

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° 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 O₃ variations were reasonably captured".

2. In section 4.1, how great, a little more quantitatively, of the BC radiative effects on the surface PM_{2.5} 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 PM_{2.5} 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 quantitatively 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 through 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

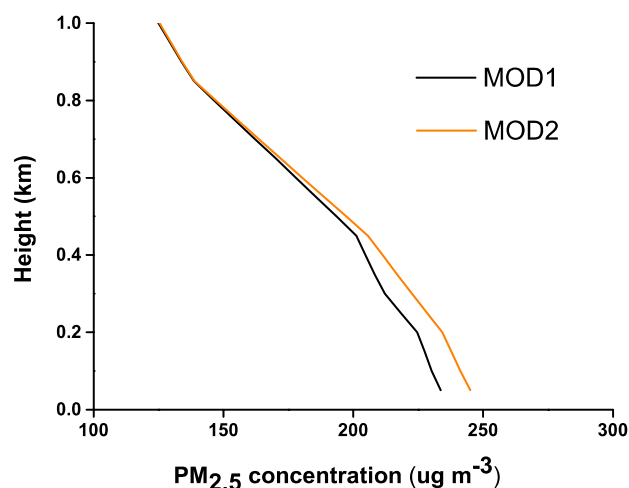


Figure R1. Vertical profiles of PM_{2.5} concentrations simulated in MOD1 and MOD2 during the daytime of Dec.7, 2013 during a haze event in Wuxi, a haze center.

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 (sulfate+nitrate+ammonium) greater than that of the PM_{2.5} (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 PM_{2.5}, meaning the enhancement of PM_{2.5} 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 PM_{2.5} 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 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 $\mu\text{g m}^{-3}$ to the PM_{2.5} enhancements, during the haze episode, while the chemical reaction contributed about 4.77 $\mu\text{g m}^{-3}$ (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, O₃ and overestimated NO₂ and SO₂ 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.