The efforts of co-authors in addressing the reviewers' comments are appreciated. There are however a couple of concerns that are not appropriately answered in my opinion.

Response: We appreciate the editor's comments which help us further improve the quality of our manuscript. We address the editor's comments below. The original comments are in black and our responses are in blue.

Both referees where concerned by the number of scenarios required to train the ERSM and there remain a couple of unsolved issues. Please provide a more detailed explanation for the reason why the number of required scenarios would be of a power of four of the number of variables in the present case. With 10 to 15 variables, that would be a maximum ensemble size of tens of thousands, not hundreds as stated in the manuscript (P4 L13). While reducing that size to 600 is a very welcomed improvement, it remains a substantial number and it cannot yet be claimed that the proposed technique is "highly economical". To address this point, and while none of the reviewer requested a more detailed quantification of the convergence rate, the authors may consider including a figure illustrating the evolution of the mean normalized error when increasing the number of simulations.

Response: We appreciate the editor's valuable comments. The number of model scenarios required to build the conventional RSM is determined to ensure that they are sufficient to accurately construct the relationship between the response variable and control variables. Specifically, we gradually increase the scenario number and build the response surface repeatedly until the prediction performance is good enough (mean normalized error $< 1\%$; correlation coefficient > 0.99). Using this method, we determined the number of scenarios required to build the conventional RSM for 2-10 control variables (shown as the dots in Figure R1). Then we fitted the dots using polynomials of 2^{nd} – 5^{th} order (shown as the lines in Figure R1). The results indicate that the equations of 2^{nd} or 3^{rd} order are not able to capture the rapid increase of the scenario number with the increase of variable number. In contrast, the 4^{th} or 5^{th} order equations fit well. Therefore, we conclude that the number of model scenarios required to build the conventional RSM depends on the variable number via an equation of fourth or higher order. We have added the explanations accordingly in the Supporting Information (from Page 1, Line 25 to Page 2, Line 11) and given a brief instruction in the main text (Page 4, Line 10-13) which refers the readers to these explanations.

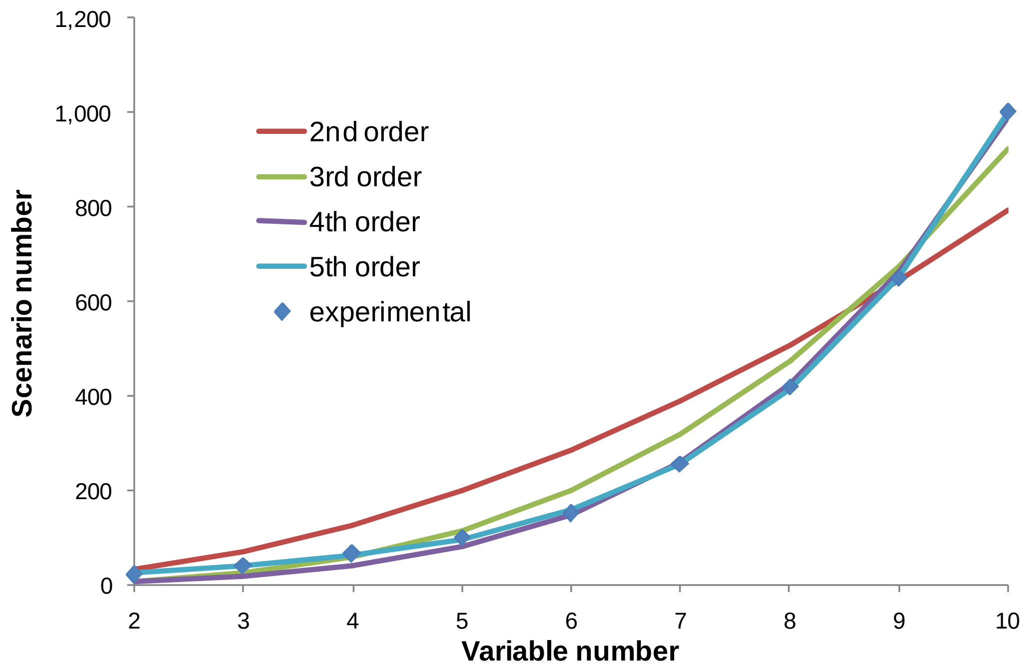

Figure R1. Number of scenarios required to build the conventional RSM based on numerical experiments (the dots) and the fits to polynomials of 2^{nd} – 5^{th} order (the lines).

We agree with the editor that with 10 to 15 variables, the required scenario number would be a maximum ensemble size of tens of thousands, not hundreds of thousands as stated in the original manuscript. Therefore, we modified that sentence as follows:
The required scenario number would be tens of thousands for over 15 variables and even hundreds of thousands for over 25 variables, which is computationally impossible for most three-dimensional CTMs. (Page 4 Line 13-16 in the revised manuscript)

We also agree with the editor that 600 scenarios remain a substantial number and it cannot yet be claimed as "highly economical". Therefore, we revised the original description as follows:
The Response Surface Modeling (RSM) technique (denoted by "conventional RSM" technique in the following text to distinguish from the ERSM technique developed in this study), has been developed by using advanced statistical techniques to characterize the relationship between model outputs and inputs. (from Page 3 Line 32

to Page 4, Line 3 in the revised manuscript)

We have discussed "convergence rate" of the conventional RSM technique in our previous paper (Xing et al., 2011). For example, Figure R2, adapted from Figure 7 of Xing et al. (2011), shows the evolution of mean normalized error and correlation coefficient with the increase of scenario number for 2, 4, and 6 control variables. It can be seen that the normalized mean error first decreases and then gradually remains stable, with the increase of scenario number. In contrast, the correlation coefficient first increases and then gradually becomes stable. We used a criterion that mean normalized error < 1% and correlation coefficient > 0.99, and thus determined the required scenario number to construct the conventional RSM. In order to determine the required scenario number for the ERSM technique, we first determine the scenario number to construct the conventional RSM for a single region, and then repeat this procedure for each region (see details in Page 6, Line 13-31 in the revised manuscript). Therefore, Figure R2 is also applicable to determine the scenario number for the ERSM technique. In the revised manuscript, we have explained the evolution of prediction performance briefly in the methodology section, and the revised text is shown as follows:

The emission control scenarios required to construct ERSM include: (1) the base case; (2) N scenarios generated by applying the LHS method for the control variables in each single region; and (3) M scenarios generated by applying the LHS method for the total emissions of gaseous precursors (NO_X and NH_3 for this case) in all regions. The scenario numbers N and M are determined to ensure that they are sufficient to accurately construct the relationship between the response variable and randomly changing control variables using conventional RSM technique. Specifically, we gradually increase the scenario number and build the conventional RSM repeatedly until the prediction performance is good enough based on the results of out of sample validation (Xing et al., 2011; Wang et al., 2011). The mean normalized error and correlation coefficients are selected as indices of prediction performance. In our previous paper (Xing et al., 2011), we showed that the normalized mean error first decreases and then gradually remains stable, with the increase of scenario number. In contrast, the correlation coefficient first increases and then gradually becomes stable. We used a criterion that mean normalized error < 1% and correlation coefficient > 0.99, and determined that 30 and 50 scenarios were required to construct the conventional RSM for 2 and 3 variables, respectively. Therefore, for the simplified case, N=50, and M=30. The required scenario number for the simplified case is therefore 1 (the base case) + 50 (scenarios for each single region) * 3 (number of regions) + 30 (scenarios for the total precursor emissions in all regions) = 181. (Page 6, Line 13-31 in the revised manuscript)

Variable number = 2

Variable number = 4

Variable number = 6

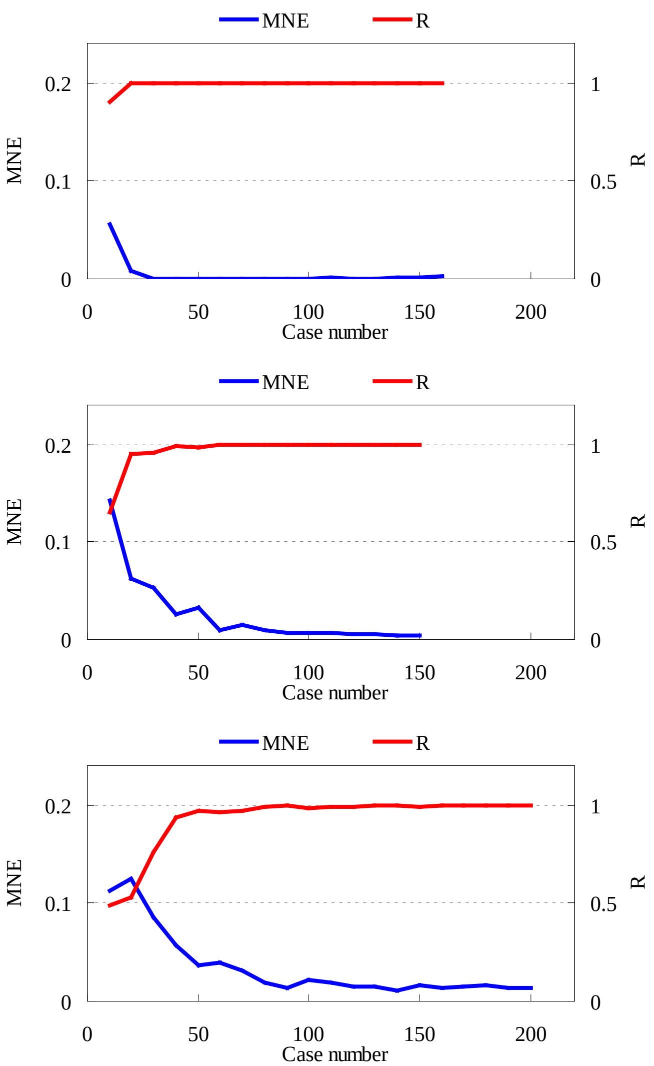

Figure R2. Evolution of the prediction performance with the increase of scenario number based on computational experiments. The figure is adapted from Figure 7 of Xing et al. (2011).

Reference:

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.

A short summary of the reason why « Xing (2011) indicated that the nonlinearity in atmospheric responses could not be captured in metropolitan regions unless fourth or higher order equations were used » (P3, L29-31) should also be included since that reference is in the grey literature, and in Chinese.

Response: We thank the editor for this comment. Xing (2011) tried to construct the relationship between O_3 concentration and the emissions of NO_X and NMVOC using polynomial equations. The general relationship is expressed by Eq. (R1) and Eq. (R2).

$$Conc\_Ozone=f(Emis\_NOx, Emis\_NMVOC) \tag{R1}$$

$$f(x,y)=\sum_{n=0}^{N}\sum_{m=0}^{n} a_{n,m}\cdot x^n y^m \tag{R2}$$

where *Conc_Ozone*, *Emis_NOx*, and *Emis_NMVOC* are the O_3 concentration, NO_X emissions, and VOC emissions in a metropolitan region, respectively.

Xing (2011) performed 30 CMAQ simulations and fitted the simulated results using polynomials of 2nd – 5th order. The relationship was also constructed using conventional RSM technique, which had been thoroughly evaluated and was used to represent actual CMAQ simulation results. Using the fitted equations, Xing (2011) predicted the O_3 concentrations in response to the continuous changes of NO_X and NMVOC emissions from zero to 200%, as shown in Figure R3. It can be seen that the equations of 2nd and 3rd order fail to reproduce the shape of the isopleths, while the 4th and 5th order equations behave fairly well. Therefore, Xing (2011) concluded that response of O_3 concentration to NO_X and NMVOC emissions could not be captured unless fourth or higher order equations are used. Considering that the isopleths of $PM_{2.5}$ in response to precursor emissions could have quite similar shapes to those of O_3 (which is also confirmed by Figure 4 of the revised manuscript), Xing (2011) believes this conclusion could be extrapolated to $PM_{2.5}$. We have added explanations above accordingly in the Supporting Information (from Page 2, Line 16 to Page 3, Line 17) and given a brief instruction in the main text (Page 3, Line 29-32) which refers the readers to these explanations.

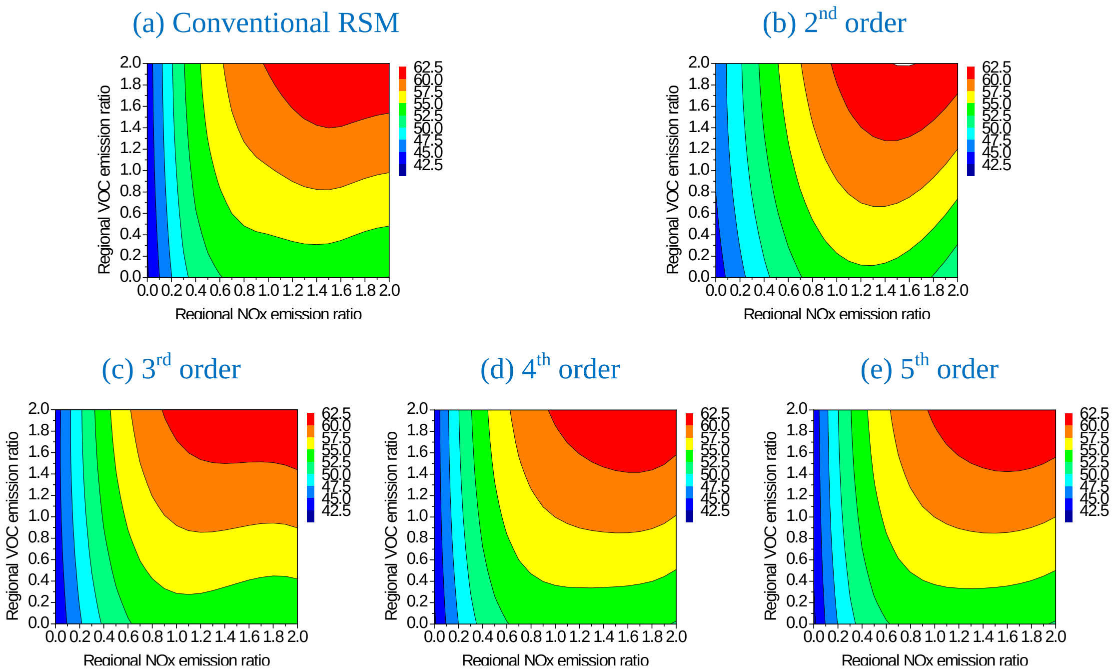

Figure R3. Comparison of the 2-D isopleths of O_3 concentrations in response to the changes of NO_X and NMVOC emissions predicted by the conventional RSM technique as well as polynomial equations of 2nd – 5th order.

Reference:

Xing, J.: Study on the nonlinear responses of air quality to primary pollutant emissions, Doctor thesis, School of Environment, Tsinghua University, Beijing, China, 138 pp., 2011 (in Chinese).

Both reviewers requested more details on the rationale and implication of neglecting interactions of transported and returning precursors, this hypothesis is indeed now better stated in the revised manuscript (P8L9-10 and P9L11-12). However, the possible implication of such a hypothesis is not given in the revised manuscript. Can you think of a possible quantification that would support neglecting this process?

Response: We appreciate the editor's valuable comments very much. We estimated the contribution of the neglected processes for the case study over the Yangtze River Delta region (see Section 2.2 for details of the case study), which proved the rationality of both assumptions mentioned in the editor's comments.

Assumption 1:

We review the 1st assumption briefly as follows. (Page 8, Line 9-15 in the revised manuscript)

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 neglected in this study.

In order to demonstrate the rationality of this assumption, we try to estimate the contribution of the “indirect” pathway to the total changes of $PM_{2.5}$ concentrations. The estimation is done in four stages. Note that the values of emissions/concentrations in the following paragraphs are all averages of January and August, 2010.

Firstly, we assume that the concentrations of NO_X, SO_2, and NH_3 in Shanghai are all reduced by 50%. Based on Eq. (2) and Eq. (3), this reduction corresponds to reductions of 55%, 62%, and 53% in the emissions of NO_X, SO_2, and NH_3 in Shanghai, respectively.

Secondly, we estimate how much the transported precursors could affect the precursor concentrations in another region (we use Jiangsu as example). Using Eq. (5) and Eq. (6), we estimate that, as a result of the reductions in Shanghai, the concentrations of NO_X, SO_2, and NH_3 in Jiangsu would decrease by about 3.0%, 1.4% and 0.1%, respectively.

Thirdly, we try to quantify how much the precursors transported to Jiangsu could in turn affect the $PM_{2.5}$ concentrations in Shanghai. The decline in precursor concentrations in Jiangsu is considered to be equivalent to a certain reduction in precursor emissions in Jiangsu. Based on Eq. (2) and Eq. (3), we estimate that the equivalent “pseudo” reductions in Jiangsu’s emissions of NO_X, SO_2, and NH_3 are 3.3%, 1.7%, and 0.1%, respectively. According to Eq. (4), such an emission reduction in Jiangsu could in turn decrease the $PM_{2.5}$ concentration in Shanghai by 0.01 $\mu g\ m^{-3}$.

Fourthly, we integrate the effects of the precursors transported to all outer regions. Similar to Jiangsu, we estimate that the decline in precursor concentrations in Zhejiang and Others could in turn reduce the $PM_{2.5}$ concentration in Shanghai by 0.02 $\mu g\ m^{-3}$ and 0.01 $\mu g\ m^{-3}$, respectively. Therefore, the total $PM_{2.5}$ reduction in Shanghai through the “indirect” pathway is estimated at about 0.04 $\mu g\ m^{-3}$, accounting for only about 1.3% of the total $PM_{2.5}$ reduction (2.67 $\mu g\ m^{-3}$).

Following the same procedure, if the precursor concentrations in Jiangsu and Zhejiang are reduced by 50%, respectively, we estimate that the "indirect" pathway would account for about 1.7% and 1.0% of the total $PM_{2.5}$ reduction, respectively. These results confirm our assumption that the "indirect" pathway is negligible. We have described the key conclusion in the main text (Page 8, Line 15-19), and provided a detailed explanation in the Supporting Information (Page 4, Line 4-31). The added text in the main text is shown as follows:

For the case study over the YRD region (see details of the case study in Sect. 2.2), we estimate that, when the concentrations of NO_X, SO_2, and NH_3 in a specific region (Shanghai, Jiangsu, or Zhejiang) are all reduced 50%, the "indirect" pathway could only account for less than 2% of the total $PM_{2.5}$ reduction (see details in the Supporting Information). This confirms our assumption that the "indirect" pathway is negligible.

Assumption 2:

We review the 2st assumption briefly as follows. (Page 9, Line 17-23 in the revised manuscript)

Strictly speaking, $[PM_{2.5}\_Trans]_{B\rightarrow A}$ and $[PM_{2.5}\_Trans]_{C\rightarrow A}$ could interact with each other. In other words, the changes of precursor emissions in Region C might affect the formation of secondary $PM_{2.5}$ in Region B, which further affects the transport of secondary $PM_{2.5}$ from Region B to Region A. Eq. (9) and Eq. (10) implies an assumption that $[PM_{2.5}\_Trans]_{B\rightarrow A}$ depends only on the precursor emissions in Region B, and is independent of precursor emissions in other regions. That is, the interaction between $[PM_{2.5}\_Trans]_{B\rightarrow A}$ and $[PM_{2.5}\_Trans]_{C\rightarrow A}$ is neglected.

In order to demonstrate the rationality of this assumption, we try to prove that the precursor emissions in Jiangsu and Others have little effect on $[PM_{2.5}\_Trans]_{Zhejiang\rightarrow Shanghai}$, i.e., the change of $PM_{2.5}$ concentration in Shanghai affected by the changes of precursor emissions in Zhejiang through the transport of secondary $PM_{2.5}$. We designed several pairs of CMAQ simulations, as summarized in Table R1. The two cases in the same pair differ in the emissions of gaseous precursor in Zhejiang. Different pairs are distinguished by different precursor emissions in Jiangsu and Others. Therefore, using the two cases in each pair, we can calculate the value of $[PM_{2.5}\_Trans]_{Zhejiang\rightarrow Shanghai}$ under certain emission rates in Jiangsu and

Others. Then, by comparing all the values $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ calculated above, we can evaluate the effect of precursor emissions in Jiangsu and Others on $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$.

Table R1. Description of the CMAQ simulations designed to test the 2nd assumption. The simulation period is August, 2010.

Pair NO.	Case NO.	Description of the cases	Objective of the cases
1	1	The CMAQ base case.	Calculate $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ when the emissions in the other regions except Zhejiang stays the base-case levels.
	2	The emissions of NO_X, SO_2, and NH_3 in Zhejiang are reduced by 50%, while the emissions in other regions remain the base-case levels.	
2	3	The emissions of NO_X, SO_2, and NH_3 in Jiangsu are reduced by 50%, while the emissions in other regions remain the base-case levels.	Calculate $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ when the emissions of NO_X, SO_2, and NH_3 in Jiangsu are reduced by 50%.
	4	The emissions of NO_X, SO_2, and NH_3 in Zhejiang and Jiangsu are reduced by 50%, while the emissions in other regions remain the base-case levels.	
3	5	The emissions of NO_X, SO_2, and NH_3 in Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	Calculate $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ when the emissions of NO_X, SO_2, and NH_3 in Others are reduced by 50%.
	6	The emissions of NO_X, SO_2, and NH_3 in Zhejiang and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	
4	7	The emissions of NO_X in Jiangsu and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	Calculate $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ when the emissions of NO_X in Jiangsu and Others are reduced by 50%.
	8	The emissions of NO_X, SO_2, and NH_3 in Zhejiang are reduced by 50%, and the emissions of NO_X in Jiangsu and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	
5	9	The emissions of SO_2 in Jiangsu and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	Calculate $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ when the emissions of SO_2 in Jiangsu and Others are reduced by 50%.
	10	The emissions of NO_X, SO_2, and NH_3 in Zhejiang are reduced by 50%, and the emissions of SO_2 in Jiangsu	

		and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	
6	11	The emissions of NH_3 in Jiangsu and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	Calculate $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ when the emissions of NH_3 in Jiangsu and Others are reduced by 50%.
	12	The emissions of NO_X, SO_2, and NH_3 in Zhejiang are reduced by 50%, and the emissions of NH_3 in Jiangsu and Others are reduced by 50%, while the emissions in other regions remain the base-case levels.	

Using Case 1-2 and Eq. (7, 8), we estimate that the change of $PM_{2.5}$ concentration in Shanghai affected by the reduction of precursor emissions in Zhejiang through the transport of secondary $PM_{2.5}$, i.e., $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$, is -3.92 μg m^{-3}. Using Case 3-4 and Eq. (7, 8), it can be estimated that, when the emissions of NO_X, SO_2, and NH_3 in Jiangsu are reduced by 50%, $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ is -3.91 μg m^{-3}. Similarly, we could estimate the values of $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ in various circumstances, as summarized in Table R2. It can be seen that the changes of precursor emissions in Jiangsu and Others could only change $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ by less than 1%. This supports our assumption that $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ depends only on the precursor emissions in Zhejiang, and is independent of precursor emissions in other regions (Jiangsu and Others). We have described the key conclusion in the main text (Page 9, Line 23-28), and provided a detailed explanation in the Supporting Information (from Page 5, Line 3 to Page 7, Line 2). The added text in the main text is shown as follows:

For the case study over the YRD region, we estimate that, the reduction of precursor emissions in Jiangsu and Others by 50% could only change $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ (i.e., the change of $PM_{2.5}$ concentration in Shanghai affected by the changes of precursor emissions in Zhejiang through the transport of secondary $PM_{2.5}$) by less than 1% (see details in the Supporting Information). This confirms the above-mentioned assumption.

Table R2. Values of $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$ in various circumstances.

Emissions in the other regions except Zhejiang	Values of $[PM_{2.5}\_Trans]_{Zhejiang \rightarrow Shanghai}$	Corresponding CMAQ simulations

The base-case levels.	-3.92	Pair 1 (Case 1-2)
The emissions of NO_X, SO_2, and NH_3 in Jiangsu are reduced by 50%.	-3.91	Pair 2 (Case 3-4)
The emissions of NO_X, SO_2, and NH_3 in Others are reduced by 50%.	-3.89	Pair 3 (Case 5-6)
The emissions of NO_X in Jiangsu and Others are reduced by 50%.	-3.91	Pair 4 (Case 7-8)
The emissions of SO_2 in Jiangsu and Others are reduced by 50%.	-3.93	Pair 5 (Case 9-10)
The emissions of NH_3 in Jiangsu and Others are reduced by 50%.	-3.89	Pair 6 (Case 11-12)