Supplement of Geosci. Model Dev., 8, 115–128, 2015
http://www.geosci-model-dev.net/8/115/2015/
doi:10.5194/gmd-8-115-2015-supplement
© Author(s) 2015. CC Attribution 3.0 License.

Geoscientific Model Development
Open Access

Supplement of

Assessing the nonlinear response of fine particles to precursor emissions: development and application of an extended response surface modeling technique v1.0

B. Zhao et al.

Correspondence to: S. X. Wang (shxwang@ tsinghua.edu.cn)

1 Supplementary information for the introduction section

1.1 Explanation to "the number of model scenarios required to build the conventional RSM depends on the variable number via an equation of fourth or higher order"

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 S1). Then we fitted the dots using polynomials of 2^{nd} – 5^{th} order (shown as the lines in Figure S1). 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.

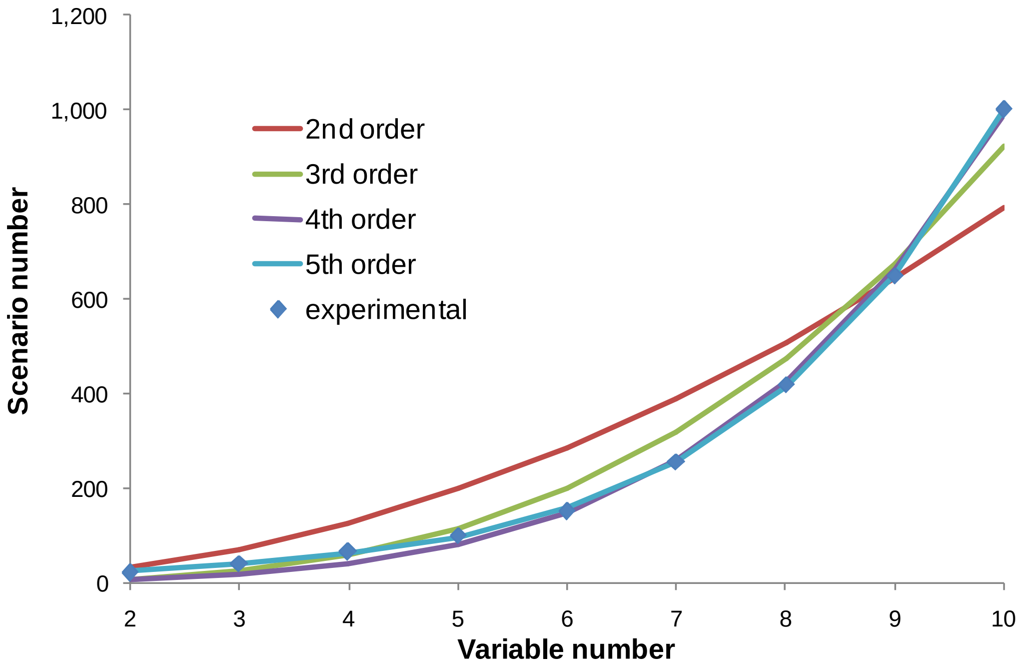


Figure S 1. Number of scenarios required to build the conventional RSM based on numerical experiments (the dots) and the fits to polynomials of 2^{nd} – 5^{th} order (the lines).

1.2 Explanation to “Xing (2011) indicated that the nonlinearity in atmospheric responses could not be captured in metropolitan regions unless fourth or higher order equations were used”

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. (S1) and Eq. (S2).

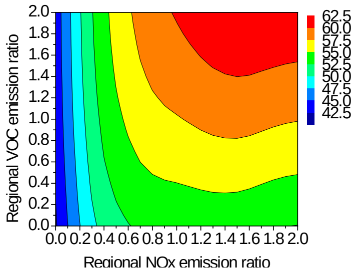
$$Conc\_Ozone=f(Emis\_NOx, Emis\_NMVOC) \quad \text{(S1)}$$

$$f(x,y)=\sum_{n=0}^{N}\sum_{m=0}^{n} a_{n,m}\cdot x^n y^m \quad \text{(S2)}$$

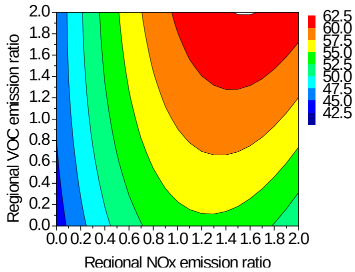
where *Conc_Ozone*, *Emis_NOx*, and *Emis_NMVOC* are the O_3 concentration, NO_X emissions, and VOC emissions in a metropolitan region, respectively.

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 S2. 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 in the main text), Xing (2011) believes this conclusion could be extrapolated to $PM_{2.5}$.

(a) Conventional RSM



(b) 2nd order



(c) 3rd order

(d) 4th order

(e) 5th order

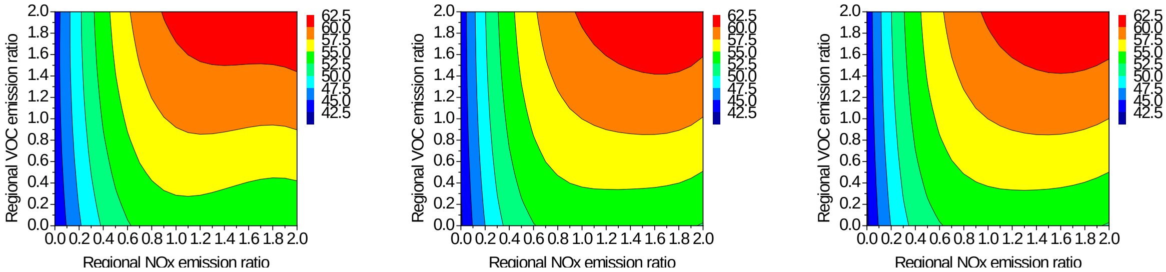


Figure S 2. Comparison of the 2-D isopleths of O_3 concentrations in response to the changes of NO_X and NMVOC emissions predicted by the conventional RSM technique as well as polynomial equations of 2nd – 5th order.

2 Supplementary information for the methodology section

2.1 Rationality of the assumption described between Eq. (6) and Eq. (7) of the main text

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 μg m^{-3} and 0.01 μg m^{-3}, respectively. Therefore, the total $PM_{2.5}$ reduction in Shanghai through the “indirect” pathway is estimated at about 0.04 μg m^{-3}, accounting for only about 1.3% of the total $PM_{2.5}$ reduction (2.67 μg 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.

2.2 Rationality of the assumption described between Eq. (10) and Eq. (11) of the main text

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 S1. 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 S 1. 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	Calculate

		reduced by 50%, while the emissions in other regions remain the base-case levels.	$[PM_{2.5}\_Trans]_{Zhejiang \to Shanghai}$ 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 $[PM_{2.5}\_Trans]_{Zhejiang \to Shanghai}$ 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 $[PM_{2.5}\_Trans]_{Zhejiang \to Shanghai}$ 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 $[PM_{2.5}\_Trans]_{Zhejiang \to Shanghai}$ 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 $[PM_{2.5}\_Trans]_{Zhejiang \to Shanghai}$ 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., $[PM_{2.5}\_Trans]_{Zhejiang \to Shanghai}$, 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 S2. 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).

Table S 2. 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)

3 Evaluation of WRF/CMAQ performance

The meteorological prediction lays the foundation for the air quality simulation. In this study, the meteorological parameters simulated by WRF were compared with the observational data obtained from the National Climatic Data Center (NCDC), where hourly or every third hour observations are available for 57 sites scattering within the innermost domain. Due to the limited observational data available, the statistical evaluation was restricted to the temperature at 2 m, wind speed and wind direction at 10 m, and humidity at 2 m. The statistical indices used include the bias, gross error (GE), the root mean square error (RMSE), and the index of agreement (IOA). A detailed explanation of these indices can be found in Baker (2004).

Table S3 lists the model performance statistics and the benchmarks suggested by Emery et al. (2001). These benchmark values were derived based on performance statistics of the Fifth-Generation NCAR/Penn State Mesoscale Model (MM5) from a number of studies over the U.S. domain (mostly at grid resolution of 12km or 4km), and have been widely accepted in many regional air quality modeling studies. We expect these standards should also be applicable in our simulation domain. For wind speed and humidity, all statistical indices are within the benchmark range. For temperature, the bias for the August simulation slightly exceeds this benchmark (-0.61K vs ±0.5K), but the bias for January, and the values of GE and IOA are all within the benchmarks, indicating an acceptable performance. While the biases for wind direction are below 10 degrees, the GEs are slightly larger than the 30 degrees benchmark value. As indicated in the previous research (Wang et al., 2010; Zhang et al., 2006), the large gross errors may result from a caveat in treating the wind direction vector as a scalar in the evaluation method, where error calculations are performed inconsistently when determining the differences between simulated and observed values. On a wind rose plot, both 0 and 360 degrees represent the direction of north. Therefore, for instance, if the observed wind is in the north direction and the predicted value is 190 degrees, the actual difference can be 190-0=190 degrees or 360-190=170 degrees. If the first value (i.e., 190) is selected in calculating the gross errors, this increases the actual difference in the gross errors by 20 degrees. The observed temperature and humidity are reproduced quite well, with all the statistical indices significantly better than the benchmark values. In summary, these statistics indicate an overall satisfactory performance of meteorological predictions.

Table S 3. Statistical results for the comparison of simulated meteorological parameters with NCDC observations.

Item	Wind speed (m/s)				Wind direction (deg)		Temperature (K)				Humidity (g/kg)			
	Bias	GE	RMSE	IOA	Bias	GE	Bias	GE	RMSE	IOA	Bias	GE	RMSE	IOA
Ref.	<±0.5		<2	>0.6	<±10	<30	<±0.5	<2		>0.8	<±1	<2		>0.6
Jan	0.41	1.16	1.52	0.81	4.02	33.00	0.46	1.35	1.74	0.93	0.28	0.56	0.76	0.85
Aug	0.40	1.13	1.47	0.78	-1.21	36.80	-0.61	1.58	2.03	0.91	0.73	1.47	1.9	0.73

During the simulation period, the Ministry of Environmental Protection of China (MEP) reported daily primary pollutant and its air pollution index (API) for 12 major cities in the innermost domain on its official website (http://datacenter.mep.gov.cn). Using each city's API and primary pollutant, it is possible to back-calculate the daily average concentration for the primary pollutant. PM_{10} is the primary air pollutant on most of the days. The simulated and

API-derived PM_{10} concentrations are therefore compared, as shown in Fig. S1. The simulated values used in the comparison are the average concentrations of the urban area (see Fig. 2 in the main text). The observation of a specific city was adopted if the API-derived PM_{10} concentrations were available for more than 70% days during the simulation period (62 days in total).

A number of statistical indices including mean observation, mean simulation, normalized mean bias (NMB), normalized mean error (NME), mean fractional bias (MFB), and mean fractional error (MFE), were calculated for the cities to give a quantitative assessment of the model performance, as shown in Table S4. The benchmarks proposed by Boylan (2005) and Morris et al. (2005) are also listed in Table S4. It can be seen that the PM_{10} concentrations are underestimated both months. This underestimation may be mainly attributable to the exclusion of fugitive dust emissions, and the underestimation of secondary organic aerosols (SOA). All the statistical indices meet the criteria, indicating a satisfactory modeling performance.

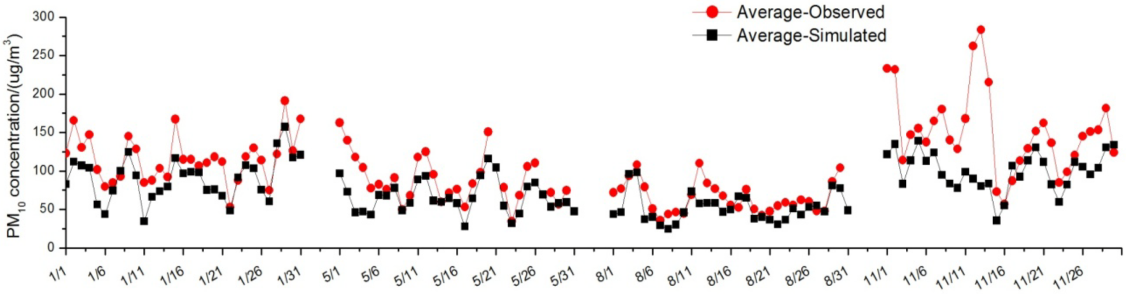


Figure S 3. Comparison of PM_{10} simulation with API-derived observation in 12 major cities

Table S 4. Statistical results for the comparison of simulated PM_{10} concentrations with API-derived observations.

Month	Mean observation ($\mu g/m^3$)	Mean simulation ($\mu g/m^3$)	Normalized mean bias (NMB)	Normalized mean error (NME)	Mean fractional bias (MFB)	Mean fractional error (MFE)
Benchmark					±50-60%	75%
Jan	116.0	90.3	-22.2%	31.7%	-26.6%	36.9%
Aug	65.3	51.7	-20.8%	36.5%	-26.9%	43.3%

The observational data of fine particles are very sparse and not publicly available during the simulation period (January and August, 2010). In order to evaluate the model performance in simulating fine particle pollution, we conducted extra simulations for two field campaign periods (July 15-30 and December 15-30) in 2011 and compared the simulated $PM_{2.5}$ concentrations with observations (unpublished data of Tsinghua University), as shown in Fig. S2. Note that the observations are not available in January for the Shanghai-Xushui site. The comparison results indicate that the modeling system can capture the temporal variation of $PM_{2.5}$ concentrations fairly well. The simulated average concentrations agree very well with observations for most periods, with NMBs ranging between -15% and +24%. Relatively large underestimation occurs in Shanghai-Pudong site during July (-38%) and relatively large overestimation occurs in Nanjing during December (+58%).

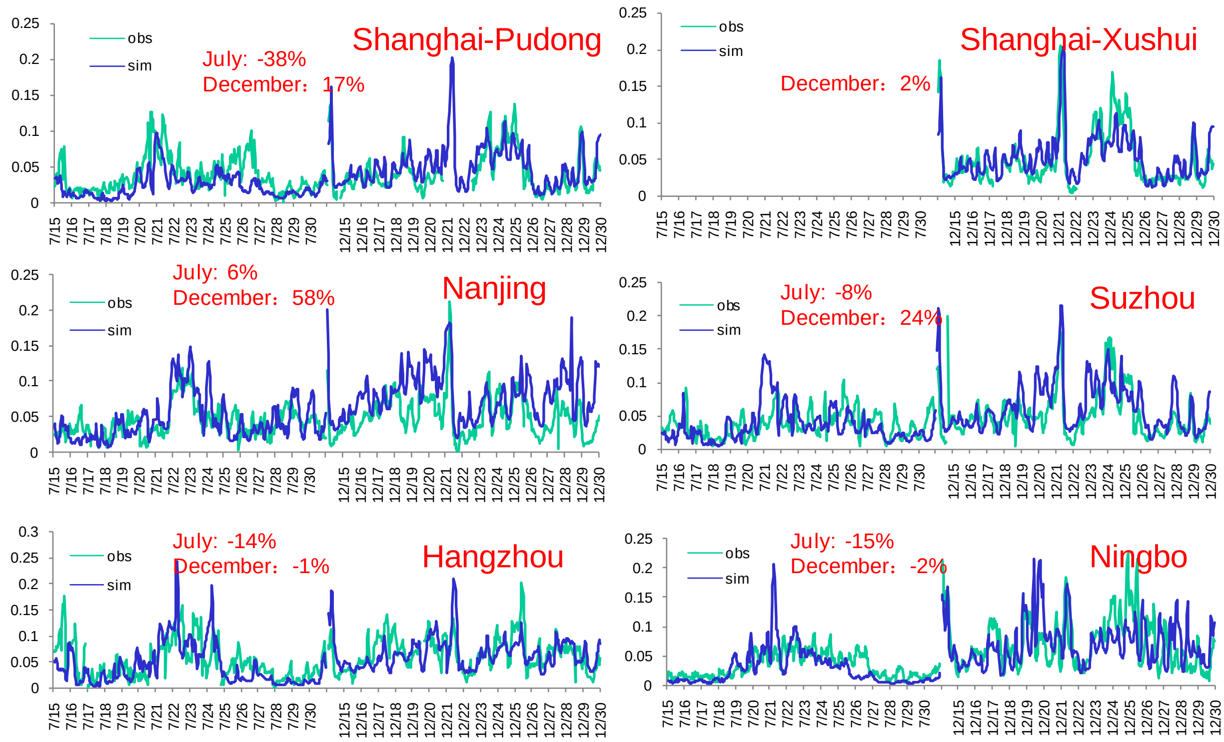


Figure S 4. Comparison of simulated hourly $PM_{2.5}$ concentrations with observations during a field campaign in 2011 (unit: $\mu g/m^3$). The percentage in the figure represents the normalized mean bias (NMB).

The simulated concentrations of inorganic aerosols are compared with the observational data at the Shanghai-Xushui site during December, 2011 (Fig. S3). It can be seen that the modeling system can capture the temporal trends of SO_4^{2-}, NO_3^-, and NH_4^+ fairly well. There is an overestimation for NO_3^- (25%), underestimation for SO_4^{2-} (-37%), and good agreement for NH_4^+ (14%). The overestimation of NO_3^- and underestimation for SO_4^{2-} to a certain extent are consistent with previous studies, probably attributable to the lack of some chemical formation pathways in the modeling system (Wang et al., 2011; Wang et al., 2013).

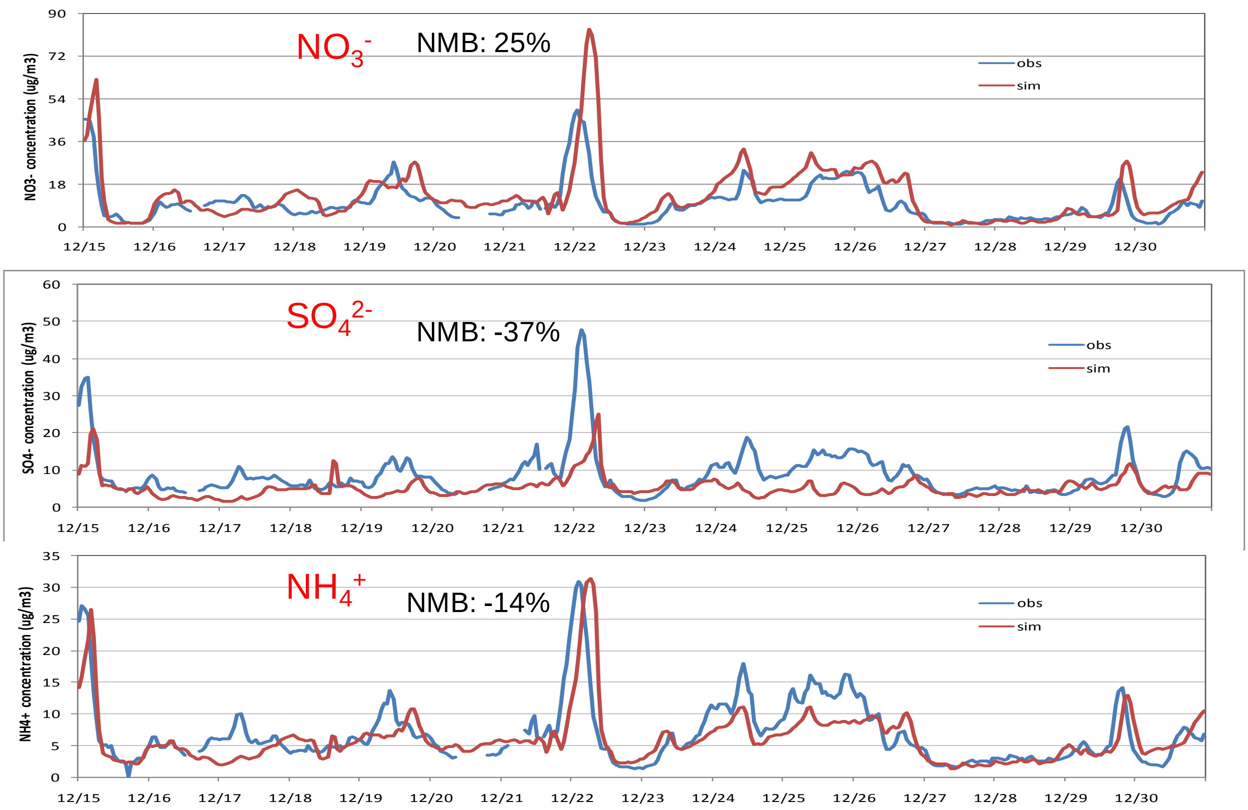


Figure S 5. Comparison of simulated inorganic aerosol concentrations with observations at the Shanghai-Xushui site during a field campaign in 2011.

4 Validation of ERSM performance

Table S 5. Description of out-of-sample scenarios

Case number	Description
1-6	Control variables of gaseous precursors in Shanghai change but the other variables stay the same as the base case. For case 1-3, the emission ratios (defined as the ratios of the changed emissions to the emissions in the base case) of all control variables of gaseous precursors in Shanghai are set to 0.1, 0.5, and 1.45, respectively. Case 4-6 are generated

	randomly by applying LHS method for the control variables of gaseous precursors in Shanghai.
7-12	The same as case 1-6 but for Jiangsu.
13-18	The same as case 1-6 but for Zhejiang.
19-24	The same as case 1-6 but for Others.
25-32	Control variables of gaseous precursors change but those of primary $PM_{2.5}$ stay the same as the base case. For case 25-27, the emission ratios of all control variables of gaseous precursors are set to 0.1, 0.5, and 1.45, respectively. Case 28-32 are generated randomly by applying LHS method for the control variables of gaseous precursors.
33-36	Control variables of primary $PM_{2.5}$ change randomly (with LHS method applied) but those of gaseous precursors stay the same as the base case.
37-40	Case 37-40 are generated randomly by applying LHS method for all control variables.

Table S 6. Comparison of $PM_{2.5}$ concentrations predicted by the ERSM technique with out-of-sample CMAQ simulations in January.

Case	ERSM prediction			CMAQ simulation			Normalized Error (NE)		
number	Shanghai	Jiangsu	Zhejiang	Shanghai	Jiangsu	Zhejiang	Shanghai	Jiangsu	Zhejiang
1	59.3	80.9	70.7	61.7	80.8	70.8	3.9%	0.1%	0.2%
2	62.9	80.6	71.1	64.3	81.0	71.2	2.2%	0.5%	0.1%
3	67.2	81.0	71.2	65.7	80.8	71.1	2.3%	0.2%	0.1%
4	63.8	80.8	71.1	63.8	80.8	71.1	0.0%	0.0%	0.0%
5	63.3	80.1	71.1	65.0	80.9	71.1	2.6%	1.0%	0.1%
6	65.0	81.2	71.4	66.2	81.2	71.4	1.9%	0.1%	0.1%
7	63.5	73.9	69.0	63.9	75.2	69.3	0.6%	1.8%	0.4%
8	64.7	78.6	70.4	64.8	80.2	70.6	0.3%	2.0%	0.3%
9	65.6	82.8	71.6	65.4	81.1	71.4	0.4%	2.0%	0.3%
10	64.5	79.1	70.4	64.6	79.2	70.6	0.1%	0.1%	0.2%
11	64.8	78.9	70.9	65.0	80.7	71.1	0.4%	2.2%	0.3%
12	65.7	81.5	71.2	65.8	82.4	71.3	0.1%	1.2%	0.1%
13	63.9	78.2	60.4	64.0	78.3	63.2	0.2%	0.2%	4.3%
14	64.8	80.0	68.0	64.9	80.1	69.0	0.1%	0.2%	1.5%
15	65.4	81.2	73.3	65.3	81.0	72.5	0.2%	0.2%	1.1%
16	64.8	80.1	68.0	64.8	80.2	68.2	0.1%	0.1%	0.3%
17	65.1	80.7	68.9	65.2	80.8	70.5	0.1%	0.2%	2.3%
18	65.2	80.6	71.5	65.2	80.6	72.0	0.0%	0.1%	0.7%
19	64.2	79.3	69.5	64.3	79.4	69.5	0.2%	0.1%	0.1%
20	64.7	80.2	70.4	64.8	80.2	70.4	0.1%	0.1%	0.1%
21	65.6	81.5	71.8	65.5	81.4	71.7	0.2%	0.1%	0.1%
22	64.8	80.4	70.6	64.9	80.5	70.6	0.1%	0.0%	0.0%
23	65.1	80.8	71.0	65.2	80.8	71.1	0.1%	0.1%	0.1%
24	65.1	80.6	70.8	65.1	80.6	70.9	0.0%	0.0%	0.0%
25	52.4	65.9	52.4	53.9	66.6	55.3	2.8%	1.1%	5.2%
26	61.2	76.5	66.2	62.7	78.0	66.7	2.5%	2.0%	0.9%
27	67.9	83.7	74.7	66.2	81.5	73.0	2.7%	2.7%	2.4%

28	63.6	77.4	67.8	64.5	79.7	68.0	1.3%	3.0%	0.4%
29	64.4	80.3	69.1	65.1	80.5	70.5	1.2%	0.3%	2.0%
30	62.5	77.6	59.4	63.6	77.7	58.6	1.7%	0.1%	1.4%
31	63.5	81.1	73.2	63.0	80.7	72.3	0.9%	0.4%	1.2%
32	64.6	78.5	70.4	65.4	81.0	71.8	1.2%	3.0%	2.0%
33	59.8	69.3	78.9	59.8	69.3	78.9	0.0%	0.0%	0.0%
34	53.9	78.0	62.8	53.9	78.0	62.8	0.1%	0.1%	0.1%
35	66.4	73.7	66.1	66.4	73.7	66.1	0.0%	0.0%	0.0%
36	58.0	82.3	72.2	58.1	82.3	72.2	0.0%	0.0%	0.0%
37	44.2	66.6	50.6	45.1	68.3	52.4	2.1%	2.4%	3.4%
38	45.6	74.3	65.4	47.7	75.2	66.1	4.5%	1.2%	1.1%
39	66.7	65.6	71.6	66.4	65.6	73.7	0.4%	0.1%	2.8%
40	61.3	83.6	67.7	61.9	82.9	67.5	1.0%	0.8%	0.3%

Table S 7. Comparison of $PM_{2.5}$ concentrations predicted by the ERSM technique with out-of-sample CMAQ simulations in August.

Case	ERSM prediction			CMAQ simulation			Normalized Error (NE)		
number	Shanghai	Jiangsu	Zhejiang	Shanghai	Jiangsu	Zhejiang	Shanghai	Jiangsu	Zhejiang
1	32.0	54.5	38.7	32.1	54.5	39.5	0.1%	0.1%	2.0%
2	36.2	55.0	39.0	36.1	55.0	39.7	0.3%	0.0%	1.7%
3	40.3	55.6	39.1	40.2	55.5	40.0	0.2%	0.1%	2.1%
4	36.2	55.1	39.2	36.0	55.1	39.7	0.4%	0.0%	1.5%
5	37.5	55.2	39.1	38.0	55.2	39.7	1.3%	0.0%	1.6%
6	38.7	55.3	39.2	38.5	55.3	39.8	0.7%	0.0%	1.4%
7	36.7	41.2	38.1	36.7	41.4	38.7	0.1%	0.5%	1.6%
8	37.8	49.0	38.6	37.8	49.0	39.3	0.0%	0.1%	1.8%
9	39.4	59.9	39.5	39.3	59.8	40.2	0.2%	0.2%	1.9%
10	38.1	50.2	38.8	38.1	50.1	39.5	0.1%	0.3%	1.9%
11	38.3	52.7	38.9	38.3	52.3	39.6	0.0%	0.8%	1.8%
12	38.2	54.3	38.9	38.2	54.4	39.5	0.0%	0.3%	1.3%
13	31.7	49.1	27.3	31.7	49.2	28.2	0.1%	0.3%	3.0%
14	34.8	52.3	33.9	34.8	52.2	33.8	0.0%	0.3%	0.3%
15	41.8	57.9	43.9	41.8	57.7	44.4	0.0%	0.4%	1.0%
16	36.6	53.6	35.7	36.6	53.6	35.6	0.2%	0.1%	0.5%
17	38.2	54.6	37.2	38.2	54.6	37.5	0.0%	0.1%	0.8%
18	34.8	52.6	37.0	34.8	52.5	36.4	0.1%	0.0%	1.5%
19	36.5	53.1	37.1	36.5	53.0	37.7	0.0%	0.2%	1.6%
20	37.5	54.2	37.9	37.5	54.1	38.7	0.0%	0.1%	1.9%
21	39.8	56.5	40.2	39.8	56.5	40.9	0.0%	0.0%	1.8%
22	38.1	54.7	38.5	38.1	54.7	39.3	0.1%	0.0%	1.9%
23	38.7	55.0	39.0	38.7	55.0	39.6	0.0%	0.0%	1.6%
24	37.7	54.6	38.1	37.6	54.6	38.7	0.0%	0.1%	1.6%
25	21.1	31.5	23.2	23.4	34.1	25.7	10.2%	7.7%	9.6%
26	30.9	44.9	31.7	30.6	44.2	32.0	1.1%	1.5%	0.8%

27	45.8	64.1	45.5	44.8	63.1	45.9	2.1%	1.6%	0.9%
28	35.4	50.4	35.6	35.3	50.4	36.4	0.4%	0.2%	2.2%
29	36.3	49.4	38.9	36.1	49.7	38.4	0.7%	0.6%	1.4%
30	33.3	48.5	28.3	32.8	48.5	28.7	1.6%	0.1%	1.2%
31	34.9	54.1	38.4	34.7	54.1	40.1	0.8%	0.0%	4.1%
32	38.7	51.9	38.2	37.9	52.0	39.0	2.0%	0.2%	2.2%
33	36.3	47.5	46.1	36.3	47.5	46.0	0.0%	0.1%	0.2%
34	31.2	53.3	33.8	31.3	53.4	33.9	0.1%	0.1%	0.2%
35	38.9	49.7	36.2	38.9	49.8	36.2	0.0%	0.0%	0.0%
36	34.4	56.4	40.5	34.4	56.4	40.4	0.0%	0.1%	0.1%
37	19.5	38.4	21.2	20.0	38.2	21.6	2.7%	0.5%	1.8%
38	24.2	41.9	32.6	23.2	42.0	32.9	4.2%	0.2%	0.9%
39	37.6	39.1	40.3	36.5	38.2	39.9	2.9%	2.5%	1.0%
40	32.0	52.9	34.1	32.0	52.8	34.7	0.0%	0.1%	1.8%

1

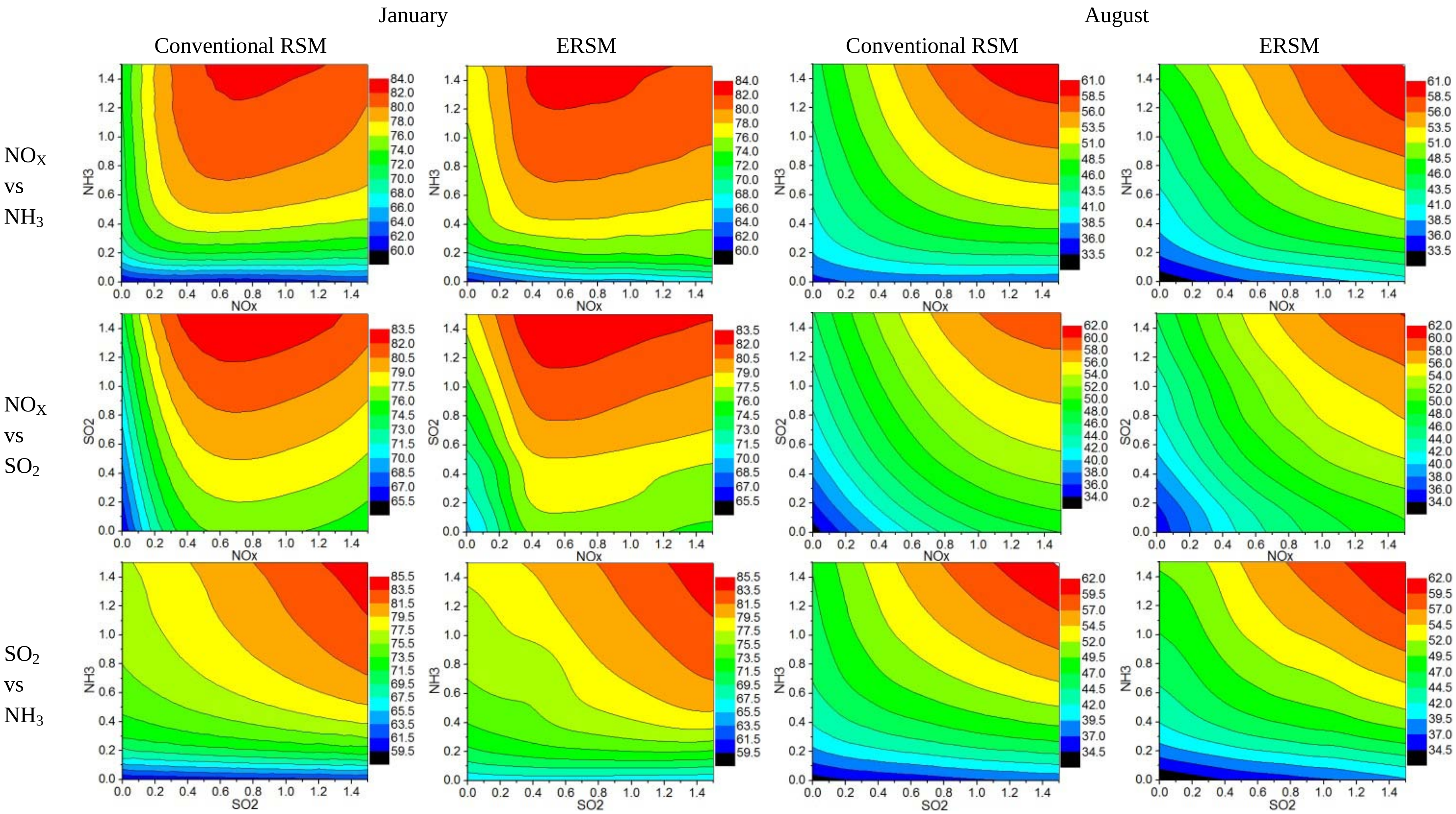


Figure S 6. The same as Fig. 4 but for the region of Jiangsu.

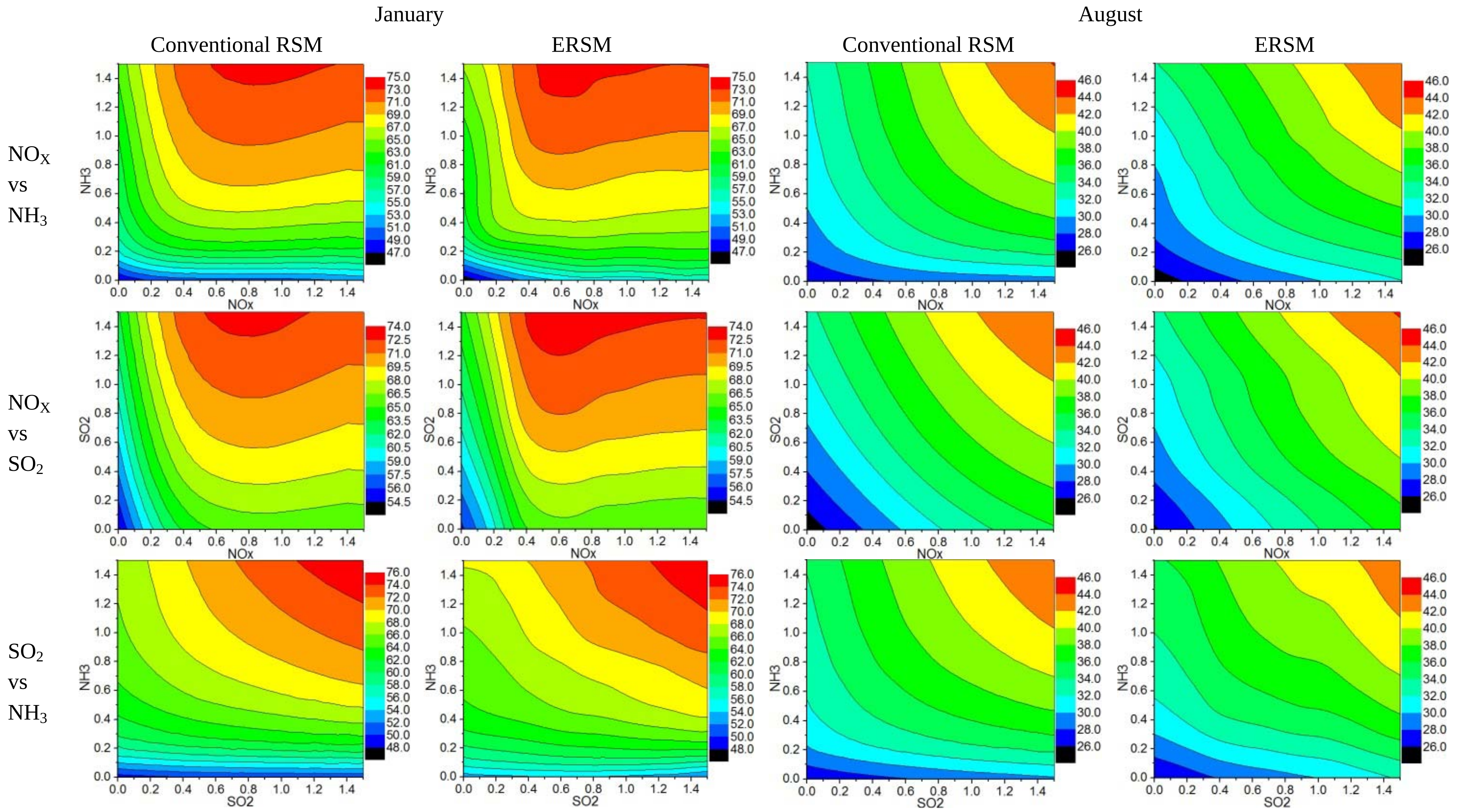


Figure S 7. The same as Fig. 4 but for the region of Zhejiang.

5 Response of $PM_{2.5}$, SO_4^{2-} and NO_3^- to precursor emissions.

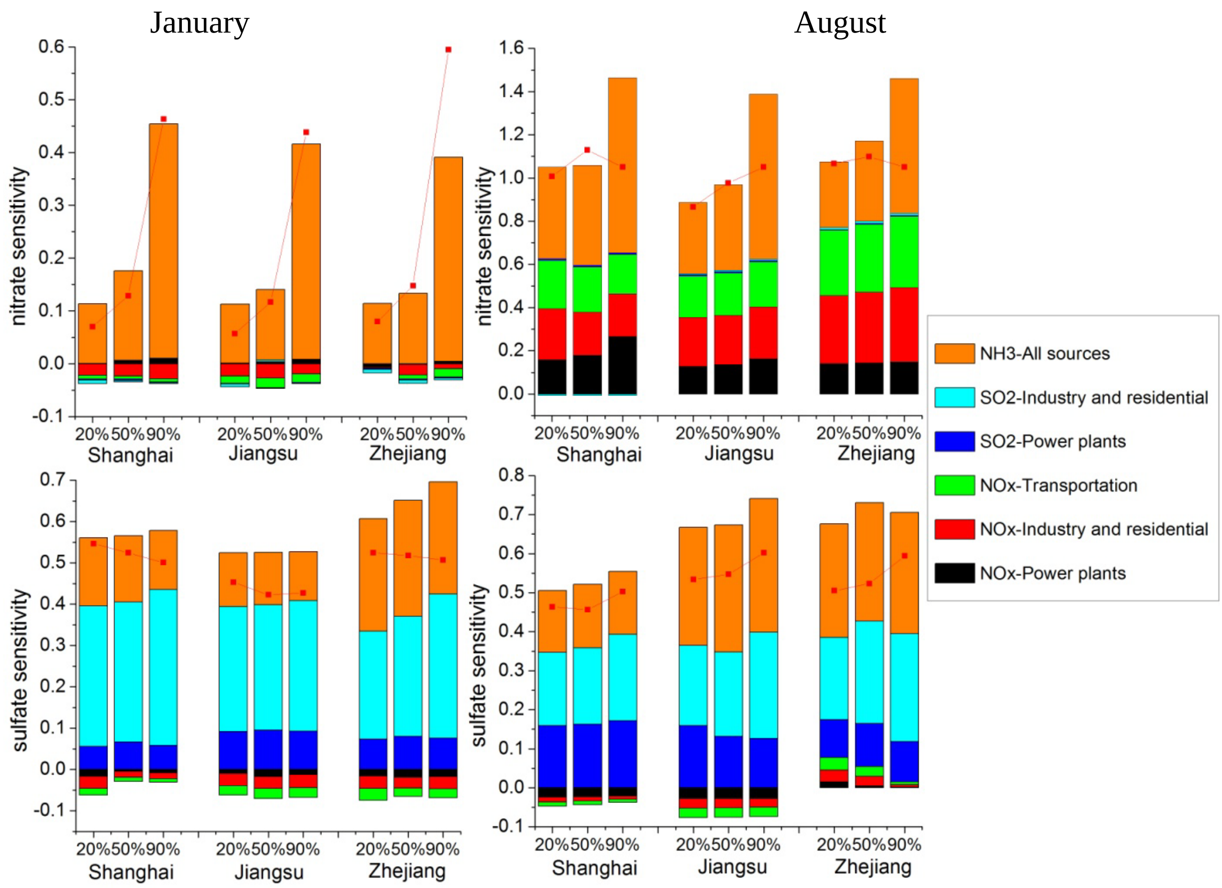


Figure S 8. Sensitivity of NO_3^- and SO_4^{2-} concentrations to the stepped control of individual air pollutants from individual sectors. The X-axis shows the reduction ratio (= 1 – emission ratio). The Y-axis shows NO_3^-/SO_4^{2-} sensitivity, which is defined as the change ratio of NO_3^-/SO_4^{2-} concentration divided by the reduction ratio of emissions. The colored bars denote the NO_3^-/SO_4^{2-} sensitivities when a particular emission source is controlled while the others stay the same as the base case; the red dotted line denotes the NO_3^-/SO_4^{2-} sensitivity when all emission sources are controlled simultaneously.

References

Baker, K.: Meteorological Modeling Protocol for Application to PM2.5/haze/ozone Modeling Projects, Lake Michigan Air Directors Consortium, Des Plaines, IL, 2004.

Boylan, J.: PM Model Performance Goal and Criteria, National Regional Planning Organizations Modeling Meeting, Denver, CO, 2005.

Emery, C., Tai, E., and Yarwood, G.: Enhanced meteorological modeling and performance evaluation for two texas episodes. Report to the Texas Natural Resources Conservation Commission, ENVIRON, International Corp, Novato, CA, 2001.

Morris, R., Koo, B., and McNally, D.: Application of Multiple Models to Simulation Fine Particulate in the Southeastern U.S., National Regional Planning Organizations Modeling Meeting, Denver, CO, 2005.

Wang, L. T., Jang, C., Zhang, Y., Wang, K., Zhang, Q. A., Streets, D., Fu, J., Lei, Y., Schreifels, J., He, K. B., Hao, J. M., Lam, Y. F., Lin, J., Meskhidze, N., Voorhees, S., Evarts, D., and Phillips, S.: Assessment of air quality benefits from national air pollution control policies in China. Part I: Background, emission scenarios and evaluation of meteorological predictions, Atmos. Environ., 44, 3442-3448, DOI 10.1016/j.atmosenv.2010.05.051, 2010.

Wang, S. X., Xing, J., Chatani, S., Hao, J. M., Klimont, Z., Cofala, J., and Amann, M.: Verification of anthropogenic emissions of China by satellite and ground observations, Atmos. Environ., 45, 6347-6358, DOI 10.1016/j.atmosenv.2011.08.054, 2011.

Wang, Y., Zhang, Q. Q., He, K., Zhang, Q., and Chai, L.: Sulfate-nitrate-ammonium aerosols over China: response to 2000-2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia, Atmos. Chem. Phys., 13, 2635-2652, DOI 10.5194/acp-13-2635-2013, 2013.

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).

Zhang, Y., Liu, P., Pun, B., and Seigneur, C.: A comprehensive performance evaluation of MM5-CMAQ for the Summer 1999 Southern Oxidants Study episode - Part I: Evaluation protocols, databases, and meteorological predictions, Atmos. Environ., 40, 4825-4838, DOI 10.1016/j.atmosenv.2005.12.043, 2006.