Response to Referee 3

This paper analyzes the individual impacts of meteorological condition and emission on summertime ozone concentration in Central Eastern China based on GEOS-Chem model. This is generally a solid study with reasonable analyzing and discussion of the model results, and the manuscript is well organized. Therefore, I would recommend the manuscript being accepted for publish if the following issues could be properly addressed

Response: we thank the reviewer for the thoughtful review and constructive comments. These comments and suggestions are very helpful for improving our manuscript. We have tried to address all of the referee's comments in the revised manuscript. Below we reply in detail to the individual comments. For clarify, the reviewer's comments are listed in black italics, while our responses and changes are highlighted in blue and red, respectively.

Major comments: *There exists significant inter-annual variability of meteorological conditions in CEC, did the authors chose these two year (2003, 2015) to conduct the simulation due to their representativeness? Additionally, it is known that China's NOx emission toped around the year 2011. So the impact of anthropogenic emission in 2011 might reach its maximum rather than in 2015. In addition, since the present work only studied one specific month (July), I personally do not think that the results can extrapolate for the whole year. Therefore, the season with concern ought to be specified in the title.*

Response: we are sorry that the original discussion may be unclear. We chose 2003 and 2015 for simulation mainly because some recent studies have reported the significant increase of summertime ozone over the CEC region (Sun et al., 2016; Ma et al., 2016). Yes, the anthropogenic emissions of NOx have been reduced since 2011, but the NMVOC emissions have continued increasing unabated. Another reason why we didn't choose 2011 for simulation is the lack of observational data for comparison and model validation. The following discussion has been added in the revised manuscript to clearly state the reason for selecting 2003 and 2015.

Page 4 Line 8: "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."

We agree with the reviewer that the season with concern should be specified in the title. We also performed the modeling analyses for August in the revision process, and the results are similar to those obtained for July. The title has been revised to "Impacts of meteorology and emissions on summertime surface ozone increases over Central Eastern China between 2003

and 2015"

Process analysis is a diagnostic tool to quantitatively provide the relative contributions from different chemical/physical processes, which is suggested to be discussed with Section 3-4 to further support the conclusion, rather than as an isolated section. For instance, ozone concentration changes due to transport and dry deposition processes may be more closely related to the circulation as well as meteorological conditions, while those due to photochemistry can be interpolated by emission change.

Response: there are indeed some relationships between Sections 3-4 and Section 6, and the budget analysis in Section 6 can quantitatively support the results discussed in Sections 3-4. Nonetheless, we still want to retain the original structure of the manuscript, as we think the current discussions of the impacts of emissions vs. meteorology and transport vs. chemistry in separate sections are also clear enough. In the revised manuscript, we have referred to the results of budget analyses in Section 6 when discussing the relative contributions from emissions and meteorological conditions in Sections 3-4.

Another suggestion is the inclusion of more in-depth analysis on of precursors' response. Specifically, information on how the changes in emission and meteorology influence spatial pattern of NOx and VOC can help better interpolate the model results.

Response: we totally agree with the reviewer that the analysis of precursors' response could help to better understand the changes of O_3. So we examined the spatial distributions of NO_2 and NMVOCs for the four modelling scenarios as well as their differences. The following discussions have been added in the revised manuscript and supplementary document.

Page 10, Line 9:

"The spatial distributions of O_3 precursors (NO_2 and NMVOCs) for the different scenarios and their differences are shown in Figure S10 and S11, which can better explain these results. Detailed description is given in the supplementary document."

Page 12, Line 18:

"The changes of NO_2 and NMVOCs also indicate the impact of emission changes larger than that of meteorological change (Figure S10 and S11)."

Supplementary document:

Spatial distributions of the modelled O_3 precursors over CEC in July 2003 and 2015

The spatial distributions of the modelled NO_2 and NMVOCs over CEC in July 2003 and 2015 and their differences are shown in Figures S10 and S11, respectively. We found that both NO_2 and NMVOCs had increased significantly over CEC from July 2003 to July 2015. The spatial distribution is in accordance with the emission inventory in Figure S15, which

shows high levels of NO_2 and NMVOCs in the eastern CEC and Sichuan basin. Comparing the results of 03E15M-03E03M and 15E03M-03E03M, we can find that the contribution from the emission change (15E03M-03E03M) is much higher than that from the meteorology change (03E15M-03E03M). This is as expected as the O_3 precursors are primary pollutants and should be governed by the anthropogenic emissions. Furthermore, the O_3 precursor concentrations over the eastern part of CEC increased much higher than the western part. Overall, the modelling results for NO_2 and NMVOCs agree well with the results of O_3 in the main context.

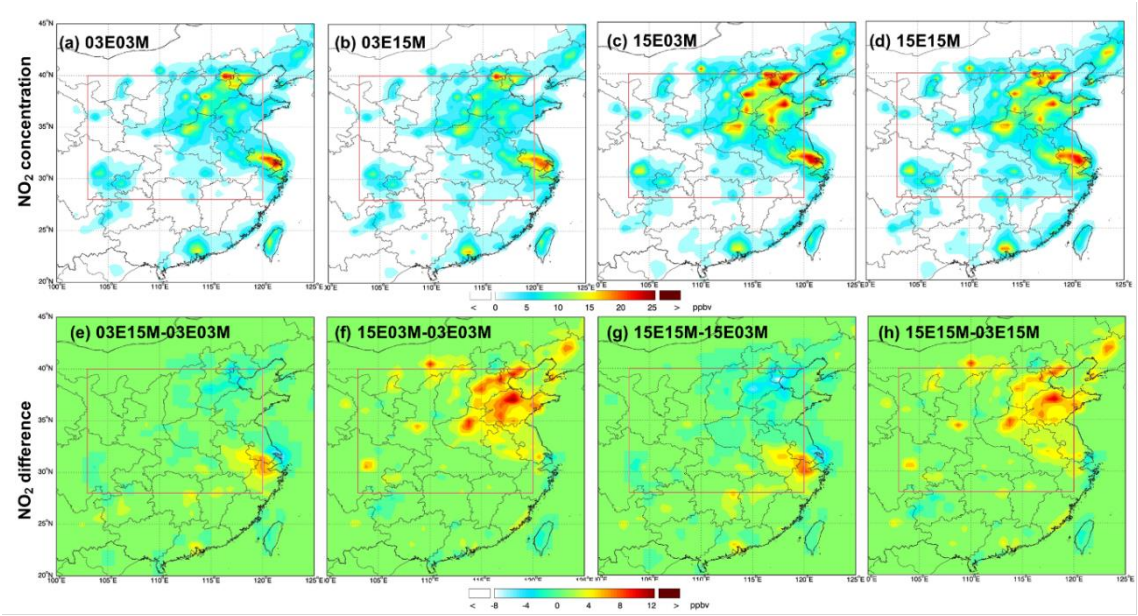

Figure S10. Monthly-mean spatial distributions of surface NO_2 in July over CEC: (a)-(d) and the differences in NO_2 concentrations between these simulations: (e)-(h). The red rectangle represents the Central Eastern China region (CEC: 103 ℃-120 ℃, 28 ℃-40 ℃).

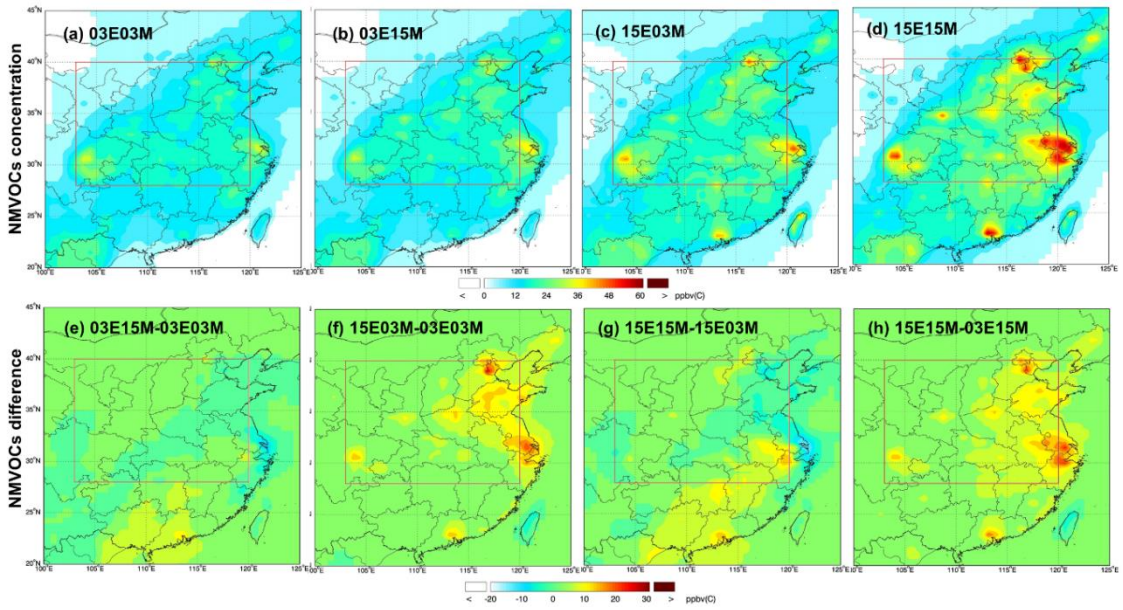

Figure S11. Monthly-mean spatial distributions of surface NMVOCs in July over CEC: (a)-(d) and the differences in NMVOC concentrations between these simulations: (e)-(h). The concentrations of NMVOCs include: ALK4 (lumped >=C4 Alkanes), Isoprene, Acetone, Methyl Ethyl Ketone, Acetaldehyde, RCHO (lumped Aldehyde >=C3), PRPE (lumped >=C3 Alkenes), Formaldehyde, Hydroxyacetone and Glycoaldehyde. The red rectangle represents the Central Eastern China region (CEC: 103 ℃-120 ℃, 28 ℃-40 ℃).

Minor corrections: Section 3.1: Technically, model evaluation should include performance on reproducing meteorology, relevant precursors as well as ozone.

Response: we have compared the observed diurnal variations of NO_2, CO and O_3 with the simulation results at the nine sites over CEC in July 2015. Overall, the model captures the observed diurnal variations of these species at these sites. We didn't get the meteorological data from the observation sites, thus we didn't compare the meteorological conditions in this study. The following model validation results and discussions have been added in the revised manuscript and supporting materials.

Page 8, Line 20: "We also compared the simulated diurnal variations of CO and NO_2 in the nine cities against the observational data (see Figures S6 and S7). Overall, the model captures most diurnal variations of CO and NO_2. The underestimation of CO by the model may be due to the underestimation of emissions and/or the excessive OH (Yan et al., 2014; Young et al., 2013). The large bias in NO_2 may be due to the effect of local emissions. Another reason for the discrepancy between observed and modelled NO_2 is the overestimation by the measurements based on catalytic conversion of other oxidized nitrogen species to NO (Xu et al., 2013)."

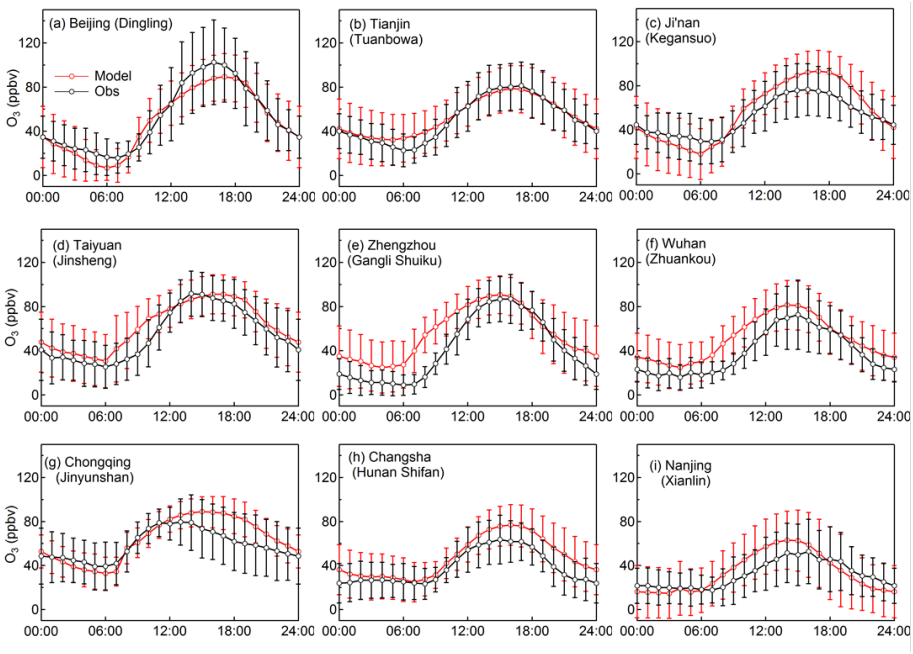

Figure S5. Observed and simulated monthly-mean diurnal variations of surface O_3 in July 2015 at representative air quality monitoring stations in nine cities.

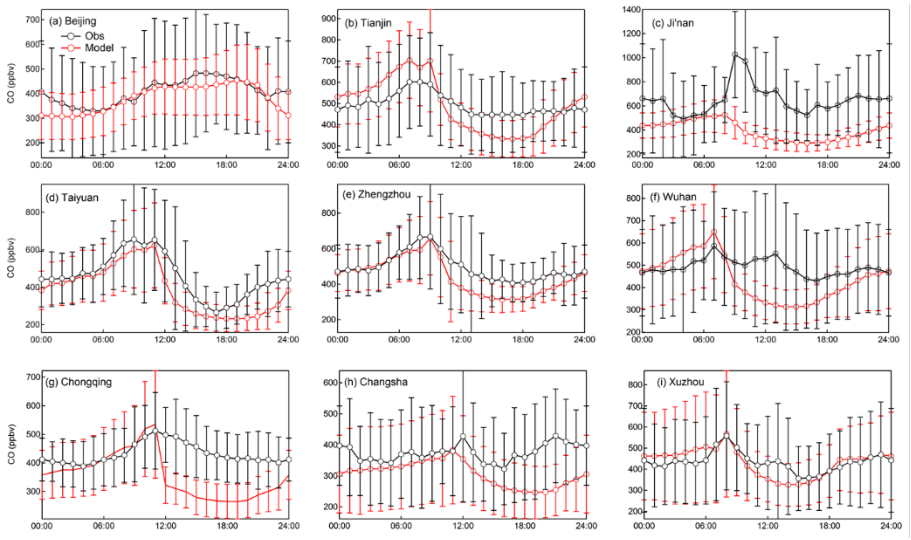

Figure S6. Observed and simulated monthly-mean diurnal variations of surface CO in July 2015 at representative air quality monitoring stations in nine cities.

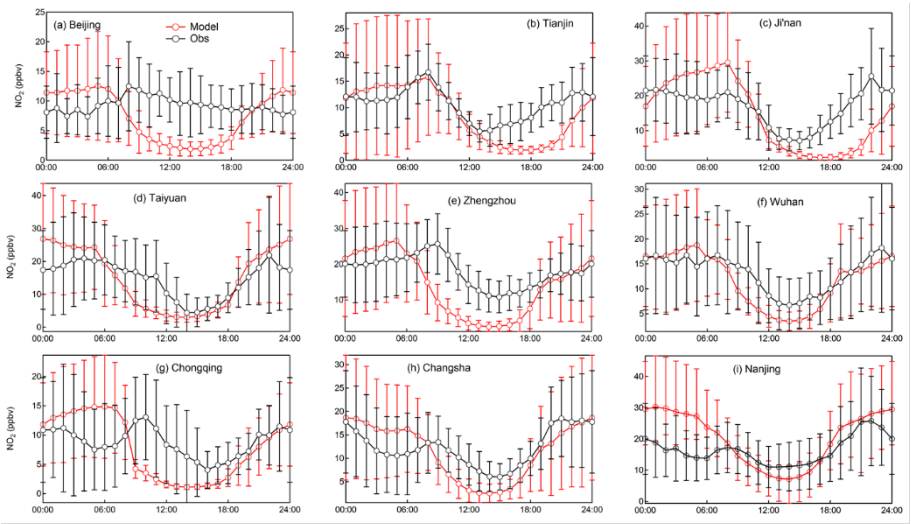

Figure S7. Observed and simulated monthly-mean diurnal variations of surface NO_2 in July 2015 at representative air quality monitoring stations in nine cities.

Table 3: What is the region for the emissions, China or global emission? It should be explained in the caption and also in the main text. Since this work mainly focused on ozone in China, I believe the comparisons of emission in China would make more sense.

Response: the emission region is the Central Eastern China. We have stated the region in the revised Table caption and the main text.

Table 4: What the values in the parenthesis stand for? Another, it is better to sum up the horizontal and vertical advection into one single term to represent the contribution of

transport.

Response: The values in the parenthesis stand for the amounts of photochemical production and loss in unit of ppbv day^{-1}. The "total transport" term represents the sum of horizontal and vertical advection, and thus represents the contribution of transport. We have elaborated this in the revised manuscript.

Figure 5: What does the white color in Figure 5 mean?

Response: We have modified the color bar of Figure 5, and the revised figure is as follows.

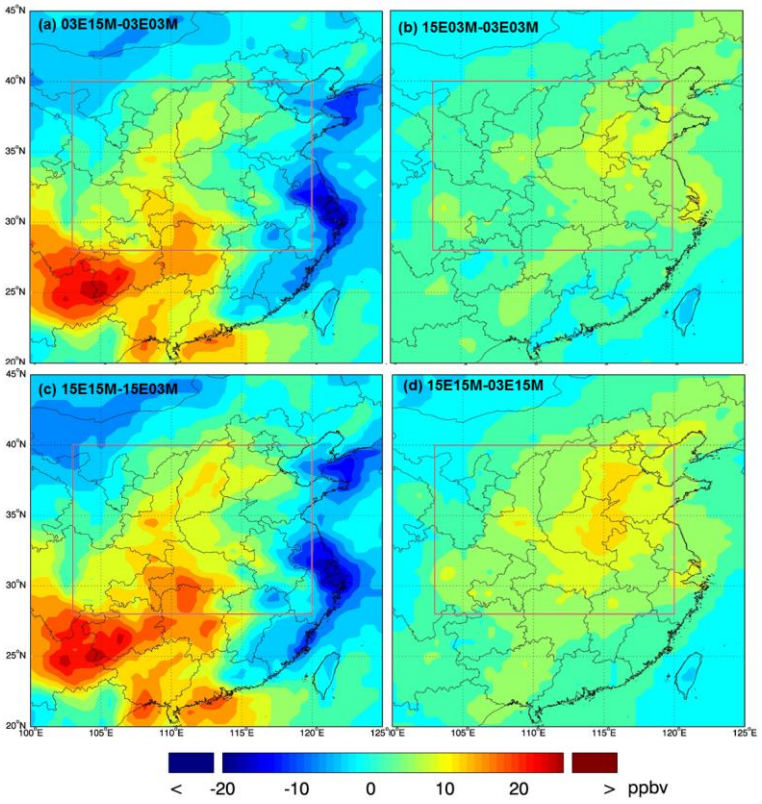

Figure 5. (a) Contributions of meteorological changes to surface MDA8 O_3, comparing 03E15M and 03E03M (2003 standard) simulations; (b) Contributions of emission changes to surface MDA8 O_3, comparing 15E03M and 03E03M (2003 standard) simulations; (c) Contributions of meteorological changes to surface MDA8 O_3, comparing 15E15M (2015 standard) and 15E03M simulations; (d) Contributions of emission changes to surface MDA8 O_3, comparing 15E15M (2015 standard) and 03E15M simulations.

Page 14 Line 25: What is difference between transboundary and long-distance transport here, and how the authors draw this conclusion based on this work?

Response: we have changed "transboundary transport" and "long-distance transport" to "transport" in the revised manuscript. We found that large-scale regional transport is an important contributor to the spatial distributions and inter-annual variations of surface O_3

over the CEC region through the O_3 transport flux analysis. The original statement has been modified as follows.

"Transport issues in local O_3 control strategies should go beyond transport from neighbouring areas (e.g., cities) and account for the long-distance transport (e.g., across provinces)."

Page 2 Line 8: which controls

Response: changed

Page 14 Line 2: "Asia" should be "Asian"

Response: changed.