Response to referee comments on "Enhanced atmospheric oxidizing capacity in simulating air quality with updated emission inventories for power plants especially for haze periods over East China"

We would like to express our sincere thanks to both referees for their careful reviews, and helpful suggestions. Below are our detailed, point-by-point replies to both referees.

To Referee #2

General comments:

This manuscript, using with and without an updated emission inventory of coal-fired power plants (UEIPP) in Multi-resolution Emission Inventory for China (MEIC) to drive WRF-Chem model, analyzes impacts of emissions and the atmospheric oxidizing capacity on particulate and ozone precursors, especially PM2.5 episode in east China's Jiangsu Province. The UEIPP developed by collecting the online monitoring data from power plants (Zhang et al., 2015) is considered as a more realistic way to obtain accurate point emission data for East China. Based on the analysis of regional air quality impacts of change in power plant emissions, the study addresses that the uncertainty in the MEIC emission data can be partly reduced by improving the accuracy of the point source emission inventory which is one of key uncertainty sources influencing modeling results in East China. Power plant is one of five sectors (power, industry, transportation, residential, and agriculture) defined in MEIC system. The subject is important for atmospheric science community and environmental protection agencies to understand the importance of development of the completed emission inventory for evaluation of the effect of air pollution control measures. The results are interesting and scientifically meaningful. However, just one mechanism (WRF-Chem/CBM-Z) applied to the study may be not enough to study atmospheric oxidizing capacity because changes in OH and VOC oxidation in the presence of NOX are sensitive to chemistry mechanisms [Jimenez et al., 2009; Stockwell et al., 2011; Knote et al., 2015; Derwent, 2017]. The authors are encouraged to use more different chemistry mechanisms to validate the results.

Response: Thanks for the reviewer's encouragement and suggestions. The main difference among existing mechanisms in the WRF-Chem lies in lumping technique used to classify organic compounds into surrogate groups with a common agreement between existing

mechanisms for O_3, and but remained differences for OH, which is a crucial oxidizing agent in atmosphere. In order to further examine the conclusion about changes in oxidizing capacity as being suggested, we have run the WRF-Chem/RADM2 simulation.

The results in O_3 and OH using RADM2 were presented below (Fig. R1), illustrating an increased pattern over Jiangsu province, which was similar to that using CBM-Z (Fig. 3o and 4b in the revised manuscript). The increases in O_3 and OH under two different mechanisms of CBM-Z and RADM2 further indicated the enhanced oxidizing capacity. We have added the results of RADM2 in the revised manuscript to further examine our conclusion.

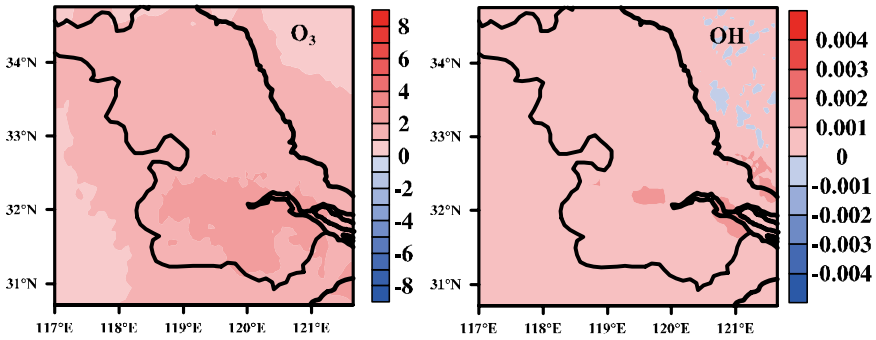

Figure R1 The same as Figure 4 in manuscript but using the chemistry mechanism RADM2.

In addition, the paper needs some English improvements for being written more precisely. Please have the manuscript examined by a native English speaker or ask for editor's help to improve the overall language of the paper. I recommend its publication basically in a revision in accordance with the review comments.

Response：Following the suggestion, the English usages in manuscript has been substantially revised. Please see the revised manuscript with the "track changes".

Major comments:

1. Please provide more references and discussions why the elevated emissions are able to lead to "more significant environmental effect through regional transport than the surface emissions" as stated at Line 80 – 83 on Page 4. Is that because elevated emitted particulate and ozone precursors get longer life cycles in upper air or are able to be regionally transported or because of the both? All the more reason why to say so. Do you think power plant emissions considered as point source is more important than on-road emissions in

studying of impacts of sources on local/regional air quality? Why?

Response: Thanks for the comments. Both elevated emitted particulate and ozone precursors getting longer life cycles in upper air and the more efficiently regional transport were thought to be the reason for more significant environment effect due to the elevated emitted particulate and ozone precursors with less deposition driving by stronger winds and well organized circulation in upper air, such as by low-level jets (Hu et al., 2013). The reasons have been provided in the revised manuscript.

As discussed above, power plant emissions might be more important on regional air quality. From this study, it could not assess which, power plant emissions or on-road emissions is more important emissions in studying of impacts of sources on local/regional air quality, which should be further studied under changing air pollutant emissions and meteorological conditions.

References:

Hu, X.-M., Klein, P. M., Xue, M., Zhang, F., Doughty, D. C., Forkel, R., Joseph, E., and Fuentes, J. D.: Impact of the vertical mixing induced by low-level jets on boundary layer ozone concentration, Atmospheric Environment, 70, 123-130, 10.1016/j.atmosenv.2012.12.046, 2013.

2. For the initial and boundary conditions (IBC) for the WRF-Chem simulation for December 1 – 31, 2013, the "default initial and boundary chemistry profiles in the model were selected" according to statement at Line 125 on Page 6. Please define what the default initial and boundary conditions are, and clarify why "the default" IBC should be used. I wonder if the authors considered the time-varying chemical boundary conditions should be used because the simulation time period of about one month is longer enough to consider the impact of inflow and background concentrations of O3 and relatively long-lived VOCs on domain solutions.

Response: The default initial and boundary chemistry conditions in WRF/Chem were based on the vertical profiles of O_3, SO_2, NO_2, CO, VOCs and other air pollutants from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM) (Liu et al., 1996). The first two-day

simulation was discarded as model spin-up. The outmost domain of modeling tests was set large enough to cover East Asia to avoid the impact of chemical boundary conditions on simulation. Furthermore, the frequent haze pollution over Eastern China is resulted from the regional pollutant emissions (Wang et al., 2015; Zhang et al., 2015) with less contribution of foreign emission to haze pollution over Eastern China. The IBC and the reasons have been clarified in the revised Section 2.1.

Refereces:

Liu, S., McKeen, S., Hsie, E. Y., Lin, X., Kelly, K., Bradshaw, J., Sandholm, S., Browell, E., Gregory, G., and Sachse, G.: Model study of tropospheric trace species distributions during PEM‐West A, Journal of Geophysical Research: Atmospheres, 101, 2073-2085, 1996.

Zhang, Q.: A Heavy Haze Episode in Shanghai in December of 2013: Characteristics, Origins and Implications, Aerosol and Air Quality Research, 10.4209/aaqr.2015.03.0179, 2015.

Wang, M., Cao, C., Li, G., and Singh, R. P.: Analysis of a severe prolonged regional haze episode in the Yangtze River Delta, China, Atmospheric Environment, 102, 112-121, 2015.

3. To understand the difference between the two emission systems, it is better to replace the absolute quantity of emissions in MEIC and UEIPP in Fig. 2 by the emission difference between MEIC and UEIPP.

Response: Following the suggestion, we have replaced the absolute quantity in the revised Fig. 2 with the emission difference, and modified the relative discussion in the revised manuscript.

4. Please provide which version of WRF-Chem model was used for this study in Subsection 2.1 Model description and configuration.

Response: The version (3.7.1) is provided in the revised manuscript.

5. The NMVOCs in UEIPP are translated to lumped VOC compounds in RADM2 mechanism according to the statements in the last paragraph of Subsection 2.3.1 on Page 8. However, WRF-Chem with CBMZ is likely used for the air quality modeling (refer to Line 117 on Page 6). I am wondering, in RADM2 and

CBMZ, which gas phase mechanism is eventually used in the study.

Response: The CBMZ is eventually used. In the revised manuscript, it has been clarified in Section 2.3.1.

6. Please clearly state which model domain (I think it should be 5-km domain) results were used for model performance evaluation in Section 3 Modeling evaluation.

Response: The results from 5-km domain were used for evaluation.

7. It is well-known that wind speed and wind direction are both determinant in the horizontal transport of air pollutants. The local concentration of air pollutants is affected by not only wind speed but also wind direction. In Subsection 3.1 Meteorological evaluation, please provide solid reason(s) why the evaluation of wind direction is missed or ignored.

Response: We have provided the evaluation of wind direction via calculating the Hit Rate (HR; Schlünzen and Sokhi, 2008) in the revised Section 3.1.

References:

Schlünzen, K., and Sokhi, R.: Overview of tools and methods for meteorological and air pollution mesoscale model evaluation and user training, Joint report by WMO and COST, 728, 2008.

8. The discussions about the relationship between the overestimated SO2 and the underestimated sulfate in Page 12 are interesting. I hope the authors continue their investigation of this issue because sulfate is one of important precursors to secondary aerosol and PM2.5. On the conversion of SO2 to sulfate, authors may refer to the paper by He et al. (2014).

Response: The conversion of SO_2 to sulfate was revealed remarkable during severe haze episodes. The paper (He et al. 2014) has been cited in the revised manuscript. We will continue to investigate the issue.

9. In subsection 4.1, I suggest the authors to take advantage of the result of VOC-limited in Section 4.2. It is helpful to understand why the enhanced O3 is caused by the increased VOC rather than decreased NO2 in UEIPP.

Response: We have presented the difference of VOC (Fig. R2) in the revised Figure 4 to explain the enhanced O_3 following the suggestion. Effect of decreased NO_2 on enhanced O_3 is reserved, reflecting that the emission decrease of NO_x is more significant than increase of VOC in UEIPP.

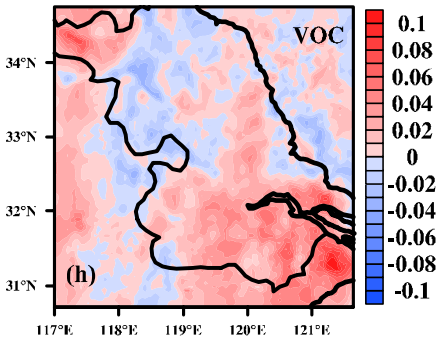

Figure R2 Difference of VOC in December 2013 (MOD2 - MOD1). Unit: "ppmv".

10. The arguments for the increased PM2.5 modeled by MOD2 in the last paragraph of Section 4.1 are very interesting but the more solid quantitative evidences are needed to support.

Response: Thanks for this suggestion. Quantitative evidences are provided in the revised Section 4.1 via performing a sensitivity simulation test MODa as same as MOD2 with closing BC emission in UEIPP. Based on the $PM_{2.5}$ differences between MOD2 and MODa regionally averaged over Jiangsu Province, it was estimated that the physical process of aerosol radiative effect stabilizing boundary layer contributed about 0.15 μg m^{-3} to the $PM_{2.5}$ enhancements, during the haze episode.

11. To further understand why modeled increase in sulfate, nitrate, and PM2.5 concentration is associated with reduction in emissions of SO2 and NOx in UEIPP, the authors may refer to a news report entitled "The Real Reasons China Is Struggling To Control Its Pollution Problem" at http://fortune.com/2017/01/10/china-red-alert-pollution-pm2-5/.

Response: The report stated that "reduction of SO_2 emissions may have had no effect on $PM_{2.5}$ overall in North China, because the reduced SO_2 may free NH_3 to react instead with

NOx creating ammonium nitrate particles", which is confirmed in our study where both ammonium and nitrate concentration was higher in MOD2 (with lower SO_2 emissions) relatively to MOD1 (Fig. 4e, f). We have added a statement in the revised conclusion as "reduction of SO_2 may free NH_3 to react instead with NOx creating ammonium nitrate particles, which would need further studying.".

Minor comments:

1. Line 37 on Page 2: "... play a determinant role in deteriorating air quality ..." might be "... play a key role in air quality,...".

Response: It has been modified.

2. Line 42 on Page 2: ''... emission inventories covering East China ..." should be "... emission inventories for East China ...".

Response：It has been modified.

3. Line 61 on Page 3: "... chemical reactions involving particle formations, SO2, NOX, O3 and oxidizing radicals." may be changed into "... chemical reactions involved in particle formation and O3 production due to emission changes in particle and ozone precursors".

Response：It has been changed.

4. Line 65 – 67 on Page 3: "An understanding of the power plant emissions in East China and subsequently a reliable evaluation of their environmental changes and effects using air quality models largely depend on the accuracy of pollutant emission inventory" is better changed into "A studying focusing on the reliable power plant emission inventory for East China used for air quality models is helpful to understand the real situation of air pollution and quantitatively assess impacts of emission sources on air quality in East China".

Response：Following the suggestion, it has been changed.

5. Line 72 – 73 on Page 3 and 4: "the inaccuracy in estimating individual power plant emissions is always a

defect that rendering intrinsic biases between observed and modeled air pollutant concentrations" might be "the inaccuracy of emissions from any local power plants can be considered as one of sources of uncertainty to lead to model bias".

Response：It has been changed.

6. Line 79 on Page 4: "shrouded this region in recent years, attracting wide scientific and governmental attention" can be written as "have shrouded this region in recent years and the poor air quality in China has attracted worldwide attention".

Response：Thanks. It has been re-written.

7. Line 86 on Page 4: "emitting over 1000 kilotons (kt) SO2 per year ...". You mean SO2 emissions for Jiangsu province was over 1000 kt per year during the period from 2005 to 2010. Am I right? The number should be clarified and the sentence needs to be rewritten.

Response：The annual SO_2 emissions were estimated with 1107 kt and 803 kt in 2005 and 2010 (not the period from 2005 to 2010), respectively. The sentence has been rewritten as "The annual SO_2 and NO_x emissions were estimated with 1107 kilotons (kt) and 626 kt in 2005, as well as 803 kt and 781 kt in 2010 in Jiangsu Province."

8. Line 89 -92 on Page 4: "China is endeavoring to control ..." might be better to change into "Source control measures focus on power generation processes and facility-related measures to reduce emissions have been widely implemented in China. These measures include flue gas desulfurization (FGD), selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR), dust collector, etc. It is important to assess any potential air quality impacts from implementation of those mitigation measures."

Response：Many thanks for the referee's careful edition. It has been changed.

9. Line 108 -110 on Page 5: For data source of NCEP reanalysis data, please cite a reference(s). It may be "Kalnay et al.,The NCEP/NCAR 40-year reanalysis project, Bull. Amer. Meteor. Soc., 77, 437-470, 1996".

Response：It has been cited there.

10. Line 292 -294 on Page 14: The authors may consider to re-written those sentences because with large

unmonitored area, from my point of view, it is hard to say differences in spatial distribution patterns caused by different emission inputs are evidences for the improvement of MOD2 simulation. The "improvements" here are still based on the results at monitoring sites. However, the evaluation of model performance over site by site has been well done and discussed.

Response: The differences are not evidences to prove the improvement in MOD2 simulation. We have revised those sentences in the paragraph.

11. Line 303 -305 on Page 14: "...,especially of O3, SO2, and NO2 according to the statistic validations and the spatial simulation performance compared with observations, which could conclude that a more realistic power emission was provided from UEIPP" can be re-written in "...,especially of O3, SO2, and NO2 according to model performance evaluation. It is therefore concluded that the power plant emissions from UEIPP is more realistic and reliable".

Response：Following the suggestion. It has been revised.

12. Line 278 – 281 on Page 13: "The underestimation of PM2.5 ...changes of R and MFE" might be replaced by "MOD2 simulations show overall improvement for all species compared to MOD1 results. Although the both MOD1 and MOD2 underestimated PM2.5, CO, O3 and overestimated NO2 and SO2 as shown in Table 3, absolute MFBs for those species are reduced by 0.07, 0.21, 10.78, 3.6, and 8.26 percent respectively from MOD1 to MOD2." The statements after this line in the entire paragraph need to be rewritten because it is little bit difficult for me to understand.

Response：Following your suggestion, the sentence has been replaced. The statements after that sentence in the entire paragraph has been rewritten as "Also, the SIAs at Nanjing were ameliorated in MOD2 simulation (Table 4). Under the unchanged meteorology between two simulations, the reduced deviations of NO_2, SO_2, CO, $PM_{2.5}$ and O_3 in MOD2 relatively to MOD1 should be attributed to emission changes in MOD2 with UEIPP as the power plant emission. However, $PM_{2.5}$ and O_3 are highly dependent on secondary formation, indicating their changes in conjunction with chemical conversion variations between the two simulations of MOD1 and MOD2, which was comprehensively investigated in Section 4.".

13. What does "accident error" mean at Line 288 on Page 13?

Response：It means some deviations induced by computer, such as different CPUs used, different compilers (PGI or Intel …) and so on between simulations. As all of those were same between MOD1 and MOD2, we have deleted the accident error in the revised manuscript.

14. Line 335 on Page 15: "Environmental Protection Agency (EPA) in China" should be "China Ministry of Environmental Protection" or "Ministry of Environmental Protection of the People's Republic of China".

Response：It has been revised to "Ministry of Environmental Protection of the People's Republic of China".

15. Line 344 – 345 on Page 16: "... the declined emissions of primary PM2.5 could not enhance the ambient PM2.5 concentration ..." might be " ... the declined emissions of primary PM2.5 could not improve the ambient P2.5 concentration ...".

Response：Thank you for the careful revision. It has been changed.