparable to those in 2003, as shown below.

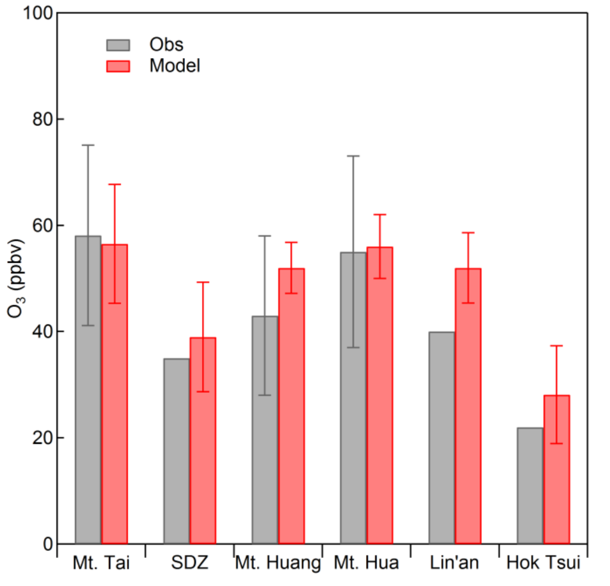

Figure S2. Comparison of observed versus simulated monthly-mean O_3 concentrations at the six rural sites in July 2004.

2) Page 7, Line 17:

Please describe more how non-urban sites were selected in this study. Based on the population density or any other information? Did the authors select one site for each city?

Response: the following descriptions have been added in the revised manuscript to clarify this issue.

"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 O_3 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 traffic roads, factories, power plants, etc.). As a result, 115 non-urban sites were selected to represent 115 cities in the East China region."

3) Page 8, Line 28

"This result is very different from the trends over the US, where summertime daytime O_3 increased over the past decades is contrast to the night-time decrease in all seasons (Yan et al., 2018a)." Yan et al. (2018a) showed that US summertime daytime O_3 decreased and nighttime O_3 increased in the past decade. Please check.

Response: the original statement has been changed as follows.

"This result is very different from the trends over the US, where summertime daytime O_3 declined over the past decades is contrast to the night-time growth in all seasons (Yan et al., 2018a)"

4) Page 10, Line 8

Please provide values of air temperature and relative humidity over CEC on Figure S7. Seen from this Figure, it seems air temperature in July 2015 was higher than that in July 2003.

Response: we have added the values of air temperature and relative humidity over CEC in the revised manuscript. Air temperature was almost the same between July 2003 and July 2015. Relative humidity in July 2015 was a little lower than that in July 2003.

"Figure S14 shows the simulated spatial distributions of air temperature and relative humidity in July 2003 and July 2015. The simulated air temperatures in 2003 and 2015 are 300.6±3.2 and 300.5±3.2 K, respectively, almost at the same level. The simulated relative humidity in 2003 was 82±10%, a little higher than in 2015 (77±12%). The average net O_3 production over CEC simulated by 03E03M (11.7 ppbv day^{-1}) is very close to that simulated by 03E15M (11.9 ppbv day^{-1}) (Table 4), suggesting that meteorological factors in 2003 and 2015 did not greatly change O_3 photochemical reactions. Therefore, neither air temperature nor relative humidity plays an important role in explaining the difference in surface O_3 between 2003 and 2015."

5) Page 10, Line 15:

What is "gradient analysis"? Please clarify.

Response: the following descriptions have been added in the revised manuscript to clarify this.

"We performed a gradient analysis, which selected different levels for the difference of MDA8 O_3 (ΔMDA8 O_3) between 2003 and 2015 standard simulations (15E15M-03E03M). The differences in MDA8 O_3 were analysed in four ways: regional mean, ΔMDA8 O_3 $\geq$0 ppbv, ΔMDA8 O_3 $\geq$5 ppbv and ΔMDA8 O_3 $\geq$10 ppbv."

6) Page 12, Figure 6:

I suggest add a figure in the Supplement showing the spatial distribution of changes in anthropogenic NMVOCs and NOx emissions between 2003 and 2015. This can provide helpful information to better interpret their resulting changes in surface O_3 as shown in

Figure 6.

Response: thanks for the good suggestion. We have added the spatial distributions of NOx and NMVOC emissions and their changes in the supplement (see below). Some brief discussions have been also added in the revised manuscript.

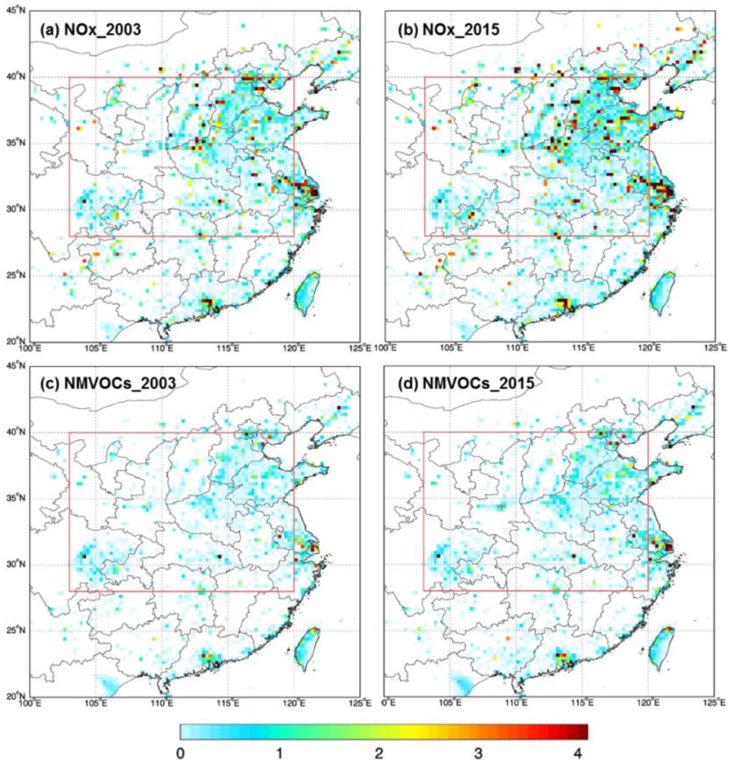

Figure S13. Anthropogenic emissions of NOx in July 2003 (a) and July 2015 (b) and NMVOCs in July 2003 (c) and July 2015 (d). Unit: $Mg/Km^2/mon$ for NOx; $Mg(C)/Km^2/mon$ for NMVOCs. The red rectangle represents the Central Eastern China region.

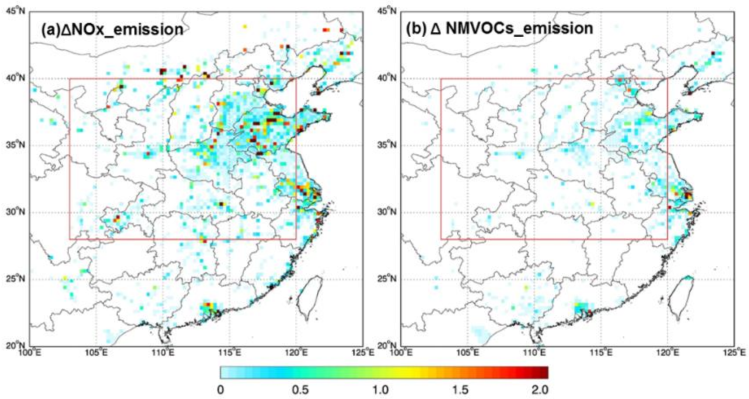

Figure S14. Spatial distributions of changes in NOx (a) and NMVOCs (b) emissions between July 2003 and July 2015 (2015-2003). Unit: $Mg/Km^2/mon$ for NOx; $Mg(C)/Km^2/mon$ for NMVOCs. The red rectangle represents the Central Eastern China region.

7) Page 13, Line 4-8:

"minus value" should be "negative value". In "15E03M (-1232 Gg mon-1)", where did

"-1232" come from? Table 4 shows "-1100"

Response: these typos have been corrected in the revised manuscript.

8) Page 13, Line 27:

"we find that the absolute value of O_3 transport flux increased by 395 Gg mon^{-1} (2015-2003)". This sentence is misleading. The absolute value of O_3 transport flux actually decreased in 2015 relative to 2003 due to less export in 2015. Please clarify.

Response: the original statement has been changed as follows in the revised manuscript.

"Comparing the results of the 2003 and 2015 standard simulations (15E15M-03E03M), we find less O_3 export from CEC in 2015 than in 2003, which means about 395 Gg mon^{-1} of O_3 was accumulated in this region in 2015."

9) Page 14, Line 2:

"Asia nested model" should be "Asian nested model".

Response: changed.

10) Page 14, Line 14:

The statement "The transport pattern in July 2015 tends to enhance O_3 levels over the central part of CEC" needs some explanation. Is that because the meteorological conditions in July 2015 favored pollution accumulation and reduced O_3 export over CEC and thus enhanced O_3 levels there?

Response: the original statement has been changed as follows to clarify this issue.

"The meteorological conditions (especially wind patterns) in July 2015 tended to accumulate pollution and reduced O_3 export over the central part of CEC and thus enhanced O_3 levels there. Air temperature and relative humidity does not promote the O_3 production in July 2015."

11) In the supplement, Figure S6 and S7:

The meteorological fields should be based on "MERRA-2" instead of "the GEOS-Chem results".

Response: changed.