Reviewer 2:

1 General comments

This is an interesting paper which develops an advanced modelling technique for particulate matter concentration responses to changes in emissions of primary particles and precursor gases. The authors apply the model to analyse the sensitivity of PM concentrations in Chinese cities to the emissions from different economic sectors and spatial origin. Given the recurring episodes of high pollution in Chinese cities, this is a very important and timely topic and fits well to this journal. Considering the still rather high number of CTM simulations needed I am not fully convinced that this is an easily applicable technique for source response modelling in general, but the example provided in the paper demonstrates feasibility of the approach for a limited set of source regions (4) and sectors.

The article is generally well written, although the methodology section is in parts rather difficult to follow. I recommend the paper to be published after addressing the comments below, mostly regarding clarifications in the methodology.

Response: We thank the reviewer for supporting the publication of our manuscript. We also appreciate his/her comments which help us improve the quality of our manuscript. We address the reviewer's comments below. The original comments are in black and our responses are in blue.

2 Detailed comments

Sect. 2.1, general question: What is the time scale for the ERSM development? Do the authors aim to develop annual mean coefficients, monthly coefficients, summer/winter coefficients? Would full-year coefficients be feasible?

Response: The ERSM technique is applicable for various time scales, ranging from a single day to several years. As a case study in the Yangtze River Delta, we developed the response surfaces for monthly mean $PM_{2.5}$ concentrations in January and August, representing winter and summer, respectively. The analysis could be extended to a full year as long as we finish

the CMAQ simulations for a full year and build the response surfaces following the same procedure. We have added the explanation accordingly in the revised manuscript. (Page 11, Line 17-21 in the revised manuscript)

p. 5055 l. 7ff: “The ERSM technique first quantifies the relationship...” - What is missing in the general description of the methodology here is a simple description of how this functional relationship is quantified. This should be explained briefly to readers who are not proficient with the RSM technique and the MPerK program.

Response: We thank the reviewer for this comment. We have added description of how the relationship is quantified in the revised manuscript. The amended texts are shown as follows.

The ERSM technique is developed starting from the conventional RSM technique; the latter characterizes the relationships between a response variable (e.g., $PM_{2.5}$ concentration) and a set of control variables (i.e., emissions of particular precursors from particular sources) following the procedures described in our previous paper (Xing et al., 2011). First, a number of emission control scenarios are generated with the Latin Hypercube Sample (LHS) method (Iman et al., 1980), a widely-used sampling method which ensures that the ensemble of random samples is representative of actual variability. Then the $PM_{2.5}$ concentration for each emission scenario is calculated with a regional CTM, and finally the RSM prediction system is developed using a MPerK (MATLAB Parametric Empirical Kriging) program (Santner et al., 2003) based on MLE-EBLUPs. The robustness of the conventional RSM technique has been validated through leave-one-out cross validation, out of sample validation and 2-D isopleths validation, as documented in our previous papers (Xing et al., 2011; Wang et al., 2011). (Page 5, Line 3-15 in the revised manuscript)

References:

Iman, R. L., Davenport, J. M., and Zeigler, D. K.: Latin Hypercube Sampling (Program User's Guide), Sandia National Laboratories, Albuquerque, NM, U.S.Technical Report SAND79-1473, 78, 1980.

Santner, T. J., Williams, B. J., and Notz, W.: The Design and Analysis of Computer Experiments, Springer Verlag, New York, U.S., 283 pp., 2003.

Wang, S. X., Xing, J., Jang, C. R., Zhu, Y., Fu, J. S., and Hao, J. M.: Impact assessment of ammonia emissions on inorganic aerosols in east China using response surface modeling technique, Environ. Sci. Technol., 45, 9293-9300, DOI 10.1021/Es2022347, 2011.

Xing, J., Wang, S. X., Jang, C., Zhu, Y., and Hao, J. M.: Nonlinear response of ozone to precursor emission changes in China: a modeling study using response surface methodology, Atmos. Chem. Phys., 11, 5027-5044, DOI 10.5194/acp-11-5027-2011, 2011.

p. 5055 l. 12ff: How about formation of secondary PM in the source region followed by transport to the target region? Is this process missed or is just the formulation unclear at this point? If it is missed, do the authors have an estimate of its contribution? How about interactions of precursor emissions from different source regions?

Response: This process is not missed but is not clearly described in our original manuscript. This process is calculated using Eq. (8). To make it clear, we have revised the original descriptions as follows:

In order to quantify the interaction among regions, we make a key assumption that the emissions of gaseous precursors in the source region affect $PM_{2.5}$ concentrations in the target region through two major processes: (1) the inter-regional transport of gaseous precursors enhancing the chemical formation of secondary $PM_{2.5}$ in the target region; (2) the formation of secondary $PM_{2.5}$ in the source region followed by transport to the target region. (Page 5, Line 20-25 of the revised manuscript)

The texts that describe how we quantify the contribution of the second process are as follows:

The contribution of the second process to $PM_{2.5}$ concentration in Region A (denoted by $[PM_{2.5}_Trans]_{B\rightarrow A}$ defined below) is then calculated by extracting the contribution of the first process (Eq. (7)) from the total (Eq. (4)), as expressed by Eq. (8).

$$[PM_{2.5}_Trans]_{B\rightarrow A}=[PM_{2.5}]_{B\rightarrow A}-[PM_{2.5}_Chem]_{B\rightarrow A} \quad (8)$$

where $[PM_{2.5}_Trans]_{B\rightarrow A}$ is the change of $PM_{2.5}$ concentration in Region A affected by the changes of precursor emissions in Region B through the transport of secondary $PM_{2.5}$ (the second process). (Page 8, Line 15-21 of the revised manuscript)

For your last question, the interaction of precursor emissions from different source regions could affect the $PM_{2.5}$ concentrations in the target region in two different ways: (1) by affecting the inter-regional transport of gaseous precursors from the source regions to the target region; (2) by affecting the inter-regional transport of secondary $PM_{2.5}$ from the source regions to the target region. The first effect is implicitly considered in Eq. (10), which sums up the changes of precursor concentrations in the target region induced by the changes of precursor emissions in every source region. The second effect is neglected, which is described in Page 9, Line 6-12 of the revised manuscript.

p. 5058 l. 4ff: Similar to above: Is something missed due to this assumption? In fact, as it is formulated it reads rather straightforward and I don't quite understand why this assumption is needed. How should PM2.5 in A be influenced by precursor gas concentration changes in A other than through local chemical formation?

Response: We thank the reviewer for this comment. The changes of precursor concentration in Region A might affect the precursor concentrations/$PM_{2.5}$ concentrations in other regions, which might in turn affect the $PM_{2.5}$ concentrations in Region A. This "indirect" pathway is neglected with the above-mentioned assumption.

We have explained this assumption in detail in the revised manuscript. The revised texts are also given below.

In order to quantify the contribution of the first process, we firstly use Eq. (5) and Eq. (6) to quantify the effect of the transport of gaseous precursors from Region B to Region A on the precursor concentrations in Region A. How much does the change of precursor concentrations in Region A enhance the chemical formation of secondary $PM_{2.5}$ in Region A? To answer this question, we introduce a straightforward assumption that the changes of $PM_{2.5}$ concentration owing to changes of precursor concentrations in the same region (described by Eq. (1)) are solely attributable to changes of local chemical formation. Strictly speaking, the changes of

precursor concentration in Region A might affect the precursor concentrations/$PM_{2.5}$ concentrations in other regions, which might in turn affect the $PM_{2.5}$ concentrations in Region A; but this "indirect" pathway is thought to be negligible in this study. (from Page 7, Line 31 to Page 8, Line 10 in the revised manuscript)

p. 5058, l. 10ff, Eqns 8 and 9: This paragraph is a bit unclear to me. If $[PM_{2.5}_Trans]_{B\rightarrow A}$ is calculated through Eq. 8, how is Eq. 9 used? Or is Eq. 8 merely the definition and Eq. 9 is in fact used for calculating the transport contribution from B to A? This should be clarified.

Response: We appreciate the reviewer's comment. When the response surface is built, $[PM_{2.5}_Trans]_{B\rightarrow A}$ is calculated using Eq. 8. But we also need to know the relationship between $[PM_{2.5}_Trans]_{B\rightarrow A}$ and the precursor emissions in Region B, which is quantified using conventional RSM technique, and is described as Eq. 9.

When we predict the $PM_{2.5}$ concentration for an arbitrary scenario using Eq. 10 or Eq. 15, $[PM_{2.5}_Trans]_{B\rightarrow A}$ is calculated using Eq. 9. In this case, it cannot be calculated using Eq. 8 because Eq. 8 holds only if the emissions in the regions other than Region B remain at the base-case levels.

In the revised manuscript, this issue has been clarified. (from Page 8, Line 15 to Page 9, Line 5 in the revised manuscript)

p 5059, l. 4ff and Eq. 11: It would be helpful if $[NOx]_{A,\,min}$ were defined explicitly. Is it certain that $[NOx]_{A\rightarrow A,\,min} = RSM_{A\rightarrow A}^{NOx}(0,\,0,\,0)$? Is the case (0,0,0) i.e. all precursor emissions equal 0 covered in any case? From a random draw it could even be higher?

Response: We have defined $[NOx]_{A,\,min}$ explicitly in the revised manuscript: $[NOx]_{A,\,min}$ is defined as the minimum NO_X concentration in Region A when the emissions from Region A change arbitrarily and those in other regions remain the base-case level. (Page 9, Line 20-22 in the revised manuscript)

We concluded $[NOx]_{A\rightarrow A,\,min} = RSM_{A\rightarrow A}^{NOx}(0,\,0,\,0)$ based on a series of numerical experiments.

In detail, we have constructed the relationship between $[NOx]_{A\rightarrow A}$ and the precursor emissions in Region A in our manuscript, as described by Eq. (2).

$$[NOx]_{A\rightarrow A}=RSM_{A\rightarrow A}^{NOx}\left(Emis_NOx_1_A, Emis_NOx_2_A, Emis_NH3_A\right) \quad (2)$$

Similar to Fig. 4 of the revised manuscript, we used Eq. (2) to plot the 2-D isopleths of $[NOx]_{A\rightarrow A}$ in response to any two of the control variables. From those isopleths we concluded that $[NOx]_{A\rightarrow A}$ is minimal when all precursor emissions equal zero.

p. 5060 l. 15 ff: δ as defined in l. 16 is not an interval. I assume the actual transition interval is $([NOx]_{A,\,min},\ [NOx]_{A,\,min}+\delta_{NOx})$ for NOx and equivalent for NH3? If so, this should be clarified in the text.

Response: Yes, this is what we meant. We have revised the texts as follows:

To assure the consistency between Eq. (10) and Eq. (15), we introduce "transition intervals" of $([NOx]_{A,\,min}, [NOx]_{A,\,min}+\delta_{NOx})$ and $([NH3]_{A,\,min}, [NH3]_{A,\,min}+\delta_{NH3})$, where $\delta_{NOx}=0.1*[NOx]_{A0}$ and $\delta_{NH3}=0.1*[NH3]_{A0}$. Eq. (10) is applied for $[NOx]_A \geq [NOx]_{A,\,min}+\delta_{NOx}$ and $[NH3]_A \geq [NH3]_{A,\,min}+\delta_{NH3}$, and we linearly interpolate between Eq. (10) and Eq. (15) for the transitional range. (from Page 10, Line 27 to Page 11, Line 2 in the revised manuscript)

I do not completely understand why the physical transport versus chemical production diagnostics module is used in the second approach outside the minimal precursor concentrations, but not within the first approach. For example, could it not be useful for distinguishing chemical and transport contributions as in Eq. 7 and 8? This should be better motivated by the authors.

Given that two different approaches are used here, the authors should comment on how smooth the transition is between the two regimes considered, i.e. how large the deviations are in the overlap interval.

Response: We thank the reviewer for this comment. Using the first approach, we could

distinguish the contributions of chemical formation and physical transport without this diagnostic module (see Eq. 7 and Eq. 8). If this module was used, we would need to develop the relationship between the chemically formed $PM_{2.5}$ and the $PM_{2.5}$ concentration, which was an extra step compared with the first approach and added to the complexity. In the second approach, we are not able to distinguish the chemical and transport contributions without the diagnostic module; this explains why that module is only used in the second approach.

Based on the case study in the Yangtze River Delta, the discrepancy between the two approaches is 1-8% in the overlap interval.

We have explained these two issues accordingly in the revised manuscript. (from Page 10, Line 20 to Page 11, Line 2 of the revised manuscript)

p. 5062 l. 15: "the cases where all control variables are controlled stringently" – If I have not overlooked something, these cases are mentioned here for the first time. They should be mentioned in the text describing the scenario generation in the last paragraph of Sect. 2.2.

Response: We agree with the reviewer's comment and have added a brief description of the cases used for out-of-sample validation in Sect. 2.2. The added text is also given as follows.

Finally, we generated 40 independent scenarios for out-of-sample validation, as described in detail in Sect. 3.1. (Page 12, Line 20-21 of the revised manuscript)

The detailed description in Sect. 3.1 is also revised more clearly as follows:

These 40 out-of-sample scenarios include 32 cases (case 1-32) where the control variables of gaseous precursors change but those of primary $PM_{2.5}$ stay the same as the base case, 4 cases (case 33-36) the other way around, and 4 cases (case 37-40) where control variables of gaseous precursors and primary $PM_{2.5}$ change simultaneously. Most cases are generated randomly with the LHS method (case 4-6, 10-12, 16-18, 22-24, 28-40), and some cases are designed where all control variables are subject to large emission changes (case 1-3, 7-9, 13-15, 19-21, 25-27). A more detailed description of the out-of-sample control scenarios is

given in Table S3. (from Page 12, Line 29 to Page 13, Line 4 in the revised manuscript)

p. 5063 l. 13 and 17: changes of total emissions: in which regions? Does this refer to all regions reducing at the same time? This should also be mentioned in the caption to Fig. 3.

Response: Yes, this refers to reducing emissions in all regions at the same time. We have revised this sentence as well as the caption of Fig. 3 (Fig. 4 in the revised manuscript) to make it clear. The revised texts are as follows.

We further evaluated the performance of the ERSM technique by comparing the 2D-isopleths of $PM_{2.5}$ concentrations in response to the simultaneous changes of $NO_X/SO_2/NH_3$ emissions in all regions derived from both the conventional RSM and the ERSM technique.

Figure 4. Comparison of the 2-D isopleths of $PM_{2.5}$ concentrations in Shanghai in response to the simultaneous changes of precursor emissions in all regions derived from the conventional RSM technique and the ERSM technique in Shanghai. The X- and Y-axis shows the emission ratio, defined as the ratios of the changed emissions to the emissions in the base case. The different colors represent different $PM_{2.5}$ concentrations.

Figures 3-5: If the authors would like to show the comparison for all three regions, then they should discuss differences and characteristics – otherwise one example would be sufficient.

Response: We agree with the reviewer that one example is sufficient. In the revised manuscript we keep only Figure 3 in the main text (Figure 4 in the revised manuscript) and move Figure 4 and Figure 5 to the Supporting Information.

Figures 6-7: Is there an easy explanation for the sometimes significant differences between effects of reducing individual pollutants, and reducing all of them together? In most cases the combined effect seems lower than the sum of individual effects, which might be explained by overlap effects of reductions in both species involved in the formation of ammonium sulfate and ammonium nitrate, but also the opposite is the case? It would be interesting if the authors

could comment on this.

Response: We appreciate the reviewer's valuable comment. The reviewer is correct that the combined effect is lower than the sum of individual effects in most cases because of the overlap effects of reductions in both species involved in the formation of ammonium sulfate and ammonium nitrate. However, it is sometimes the other way around in January, as shown in the left part of Figure 7 (Figure 6 of the revised manuscript, as shown below). This is because, in January, the response of $PM_{2.5}$ to NO_X emissions is negative for relatively small reductions (< 40-70%), but becomes positive for large reductions (> 40-70%). The reduction of NO_X emissions of a single emission sector leads to the increase of $PM_{2.5}$ even at large reduction ratio since the emission reduction is small compared with the total NO_X emissions. Therefore, when the NO_X emissions from each sector are reduced individually (the bars), we sum up the negative effects. In contrast, when all pollutants from all sectors are reduced simultaneously (the red dotted line), the NO_X emission reduction at large ratio could have positive effect on $PM_{2.5}$ reduction. This is why the combined effect sometimes exceeds the sum of individual effects in January. We have added the explanations accordingly in the revised manuscript. (from Page 15, Line 24 to Page 16, Line 3 in the revised manuscript)

January August

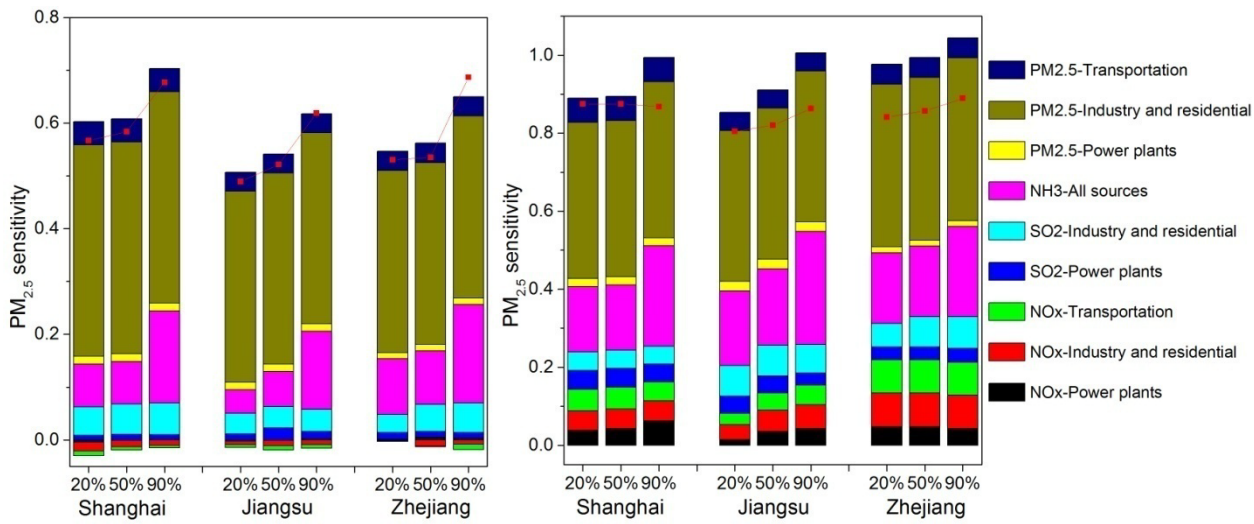

Figure 7 (Figure 6 of the revised manuscript). Sensitivity of $PM_{2.5}$ concentrations to the stepped control of individual air pollutants from individual sectors. The X-axis shows the reduction ratio (= 1 – emission ratio). The Y-axis shows $PM_{2.5}$ sensitivity, which is defined as the change ratio of concentration divided by the reduction ratio of emissions. The colored bars denote the $PM_{2.5}$ sensitivities when a particular emission source is controlled while the others stay the same as the base case; the red dotted line denotes the $PM_{2.5}$ sensitivity when all emission sources are controlled simultaneously.

3 Technical corrections

p. 5056 l. 29: “the same as NH3”: I assume what is meant is “equivalent for NH3”.

Response: Revision has been made. (Page 7, Line 8 of the revised manuscript)

p. 5058 l. 21: “changes”: change

Response: Revision has been made. (Page 8, Line 27 of the revised manuscript)

p. 5059, l. 9: “the same as NH3”: I assume what is meant is “equivalent for NH3”.

Response: Revision has been made. (Page 9, Line 18 of the revised manuscript)

p. 5060 l. 17: “interpolates”: interpolate

Response: Revision has been made. (Page 10, Line 30 of the revised manuscript)

Figures 3-5: Axes font sizes are very small and hardly readable, I suggest to increase them. Also, I wonder whether there is a reason for the upside-down colorbars, otherwise I’d suggest inverting them.

Response: We have increased the font sizes and inverted the colorbars. The revised Figure 3 (Figure 4 in the revised manuscript) is shown below.

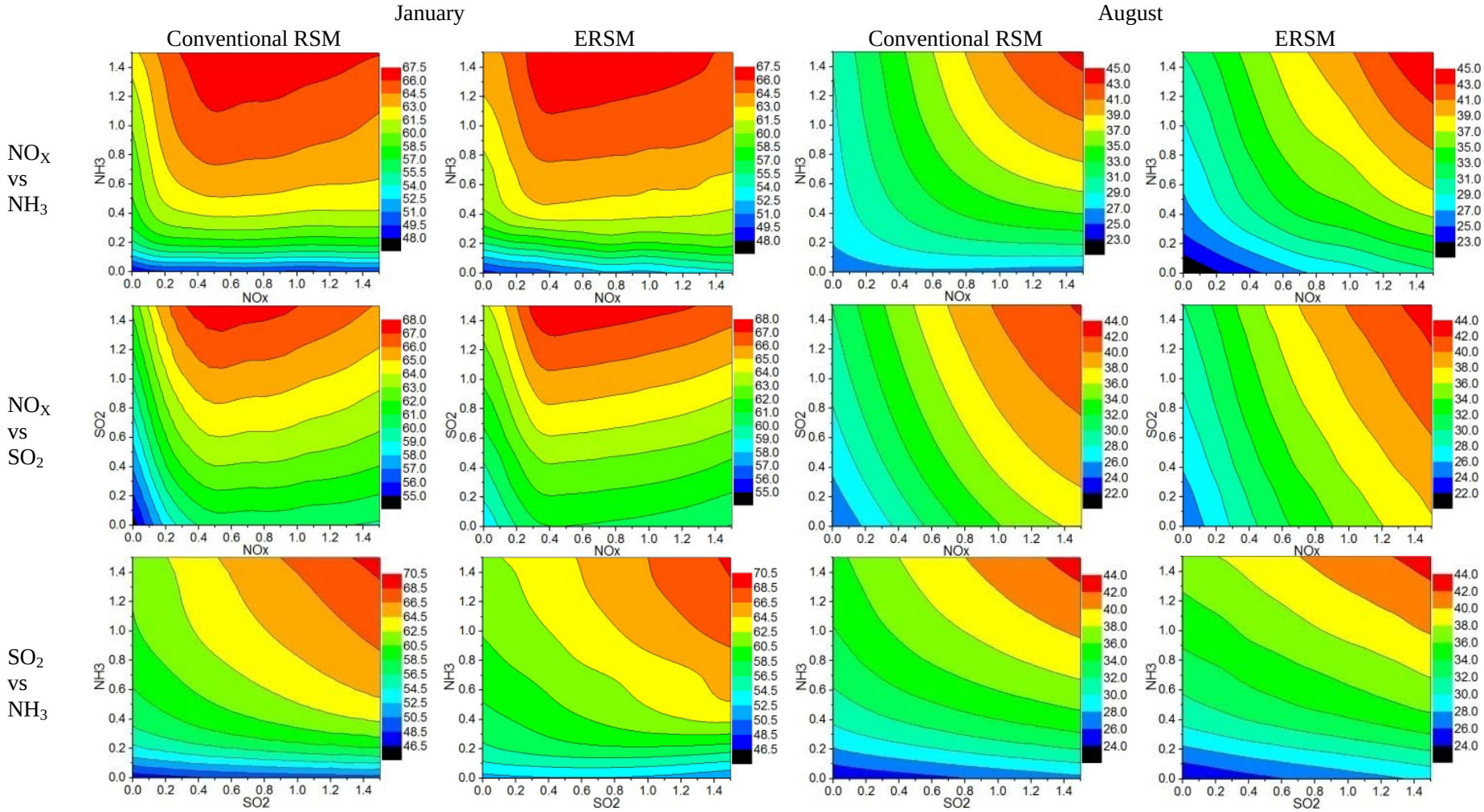

Figure 4. Comparison of the 2-D isopleths of $PM_{2.5}$ concentrations in Shanghai in response to the simultaneous changes of precursor emissions in all regions derived from the conventional RSM technique and the ERSM technique. The X- and Y-axis shows the emission ratio, defined as the ratios of the changed emissions to the emissions in the base case. The different colors represent different $PM_{2.5}$ concentrations (unit: $\mu g\ m^{-3}$).