Supplementary information for the manuscript *Development and application of observable response indicators for design of an effective ozone and fine particle pollution control strategy in China*

Text S1: Definitions of the degree of sulfate neutralization (DSN), gas ratio (GR), and Adjusted Gas Ratio (AdjGR), as follows (Wang et al., 2011):

$$DSN = \frac{\left[NH_4^+\right] - \left[NO_3^-\right]}{\left[SO_4^-\right]} \tag{S1}$$

$$GR = \frac{[TA] - 2 \times [TS]}{[TN]} = \frac{\left(\left[NH_3\right] + \left[NH_4^+\right]\right) - 2 \times \left[SO_4^{2-}\right]}{\left[NO_3^-\right] + \left[HNO_3\right]} \tag{S2}$$

$$AdjGR = \frac{[TA] - DSN \times [TS]}{[TN]} = \frac{\left[NH_3\right] + \left[NO_3^-\right]}{\left[NO_3^-\right] + \left[HNO_3\right]} \tag{S3}$$

where $[TA]$, $[TN]$ and $[TS]$ are the total molar concentration of ammonia ($\left[NH_3\right] + \left[NH_4^+\right]$), nitrate ($\left[NO_3^-\right] + \left[HNO_3\right]$), and sulfate ($\left[SO_4^{2-}\right]$), respectively.

Text S2: Definitions of the PR for $PM_{2.5}$ chemistry.

For $PM_{2.5}$, the PR is the NO_x emissions that produce the maximum $PM_{2.5}$ concentrations under baseline VOC emissions. A PR of < 1 (i.e., baseline) indicates that the baseline condition is strongly NO_x-saturated, and the control of NO_x leads to an increase in $PM_{2.5}$ levels. Similarly to that for O_3, the PR for $PM_{2.5}$ can be directly calculated from the polynomial function:

$$PR = 1 + \Delta E_{NOx}|_{\frac{\partial \Delta Conc_{PM2.5}}{\partial \Delta E_{NOx}}=0} \qquad \Delta E_{NOx} \epsilon [a, b] , \qquad \text{(S4)}$$

where $\frac{\partial \Delta Conc_{PM2.5}}{\partial \Delta E_{NOx}}$ is the first derivation of the $\Delta Conc_{PM2.5}$ to ΔE_{NOx}; the ΔE_{NOx} can be derived by solving the following equation:

$$4 * X_{14} * {\Delta E_{NOx}}^3 + 3 * X_{13} * {\Delta E_{NOx}}^2 + 2 * X_{12} * \Delta E_{NOx} + X_{11} = 0 \quad \text{(S5)}$$

Table S1. WRF performance against observation in 2017 (Ding et al., 2019b)

Meteorological parameter	Statistic	Unit	North	Northeast	East	Central	South	Northwest	Southwest
Wind Speed	MeanOBS	(m/s)	2.92	2.91	2.92	2.33	2.70	2.43	2.08
	MeanPRD	(m/s)	3.00	3.10	2.78	2.25	2.70	2.97	2.64
	Bias	(m/s)	0.08	0.19	-0.15	-0.08	0.00	0.53	0.56
	GrossError	(m/s)	1.18	1.02	1.10	0.91	1.03	1.46	1.30
	RMSE	(m/s)	1.56	1.30	1.58	1.25	1.37	1.95	1.75
	SysRMSE	(m/s)	0.99	0.86	1.10	0.93	0.92	1.27	1.03
	UnsysRMSE	(m/s)	1.20	0.98	1.12	0.81	1.00	1.47	1.41
	IOA		0.79	0.81	0.80	0.73	0.79	0.69	0.67
Temperature	MeanOBS	(K)	282.14	279.37	291.02	290.22	296.38	281.69	287.07
	MeanPRD	(K)	281.59	278.75	290.60	289.62	296.06	281.54	285.20
	Bias	(K)	-0.55	-0.62	-0.41	-0.60	-0.32	-0.15	-1.87
	GrossError	(K)	1.64	1.56	1.50	1.52	1.31	2.28	2.72
	RMSE	(K)	2.09	1.99	2.05	2.01	1.63	2.97	3.76
	SysRMSE	(K)	0.74	0.98	0.70	0.76	0.60	0.74	1.89
	UnsysRMSE	(K)	1.92	1.69	1.90	1.83	1.50	2.86	3.23
	IOA		0.97	0.96	0.94	0.92	0.93	0.96	0.94
Humidity	MeanOBS	(g/kg)	5.15	5.52	11.12	10.46	15.28	4.79	9.67
	MeanPRD	(g/kg)	5.24	5.43	10.81	9.82	14.71	4.64	8.73
	Bias	(g/kg)	0.08	-0.09	-0.31	-0.64	-0.57	-0.15	-0.94
	GrossError	(g/kg)	0.86	0.76	1.19	1.17	1.32	1.07	1.91
	RMSE	(g/kg)	1.20	0.98	1.58	1.47	1.65	1.48	2.91
	SysRMSE	(g/kg)	0.56	0.48	0.77	0.93	0.95	0.46	2.00
	UnsysRMSE	(g/kg)	1.04	0.84	1.35	1.09	1.32	1.38	2.09
	IOA		0.88	0.83	0.87	0.81	0.78	0.86	0.85

Table S2. Control matrix of CMAQ scenarios for RSM design

Cases	ΔE_{NOx}	ΔE_{SO2}	ΔE_{NH3}	ΔE_{VOCs}
Baseline	0	0	0	0
Scenario 1	-98.0%	-50.0%	11.1%	28.0%
2	-95.5%	50.0%	-90.1%	-28.0%
3	-92.0%	-87.5%	58.0%	68.0%
4	-87.5%	-12.5%	60.5%	-98.7%
5	-82.0%	12.5%	-60.5%	-53.9%
6	-75.5%	87.5%	-58.0%	54.9%
7	-68.0%	-96.9%	90.1%	-3.7%
8	-59.5%	-53.1%	-98.9%	79.5%
9	-50.0%	-21.9%	9.7%	-94.9%
10	-39.5%	71.9%	29.8%	-37.3%
11	-28.0%	-71.9%	-82.4%	84.3%
12	-15.5%	21.9%	85.5%	18.1%
13	-2.0%	53.1%	72.6%	88.5%
14	12.5%	96.9%	-46.2%	-88.5%
15	28.0%	-99.2%	-32.8%	-18.1%
16	44.5%	-75.8%	95.6%	-84.3%
17	62.0%	-36.7%	-95.6%	37.3%
18	80.5%	61.7%	32.8%	94.9%
19	100.0%	-80.5%	46.2%	-79.5%
20	-80.5%	5.5%	-72.6%	3.7%
21	-62.0%	32.0%	-85.5%	-54.9%
22	-44.5%	93.0%	82.4%	53.9%
23	-28.0%	-93.0%	-29.8%	98.7%
24	-12.5%	-32.0%	-9.7%	-99.9%
25	2.0%	-5.5%	98.9%	-65.4%
26	15.5%	80.5%	-99.9%	33.2%
27	28.0%	-61.7%	-4.4%	-22.9%
28	39.5%	36.7%	17.6%	70.5%
29	50.0%	75.8%	-87.8%	-98.2%
30	59.5%	99.2%	66.9%	-50.8%
31	68.0%	-99.8%	64.8%	60.6%
32	75.5%	-87.7%	-56.0%	0.9%
33	82.0%	-43.6%	-49.4%	81.5%
34	87.5%	56.1%	92.2%	-93.8%
35	92.0%	-84.2%	-98.0%	-33.6%
36	95.5%	-3.3%	17.2%	90.5%
37	98.0%	22.1%	35.5%	22.1%
38	99.5%	90.4%	-79.4%	90.0%
39	100.0%	-95.1%	95.1%	-86.9%
40	-100.0%	-42.4%	76.1%	-13.9%

Table S3. Fitting coefficients of terms in the pf-RSM for population-weighted O_3 concentration (annual daytime averages)

Province*	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}	X_{11}	X_{12}	X_{13}	X_{14}
	NO_x^5	NO_x^4	NO_x^3	NO_x^2	NO_x	VOC	VOC^2	VOC^3	NO_xVOC	NO_xVOC^3	NO_x^5VOC	NO_x^2VOC	SO_2	NH_3
BJ	15.6331	-15.8197	0.3295	-1.7909	-11.1997	16.1798	-0.3235	0.5200	5.5614	-2.0192	4.3407	-11.0443	-0.0268	-0.1451
TJ	17.3346	-17.1611	-0.8376	1.1536	-15.7518	16.8804	-0.3917	0.4903	4.5396	-2.3686	4.9447	-11.4644	-0.0979	-0.2838
HEB	20.4479	-19.1223	-4.0097	2.6044	-14.1593	15.6764	-0.4857	0.3041	3.6528	-1.2845	4.8112	-10.7600	-0.1756	-0.3235
SX	16.8306	-14.8351	-5.5225	2.2237	-8.6383	9.2259	-0.5845	0.1108	2.0626	0.1330	2.6040	-5.8103	-0.1776	-0.3086
IM	8.1159	-8.4605	-1.9157	1.2122	-2.9017	4.2849	-0.0467	0.1621	0.1106	0.1583	1.8939	-2.4732	-0.1352	-0.2103
LN	11.2944	-11.7860	-2.1728	3.2955	-9.7696	9.4901	0.1070	0.3875	0.3383	-0.9459	3.8321	-5.5380	-0.1684	-0.3164
JL	6.9481	-7.8974	0.2604	-0.1701	-3.7931	6.4701	-0.1097	0.3056	1.5819	-0.7334	2.3809	-4.1174	-0.1079	-0.1453
HLJ	8.3741	-7.1333	-4.2578	3.3045	-5.1286	4.3850	0.0667	0.1289	-0.5116	-0.3794	1.8777	-2.3095	-0.0331	-0.1345
SH	12.4808	-9.1289	-0.2042	-6.9328	-9.3299	20.2472	-2.3347	-0.0417	10.2219	-2.8474	1.5583	-10.4248	0.1382	-0.0062
JS	14.0901	-10.1936	0.4790	-10.2577	-7.9685	18.5893	-2.7836	-0.2433	11.9375	-1.2656	0.3482	-10.2163	0.0691	0.0249
ZJ	10.1515	-5.7401	-0.2942	-11.6081	1.2486	15.2495	-3.8427	-0.6984	12.4704	-0.4918	-1.1202	-6.5267	-0.0750	-0.0643
AH	14.9767	-12.3072	-0.0134	-6.5191	-9.4715	15.4040	-2.0393	-0.0674	8.5031	-0.7564	1.2169	-9.6501	-0.0524	-0.0519
FJ	6.2076	-6.0441	1.7370	-7.2390	-0.4611	8.4330	-2.0350	-0.0371	6.3072	0.8887	0.0050	-3.5297	-0.2804	-0.0563
JX	11.5044	-9.6342	-0.3953	-7.2507	-0.4884	10.9546	-2.3108	-0.1763	7.5657	0.5231	0.2831	-5.7267	-0.2976	-0.1268
SD	14.8206	-12.8542	-0.0711	-4.2985	-11.4392	15.5321	-1.2732	0.0856	6.7294	-1.1118	2.4226	-10.1057	-0.0457	-0.2897
HEN	21.6638	-20.1066	-2.5469	-0.7835	-13.0395	17.2070	-0.9852	0.3954	5.7676	-0.9850	4.3705	-12.3815	-0.1656	-0.2319
HUB	19.6246	-16.8693	-4.2339	-1.1467	-8.6650	14.6338	-1.5539	0.1002	5.6259	-0.3513	2.8354	-9.4989	-0.1324	-0.2600
HUN	13.1882	-9.9947	-0.8566	-8.5519	-0.9221	12.1622	-2.4241	-0.1714	8.8038	0.1527	-0.1103	-6.7420	-0.2998	-0.2418
GD	11.7236	-10.1782	1.2713	-8.2577	-4.2583	14.9302	-2.0142	0.1313	8.6205	-0.4269	0.8396	-8.6753	0.0098	0.1519
GX	7.4335	-7.1958	0.7881	-6.7507	3.9491	6.9116	-1.8417	-0.0743	5.5165	1.1276	-0.0001	-2.9113	-0.4194	-0.1539
HN	2.6670	-2.9667	0.3860	-3.6930	7.1018	2.9134	-1.3385	0.0608	3.0393	1.0371	-0.0499	-0.8197	-0.4314	-0.0178
CQ	25.2938	-21.0970	-8.7884	2.9114	-11.6974	14.0587	-0.5025	0.7163	4.1000	-1.3939	3.5693	-9.7821	-0.4523	-0.7005
SC	15.0487	-14.6018	-1.5488	-5.6843	0.7248	11.8284	-2.0989	0.1973	7.0802	1.0009	1.9235	-6.0528	-0.6658	-0.5191
GZ	8.6081	-7.5577	-0.9885	-4.9316	3.2954	5.4403	-1.2726	0.1432	4.4792	0.3585	-0.0443	-2.5443	-0.2719	-0.4527
YN	3.3274	-3.0550	0.4774	-3.4324	3.0002	2.5747	-0.2067	0.1823	1.8914	-0.3830	0.1115	-1.3120	-0.1903	-0.2905
TB	0.3274	-0.3954	0.1874	-0.4639	-0.2829	0.1447	0.0002	-0.0171	0.0170	0.0249	0.0434	-0.0576	-0.0175	-0.0631
SHX	16.4028	-16.4373	-2.9256	1.1616	-7.8517	10.4015	-0.5329	0.3826	2.3335	0.0936	3.5975	-6.8185	-0.4573	-0.4309
GS	5.6016	-6.5615	0.1345	-1.8024	2.1907	3.3143	-0.2495	0.1729	1.0539	0.4111	1.1930	-1.7939	-0.2391	-0.2868
QH	6.0360	-5.7068	-1.6932	-0.3074	-0.6021	2.3646	0.0950	0.0439	0.3671	-0.1395	1.0207	-1.3353	-0.3096	-0.4090
NX	8.6833	-8.8175	-2.0601	0.7320	-2.7804	3.5572	-0.2324	0.1717	0.3208	0.5012	1.3932	-2.0604	-0.1822	-0.3161
XJ	5.1153	-5.3064	-0.9088	0.6578	-3.1813	2.8132	0.2195	0.0593	-0.3068	-0.1290	1.3803	-1.7406	-0.0537	-0.1303

* BJ-Beijing; TJ-Tianjin; HEB- Hebei; SX-Shanxi; IM-Inner Mongolia; LN- Liaoning; JL- Jilin; HLJ-Helongjiang; SH- Shanghai; JS-Jiangsu; ZJ-Zhejiang; AH- Anhui; FJ- Fujian; JX- Jiangxi; SD- Shandong; HEN- Henan; HUB-Hubei; HUN- Hunan; GD-Guangdong; GX- Guangxi; HN-Hainan; CQ-Chongqing; SC- Sichuan; GZ-Guizhou; YN- Yunnan; TB- Tibet; SHX-Shaanxi; GS-Gansu; QH-Qinghai; NX- Ningxia; XJ-Xinjiang.

Table S4. Fitting coefficients of terms in the pf-RSM for population-weighted $PM_{2.5}$ concentration (annual 24-h averages)

Province*	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}	X_{11}	X_{12}	X_{13}	X_{14}
	VOC	NH_3	NH_3^2	NH_3^3	SO_2	VOC^2	NO_xVOC	NO_x^2VOC	NO_x^4VOC	NO_xNH_3	NO_x	NO_x^2	NO_x^3	NO_x^4
BJ	7.7556	7.4054	-8.7095	2.9863	4.0483	-2.8646	8.1551	-3.8546	-0.4442	6.7633	2.0520	-1.9794	8.7052	-11.0668
TJ	9.5593	9.7907	-10.9550	3.9230	5.2622	-3.5155	9.9360	-5.7595	-0.0747	7.5623	1.1214	-2.4473	10.7841	-13.6535
HEB	9.3249	6.6827	-10.1831	5.3916	4.9079	-3.1422	9.1783	-6.2288	0.8058	6.9840	-0.0588	0.3997	11.9043	-15.2199
SX	3.3916	5.9827	-5.0190	1.9787	3.8172	-1.4826	4.0957	-1.5934	-0.6231	5.3642	1.1224	-0.0181	6.0734	-7.5406
IM	1.7305	1.0566	-1.9597	2.1441	1.8999	-0.4328	1.0563	-1.5130	0.3254	1.8026	-0.6166	1.6555	3.2925	-4.1368
LN	4.8197	5.3529	-4.9629	2.6101	3.1796	-1.4505	3.9294	-3.4433	0.6468	4.3699	0.3818	0.6248	5.9124	-7.4582
JL	3.8126	3.3371	-3.5163	1.9600	2.4967	-1.1247	3.2384	-3.8413	1.1917	3.5219	0.1496	0.6977	5.7623	-6.5905
HLJ	3.8981	1.1602	-2.9603	3.0550	2.7423	-0.9233	2.3742	-4.7053	2.0048	2.1677	-2.0429	2.3587	5.5937	-6.7466
SH	4.3130	5.8667	-3.9630	0.8238	3.3826	-1.5488	4.0451	-2.5729	0.4704	3.8270	2.2659	-0.8441	4.0439	-5.1297
JS	6.2330	6.2469	-7.2064	3.8650	3.7971	-2.3532	7.1307	-3.8114	0.8037	6.2275	4.7859	0.2224	7.8850	-10.8542
ZJ	4.0687	6.9259	-4.9115	0.7605	3.2240	-1.9214	5.1829	-1.9586	0.2797	5.3303	4.1839	-2.2089	4.4253	-5.5549
AH	5.9338	9.2555	-8.6049	2.4512	4.5626	-2.9592	8.3714	-2.9025	0.4882	8.0679	5.6391	-2.3932	7.8228	-10.4374
FJ	0.9813	3.9087	-1.7869	0.1173	2.8264	-0.5199	1.5197	0.3777	-0.7220	3.0442	2.1536	-0.6957	1.8108	-1.6368
JX	3.0115	9.2906	-5.0933	0.1243	4.4682	-2.0885	5.8731	-0.7971	-0.0248	7.5403	5.1472	-2.6149	5.8525	-6.9619
SD	6.6103	11.1616	-10.5571	3.1067	5.5037	-3.3303	8.7943	-2.5163	-1.2604	8.5107	4.3018	-2.4442	9.0008	-11.5291
HEN	9.2044	9.3643	-12.7469	5.6423	6.5107	-3.6218	10.8052	-5.9387	0.6254	9.0837	4.1492	-1.6191	12.3585	-15.7242
HUB	6.7413	8.8930	-10.6457	5.0903	6.1528	-3.0865	9.5607	-3.9428	0.8923	9.0598	4.7793	-1.3904	10.1912	-12.7410
HUN	4.3557	11.6956	-9.5571	1.5297	5.8428	-3.2041	9.2581	-0.6912	-0.8620	9.4699	6.5217	-3.3695	8.5250	-10.4046
GD	2.5866	4.6811	-2.7435	0.4128	3.9678	-1.0115	2.6796	-0.8034	-0.9537	3.9233	2.0070	-0.5193	3.7188	-3.7987
GX	0.8613	5.1114	-2.2180	-0.1399	4.5106	-0.6557	2.2019	-0.3470	-0.2501	4.5354	3.4683	-0.6587	4.1333	-3.5222
HN	0.4085	2.1675	-0.8492	-0.2276	2.5496	-0.1965	0.5176	0.3188	-0.6984	1.8673	1.7495	-0.4705	0.9878	-0.4613
CQ	5.3752	6.9700	-10.6521	6.1199	8.9927	-2.1747	7.6358	-4.6775	2.4996	6.9991	4.6190	0.0145	9.6148	-11.9478
SC	2.5696	5.3600	-7.8325	4.1948	6.6754	-1.3951	5.5957	-1.6891	2.3151	6.7589	8.3160	-0.1841	5.7384	-8.2599
GZ	0.8093	5.4918	-3.0000	0.2785	5.3099	-0.1956	1.8927	-1.8164	1.2759	3.3880	2.9560	-2.8713	2.8504	-0.9135
YN	0.3964	1.7844	-1.4075	0.4204	2.5055	-0.0292	0.4442	-1.5399	1.1809	0.8036	0.8854	-0.8659	0.7439	0.0615
TB	0.0073	0.0350	-0.0120	0.0202	0.0313	-0.0009	0.0009	0.0072	-0.0088	0.0239	0.0181	0.0175	0.0131	-0.0272
SHX	2.1404	3.9888	-3.5280	1.7244	4.3501	-0.8223	2.6566	-1.4880	0.4632	3.9755	1.7424	0.6461	4.6422	-5.4597
GS	0.5605	1.5892	-1.3864	0.7272	1.7895	-0.2005	0.6275	-0.2931	-0.1610	1.7863	1.2454	0.1539	1.5287	-1.7752
QH	0.4616	0.9877	-1.1674	0.6782	1.3458	-0.1399	0.3300	-0.5065	0.1401	1.0384	0.7092	0.5767	1.0397	-1.4389
NX	0.6379	1.3231	-1.2208	1.0114	2.4299	-0.1821	0.5380	-0.5124	-0.0670	1.3810	0.1574	0.7569	2.1130	-2.2976
XJ	0.6295	0.3256	-0.6488	0.7537	0.7105	-0.0953	0.3013	-0.8843	0.3476	0.4498	-0.5093	0.8907	1.3758	-1.7630

* BJ-Beijing; TJ-Tianjin; HEB- Hebei; SX-Shanxi; IM-Inner Mongolia; LN- Liaoning; JL- Jilin; HLJ-Helongjiang; SH- Shanghai; JS-Jiangsu; ZJ-Zhejiang; AH- Anhui; FJ- Fujian; JX- Jiangxi; SD- Shandong; HEN- Henan; HUB-Hubei; HUN- Hunan; GD-Guangdong; GX- Guangxi; HN-Hainan; CQ-Chongqing; SC- Sichuan; GZ-Guizhou; YN- Yunnan; TB- Tibet; SHX-Shaanxi; GS-Gansu; QH-Qinghai; NX- Ningxia; XJ-Xinjiang.

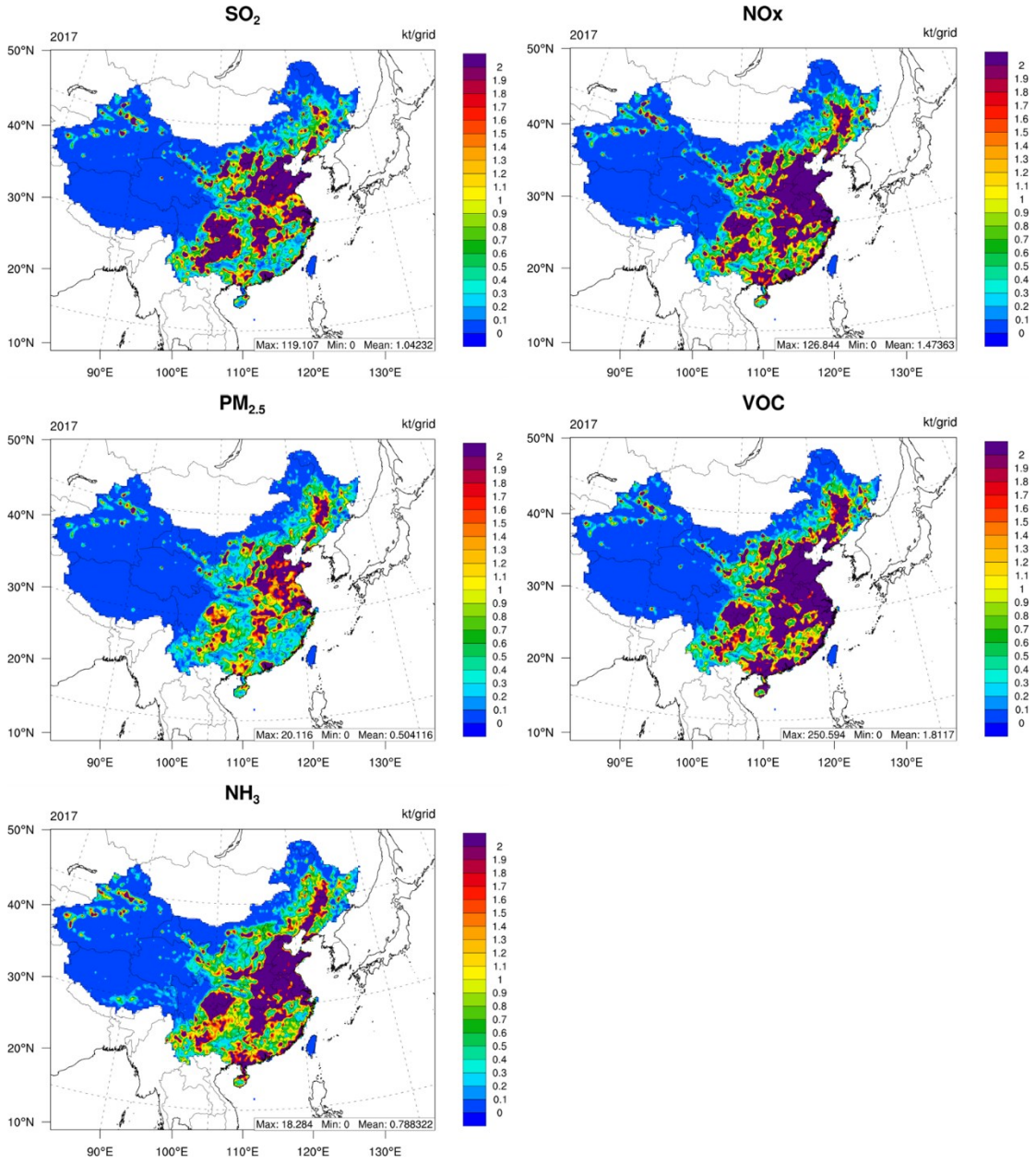

Figure S1. Spatial distribution of annual anthropogenic emissions in 2017 (Ding et al., 2009b)

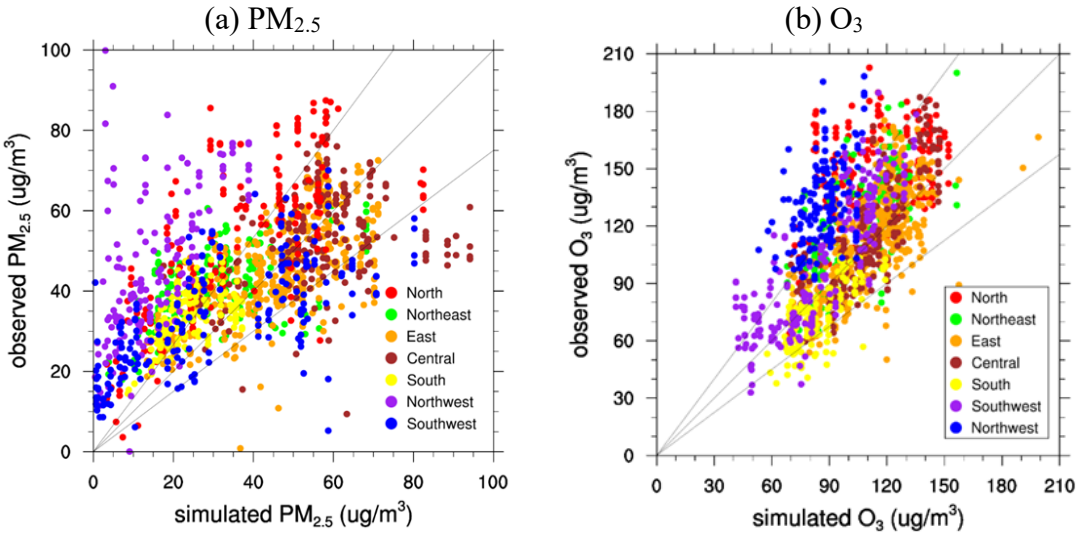

Figure S2. CMAQ performance in simulating $PM_{2.5}$ (annual) and O_3 (July) levels in 2017 (Ding et al., 2019a, b).

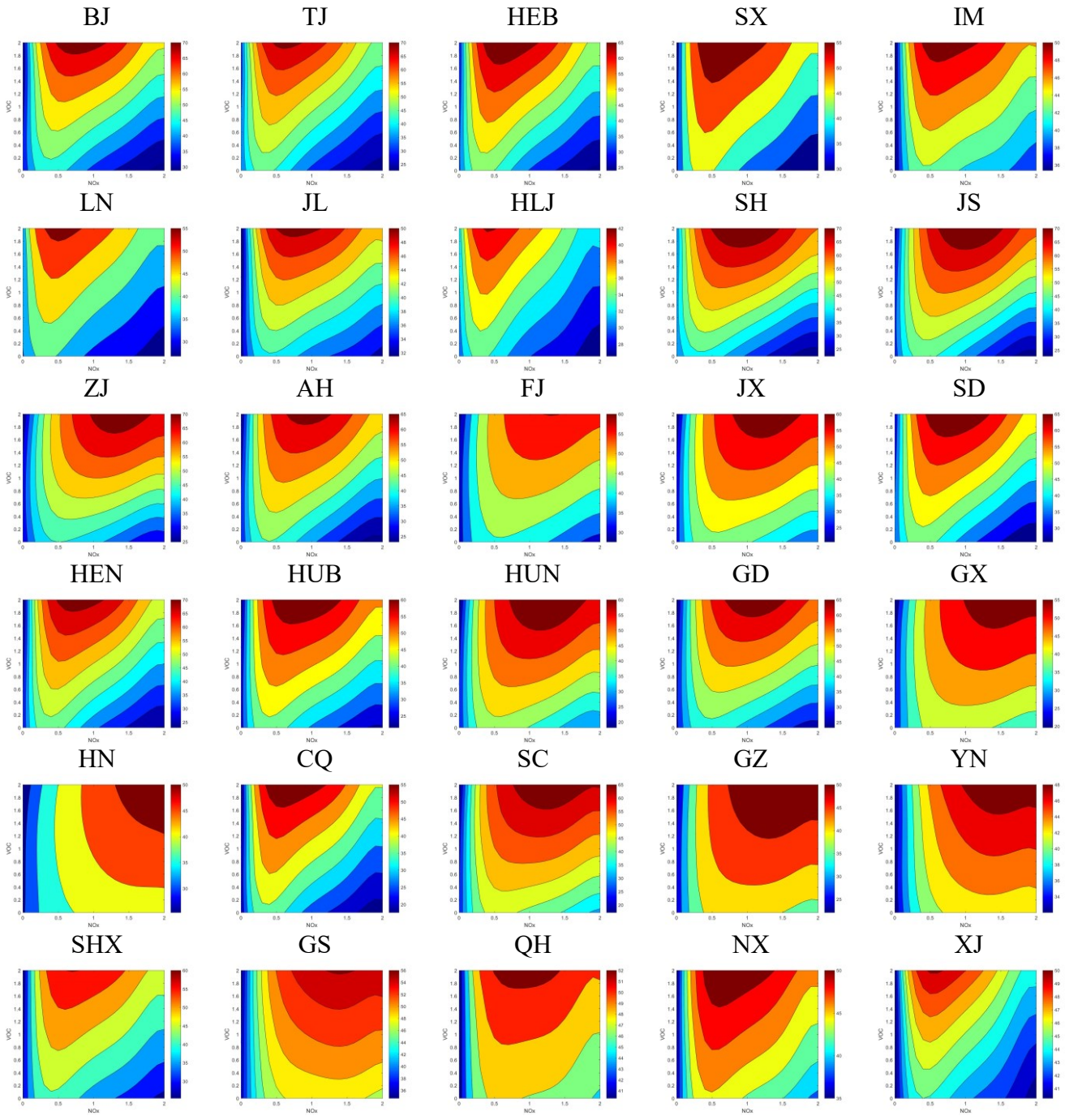

Figure S3. Isopleth of population-weighted daytime O_3 to NO_x (x- axis) and VOC (y- axis) emission changes in different months (the x- and y- axes represent that the precursor emission rates, background colors represent the population-weighted daytime O_3 concentrations in China; the plot for Tibet is unavailable because of low change in O_3 levels)

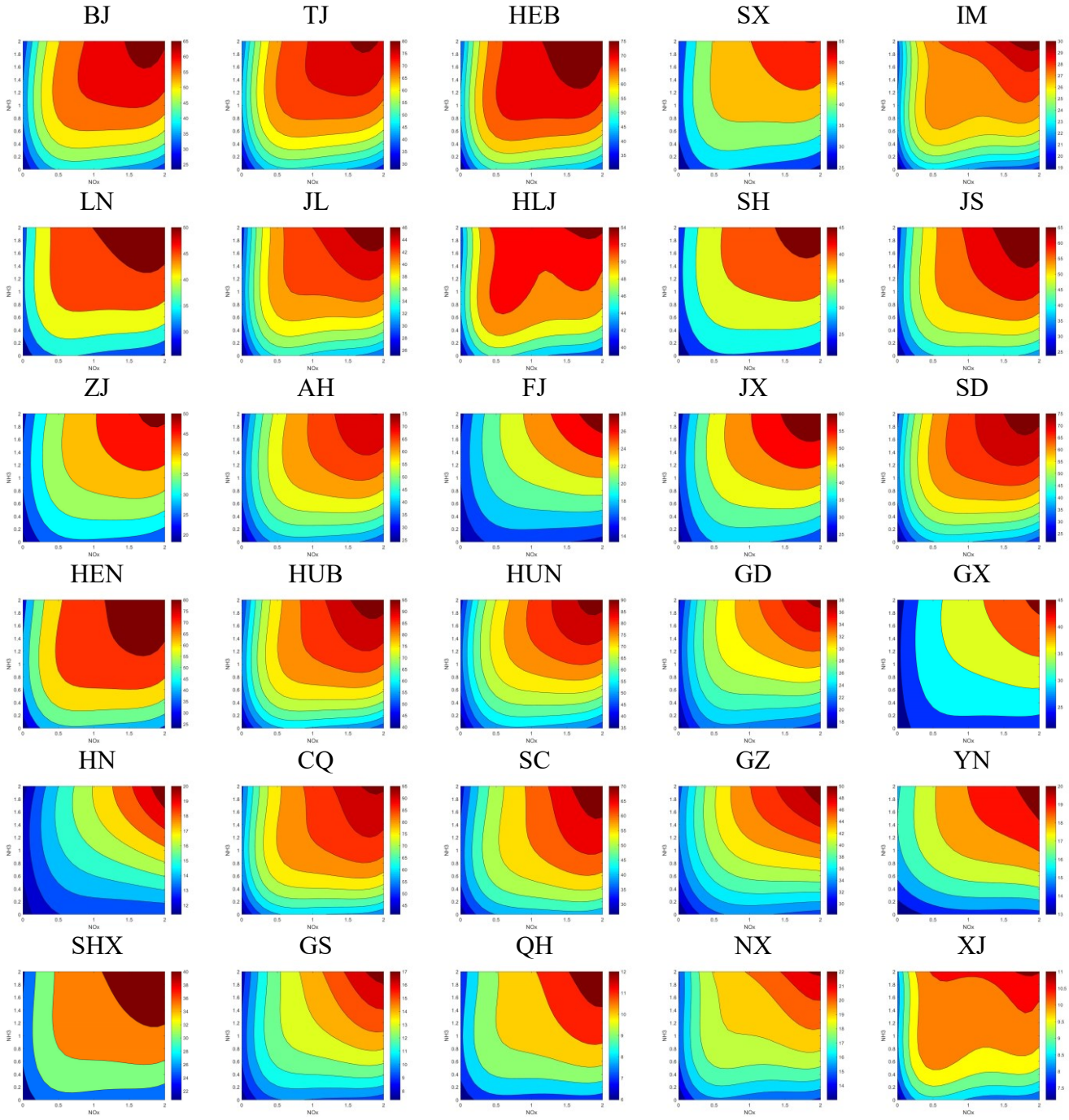

Figure S4. Isopleth of population-weighted $PM_{2.5}$ to NO_x (x- axis) and NH_3 (y- axis) emission changes in different months (the x- and y- axes represent that the precursor emission rates, background colors represent the population-weighted $PM_{2.5}$ concentrations in China; the plot for Tibet is unavailable because of low change in $PM_{2.5}$ levels)

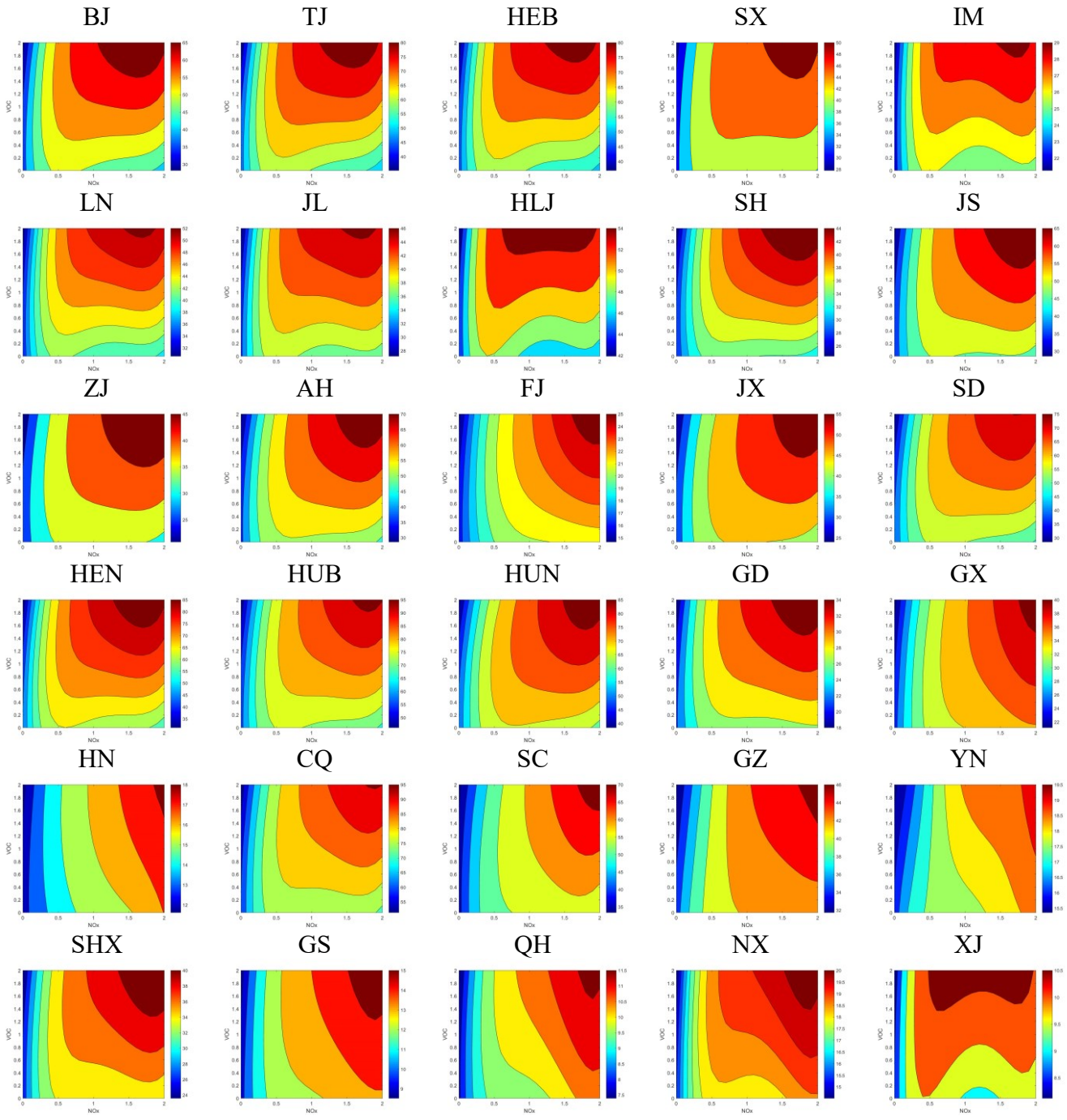

Figure S5. Isopleth of population-weighted $PM_{2.5}$ to NO_x (x- axis) and VOC (y- axis) emission changes in different months (the x- and y- axes represent that the precursor emission rates, background colors represent the population-weighted $PM_{2.5}$ concentrations in China; the plot for Tibet is unavailable because of low change in $PM_{2.5}$ levels)

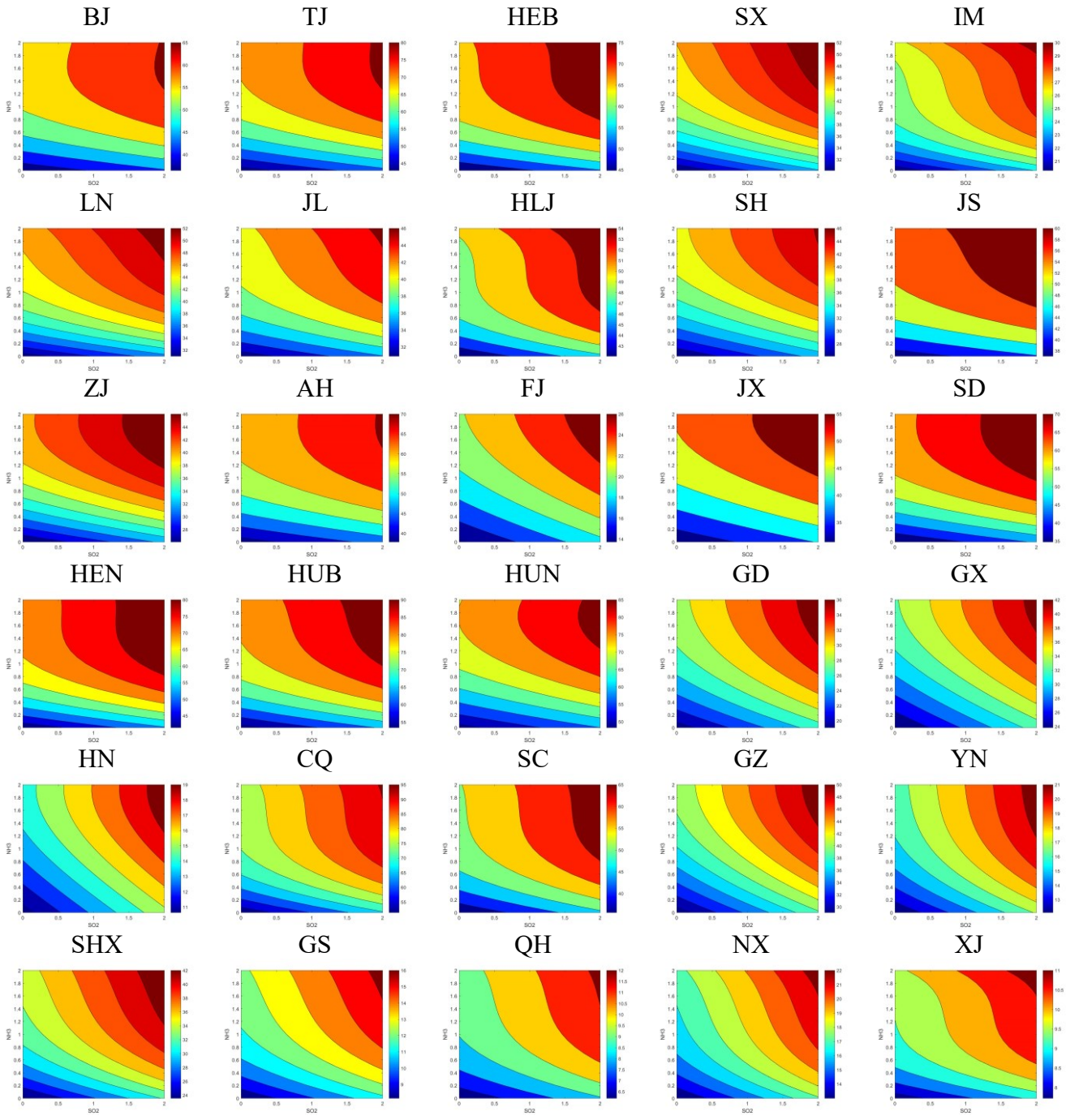

Figure S6. Isopleth of population-weighted $PM_{2.5}$ to SO_2 (x-axis) and NH_3 (y- axis) emission changes in different months (the x- and y- axes represent that the precursor emission rates, background colors represent the population-weighted $PM_{2.5}$ concentrations in China; the plot for Tibet is unavailable because of low change in $PM_{2.5}$ levels)

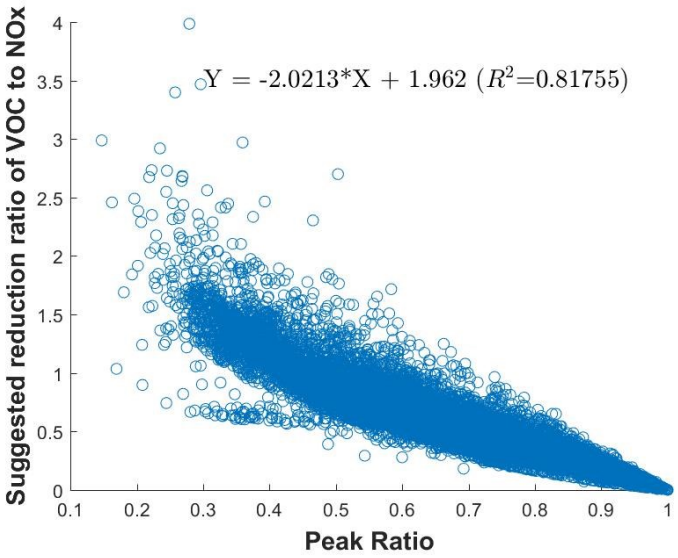

Figure S7. Correlations between the PR and VNr.

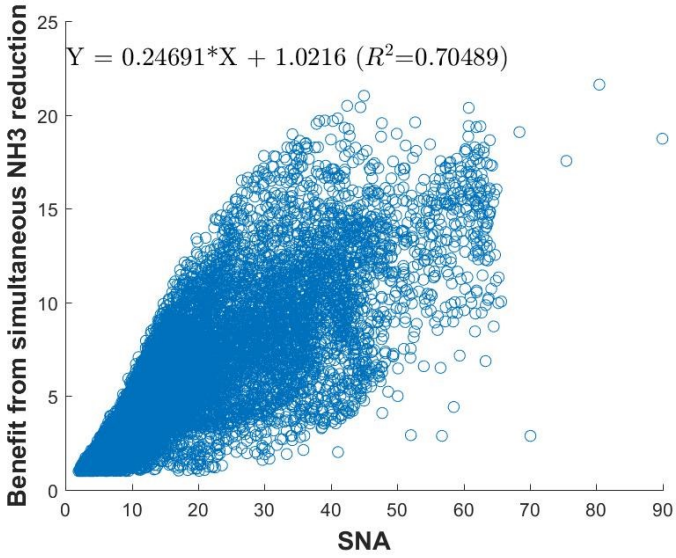

Figure S8. Correlations between concentrations of secondary inorganic aerosol (SNA, μg m^{-3}) and the benefit in reducing $PM_{2.5}$ from simultaneous NH_3 reduction (ΔC_NH_3, μg m^{-3})

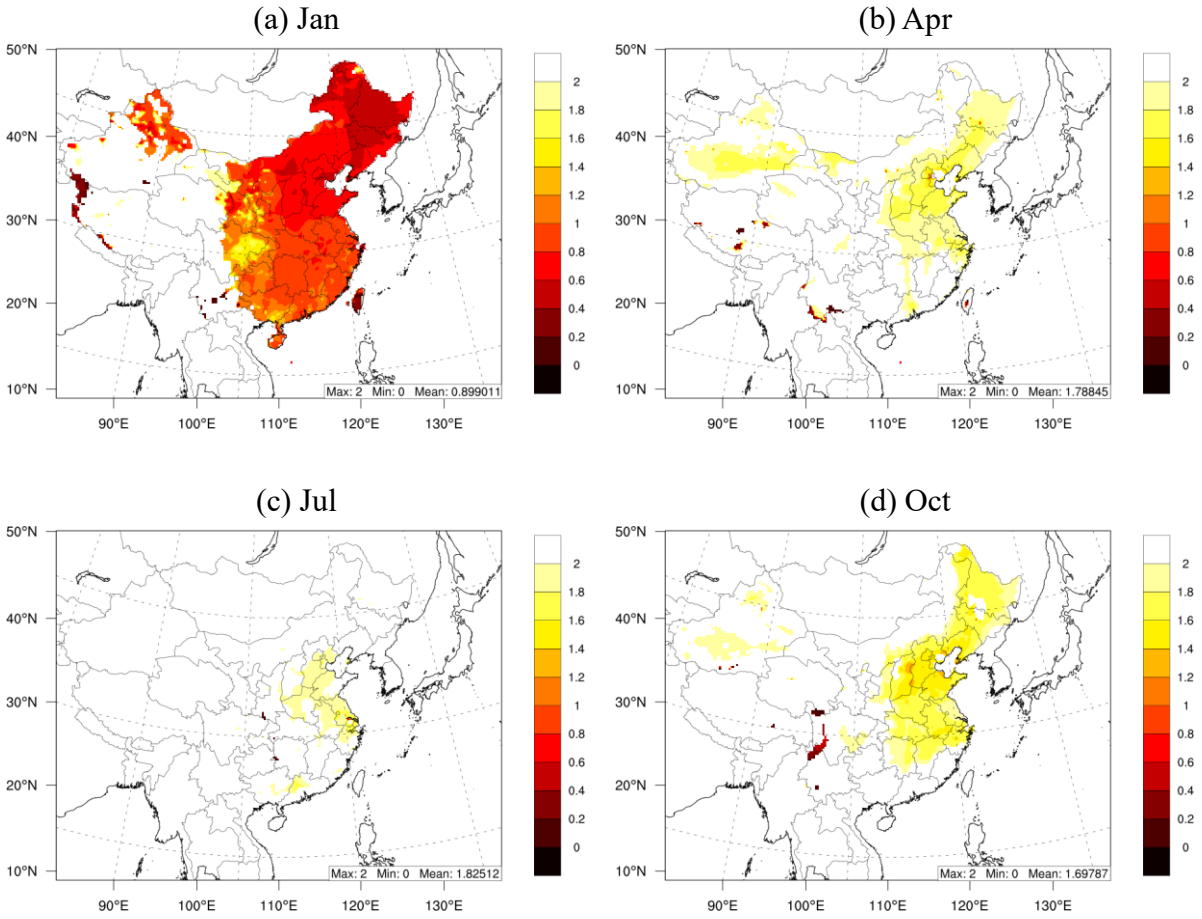

Figure S9. Spatial distribution of the PR to identify $PM_{2.5}$ sensitivity to NO_x emission.