

***Interactive comment on* “Nonlinear response of ozone to precursor emission changes in China: a modeling study using response surface methodology” by J. Xing et al.**

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We would like to thank Reviewer 1 for a very thoughtful and detailed review of our manuscript that helped to improve the paper. Here we address all the points raised by the respectful reviewer as follows. We basically followed all the comments and revised manuscript accordingly. Revised manuscript with revised part in yellow background was also uploaded as a supplement.

1. it is necessary to demonstrate that the model reproduces ozone well, but also reproduces ozone precursors well. Typically, emissions inventories themselves are highly uncertain, and that leads to strong uncertainties in the ozone sensitivity. At least, the

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authors should discuss these uncertainties and not give the impression that ozone sensitivity is known with precision. At most, the authors should use their model to quantify uncertainties and explore how robust their predictions are. If there is other evidence showing that the sensitivities in this model agree or disagree with other models, or based on observational studies, those should be discussed in this context.

We agree that it's necessary to address the uncertainties. And we will revise it as follows.

(Page 4 line 23) "Compared to other ones available in literature, e.g. Streets et al. (2003), Zhang et al. (2009), the uncertainties in our base year emissions are lower. The uncertainties (i.e., 95% confidence intervals around the central estimates) of NO_x and VOC emission inventory used in this study are -10%~36% (Zhao et al., 2010) and -44%~109% (Wei et al., 2008). "

(Page 20 line 6) "The ozone sensitivities may still suffer the uncertainties of emission inventory. For example, when NO_x reduced by 10% and VOC increased by 100%, it's more likely to have the translation from VOC-limited regime to NO_x-limited regime, see the 2-D isopleths of ozone in Fig 5. Therefore, it's quite important for the future work to better understand the precursors' emission inventory, especially for VOC emissions."

Reference

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, 108, 8809, 2003

Wei, W., Wang, S. X., Satoru, C., Klimont, Z., Cofala, J., Hao J. M., 2008. Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China, *Atmospheric Environment*, 42(20), 4976-4988.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont,

Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, *Atmos. Chem. Phys.*, 9, 5131-5153, 2009.

Zhao, Y., Nielsen, C.P., Lei, Y., McElroy, M.B., and Hao, J.M.: Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China, *Atmos. Chem. Phys. Discuss.*, 10, 29075-29111, 2010

2. I also do not understand whether the model has been run over a big domain (Fig 2) or three small domains (Fig 8).

The model has been run over a big domain as shown in Fig. 2. In the modeling domain, we chose three sub-areas (Beijing, YRD and PRD) as target areas for analysis. We revise the manuscript as follows.

(Page 5 line 8) “A one-way nested technique is employed in this study. Modeling domain 1 covers almost entire China with a 36×36 km horizontal grid resolution and generates the boundary conditions for nested domain at 12-km resolution over popular Eastern China, as shown in Fig. 2a. Three sub-areas (i.e., Beijing, YRD and PRD) are selected for analysis.”

3. The figures are extremely small, where often the legends or axis labels or color scales cannot be read without being magnified, and the figure captions do not adequately explain the figures. I suggest removing unnecessary labels on the figures (for example the words on top of Figure 2b), making the individual panels larger, and making sure all text on the figure is large enough to read. Similarly, the figure captions need to be much more descriptive as these multi-panel figures are extremely complex.

We followed the reviewer’s suggestion and re-plotted the Fig 2b and Fig 8 in the revised manuscript.

4. Finally, the English writing is not bad, but is difficult enough to get through that it hinders communication. Since one of the authors is based in the US, I hope that

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he/she can help improve the writing.

The language of the manuscript was edited by a native English speaker.

5. p. 29810, l. 18-19 “Effectiveness of NO_x emission control is growing along with stricter control efforts” – I don’t know how to interpret this, nor how it relates to the rest of the sentence. Does it mean that NO_x emissions are going down due to emission controls? Or are those reductions now more effective at reducing ozone? Or something else?

To avoid confusion, we modified the sentence in the revised manuscript as follows.

(Page 1 line 27) “Stricter NO_x emission control has higher effects on ozone reduction because of the shift from VOC-limited regime to NO_x-limited regime. Therefore, NO_x emission control should be significantly enhanced to reduce the ozone pollution in China.”

6. p. 29815, l. 6-25. I do not understand the “area of influence analysis”. How were the “ratios of inner-influence among the three regions” determined and what do they mean? In what way does fig. 2b show “interactions”? What was the model simulation on which this was based?

The purpose of “area of influence analysis” is to testify the independence of three cities (i.e., Beijing, Shanghai and Guangzhou) and their emissions won’t affect each other. The “ratios of inner-influence among the three regions” are conducted through the sensitivity analysis, that is, the differences between the baseline simulation and the controlled simulation which zeroed out all emission in selected cities, as shown in figure 2b. The figure 2b gives the influence scope of three cities. It can be seen that the interactions of their emission impacts are less than 0.5 ppb, which is negligible. We will make the explanation clear in the revised manuscript, as follows.

(Page 6 line 22) “In this study, the particular urban areas selected are Beijing, Shanghai and Guangzhou. Local versus regional impacts have been teased out for the three

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urban areas. The local emissions in those three cities are grouped together as one region (Region A), and the other areas are grouped as another region (Region B). In our analysis, Region A represents the local emission of each city. To testify the independence of three cities, sensitivity analysis was conducted to calculate the impact of one city's emission on the other two, which is given by the differences between the baseline simulation and the controlled simulation which zeroed out all emission in selected urban areas, as shown in figure 2b. The figure 2b gives the influence scope of three cities. It can be seen that the interactions of their emission impacts are less than 0.5 ppb, which is negligible.”

7. p. 29817, l. 10-25. “emission ratios” are not defined – I think this is the factor multiplier for emissions describing the range over which emissions are varied. Then the “weight coefficients” are not defined nor does it describe where the values come from. I think all that is happening here is: $(tNO_x) (RtNO_x) = \text{SUM} (NO_{xi})(RNO_{xi})$, and so the weight coefficients are just the NO_x emissions, but this presentation doesn't make that clear.

In the revised manuscript, the “emission ratios” are defined as the ratio of the changed emission compared to the baseline emissions. For example, the emission ratio is 1 for baseline emissions, and the emission ratio is 0.6 for 40% emission reduction.

The “weight coefficients” reflects contribution from each emission source, which is defined by the following equations.

$$(NO_{xi}) = (tNO_x) (A_i)$$

$$(tNO_x) (RtNO_x) = \text{SUM} (NO_{xi})(RNO_{xi}) = \text{SUM} (tNO_x) (A_i) (RNO_{xi}), \text{ then}$$

$$(RtNO_x) = \text{SUM} (A_i) (RNO_{xi}), \text{ and } \text{SUM} (A_i) = 1.$$

In the preliminary experiments, the “weight coefficients” (A_1, A_2, \dots, A_n) were set to be 1:2:3...:N, with sum as 1.

We have explained these two terms in the revised manuscript (see Page 7 line 10 and

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Page 9 line 8).

8. p. 29820 – Methods like the LOOCV should be described more completely, possibly through a simple illustration

We will give the explanation in the revised manuscript as follows.

(Page 10 line 14) “The definition of LOOCV is to use a single observation from the original sample as the validation data, and the remaining observations as the training data to build prediction RSM. Each sample in the datasets is used once as the validation data. For example, for N training data (d1, d2. . . .dN), the sample i (di) has been selected as the validation data, and the remaining samples (d1, d2. . .d(i-1), d(i+2). . .dN) are used to build RSM to predict the sample i and make comparison.”

9. p. 29822, l. 22-27. Here it is not clear how the authors determine that one indicator is “more robust” than another – what is being compared with what to reach that conclusion?

We agree it’s not sufficient to determine which indicator is more robust than another in this study. We have deleted the discussions on this point in the revised manuscript.

10. p. 29829, l. 17. There is no discussion of costs of emission reductions in the paper, so I don’t think the authors can conclude about “cost-effectiveness”. Should only judge “effectiveness”.

We agree to change the cost-effectiveness to effectiveness. We delete it in the revised manuscript.

11. Table 4 – Why are these combinations of reductions selected?

We will give the explanation in the revised manuscript as follows.

(Page 18 line 24) “RSM allows us to calculate the emission reduction ratio to attain a certain target concentration (i.e., 80ppb, 1-h maximal ozone). In order to attain this target, several optional control scenarios with various control ranges are designed ac-

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ording to the RSM results (HSS6-200 case). The reduction efforts are different when controlling different sources, as shown in Table 4.”

12. Fig. 3b – Why is “sum 4 variables” shown here and what is its relevance for this study? Is this the sum of 4 numbers, each randomly selected from the same distribution (0-1)? If so, then wouldn’t values greater than 1 be possible?

It is not the “sum 4 variables”. We modified it to “weighted mean of 4 variables” in the revised manuscript.

The purpose of this discussion is to explain why margin processing is necessary in this study. For example, in our RSM design, four types of NO_x emission sources were considered. The “weighted mean of 4 variables” represents the change ratio of total NO_x emission, which equals the change ratio of each individual NO_x source multiply its “weight coefficient” which were set to be 1:2:3...:N, with sum as 1. Therefore, its value range is from 0 to 1. We add more explanation in the revised manuscript in figure 3.

13. Fig. 5a – what method is used for this plot?

Fig.5a was plotted based on the HSS6-200 method. We have added this point in the revised manuscript.

14. Fig. 6b – Is that correct that all symbols mean $n=160$?

It’s all $n=160$, but different LHS samples. We changed it to sample 1,2...6.

15. Fig. 7 – I do not know what the labels of the figures mean, such as “8vs2 – scale6”

Sorry for the confusion, it means there’re 8 types of NO_x sources and 2 types of VOC sources with marginal process level as 6. We modified it in the revised manuscript.

16. Fig. 9 – Consider plotting height on the vertical axis.

We followed the reviewer’s suggestion to re-plot the figure in the revised manuscript.

17. Fig. 10 – This is a very complicated figure that isn’t explained. It seems that reduc-

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tions of NO_x have little effect. Sorting days high->low is fine but should be explained. Based on which RSM are these results? Are b and c figures for averages of multiple days?

The reductions of NO_x have little effects because of relative low control level. The purpose of this analysis is to verify the ozone chemistry. The figure 10b and 10c are for the averages of multiple days, based on the results of RSM-HSS6-200. To avoid confusion, we delete the figure 10.a and focus on the discussion for Fig 10b c, as follows.

(Page 15 line 7) “Large differences are found in the comparison of averaged ozone isopleths for high ozone days (>70ppb) and lower ozone days (<30ppb) in three cities, as shown in Fig. 10. During the days when higher ozone (>70ppb) occurs under favorable meteorological condition for photochemical production of ozone, the ozone response is mostly NO_x-limited, with peak ratio larger than 0.8. The NO_x emission control is benefit for Ozone reduction. However, in the days with lower ozone mixing ratio (<30ppb), usually the effects of NO_x controls are negative for ozone, with peak ratio lower than 0.5, mainly because negative photochemical production leads to due to NO titration of ozone under high NO_x emissions (NO_x-rich conditions).”

18. Fig. 11 – Would it be more straightforward to plot d(O₃ concentration) against Emissions, rather than d(O₃)/(1-dEmis)? I find it difficult to understand what this plot means. Plots b and c then present the effects of VOC and NO_x “with synchronic control” and “with control of NO_x from power plants”, but I don’t see where synchronic control is defined and I don’t know how to interpret “with power plants” since power plants are the yellow and orange bars.

It is a typo. Actually this plot is d(O₃) against d(Emis).

Synchronic control means that all type of emission sources are controlled by same ratio. The impact of each source “under the synchronic control of other sources” is evaluated through the difference between the synchronic control scenario of all sources

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and the scenario that an individual source doesn't change.

The sub-title of Fig.11c should be "Impacts of each source with zero NO_x emissions from power plants". The purpose of this figure is to see the ozone response to other emission sources when the NO_x emissions from power plants were set to zero.

We have modified them in the revised manuscript, as follows.

(Page 15 line 18) "Following other sensitive studies (Yarwood et al, 2005; Koo et al., 2009), we defined the "Ozone response" as the change ratio of ozone to the change ratio of emission, to evaluate the control effects of each source. From RSM results, the "Ozone response" can be calculated in a large range of emission variation (from 10% control to 100% control), as seen in Fig.11."

(Page 17 line 2) "In order to explore the nonlinear effects among different sources, we also compared the impact of each source under the synchronic control of other sources. Synchronic control means that all type of emission sources are controlled by same ratio. The impact of each source under the synchronic control of other sources is evaluated through the difference between the synchronic control scenario of all sources and the scenario that an individual source doesn't change."

(Page 17 line 16) "One example is the control of NO_x from power plants. When the NO_x emission from power plants was set to zero, the ozone response to other NO_x emission has been considerably enhanced, as shown in Fig 11c."

References

Yarwood, G., G. Wilson, R. Morris: DEVELOPMENT OF THE CAM_x PARTICULATE SOURCE APPORTIONMENT TECHNOLOGY (PSAT), final report, ENVIRON International Corporation, 2005

Koo, B., G.M. Wilson, R.E. Morris, A.M. Dunker, G.Yarwood: Comparison of Source Apportionment and Sensitivity Analysis in a Particulate Matter Air Quality Model, Environ. Sci. Technol., 43, 6669–6675, 2009.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/10/C14560/2011/acpd-10-C14560-2011-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 29809, 2010.

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