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 #1

This study attempts to examine the influence of a more accurate emission inventory of coal-fired power plant, which was derived from online monitoring data and implemented in the Multi-resolution Emission Inventory for China, on the simulation of air quality during haze events. The authors find that the updated emission inventory improves the simulation of the ambient concentrations of the primary air pollutants and strengthens the formation of aerosols by increasing oxidizing agents like O3 and OH. This study sheds some light on how important of the use of more accurate emission inventory in reducing the uncertainty of air pollution prediction. Below are some issues which need sufficient revision.

1. In section 3, could the authors provide additional statistical significance tests for model validation? For example, when making comparison of observation and simulations, one of the statistical parameters the authors looking at is the correlation coefficients (see Tables 2 and 3). But, are these correlations between observation and simulation statistically significant? A quick check for this concern could be made by examining the p-values when doing linear regression. Also, the difference in most statistics (e.g. R, MFB, MFE, and so on) between MOD1 and MOD2 are relatively small (Tables 3 and 4), and it is difficult to evaluate how significant of the changes in concentrations of aerosol compositions presented in Table 5. Therefore, the authors may also need to perform some statistic tests to see the significance of the improvements in prediction of atmospheric chemical species when introducing with UEIPP into MEIC emission inventory.

Response: Thanks for the suggestions. The significance test of correlation coefficients R between observation and simulation was checked with p-values. In the revised manuscript, R values in Table 2 and 3 were labeled with p-value < 0.001.

The bootstrap confidence interval (DiCiccio and Efron, 1996) test was used to see the significance of the improvements of atmospheric chemical species. Results of the significance test for R, MFB, MFE were shown in Table 3. Discussions about the significance of the improvements in atmospheric chemical species were added in the revised manuscript in lines 294-299. Improvements of the aerosol compositions in Table 4 didn't passed the confidence level of 90 % with 31 samples for each composition in the daily observation.

References:

DiCiccio, T. J., and Efron, B.: Bootstrap confidence intervals, Statistical science, 189-212, 1996.

Following are other two minor issues about the statistics and their evaluation criteria. In lines 238-239, what's the detailed criteria for a "good" model performance proposed by Emery et al. (2001)? The MFB and MFE values for O3 in Table 3 appear much greater than the "satisfactory" criteria values (60% and 75%, respectively) proposed by Morris et al. (2005). Does this contradict with the statement in lines 253-254, i.e. O3 hourly variations were well captured?

Response: (1) Emery et al. (2001) proposed that good model performance would be classified as temperature bias smaller than 0.5^{o} and wind speed RMSE smaller than 2 m s^{-1}, without indicating the R-ranges. We revised the sentence in lines 236-239 to "the variations of wind speed were generally captured by the model with the R varying from 0.51 to 0.77 (p-value < 0.001). The RMSE ranging from 1.8 m s^{-1} to 2.1 m s^{-1}, basically conforming to the "good" model performance criteria for wind speed (Less than 2.0 m s^{-1}; Emery et al., 2001).".

(2) In the revised manuscript, the statement in lines 253-254 was changed to "hourly O_3 variations were reasonably captured".

2. In section 4.1, how great, a little more quantitively, of the BC radiative effects on the surface PM2.5 concentration? It seems the both 2 m temperature and boundary layer height (BLH) change a little in MOD2 relatively to MOD1. The signals in atmospheric warming and BLH reduction are too weak. Maybe focusing on haze episodes only could give stronger signals induced by BC absorbing. Also, try to check the vertical profiles of PM2.5 under different emission conditions, which might provide some insights of the relationship between surface PM concentration and the BLH, given that aerosols are well mixed in well-developed boundary layer. If necessary, additional simulation could be performed, in which UEIPP is used but BC radiative effects turned off. This sort of control experiments might help the authors to more quantitively evaluate the perturbation of surface PM concentration due to BC radiative effects.

Response: Sincere thanks for this suggestion. Although a small fraction of BC in the ambient $PM_{2.5}$, due the strong radiative absorption of BC, the BC radiative effects on the ambient $PM_{2.5}$ variation could be significant trough changing boundary layer structure (Ding et al., 2016). Regionally averaged, the both 2 m temperature and boundary layer height changed a little in MOD2 relatively to MOD1. The decreases in downward short wave flux at ground, and 2m air temperature as well as BLH reductions focusing on haze episodes and center could give stronger signals induced by BC absorbing (Table R1). The vertical profiles of $PM_{2.5}$ under different emission conditions in MOD1 and MOD2 provide some insights of the relationship between surface PM concentration and the BLH (Fig. 1R). This relative analysis has been added in lines 358-364 in the revised manuscript.

Table R1. The changes of downward short wave flux at ground (SWDOWN), 2m air temperature (T2) and boundary layer height (BLH) from MOD1 to MOD2 in the daytime of Dec.7, 2013 during a haze event in Wuxi, a haze center.

	SWDOWN (W m^{-2})	T2 (K)	BLH (m)
Changes (MOD2-MOD1)	-11.8	-0.30	-26.4

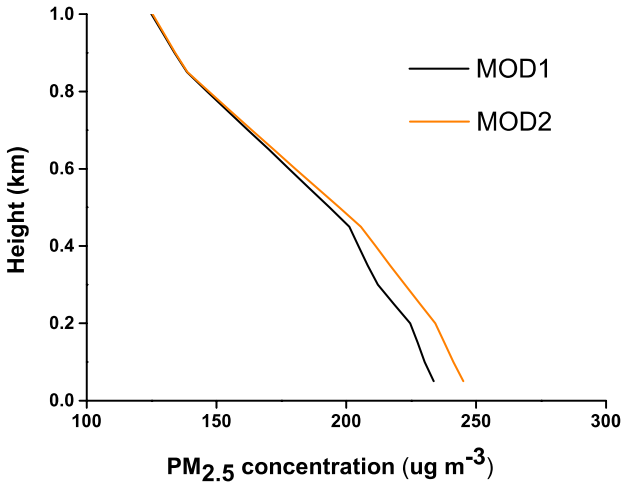

Figure R1. Vertical profiles of $PM_{2.5}$ concentrations simulated in MOD1 and MOD2 duing the daytime of Dec.7, 2013 during a haze event in Wuxi, a haze center.

References:

Ding, A. J., Huang, X., Nie, W., Sun, J. N., Kerminen, V. M., Petäjä, T., Su, H., Cheng, Y. F., Yang, X. Q., Wang, M. H., Chi, X. G., Wang, J. P., Virkkula, A., Guo, W. D., Yuan, J., Wang, S. Y., Zhang, R. J., Wu, Y. F., Song, Y., Zhu, T., Zilitinkevich, S., Kulmala, M., and Fu, C. B.: Enhanced haze pollution by black carbon in megacities in China, Geophysical Research Letters, 43, 2873-2879, 10.1002/2016gl067745, 2016.

3. In section 4.2, what's the reason for the enhancement in concentration of SIAs (surfate+ nitrate+ammonium) greater than that of the PM2.5 (see Table 5 and lines 392- 392)? I would expect that both the increases in SIAs and BC/EC should contribute to the increase of PM2.5, meaning the enhancement of PM2.5 should be larger than SIAs'.

Response: Compared to the MOD1, the lower emission of primary $PM_{2.5}$ in UEIPP lead to the less concentrations of primary $PM_{2.5}$ in MOD2 (Table 1), such that the enhancement in concentrations of SIAs was partially offset by the lower primary concentrations of $PM_{2.5}$. This explanation is added in Lines 424-426 in the revised manuscript.

In addition, which process, the physical process like BC radiative effect stabilizing boundary layer or the

chemical reaction like intensified SIA formation, is more dominant in the PM2.5 enhancement observed in this study when using UEIPP as coal-fired power plant emission inventory?

Response: In order to quantify the radiative effects induced by BC emission change, a sensitivity simulation test MODa as same as MOD2 with closing BC emission in UEIPP was performed. Based on the PM2.5 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, while the chemical reaction contributed about 4.77 μg m^{-3} (Table 5) to the $PM_{2.5}$ enhancements during the haze episode, reflecting that the chemical reaction was more dominant in the $PM_{2.5}$ enhancements in our study. This conclusion is added in lines 453-456 in the revised manuscript.

Minor comments:

Line 24: please expand the term of NMVOCs.

Response： It has been expanded in the revised abstract.

Line 61-63: What the refs for this statement that power plant emission is the most important source of pollutant?

Response： We have added the reference (Zhao et al. 2010) in the revised manuscript.

Lines 266-267: Two related studies recently published (Wang et al., PNAS, 2016; Cheng, Y., et al., Science Advances, 2016) should be cited here.

Response: Thanks for the suggestion. The two papers have been cited.

Line 273: Pls change "reasonable" to "reasonably".

Response： It has been changed.

Line 279: The overestimates or underestimates are still present in MOD2. Use another word instead of "diminished".

Response： The sentence has been re-written as "Although the both MOD1 and MOD2 underestimated $PM_{2.5}$, CO, O_3 and overestimated NO_2 and SO_2 as shown in Table 3, the MFBs for those species are reduced by 0.07, 0.21, 10.78, 3.6, and 8.26 % respectively from MOD1 to MOD2"

Line 290: 16.38% should be -16.38%.

Response： This mistake has been corrected.

Line 389: Fig. 6 should be Fig. 5?

Response： It should be Fig. 4f and has been changed in the revised manuscript.

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.

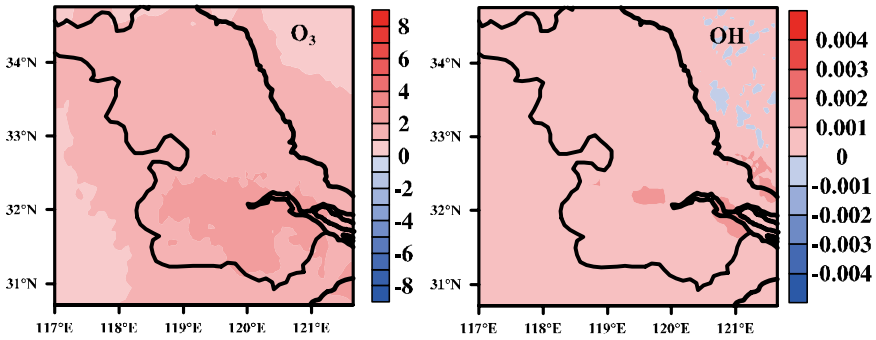

Figure R1 The same as Figure 4 in manuscript but using the chemistry mechanism RADM2.

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.

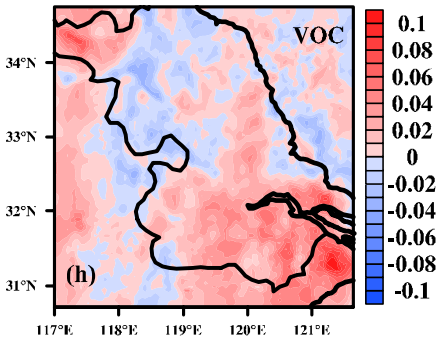

Figure R2 Difference of VOC in December 2013 (MOD2 - MOD1). Unit: "ppmv".

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.

Enhanced atmospheric oxidizing capacity in simulating air quality with updated emission inventories for power plants especially for haze periods over East China

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Abstract. Air pollutant emissions play a determinant role in deteriorating air quality. However, an uncertainty in emission inventories is still the key problem for modeling air pollution. In this study, an updated emission inventory of coal-fired power plants (UEIPP) based on online monitoring data in Jiangsu Pprovince of East China for the year of 2012 was implemented in the widely used Multi-resolution Emission Inventory for China (MEIC). By employing the Weather Research and Forecasting Model with Chemistry (WRF-Chem), two simulation experiments were executed to assess the atmospheric environmental change by using the original MEIC emission inventory and the MEIC inventory with the UEIPP. A synthetic analysis shows that (1) power compared to the power emissions of MEICemissions of, $PM_{2.5}$, PM_{10}, SO_2 and NO_x were lower, and CO, black carbon (BC), organic carbon (OC) and NMVOCs (Non-methane volatile organic compounds) were higher in the UEIPP relatively to those in MEIC, reflecting a large discrepancy in the power emissions over East China; (2) In accordance with the changes of UEIPP, the modeled concentrations were reduced for SO_2 and NO_2, and increased for

most areas of primary OC, BC and CO, whose concentrations in atmosphere are highly dependent on emission changes. (3) Interestingly, when the UEIPP was used, the atmospheric oxidizing capacity significantly reinforced, reflecting by increased oxidizing agents, e.g. O_3 and OH, thus directly strengthened the chemical production from SO_2 and NO_x to sulfate and nitrate, which offset the reduction of primary $PM_{2.5}$ emissions especially in the haze days. This study indicated the importance of updating air pollutant emission inventories in simulating the complex atmospheric environment changes with the implications on air quality and environmental changes.

Keyword: Emission inventory; haze pollution; air quality modeling; secondary aerosols; oxidizing agents

1 Introduction

East China is one of the regions with serious air pollution and frequent haze. In these highly polluted regions, air pollutant emissions play a ~~determinant~~ key role in ~~deteriorating~~ air quality, and their variations can cause a large uncertainty in air pollution modeling and prediction. It is also crucial for air pollution mitigation ~~agency~~ to comprehensively understand ~~anthropogenic~~ air pollutant emissions and their impacts on atmospheric environment. Emission inventories are essential for atmospheric environment research, especially for modeling study and air quality policy making.

During past decades, emission inventories ~~covering~~ for ~~East~~ China were established by several groups. These include the global-scale work, such as the Reanalysis of the Tropospheric chemical composition (RETRO) (Schultz, 2007; Zheng et al., 2009), the Hemispheric Transport of Air Pollution (HTAP) (Janssens-Maenhout et al., 2015), and the Emission Database for Global Atmospheric Research (EDGAR), and the national-scale studies including the Transport and Chemical Evolution over the Pacific mission (TRACE-P) (Olivier et al., 2005), the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) (Zhang et al., 2009), the Regional Emission inventory in Asia (REAS) (Ohara et al., 2007)

and the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/). Owing to less measurements, several of these studies were based on "top-down" algorithm, which rendered the uncertainties in estimating of emission ~~budgets~~ and subsequently decreased the accuracy in ~~the~~ modeling ~~study of~~ atmospheric environment. For example, previous studies showed a difference of 30 % in CO emission among various emission inventories over East Asia, leading to an up to 8 % simulated deviation (Amnuaylojaroen et al., 2014). Regional emission inventories were developed recently in China, for the regions of Yangtze River Delta (Huang et al., 2011; Fu et al., 2013), North China Plain (Wang et al., 2010) and Pearl River Delta (Zheng et al., 2009), as well as several provincial and urban areas (Zhao et al., 2015; Jing et al., 2016; He et al., 2016), with more underlying data for activity levels, emission factors, energy combustion and traffic database obtained.

Air pollution in East China is changing from coal-smoke to mix-source polluted type, particularly the secondary aerosols surging in severe haze episodes (Huang et al., 2014a), with more complicated chemical reactions ~~involving~~ involved in interaction of particle formation~~s, SO_2, NO_x, O_3 and oxidizing radicals~~ and O_3 production ~~due to emission changes in particle and O_3 precursors~~. As ~~a single and~~ the largest coal-fired sector of emission framework in China, electric power generation is believed to be the most important source of atmospheric pollutant emissions (Zhao et al., 2010). The power plant emissions accounted for 31-59 % of national anthropogenic emissions of SO_2 and 21-44 % of NO_x (Zhao et al., 2008; Wang et al., 2012). ~~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. 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.~~ The pollutant emissions from coal-fired power plants were usually estimated by the widely adopted

"bottom-up" approach (Hao et al., 2002; Zhang et al., 2007b; Zhang et al., 2007a; Ohara et al., 2007; Zhao et al., 2008). However, due to limited access to specific information about power plants, such as the mass of pollutant emitted per unit fuel consumption or per unit industrial production, coal-fired boiler types or accurate location of power plants (Wang et al., 2012), ~~the inaccuracy in estimating individual power plant emissions is always a defect that rendering intrinsic biases between observed and modeled air pollutant concentrations.~~ the inaccuracy of emissions from ~~any~~ local power plants can be ~~considered as~~ one of ~~sources of~~ uncertainty sources to lead to model bias. ~~Therefore, t~~The validations of ~~the~~ power plant emissions ~~and their~~with the impacts on atmospheric environment particularly in haze episodes are still a gap.

Jiangsu Province is one of the most developed areas in East China, providing living place for a population of 79.2 million with the highest gross domestic production (GDP) per capita in China (NBSC, 2013a; JSNBS, 2013). Severe air pollution episodes of haze and photochemical pollution ~~repeatedly shrouded this region in recent years, attracting wide scientific and governmental attention~~have ~~repeatedly~~ shrouded this ~~region~~province in recent years ~~and the poor air quality in China has attracted worldwide attention~~ (Fu et al., 2008; Wang et al., 2014; Qi et al., 2015). As elevated emission source, ~~the~~ power plants emit air pollutants with longer life cycles in upper air and more efficiently regional transport because of ~~with high inject height in the atmosphere~~less deposition driving by stronger winds and well organized circulation in upper air, such as by low-level jets (Hu et al., 2013), leading to ~~leading to~~ more significant environmental effect ~~through regional transport~~ than ~~the~~ surface emissions (e.g. ~~vehicle~~ on-road emission), reflecting ~~a~~ potential importance of accurately estimating the power plant emissions and their influences on air quality. Based on unit-based methodology, Zhao et al. (2008) developed an inventory of coal-consuming power plants for all the provinces in China~~,~~. 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

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2010 in Jiangsu Provinceamong which five provinces including Jiangsu take up the largest coal consumption, emitting over 1000 kilotons (kt) SO_2 per year and NO_x emitted from Jiangsu province with about 626 kt and 781 kt in 2005 and 2010, respectively. Alternately, another study estimated the NO_x emission in the province from Jiangsu's power plants was estimated aboutat about 748 kt in 2005 by (-Wang et al., (2012), reflecting the uncertainties in the estimation of NO_x emission from power plants. China is endeavoring to control air pollution that the recent measures including construction, transfer, and implementation of techniques (e.g. flue gas desulfurization (FGD), selective catalytic reduction (SCR)/selective non-catalytic reduction (SNCR), and dust collectors, etc.) to power plants need to be assessed for their effects on mitigating air pollution. Source control measures focusing 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 anythe potential air quality impactschanges fromimplementation of those mitigation measures.

Zhang et al. (2015) established annn emission inventory of coal-fired power plants (UEIPP) by collecting the online monitoring data from power plants in atmospheric verifiable accounting tables of Jiangsu Ppprovince for 2012. The volumes of flue-gas and pollutant concentrations were measured in-site for each unit, providing the more realistic data for calculating power plant emissions than that those used in previous studies. As a major objective of this studyIn the current work, the UEIPP was integrated into MEIC to evaluate the impact of emission change update on provincial regional atmospheric environment with an air quality model, which was a major objective of this study. We presented the details of model settings, observational data and emission inventories in Sect. 2 and the modeling evaluation in Sect. 3. The impact of emission change change on atmospheric environment and the underlying mechanism were discussed in Sect. 4. The study was summarized in Sect. 5.

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2 Data and method

2.1 Model description and configuration

In this study,The period from A severe haze event overDecember 1November 29 to – December 31, 2013 werewas selectedchosen as the modeling period, covering a severe haze period (from 3 to 8 December 2013) in Jiangsu Province. In this modeling study. for simulationsimulation period, since severe haze pollution events occurred frequently in this monththe December, the The online coupled Weather Research and Forecasting Model with Chemistry (WRF- Chem, – in version 3.7.1) model was configured in three nesting domains with the horizontal resolutions respectively –ofin 45 km covering most area of East Asia, the resolution of 15 km covering Eastern China and surrounding areas, and 5 km the resolution of 5 km covering Jiangsu province Province and surrounding areas (Fig. 1a). Vertically, there were 35 full eta levels from surface up to 100 hPa with 7 levels below 1 km. The National Center for Environmental Prediction Final Global Forecast System operational analysis data (Kalnay et al., 1996) was utilized for providing the initial and lateral meteorological conditions to WRF-Chem. Grid nudging (Stauffer and Seaman, 1990) was employed for the outmost domain every 6 hours, treating temperature, horizontal wind, and water vapor, to guarantee the precision of large-scale meteorology during the simulations.

The selected physics physical configurations included Morrison double-moment microphysics scheme (Morrison et al., 2009), RRTMG (Rapid Radiative Transfer Model for GCMs (Global Climate Models)) long and short wave radiation scheme (Iacono et al., 2008), Grell 3D cumulus parameterization, Yonsei University planetary boundary layer scheme (Hong et al., 2006), and Noah land surface model. For chemistry and aerosol mechanism, the CBM-Z (Carbon Bond Mechanism; Zaveri, 1999) coupling with the 8-bin sectional MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) model with aqueous chemistry (Zaveri et al., 2008) was used. The MOSAIC, treating all the

important aerosol components, including nitrate, sulfate, ammonium, black carbon, and primary organic aerosols and other inorganic aerosols, is efficient without compromising accuracy and widely used in air quality and regional/global aerosol models (Zaveri et al., 2008). Since the MOSAIC is incapable of simulating secondary organic aerosols (SOAs), the simulated organic aerosols mentioned hereinafter all refers to primary organic aerosols. The crucial processes of radiation feedback, aerosol and cloud interaction, dry deposition, wet scavenging and cloud chemistry were turned on. Biogenic emissions were calculated online with~~by~~ the MEGAN ~~model~~ (Model of Emission of Gases and Aerosol from Nature) (Guenther et al., 2006). The initial and boundary chemistry conditions were based on the vertical profiles of O_3, SO_2, NO_2, 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~~the spin-up. The outmost domain of modeling tests was set large enough to cover East Asia to avoid the ~~impact~~influence 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, 2015) with ~~the~~ less contribution of foreign emission to haze pollution over Eastern China.~~Default initial and boundary chemistry profiles in the model were selected. Biogenic emissions were calculated online by the MEGAN model (Model of Emission of Gases and Aerosol from Nature) (Guenther et al., 2006).~~

~~December 1 – 31, 2013 were selected for simulation period, since severe haze pollution events occurred frequently in this month. To alleviate the effects of initial conditions, simulation of the first 46 hours as the spin-up was discarded.~~

2.2 Observational data

Meteorological fields simulated by ~~WRF~~ the model are crucial for the accuracy of air quality ~~simulation~~modeling. In the south, middle and north parts of Jiangsu P~~p~~rovince~~In this study~~, we selected

three prefecture-level cities of Nanjing, Yancheng and Lianyungang~~, located in the south, middle and north parts of Jiangsu province~~, respectively, to evaluate the overall perspective of meteorological simulation with the available observations. The observed meteorological data, ~~consisting of~~involving 2-m temperature, 2-m relative humidity and 10-m wind speed and direction was collected from the Jiangsu Provincial Meteorological Bureau and Meteorological Information Comprehensive Analysis and Process System (MICAPS) of China Meteorological Administration (CMA). Hourly surface concentrations of chemical constituents in 13 cities of Jiangsu, including SO_2, NO_2, $PM_{2.5}$, CO and O_3, were obtained from Jiangsu Environmental Protection Bureau. Daily secondary inorganic aerosols (SIAs; sulfate, nitrate, ammonium) in $PM_{2.5}$ were ~~observed~~ measured using MARGA (Online Analyzer of Monitoring of Aerosol and Gases) ~~in~~ at Jiangsu Environmental Monitoring Center~~ (Phoenix West Street, Nanjing)~~. MARGA is a fully autonomous sampling and measurement system that continuously measures the gases (HCl, HNO_3, HNO_2, SO_2, and NH_3) and aerosol components (CL^-, NO^{3-}, SO_4^{2-}, NH^{4+}, K^+, Ca^{2+}, and Mg^{2+}) by ion chromatography with internal standard eliminates calibration.

2.3 Air pollutant emission inventory

2.3.1 Two inventories for power plant emissions

This study utilized the MEIC inventory as the default anthropogenic emissions~~. This inventory included~~ including the emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), NH_3, black carbon (BC), organic carbon (OC), non-methane volatile organic compounds (NMVOCs), $PM_{2.5}$, and PM_{10} by five sectors of power, industry, transportation, residential, and agriculture.

The UEIPP in Jiangsu Province for the year of 2012~~, actually~~ consisting of six online species (SO_2, NO_x, $PM_{2.5}$, PM_{10}, BC and OC), was established ~~by Zhang et al. (2015)~~ using the online monitoring data ~~, which include daily concentrations~~ of three pollutants (SO_2, NO_x and total suspended particles (TSP)) and

volume of flue gases at unit and daily level (Zhang et al.,2015). Atmospheric verifiable accounting tables, comprising accurate locations, boiler type, coal consumption, and control policies for individual plant, were adopted to calculate the CO and NMVOCs emissions in UEIPP (Zhang et al.,2015), ~~by Zhang et al., (2016)~~.

The SO_2, NOx and TSP emissions were reckoned directly from online concentrations and volumes of flue gases as follows:

$$E_{i,j,k} = A_{i,j,k} \cdot c_{i,j,k}, \quad (1)$$

where $A_{i,j,k}$ is the daily emitted volume of flue gas, and $c_{i,j,k}$ is the daily concentration, with i, j, and k representing the pollutant species, individual plant, and day, respectively. The emissions of $PM_{2.5}$, PM_{10}, BC and OC were then calculated using the online TSP emissions:

$$E_{i,j,k} = T_{i,j,k} \cdot P_i, \quad (2)$$

where $T_{i,j,k}$ stands for the online TSP emissions; P_i represents the $PM_{2.5}$, PM_{10}, BC and OC mass ratios to TSP. The online monitoring system is currently incapable of providing the mass ratios at unit level and thus, it's given as a unified value referring to the work of Zhang et al. (2006) for each the four species, which was 52.7 % ($PM_{2.5}$), 80.4 % (PM_{10}), 8.6 % (BC), and 6.1 % (OC), respectively.

The annual emissions of CO and NMVOCs were calculated using Eq. (3):

$$E_{i,j,k} = A_{i,j,k} \cdot EF_{i,j,k} \cdot (1-\eta_{i,j,k}), \quad (3)$$

where $A_{i,j,k}$ is the activity level, $EF_{i,j,k}$ is the uncontrolled emission factor, and $\eta_{i,j,k}$ is the removal efficiency of air pollutant control device. In refer to previous studies (Wang et al., 2005; Streets et al., 2006; Bo et al., 2008; Huang et al., 2011), the $EF_{i,j,k}$ of CO and NMVOCs was set at 4.03 g kg^{-1} and

205 0.12 g kg^{-1} respectively, and $\eta_{i,j,k}$ was set to 0.

206 Following the method used by Li et al., (2014) and the mechanism-dependent mapping tables
207 developed by Cater (2013), the NMVOCs in UEIPP were specified to individual constituent in Regional
208 Acid Deposition Model chemical mechanism (RADM2, Stockwell et al., 1990), which could be adapted
209 to the WRF-Chem/CBMZ mechanism used in this study. The primary distinction, between UEIPP and the
210 power emission inventory estimated in previous China studies, lies in the different data used and
211 subsequently the estimation algorithm as well as the temporal resolution. Previously, power emission
212 inventory was mostly estimated using various data such as activity levels, boiler types, fuel types,
213 control policies and emission factors, and the activity levels were usually collected at annually or monthly
214 level. In the UEIPP, the emissions of SO_2, NO_x, $PM_{2.5}$, PM_{10}, BC, and OC were calculated using the
215 online pollutant concentrations and volume of flue gases at daily level. Rejection heights of the UEIPP
216 and the original power emission in MEIC were set at about 100 m and 200 m above ground, corresponding
217 to the second and third model levels for this WRF-Chem simulation.

218 **2.3.2 Differences between two power emission inventories**

219 The UEIPP~~,~~ and MEIC power emissions of major air pollutants with ~~and~~ their ~~share~~ fractions ~~of~~ in
220 the total emissions over Jiangsu Province in 2012 were presented in Table 1 for contrastive analysis.
221 Appreciable differences between the two power emission inventories were revealed. ~~Firstly, in the MEIC,~~
222 the power emissions of SO_2, $PM_{2.5}$, PM_{10} and NO_x in the MEIC are 367.8 kt, 72.2 kt, 103.7 kt, and 733.8
223 kt ~~respectively, and~~with ~~reduce~~ reducing to 105.6 kt, 21.6 kt, 32.6 kt and 277.9 kt~~,~~ respectively in the
224 UEIPP. The notable reductions of SO_2 and NO_x may largely due to comprehensive implementation of
225 FGD and SCR/SNCR in Jiangsu Province, which was not ~~promptly~~ captured in national inventory.
226 Application rate and average SO_2 removal efficiency of FGD in coal-fired power plants were obviously

higher than those in other sectors (Zhou et al., 2016), further confirmed by abrupt decrease of SO_2 ~~'s~~ power emission in China since 2006 (Liu et al., 2015). In addition, differences existed in estimation of NO_x removal efficiency of SCR/SNCR for Jiangsu's power plants in 2012 among different studies, reflecting 37 % (average of SCR/SNCR) calculated by Zhou et al. (2016) while 70 % (SCR) and 25 % (SNCR) calculated by Tian et al. (2013). In addition, due to inconsistent penetration rates and removal efficiencies of dust collectors determined at national and provincial levels, there also remained discrepancies in estimation of $PM_{2.5}$ and PM_{10} emissions (Xia et al., 2016; Zhou et al., 2016). However, the bias could be avoided in the UEIPP (see Section 2.3.1 for detail). The UEIPP produced higher CO (582.0 kt), BC (3.6 kt), OC (2.5 kt) and NMVOCs (17.3 kt) emissions compared to the MEIC. The power plants in the MEIC ~~produce~~ present the very low emissions of BC and OC, particularly for OC with 0.0 kt, resulting largely from the high uncertainties in the emission factor of these species (Zhao et al., 2015; Zhao et al., 2011; Zhou et al., 2016). ~~Secondly~~ Furthermore, in the MEIC ~~inventory, the~~ power emissions of SO_2, $PM_{2.5}$, PM_{10} and NO_x shared ~~larger (~~with the larger fractions of 28.8 %, 11.3 %, 11.6 % and 35.4 %, ~~respectively)~~ relatively to CO, BC, OC and NMVOCs ~~(~~with the fractions of 3.7 %, 0.2 %, 0.0 % and 0.4 %, ~~respectively) of~~in the total emissions (Table 1). When the UEIPP was introduced to MEIC by replacing the original power emission, ~~the shares changed. The~~ the UEIPP contributed 10.4 %, 3.7 %, 4.0 %, 17.2 %, 6.2 %, 4.3 %, 1.7 % and 0.9 %, to the total emissions of SO_2, $PM_{2.5}$, PM_{10}, NO_x, CO, BC, OC, and NMVOCs, respectively. ~~–~~The ratios of $PM_{2.5}$ (3.7 %) and PM_{10} (4.0 %) of the UEIPP were ~~approximate~~ comparable to the ratios of 4 % for $PM_{2.5}$ and 6 % for PM_{10} calculated ~~in~~ by Zhou et al. (2016).

~~Figure 2 shows the~~The spatial ~~distributions~~ difference of ~~the~~ two emission inventories ~~in 2012~~ over Jiangsu was shown in ~~f~~Figure 2, as well as their absolute ~~quantity~~values in ~~f~~Figure S1. ~~In most areas of Jiangsu Province, t~~The UEIPP presented the lower emissions of SO_2, $PM_{2.5}$, PM_{10} and NO_x in the most

areas ~~of the province~~ (Fig. 2a-d), and the high~~er~~ emissions of CO, BC, OC and NMVOCs in urban areas over the province (Fig. 2e-h). T~~he t~~wo inventories exhibited the similar spatial distribution patterns with large emissions in southland and low emissions in midland and northland (Fig. S1). The power plants around Xuzhou, an industrial city, formed a high emission center over the northwestern Jiangsu (Fig. S1). ~~In most areas of Jiangsu Province, the UEIPP presented lower emissions of SO_2, $PM_{2.5}$, PM_{10} and NO_x (Fig. 2a-d), and higher emissions of BC, OC and NMVOCs (Fig. 2f-h).~~

To assess the simulation performance ~~of~~ with the UEIPP and ~~characterize~~ changes of atmospheric environment ~~in~~ over Jiangsu Province under the updated emission condition~~s~~, two simulations with the original MEIC emission inventory (hereinafter referred as MOD1 simulation) and the updated MEIC emission inventory with the power generation replaced by UEIPP (hereinafter referred as MOD2 simulation) were carried out. The ~~chemical differences~~difference of chemical components between MOD1 and MOD2 simulations were used to assess atmospheric environmental changes in the following sections.

3 Modeling evaluation

3.1 Meteorological evaluation

An evaluation of the meteorological simulations ~~from~~over the ~~5 km~~ domain with 5 km horizontal resolution was carried out ~~with~~ in respects of temperature, relative humidity (RH), ~~and~~ wind speed~~s~~ and ~~wind~~ direction in Nanjing, Yancheng and Lianyungang ~~in southern, central and northern Jiangsu Province~~. The evaluation statistical parameters included mean bias (MB), correlation coefficient (R) and root mean square error (RMSE) (Table 2). The R and RMSE of temperature in the three cities ranged from 0.86 to 0.94 (p-values < 0.001) and 2.0 ^{o}C to 3.0 ^{o}C, showing a close agreement between the simulation and observations. MB values of temperature manifested a slight underestimate in Nanjing (-1.0 ^{o}C) and Lianyungang (-0.5 ^{o}C), and overestimate in Yancheng (1.1 ^{o}C). The R of RH was 0.79, 0.79 and 0.82

(p-values < 0.001), with the RMSE values of 14.6%, 14.4% and 18.2%, respectively in Nanjing, Yancheng and Lianyungang, which were comparable to previous studies (Gao et al., 2016; Liu et al., 2016). The MB of RH was positive in Nanjing and Lianyungang, but negative in Yancheng. ~~Although a slight overestimate, t~~The variations of wind speed were ~~well~~ generally captured by the model with the R varying from 0.51 to 0.77 (p-values < 0.001). ~~and the~~The RMSE ranging from 1.8 m s^{-1} to 2.1 m s^{-1}, basically conforming to the "good" model performance criteria for wind speed ~~prediction proposed by~~ (Less than 2.0 m s^{-1}; Emery et al., ~~(~~2001). Wind directions were evaluated via Hit Rates (HR; Schlünzen and Sokhi, 2008) with desired accuracy between $\pm 45^{o}$. The HR values were 63 %, 64 % and 49 %, respectively in Nanjing, Yancheng and Lianyungang, indicating that variations of wind direction were basically captured. ~~Overall,~~Generally, the meteorological fields in Jiangsu P~~p~~rovince were ~~well~~ reasonably reproduced by WRF-Chem during the simulation period.

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3.2 Chemical evaluation

The surface observations of $PM_{2.5}$, CO, NO_2, O_3 and SO_2 at 13 urban sites in Jiangsu (Fig. 1b) were collected for evaluating the chemical simulation over the domain with 5 km horizontal resolution ~~from the 5-km domain~~ in MOD1 and MOD2. Three evaluation statistical parameters of R, mean fractional bias (MFB) and mean fractional error (MFE) were presented in Table 3. MFB and MFE could normalize bias and error for simulated-observed pair ranging from -200% to 200%, and from 0% to 200% respectively, indicating their appropriateness to evaluate performance over a wide range of concentrations (Boylan and Russell, 2006). Normalized mean bias (NMB) and normalized mean error (NME) by individual site and air pollutant were additionally presented in Table S1 in the Supplement. As shown in Table 3, the values of MFB and MFE indicated that the hourly variations of $PM_{2.5}$, CO and NO_2 were reasonably captured ~~in~~ by both MOD1 and MOD2 simulations, conforming to the "satisfactory" criteria

proposed by Morris et al. (2005) that MFB is within ±60 % and MFE is below 75 %. Given a high dependence on emissions, the deviations of CO and NO_2 ~~seemed largely due to~~concentrations could be largely resulted from their emission uncertainties. The high~~er~~ R and the negative MFB of O_3 indicated the hourly variations were ~~well~~ reasonably captured but undervalued systematically, especially at night (Fig. 9b; Fig. ~~S1~~S2). The CBM-Z scheme and the outdated land-use data from United States Geological Survey (USGS) were prone to undervalue the surface O_3 concentrations ~~near surface, due to~~ in association with ~~produce~~ producing high NO-titration and dry deposition ~~rates~~, respectively (Balzarini et al., 2015; Park et al., 2014). Similar underestimations were previously simulated in Eastern China (Gao et al., 2015; Liao et al., 2015; Wang et al., 2016). The mean NMB and NME of O_3 in MOD1 ~~simulation, calculated~~ at -53.97 % and 67.00 % respectively (Table S1), were comparable to previous China studies (Li et al., 2012; Tang et al., 2015; Wang et al., 2016; Zhou et al., 2016), ~~and~~ while the mean NMB and NME of O_3 in MOD2 were ameliorated respectively to -45.83 % and 63.61 % ~~in MOD2 simulation, indicating the WRF-Chem modeling deviations of O_3 were acceptable~~. The SO_2 changes ~~was~~ were generally captured in the two simulations in terms of MFB and MFE, but with an overestimation and ~~lower~~ the low R~~, which may not purely due to uncertainty in emissions~~. In addition of emissions, absence of pathways converting SO_2 to sulfate in current WRF-Chem model, such as aqueous phase oxidation of dissolved $S_{(IV)}$ (the sum of hydrated SO_2 ($SO_2 \cdot H_2O$), bisulfite (HSO_3^-), and sulfite (SO_3^{2-})) by dissolved NO_2 under conditions of high ammonia (NH_3) and NO_2 concentrations (Huang et al., 2014b; Xue et al., 2016; He et al., 2014), was partially responsible for the simulation deviations of SO_2 and NO_2. Aerosols in East Asia ~~were often~~are ~~documented~~featured with low acidity due to the high NH_3 and mineral dust emissions ~~there (Cheng et al., 2016), and are expected to~~ capture~~ing~~ more acidic gases (SO_2 and NO_x) under ~~high RH~~moist air conditions ~~to strengthen severe~~duri~~i~~ng haze ~~pollution~~episodes (Cheng et al., 2016; Wang et al., 2016). The modeled SIAs at Nanjing site (Fig. 1b) were assessed in addition. As can be seen from Table 4 and

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Fig. S3~~2~~a, the simulated sulfate concentrations were obviously underestimated, providing a further evidence for ~~the~~ the above-mentioned speculation~~ above~~. Similar ~~–~~underestimation of sulfate was also ~~modeled~~ found in North China Plain ~~(by~~ Gao et al.~~(~~, 2016a~~,~~, 2016b). The observed nitrate and ammonium concentrations were comparatively well caught, particularly the NMBs of nitrate ranged within ± 20 % in the two simulations. ~~Thus, in~~ In general, ~~, the~~the chemical observations were ~~two simulations were reasonable~~ reasonably ~~compared with the observation data~~captured by ~~the two~~both simulations of MOD1 and MOD2.

The R, MFB and MFE in the MOD1 and MOD2 simulations were ~~compared~~ presented in Table 3 ~~to give~~with an overall assessment of simulation with the UEIPP. A better simulation performance ~~would be~~is reflected by higher R, smaller absolute value of MFB and ~~smaller~~ lower MFE, respectively tagged with upward arrows in Table 3. Additionally, significance of the improvements between the statistical indices was checked via using the method of bootstrap confidence interval (DiCiccio and Efron, 1996; He et al., 2017). In response to the introduction of UEIPP, chemical simulation showed a comprehensive improvement in MOD2 (see the upward arrows in Table 3). ~~The underestimation of $PM_{2.5}$, CO and O_3, and overestimation of NO_2 and SO_2 were simultaneously diminished in terms of MFB, respectively by 0.07, 0.21, 10.78, 3.6 and 8.26 percentage points in MOD2, where the improvements of modeling NO_2, O_3 and SO_2 were more remarkable by the changes of R and MFE. MOD2 simulations show overall improvement for all species compared to MOD1 results.~~ Although ~~the~~ both MOD1 and MOD2 underestimated $PM_{2.5}$, CO, O_3 and overestimated NO_2 and SO_2 as shown in Table 3, ~~absolute~~the MFBs for those species ~~are~~ reduced by 0.07, 0.21, 10.78, 3.6, and 8.26 % ~~percent~~ respectively from MOD1 to MOD2 ~~(see the up arrows in Table 3)~~. The improvements of O_3, SO_2 and NO_2 were statistically significant ~~(see asterisks in~~ Table 3). For the O_3 simulation, the improvements of the ~~three statistics (~~R, MFB and MFE~~)~~ ~~were~~ significantly passed ~~wi~~the confidence level ~~at~~of 99 %. For the SO_2 simulation, the

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improvements of MFB and MFE were significant withpassed the confidence levels atof 99 % and 90 %, respectively. Improvement in MFB of NO_2 was significant at 95 % confidence level. The reason, towards primary pollutants, was the concentrations of NO2, SO2 and CO were prone to be determined by emissions under similar meteorology and thus, the deviations of NO2 and SO2 in MOD2 were mitigated more obviously due to the larger emission changes in the UEIPP (aforementioned in Section 2.3.2). In respect of secondary formation, it is difficult to explicitly characterize the evolution of PM2.5 and O3 through perspective of emission changes, yet their simulations in MOD2 were improved as well. The improvement of PM2.5 was seemingly limited when using the UEIPP, making it hardly to affirm it's attributed to the emission improvement rather than accidental error of WRF-Chem model. However, as can be seen from Table 4, the SIAs in PM2.5 at Nanjing site were ameliorated, especially the nitrate and ammonium with the NMBs changed from -19.47 % to -16.38 %, and from -55.12 % to -53.47 % respectively. A logical inference was that the UEIPP provided a more realistic power emission, at least around NanjingAlso, the modleeledd 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 shcould be attributed to the 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 theiir changes in conjunction withof chemical conversio variations between the twoin in the simulations of MOD1 and MOD2, which was more comprehensively investigated in Section 4.

Spatially,The spatial patterns in MOD1 and MOD2 simulations as well as their differences (Fig. 3) provided the further evidences for the improvement in MOD2 simulation, which could be confirmed with the improvement in the difference between observation and simulation in MOD2 in Fig. 3. The overestimates of SO_2 in MOD1 were mainly occurred in south urban areas areas and the vicinity of Xuzhou (Fig. 3a), where the SO_2 overestimates were mostly improved in MOD2 (Fig. 3k, f). For NO_2

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simulation, the overestimates lay in the majority cities throughout Jiangsu Pprovince with a few cities underestimated such as Suzhou (Fig. 3b), and were mitigated correspondingly in MOD2 as well (Fig. 3l, g). As a common feature of MOD1 simulation, CO, $PM_{2.5}$ and O_3 were undervalued throughout the province most city sites (Fig. 3c-e), while in response to the usage of UEIPP, their concentrations in MOD2 were comprehensively improved (Fig. 3m-o, h-j).

As above mentioned above, due to the introduction of UEIPP, the MOD2 with the introduction of UEIPP–, improved the simulation of air pollutants, especially of O_3, SO_2, and NO_2 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. especially of O_3, SO_2, and NO_2 according to model performance evaluation. It is therefore concluded that the power plant emissions fromof UEIPP is more realistic and reliable.

4 Environment changes under updated emission conditionwith two emission inventories

4.1 Influence of emission changes on air pollutant modeling

Aside from estimating introducing more accuratethe updated emission inventories, another important and meaningful work in this study is to explore how the emission changes affect the atmospheric environment especially in severe pollution haze episodes for better understanding the complexity of atmospheric environment. To this end, we presented the differences of some otheratmospheric compositions simulated in with MOD1 and MOD2 in Figure 4. Consistently with the emission changes (Fig. 2), the concentrations wereis reduced for SO_2 and NO_2 (Figs. 3k and 3l), and enhanced for OC and BCBC and OC (Figs. 4c and 3d4d) in the most areas. However, it's not clear yet what was responsible for the enhanced O_3 concentration in MOD2, and why the $PM_{2.5}$ concentration enhanced when the $PM_{2.5}$ primary emission largely reduced, which were urgently needed to address especially for the abnormal

385 increase of $PM_{2.5}$, since more implemented restrictions on power plants are being executed in East China.

386 As a secondary air pollutant in the boundary layer, O_3 is highly subjected to its precursors~~, solar~~
387 ~~radiation and process of planet boundary layer~~ (Ou Yang et al., 2012; Gao et al., 2005). The Yangtze
388 River Delta was characterized of VOC-limited, especially in winter, indicating O_3 concentrations were
389 depressed by NO_x and sensitive to VOC (Liu et al., 2010;Wang et al., 2008; Tie et al., 2013). The ratio
390 of $HCHO/NO_y$, an indicator to differentiate VOC-limited ($HCHO/NO_y < 0.28$) and NO_x-limited
391 ($HCHO/NO_y > 0.28$) conditions, verified the Jiangsu Province as the VOC-limited region during the
392 modeling period (Fig. 6). Therefore, either the increase of VOC or the ~~reduction~~ decrease of NO_x could
393 enhance the surface O_3 level. Coincidentally, the concentrations of VOC and NO_2 increased and
394 decreased respectively (Fig. 4h and 3l), following their emission changes in UEIPP (Table 1)~~ the lower~~
395 ~~NO_x emissions and the stronger VOC emissions are found in the UEIPP (Table 1Fig. 2)~~. In addition, a
396 high anti-correlation relationship ~~exists~~ existed between the spatial ~~difference patterns~~changes of O_3 and
397 NO_2 (Figs. 3l and 3o) as well as the diurnal ~~difference pattems~~changes (Figs. 9a-b). Therefore, we could
398 partially attribute the underestimated O_3 to the emission ~~overestimation of~~uncertainties of VOC and NO_2
399 in the original MEIC~~,~~. Furthermore, as a precursor of O_3, high CO concentrations in MOD2 with the
400 updated MEIC emission inventory would contribute to the enhancement of O_3 concentration as well.~~and~~
401 ~~the O_3 simulation was improved by the UEIPP, which could indicating reflect the complexity of air quality~~
402 ~~control in this region.~~

403 Quite surprising to us, the surface $PM_{2.5}$ concentrations ~~(Fig. 3n)~~ didn't follow the reducing
404 emissions of primary $PM_{2.5}$, but increased over almost all the province (Fig. 3n). As $PM_{2.5}$ is highly
405 dependent on three factors of primary emissions, physical processes and chemical reactions in the
406 atmosphere, the latter two factors were more likely to dominate in the simulation of ~~increasing~~ increased

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PM$_{2.5}$ concentrations. ~~It was deemed that the interesting phenomenon would be induced by some physical processes.~~ Due to the strong absorbing effects of BC to solar radiation and the higher rejection height of power plants, as ~~(~~the chimney height of new constructed coal-consuming power plants of larger than 300 MW is higher than 200 m according to ~~–~~the national standard ~~Environmental Protection Agency (EPA) in China~~ ~~Ministry of Environmental Protection of the People's Republic~~ of China; ~~China Ministry of Environmental Protection;~~, and ~~–~~the rejection height was set at two model levels of 100 m and 200 m in this study~~)~~, the enhancement of BC concentrations (Fig. 4c~~f~~; Table 5) would ~~result in an elevated warmer layer in the atmosphere~~reduce solar radiation to ground, and thus suppressing vertical diffusion ~~below it~~ and accumulating more pollutants near surface. The speculation could be verified by ~~changes~~ reductions of downward short wave flux at ground surface (SWDOWN) ~~and~~, 2 m air temperature ~~as well as~~and boundary layer height (BLH) ~~could be also used to interpret the speculation~~. Regional averaged over Jiangsu Province during Dec. 1-31, ~~The~~ SWDOWN, 2 m temperature and BLH reduced by ~~reduced~~ 0.65 W m^{-2}, ~~2 m air temperature decreased about~~ 0.005 °C~~K,~~ and ~~BLH reduced~~ 0.4 m, respectively, and the reductions became more significant to 11.8 W m^{-2}, 0.3 °C and 26.4 m respectively, during the daytime of Dec. 7, 2013 in Wuxi, a haze center~~,~~, ~~averaged over the province in MOD2 simulation, which revealed the increased~~ increasing air stability for more air pollutant accumulation. In order to quantify the radiative effects induced by BC emission change, a sensitivity test MODa as same as MOD2 with closing BC emission in UEIPP was performed. Based on the PM$_{2.5}$ differences between MOD2 and MODa regionally averaged over Jiangsu Province, it was estimated that the ~~physical process of~~ BC aerosol radiative effect stabilizing boundary layer contributed only about 0.15 μg m^{-3} to the PM$_{2.5}$ enhancements, during the haze episode.

4.2 Reinforcing atmospheric oxidation capacity and enhancing secondary inorganic aerosols

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As descripted in Sect 4.1, ~~the declined emissions of primary $PM_{2.5}$ could not enhance the ambient $PM_{2.5}$ concentrations~~ the declined emissions of primary $PM_{2.5}$ could not improve the ambient $PM_{2.5}$ concentration and the feedback aerosol radiative effects ~~stabilizing boundary layer~~on aerosol change were ~~a little~~ weak, implying the contribution of chemical production to the ~~ambient~~ $PM_{2.5}$ enhancement. In this section, ~~We here studied~~ the chemical production of SIAs was analyzed, since the CBM-Z/MOSAIC used in the WRF-Chem is incapable of simulating SOA~~s with missing pathways of SOAs~~ formation ~~in the WRF-Chem~~. Previous studies had revealed that SIAs played an important role in $PM_{2.5}$, particularly in the haze pollution over Eastern China (Huang et al., 2014a; Wang et al., 2014; Gao et al., 2016). Given the reduction of SO_2 and NO_x emissions in the UEIPP, the simulated sulfate and nitrate should be lower from the oxidation of SO_2 and NO_2. However, as shown in Fig. 4, both sulfate and nitrate are increased in MOD2, with more significantly during haze episode (Dec.3-8; Fig. 5). In the atmosphere, sulfate is formed through oxidation of SO_2 by gas-phase reactions with OH (Stockwell and Calvert, 1983; Blitz et al., 2003) and stabilized Criegee intermediate ~~(which is~~ formed by O_3 and alkenes~~)~~ (Mauldin III et al., 2012) as well as by heterogeneous reactions with H_2O_2, O_3, OH, organic peroxides, and various oxides of nitrogen in clouds (Seinfeld and Pandis, 2012). Nitrate is mostly formed from the gas-phase reactions of NO_2 with OH during daylight and heterogeneous reactions of nitrate radical (NO_3) at night. Therefore, the formations of secondary sulfate and nitrate depend not only on their precursors, but also on oxidizing capacity of atmosphere.

As~~In this section, the variations of O_3,~~ ~~a~~ major oxidizing agents in atmosphere, O_3 and OH were increased from MOD1 to MOD2 (Fig. 3o and 4b), ~~were employed to evaluate the changes~~indicating the enhanced ~~of atmospheric~~ oxidizing capacity ~~between~~ in ~~MOD1~~ MOD2 ~~and MOD2~~relatively to MOD1 ~~.~~ ~~Figure 6 showed the ratios of surface HCHO/NO_y in Jiangsu province averaged over the simulation. The ratio of HCHO/NO_y concentrations increased from 0.039 in MOD1 to 0.047 in MOD2 when using the~~

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~~UEIPP with lower NO_x and higher VOC emissions in the UEIPP.~~] ~~The atmospheric environment in VOC-limited condition indicates that the MOD2 condition could level up O_3 concentration. Additionally, as a precursor of O_3, high CO concentrations in MOD2 would contribute to the build-up of O_3 as well. The OH radical concentrations were also increased (Fig. 4b) because of~~with its high dependence on O_3, ~~thus resulting in enhanced oxidizing capacity of the atmosphere~~ in Jiangsu region. A WRF-Chem/RADM2 simulation was performed as well because ~~As~~ changes in OH and VOC oxidation in the presence of NO_x are sensitive to chemistry mechanisms (Derwent, 2017; Knote et al., 2015; Stockwell et al., 2011; Jimenez et al., 2003)~~, another couple runs using chemistry mechanism RADM2 was performed being otherwise the same as the runs of CBM-Z~~. The ~~S~~similar ~~response to the emission change from MEIC to UEIPP was found that~~~~increased~~pattern of increasing O_3 and OH were ~~both increased~~found over the province (Fig. S~~4~~4), which could further ~~validate the analysis of enhanced oxidizing capacity~~ indicate the enhanced oxidizing capacity.

To evaluate how the formation of secondary aerosols responded to the enhanced oxidizing capacity, we analyzed the BC-scaled concentrations for sulfate and nitrate. The purpose was to eliminate the influence of air pollutant dilution and mixing in atmospheric physical process. Since BC is quite inertial to chemical reactions, its variations could well reflect the atmospheric physical processes. Thus, the BC-scaled concentration ~~will~~ could better represent the contribution of chemical reaction (Zheng et al., 2015). Figure 7 presents the daily averaged variation of BC-scaled concentrations for sulfate and nitrate, appending the differences of O_3 as an indicator for the change of atmospheric oxidizing capacity. As revealed in~~can be seen from~~ Fig. 7, the enhancements of chemical production simulated in MOD2 ~~was~~ were consistent well with the variations of O_3 difference between MOD2 and MOD1. During the haze episode ~~of~~over Dec. 3-8, 2013 the chemical production of sulfate and nitrate enhanced obviously, which ~~exactly~~ in accordance with the rapid build-up of O_3, ~~which indicated~~ indicating the chemical production is

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intensified by strengthen of oxidizing capacity during the episode.

The SOR (molar ratio of sulfate to sum of sulfate and SO_2) and NOR (molar ratio of nitrate to sum of nitrate and NO_2) were used as indicators of secondary transformation (Sun et al., 2006), since the BC-scaled concentrations just represent the intensity of chemical reaction effects with overlooking the precursors for individual compounds. The SOR and NOR would give insights to the chemical transformation of SO_2 and NO_2. As shown in Figure 8, aside frombesides in the haze period of Dec. 3-8, 2013, the chemical transformations from SO_2 and NO_x to sulfate and nitrate were always strengthened during the whole month in MOD2. That, that could be why the chemical production of sulfate and nitrate in MOD2 increased (Figs. 7a-b) even with less precursor concentrations. Additionally, in response to the enhanced atmospheric oxidizing agents, the secondary ammonium was also increased (Fig. 6 4f) under the same NH_3 emission conditions in MOD1 and MOD2.

As shown in Table 5, the total concentration of sulfate, nitrate and ammonium increased by 1.32 μμg m^{-3} during the whole month (of Dec.1-31), 2013 and even reached up to 4.77 μμg m^{-3} in the haze episode of Dec. 3-8, 2013, higher than the increment of $PM_{2.5}$ as well as the total increment of BC and OC, which could clearly reveal that the enhancement of SIAs in response to the reinforced atmospheric oxidizing capacity contributed the majority to the increased $PM_{2.5}$ concentrations. This conclusion could bewas verified by an emission sensitivity study in the North China Plain performed by Wang et al., (2016), who found that the 30 % emission reduction of NO_x led to a notable increase in $PM_{2.5}$ concentrations contributing to NH_3-rich and VOC-limited conditions in the winter there.

It should interpreted the larger reasonlargerenhancement in concentrations of SIAs greater than that of the $PM_{2.5}$ in in Table 51. cCompared to the MOD1, the lower emission of primary $PM_{2.5}$ in UEIPP (Table 1) lead to the less concentrations of primary $PM_{2.5}$ in MOD2, such thatand the enhancement

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in concentrations of SIAs was partially offset by the lower primary concentrations of $PM_{2.5}$.

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5 Conclusions

Power plant, as a major air ~~pollution~~ pollutant source in China, had been imposed restrictions by the government in response to the increasing air pollution, which led power plant emissions to large variations during ~~the~~ past few years. Due to various underlying data and approaches, there remained uncertainties in estimating the power plant emission ~~inventorys~~. In the present study, the UEIPP in Jiangsu Pp~~r~~ovince for 2012 was introduced in the MEIC emission inventory as the major point sources ~~of emissions~~. The variation and complexity of atmospheric environment in response to the change of power plant emissions over Jiangsu were studied, by executing ~~two~~ the WRF-Chem simulations using the original emissions of MEIC and the MEIC with its power emission inventory updated by the UEIPP.

The study revealed the uncertainty ~~and complexity~~ in estimating the power plant emissions ~~due to various data and strategies, such as implementation of FGD, SCR/SNCR and dust collectors over recent years in East China~~. In the UEIPP, the emission amounts of SO_2, $PM_{2.5}$, PM_{10}, NO_x, CO, BC, OC and NMVOCs were 105.6 kt, 21.6 kt, 32.6 kt, 277.9 kt, 582.0 kt, 3.6 kt, 2.5 kt, and 17.3 kt, respectively, manifesting obvious difference with the MEIC emission inventory. ~~The reduction of SO_2, NO_2, $PM_{2.5}$ and PM_{10} in the UEIPP quantified the effects of FGD, SCR/SNCR and dust collectors, which partially omitted in national works.~~

The UEIPP drove the simulation performance superior to the original power emission of MEIC inventory in terms of the proximity between simulated and observed air pollutant concentrations, suggesting a more realistic power emission inventory ~~was provided~~. The complexity of atmospheric environment ~~and difficulty in policy making to protect air quality~~ were revealed as well, through comparing the changes of various primary and secondary compositions in atmosphere. Atmospheric

oxidizing capacity was reinforced in response to the enhancement of O_3 and OH, which was largely due to higher VOC emissions and lower NO_x ~~and higher VOC emissions~~ in the UEIPP ~~and the VOC-dominated conditions~~. $PM_{2.5}$ increased almost all over the ~~province~~ Jiangsu Province even the primary emission reduced by 7.6%. This anomaly phenomenon was mainly attributed to the enhanced formation of SIAs, induced by the enhanced atmospheric oxidizing capacity, revealing the complex mechanism of air pollution from fine particulate matter to atmospheric oxidants~~.~~. Reduction of SO_2 may free NH_3 to react instead with NO_x creating ammonium nitrate particles, which would need further studying.

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Our study also quantified the ~~revealed the reaction of physical processes~~$PM_{2.5}$ enhancement ~~in~~ response to ~~the chemical changes~~ BC radiative effect stabilizing boundary layer~~when using UEIPP as core-fired plant emission inventory~~through investigating the changes in surface solar radiation, ~~temperature and boundary layer height. However, comparison indicated~~ The chemical reaction was more dominant ~~than the BC radiative effect~~ in $PM_{2.5}$ enhancement than the BC radiative effect.~~.~~

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Given the complicated processes in environmental change, the restrictions of emissions should be comprehensively considered rather than one single factor. Furthermore, the effects of emission inventories on ~~seasonal variation in~~ air quality variations could be assessed based on long-term observation and simulation studies, and formation of SOAs would be also enhance due to the reinforced atmospheric oxidizing capacity and higher VOC emissions, which needs to be addressed in future studies.

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Table 1 The UEIPP, and MEIC's power emissions of major air pollutants inventory and as well as their ratio in total emission inventory over Jiangsu Province in 2012.

	UEIPP		MEIC power emission	
	Emission (kt year^{-1})	Ratio in total (%)	Emission (kt year^{-1})	Ratio in total (%)
SO_2	105.6	10.4	367.8	28.8
$PM_{2.5}$	21.6	3.7	72.2	11.3
PM_{10}	32.6	4.0	103.7	11.6
NO_x	277.9	17.2	733.8	35.4
CO	582.0	6.2	343.5	3.7
BC	3.6	4.3	0.1	0.2
OC	2.5	1.7	0.0	0.0
NMVOCs	17.3	0.9	7.2	0.4

Table 2 Statistics between observed and simulated meteorology.

	Nanjing					Yancheng					Lianyungang				
	Obs.	Mod.	MB	R	RMSE	Obs.	Mod.	MB	R	RMSE	Obs.	Mod.	MB	R	RMSE
T (℃)	4.8	3.8	-1.0	0.86	3.0	4.4	5.5	1.1	0.94	2.0	3.4	2.9	-0.5	0.90	2.3
RH (%)	63.0	63.6	0.6	0.79	14.6	64.3	58.6	-5.7	0.79	14.4	56.4	58.2	1.8	0.82	18.2
WS (m s^{-1})	2.0	3.1	1.1	0.51	1.8	3.0	4.3	1.3	0.77	2.0	2.0	3.8	1.4	0.56	2.1

* T: temperature at 2m; RH: relatively humidity at 2m; WS: wind speed at 10m; MB: mean bias; R: correlation coefficient; RMSE: root mean square error; R values were labeled green when p-values less than 0.001 R values passing through the significant level of 0.0501 were labeled green.

Table 3 Statistics variables between observed and simulated $PM_{2.5}$, CO, NO_2, O_3 and SO_2.

	R		MFB (%)		MFE (%)	
	MOD1	MOD2	MOD1	MOD2	MOD1	MOD2
$PM_{2.5}$	0.568	0.571 ↑	-8.31	-8.24 ↑	45.40	45.65
CO	0.516	0.515	-36.05	-35.84 ↑	52.05	52.10
NO_2	0.456	0.466 ↑	14.08	10.48 ↑ **	39.37	38.77 ↑
O_3	0.600	0.625 ↑ ***	-121.46	-110.68 ↑ ***	131.74	124.67 ↑ ***
SO_2	0.260	0.261 ↑	24.88	16.62 ↑ ***	65.18	63.20 ↑ *

* R values passing through the significant level of 0.05 were labeled green when p-values less than 0.001; Up arrows indicate the chemical simulation results in MOD2 are better improved; "***", "**" and "*" indicates the improvements are statistically significant with confidence level at 99 %, 95 % and 90%, respectively than those in MOD1. Equations of R, MFB and MFE were presented in Supplement.; R, MFB and MFE are calculated using the following equations (P and O are simulation and observation, respectively; N is the number of measurements, and M is the number of monitoring sites):

$$R = \frac{1}{M}\sum_{j=1}^{M}\frac{\sum_{i=1}^{N}(P_{ij}-\overline{P_j})(O_{ij}-\overline{O_j})}{\sqrt{\sum_{i=1}^{N}(P_{ij}-\overline{P_j})^2}\sqrt{\sum_{i=1}^{N}(O_{ij}-\overline{O_j})^2}}$$

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$$MFB = \frac{1}{N \cdot M} \sum_{i=1}^{N} \sum_{j=1}^{M} \left(2 \cdot \frac{P_{ij} - O_{ij}}{P_{ij} + O_{ij}}\right) \cdot 100\%; \quad MFB = \frac{1}{N \cdot M} \sum_{i=1}^{N} \sum_{j=1}^{M} \left| 2 \cdot \frac{P_{ij} - O_{ij}}{P_{ij} + O_{ij}} \right| \cdot 100\%.$$

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Table 4 ~~Performance~~Validation ~~-~~statistics of SIAs in $PM_{2.5}$ at Nanjing~~site~~.

	NMB (%)		NME (%)	
	MOD1	MOD2	MOD1	MOD2
Sulfate	-87.61	-87.29 ↑	87.61	87.29 ↑
Nitate	-19.47	-16.38 ↑	29.06	28.61 ↑
Ammonium	-55.12	-53.47 ↑	55.12	53.47 ↑

* Up arrows indicate the same meaning as in Table 3. Equations of NMB and NME were presented in Supplement.~~NMB and NME are calculated using the following equations:~~

~~$$NMB = \frac{\sum_{i=1}^{N}(P_i - O_i)}{\sum_{i}^{N} O_i} \cdot 100\%; \quad NME = \frac{\sum_{i=1}^{N}|P_i - O_i|}{\sum_{i}^{N} O_i} \cdot 100\%.$$~~

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Table 5 Increased concentrations (~~u~~g m^{-3}) ~~and percentages for the (1)~~of $PM_{2.5}$, ~~(2)~~ total of BC and OC, and ~~(3)~~ total of SOA (sulfate, nitrate and ammonium) from MOD1 to MOD2~~,~~ averaged over Jiangsu P~~p~~rovince with the percentage ~~the relative~~ differences ~~in percentages~~ between MOD1 and MOD2 compared with ~~the~~ MOD1~~2~~ ~~simulations. in MOD2 simulation from that in MOD1 simulation.~~

	$PM_{2.5}$	BC+OC	Sulfate+Nitrate+Ammonium
Hazy days (Dec.3-8)	3.38 (1.81 %)	1.04 (3.06 %)	4.77 (5.11 %)
The whole (Dec.1-31)	1.03 (0.83 %)	0.58 (2.38 %)	1.32 (3.96 %)

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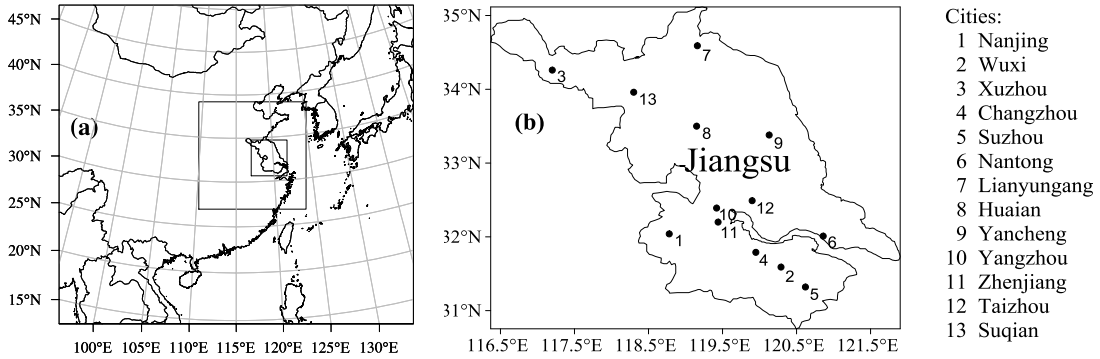
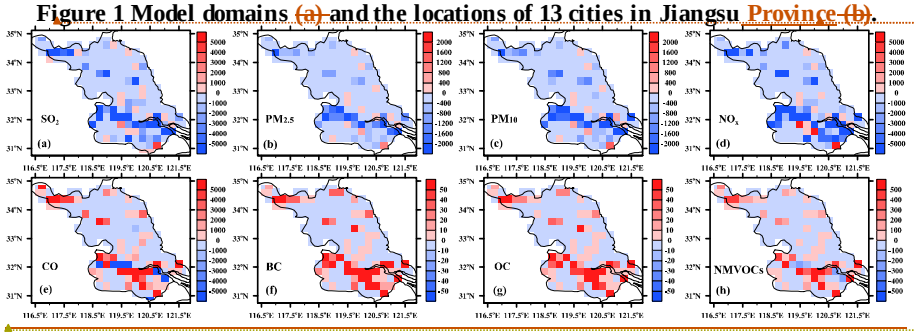


Figure 1 Model domains ~~(a)~~ and the locations of 13 cities in Jiangsu Province ~~(b)~~.



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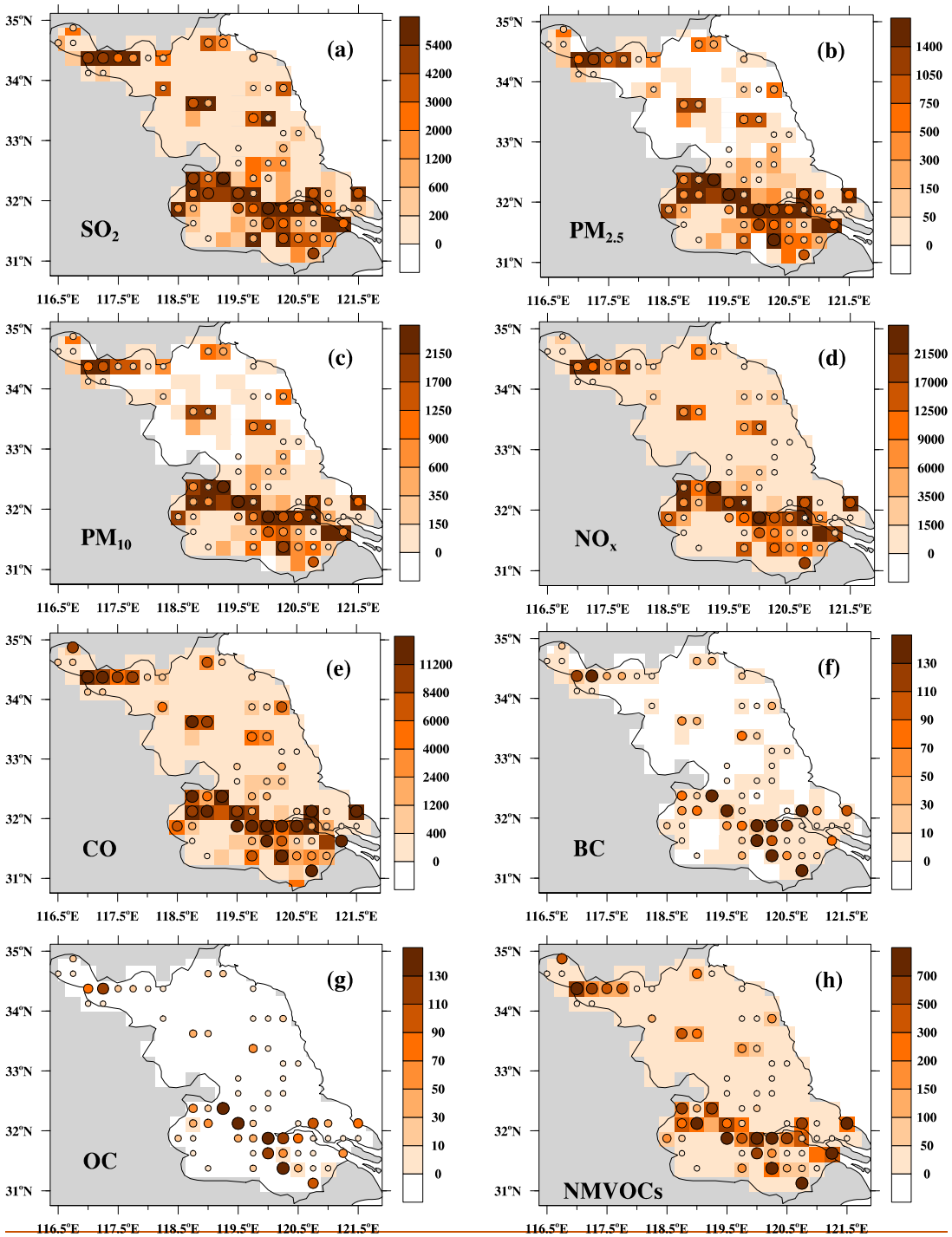

Figure 2 Differences of pPower plant emission ~~differences~~ between MEIC and UEIPP in 2012 (~~UEIPP – MEIC;~~ units: tons).~~Spatial distributions of power plant emissions of MEIC (shaded grids) and the UEIPP (shaded circles) in 2012 (unit: tons); the UEIPP has been mapped into 0.25°×0.25° grids, consistent with the spatial resolution of MEIC.~~

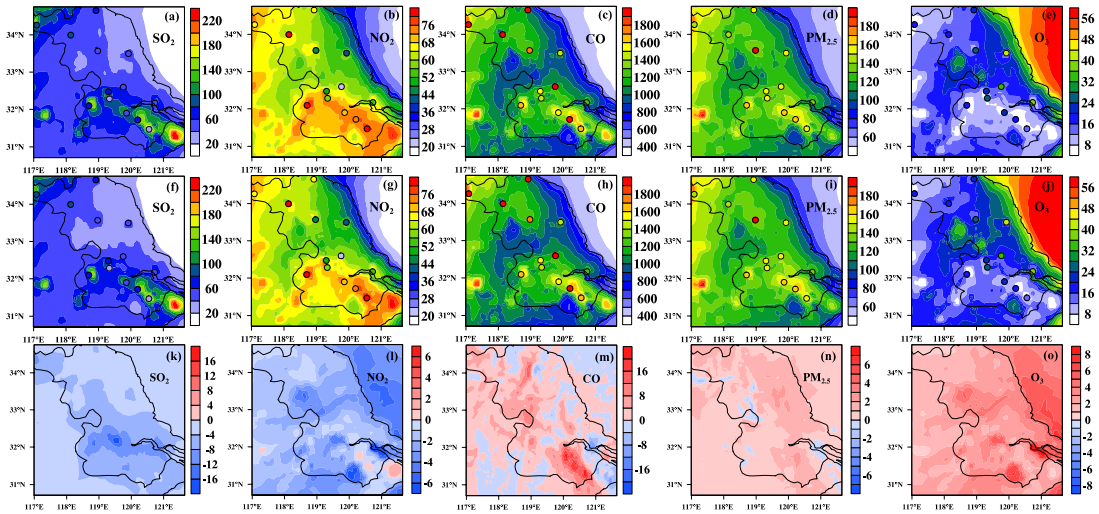

Figure 3 The spatial distributions of near-surface SO_2, NO_2, CO, $PM_{2.5}$ and O_3 ~~mean~~ concentrations (μg m^{-3}) from ~~MOD1~~ (a-e) MOD1, ~~MOD2~~ (f-j) MOD2 and (k-o) the~~ir~~ differences ~~(~~between MOD2~~-~~ and MOD1~~1~~ ~~;~~ k-o) in~~ averaged over December 2013~~;~~. ~~t~~The observed ~~mean~~ concentrations were indicated by shaded circles. ~~Unit: ug m^{-3}~~.

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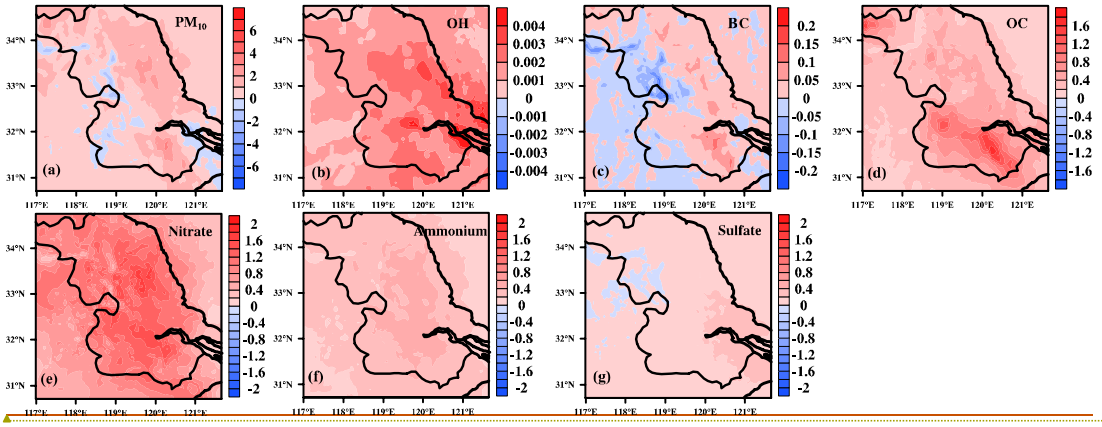

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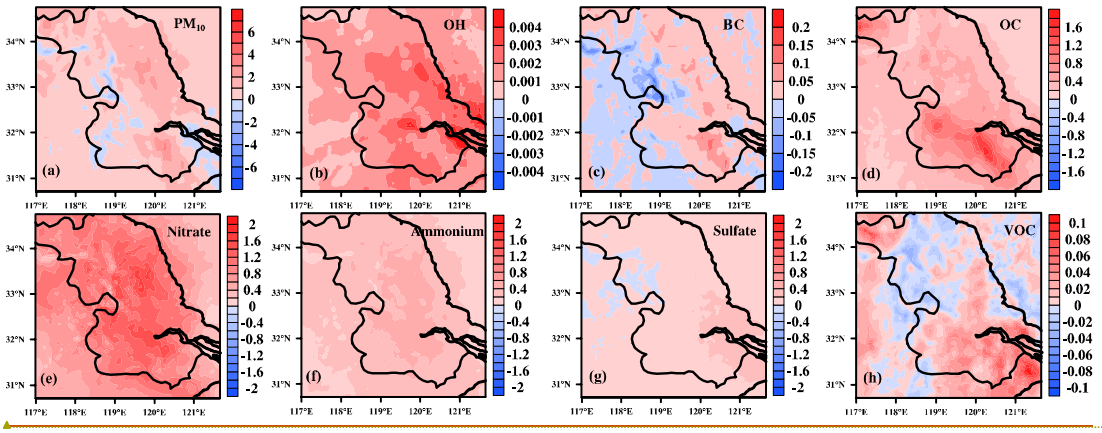

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Figure 4 Differences of ~~chemical constituents~~chemical species between MOD2 and MOD1 ~~–~~in December

2013 ~~(MOD2 – MOD1)~~. Unit: "pptv" for OH; "ppmv" for VOC; "~~μ~~g m^{-3}"for the others.

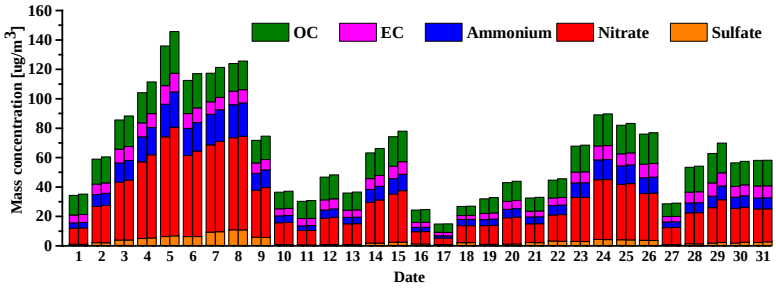

Figure 5 Chemical species of $PM_{2.5}$ simulated in MOD1 (left column) and MOD2 (right column) in December 2013

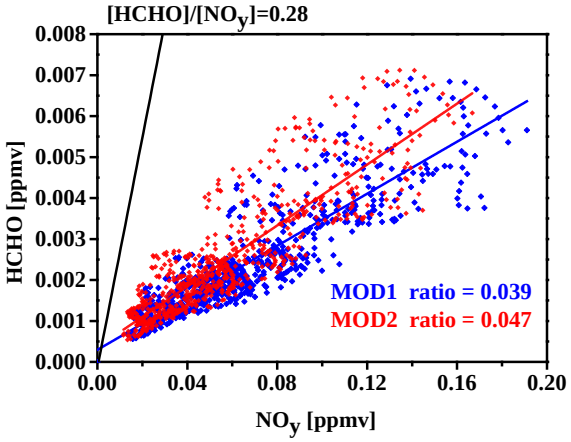

Figure 6 Ratios of $HCHO/NO_y$ simulated in MOD1 (blue) and MOD2 (red~~);~~). ~~(the threshold ratio of VOC-limited and NO_x-limited is 0.28.)~~

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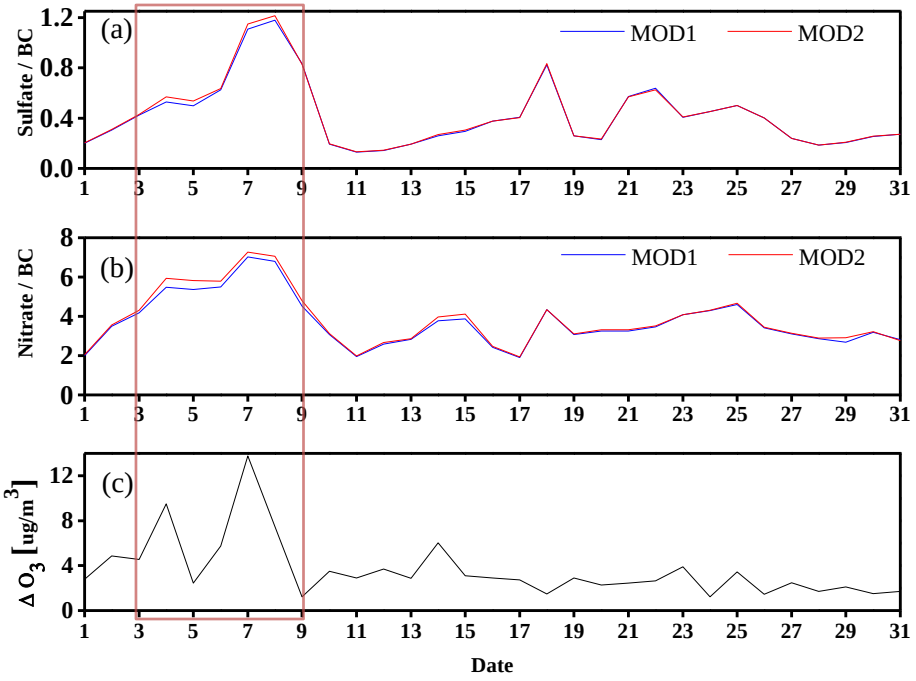

Figure 7 Daily variations of (a) sulfate/BC ~~(a)~~, (b) nitrate/BC ~~(b)~~ and (c) difference of O_3 (~~(c);~~ MOD2 - MOD1) averaged over Jiangsu with the ~~two~~ red rectangular column~~s~~ marking ~~two~~ the severe haze episode (Dec. 3-8)~~s~~; the increase of sulfate/BC and nitrate/BC suggests enhanced chemical productions.

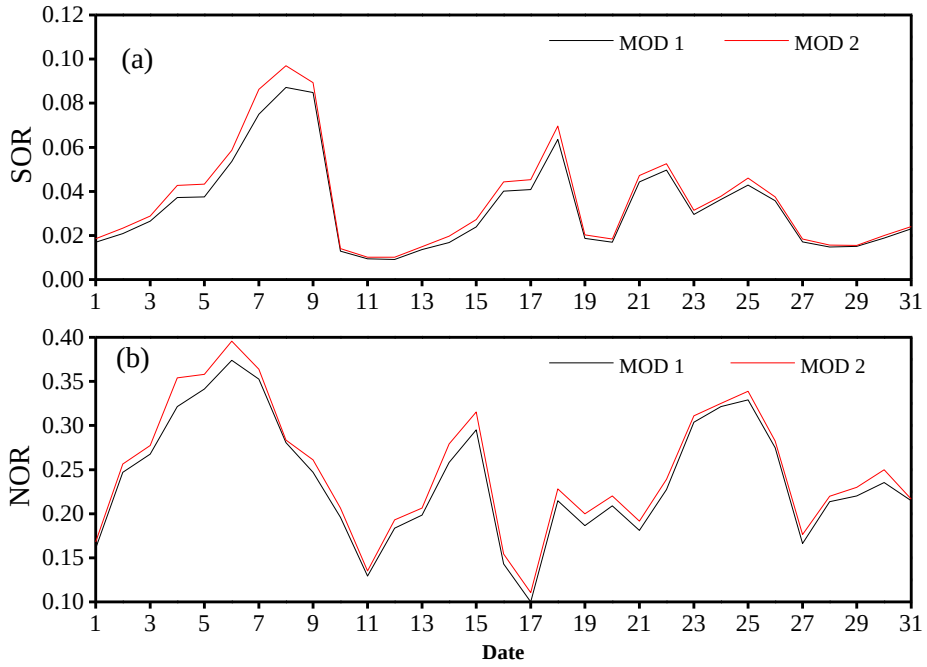

Figure 8 Daily variations of (a) SOR ~~(a)~~ and (b) NOR ~~(b)~~.

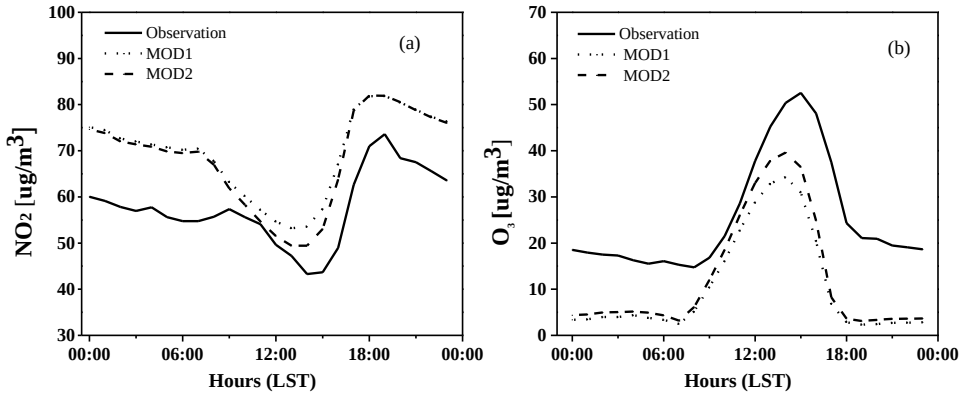

Figure 9 Diurnal variations of (a) NO_2 ~~(a)~~ and (b) O_3 ~~(b)~~ averaged over 13 cities in Jiangsu (Fig. 1b)

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