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Comprehensive analyses of source sensitivities and apportionments of $PM_{2.5}$ and ozone over Japan via multiple numerical techniques

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Table S1: Annual total emission amounts (Mg/year) of each source group for the 2016 fiscal year in d02.

Group	CO	SO_2	NO_X	NH_3	NMVOC[*1]	$PM_{2.5}$	EC[*2]	OC[*3]
s01	1,171,555	910	411,047	15,576	122,712	23,850	6,091	5,361
s02	59,862	279,102	682,708	0	20,593	55,988	10,766	16,045
s03	164,110	3,057	97,754	0	11,404	4,479	2,503	1,421
s04	1,230,684	356,649	761,388	0	226,562	19,196	1,661	3,459
s05	69,836	369	4,924	943	8,016	10,028	586	6,997
s06	46,034	840	33,421	0	11,938	1,770	190	619
s07	0	0	0	0	587,827	0	0	0
s08	0	0	0	389,915	55,714	1,811	0	0
s09[*4]	404,322	1,534,513	58,700	21,732	3,954,064	0	0	0
	132,359	1,534,513	9,940	21,732	1,302,798	0	0	0
s10	13,152,939	1,168,210	2,990,211	649,985	2,108,940	736,968	112,112	241,057

[*1] Non-methane volatile organic compound
[*2] Elemental carbon
[*3] Organic carbon
[*4] Lower values indicate amounts within Japan only

Table S2: Statistics of the model performance on the MDA8O3 and daily mean $PM_{2.5}$ concentrations for the entire 2016 fiscal year 2016 in the regions.

Species	Region	Number[*1]	Obs.[*2]	Sim.[*3]	NMB[*4]	NME[*5]	R[*6]
MDA8O3 (ppb)	JP	1150	42.7	46.0	7.79%	20.0%	0.860
	KO	151	43.4	48.4	11.3%	24.0%	0.820
	CS	143	44.4	46.8	5.27%	20.7%	0.834
	KS	176	43.1	46.2	7.14%	20.0%	0.868
	TH	201	43.6	47.4	8.69%	18.9%	0.869
	KK	369	41.5	44.3	6.94%	19.5%	0.872
	HT	110	39.4	44.1	11.7%	21.2%	0.797
	OH	108	42.6	45.2	6.10%	20.7%	0.864
	AM	75	42.6	45.6	6.89%	19.3%	0.874
	ST	196	40.8	42.9	5.21%	19.7%	0.880
$PM_{2.5}$ ($\mu g/m^3$)	JP	820	11.9	7.62	-35.9%	41.6%	0.852
	KO	127	14.2	10.3	-27.2%	36.6%	0.860
	CS	113	13.6	9.26	-30.3%	38.9%	0.853
	KS	134	12.0	7.77	-35.5%	39.5%	0.862
	TH	132	10.7	6.62	-38.2%	42.0%	0.855
	KK	243	11.3	6.48	-42.8%	46.2%	0.836
	HT	71	9.01	5.41	-39.9%	46.0%	0.827
	OH	71	12.5	8.30	-33.8%	38.2%	0.863
	AM	43	11.6	7.40	-36.3%	40.0%	0.855
	ST	156	12.0	6.68	-44.5%	46.8%	0.839
SO_4^{2-} ($\mu g/m^3$)	JP	154	2.73	2.64	-3.29%	40.9%	0.710
NO_3^- ($\mu g/m^3$)	JP	154	0.641	1.01	57.1%	121%	0.441
NH_4^+ ($\mu g/m^3$)	JP	154	1.11	1.08	-3.07%	41.1%	0.704
EC ($\mu g/m^3$)	JP	136	0.757	0.345	-54.4%	58.3%	0.477
OC ($\mu g/m^3$)	JP	151	2.58	0.958	-62.8%	66.0%	0.487

[*1] Number of monitoring stations
[*2] Observed values
[*3] Simulated values
[*4] Normalized Mean Bias
[*5] Normalized Mean Error
[*6] Correlation coefficient

Table S3: Source sensitivities of the annual mean ozone and $PM_{2.5}$ concentrations simulated in the regions. The upper table shows ratios (%) against the simulated concentrations. The lower table shows their normalized ratios (%).

(a1) O_3 (not normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	-0.8	-1.0	-0.5	-1.6	-1.1	-1.7	-0.1	-5.2	-5.9	-8.0
s02	0.6	0.7	-0.1	0.5	0.8	0.7	0.8	-1.7	-0.3	-0.5
s03	-0.2	-0.1	-0.1	-0.4	-0.3	-0.5	0.0	-1.7	-1.7	-2.5
s04	-0.6	-0.7	-1.2	-2.0	-0.6	-1.2	0.4	-8.3	-7.8	-9.6
s05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
s06	-0.2	-0.1	-0.1	-0.3	-0.2	-0.3	-0.1	-1.0	-0.8	-1.6
s07	0.6	0.5	0.7	0.9	1.0	1.1	0.2	1.5	1.8	2.3
s08	0.0	-0.1	0.0	0.0	0.0	-0.1	0.0	0.0	0.0	-0.1
s09	1.0	1.2	1.5	1.6	1.4	1.2	0.4	2.3	2.0	1.9
s10	1.1	0.2	0.6	1.1	1.4	1.5	1.3	1.2	1.5	1.5
s11	77.5	77.7	75.1	75.1	73.3	75.9	81.0	81.7	80.0	85.3
s12	-0.2	-0.2	-0.2	-0.2	-0.2	-0.2	-0.1	-0.3	-0.3	-0.3
Sum	79.0	78.0	75.6	74.6	75.4	76.3	83.8	68.4	68.5	68.4

(a2) O_3 (normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	-1.0	-1.3	-0.7	-2.1	-1.5	-2.3	-0.1	-7.6	-8.6	-11.7
s02	0.8	0.9	-0.1	0.7	1.1	0.9	1.0	-2.5	-0.4	-0.8
s03	-0.2	-0.2	-0.1	-0.6	-0.4	-0.6	0.0	-2.5	-2.5	-3.6
s04	-0.7	-0.9	-1.6	-2.7	-0.9	-1.6	0.4	-12.1	-11.4	-14.0
s05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
s06	-0.2	-0.1	-0.1	-0.4	-0.2	-0.4	-0.1	-1.5	-1.1	-2.4
s07	0.8	0.6	0.9	1.2	1.3	1.4	0.2	2.2	2.6	3.3
s08	0.0	-0.1	0.0	0.0	0.0	-0.1	0.0	0.0	0.0	-0.1
s09	1.3	1.5	2.0	2.1	1.8	1.6	0.5	3.4	2.9	2.7
s10	1.4	0.3	0.8	1.4	1.9	1.9	1.5	1.7	2.2	2.2
s11	98.2	99.6	99.3	100.6	97.1	99.5	96.6	119.4	116.9	124.7
s12	-0.2	-0.3	-0.3	-0.3	-0.3	-0.3	-0.2	-0.4	-0.4	-0.4

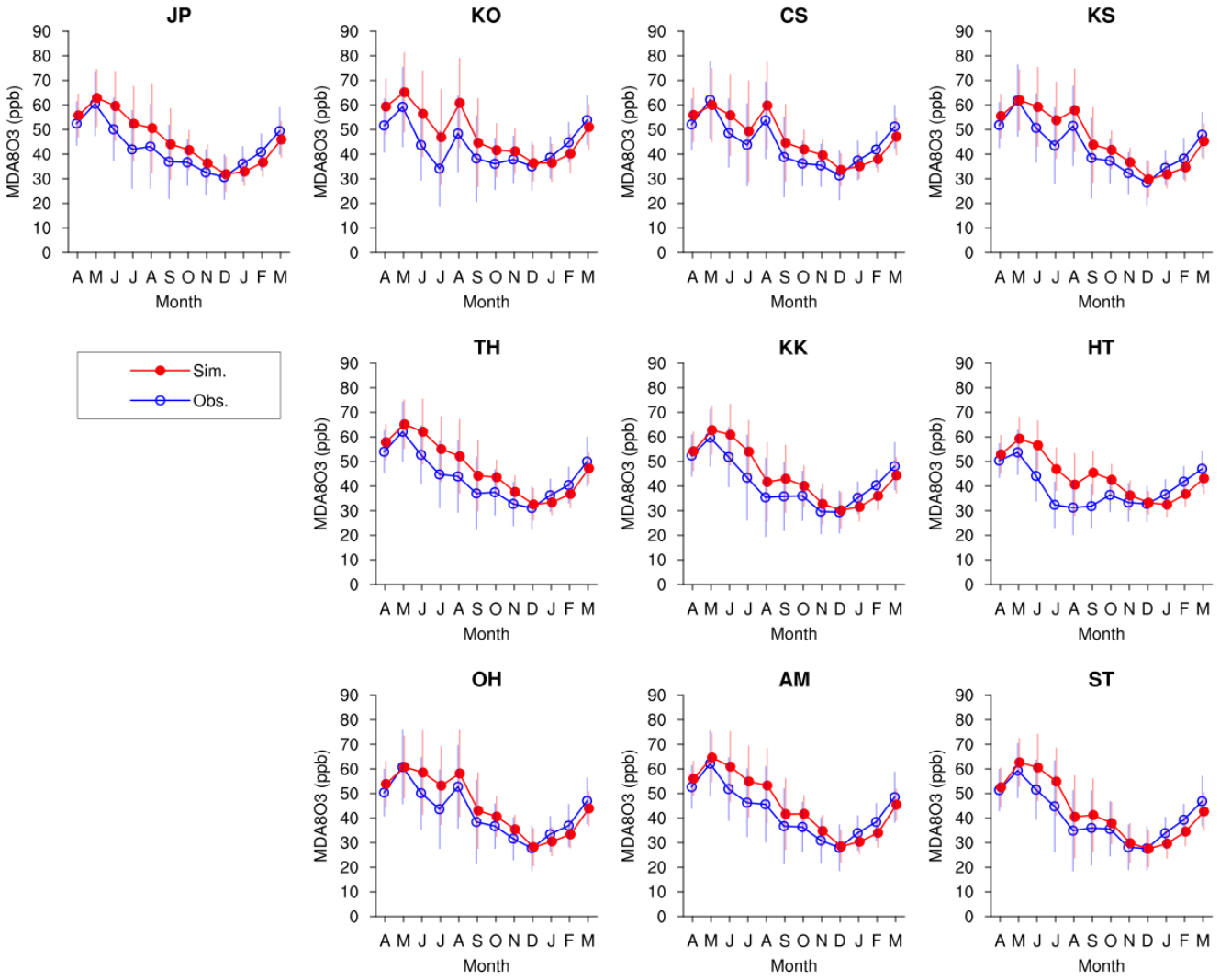
Table S3: ***Cont'd.***

(b1) $PM_{2.5}$ (not normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	4.6	3.8	3.2	5.5	6.1	8.3	3.1	9.1	10.6	12.6
s02	5.9	5.9	6.9	7.8	7.5	6.2	4.0	10.1	10.7	9.4
s03	0.7	0.5	0.4	0.8	1.0	1.5	0.5	1.5	1.8	2.6
s04	8.2	6.0	8.0	9.4	11.0	12.2	6.6	10.5	13.9	14.2
s05	0.9	0.6	0.4	1.0	1.0	1.7	1.0	2.0	2.0	3.8
s06	0.3	0.2	0.1	0.2	0.3	0.6	0.4	0.3	0.5	0.8
s07	0.3	0.1	0.3	0.4	0.5	0.9	0.1	0.7	1.0	2.0
s08	10.8	12.5	10.7	10.4	10.6	13.6	8.5	10.0	13.3	13.5
s09	7.6	11.7	9.1	8.4	7.9	7.6	3.9	7.1	6.1	5.4
s10	11.6	11.6	13.2	11.9	11.0	9.6	11.8	10.7	9.6	7.2
s11	66.2	65.9	65.4	61.8	60.4	59.7	73.1	56.8	55.6	56.4
s12	-3.9	-4.7	-4.4	-4.4	-4.3	-4.2	-2.8	-4.3	-5.4	-4.4
Sum	113.2	114.0	113.3	113.4	113.0	117.6	110.3	114.5	119.6	123.4

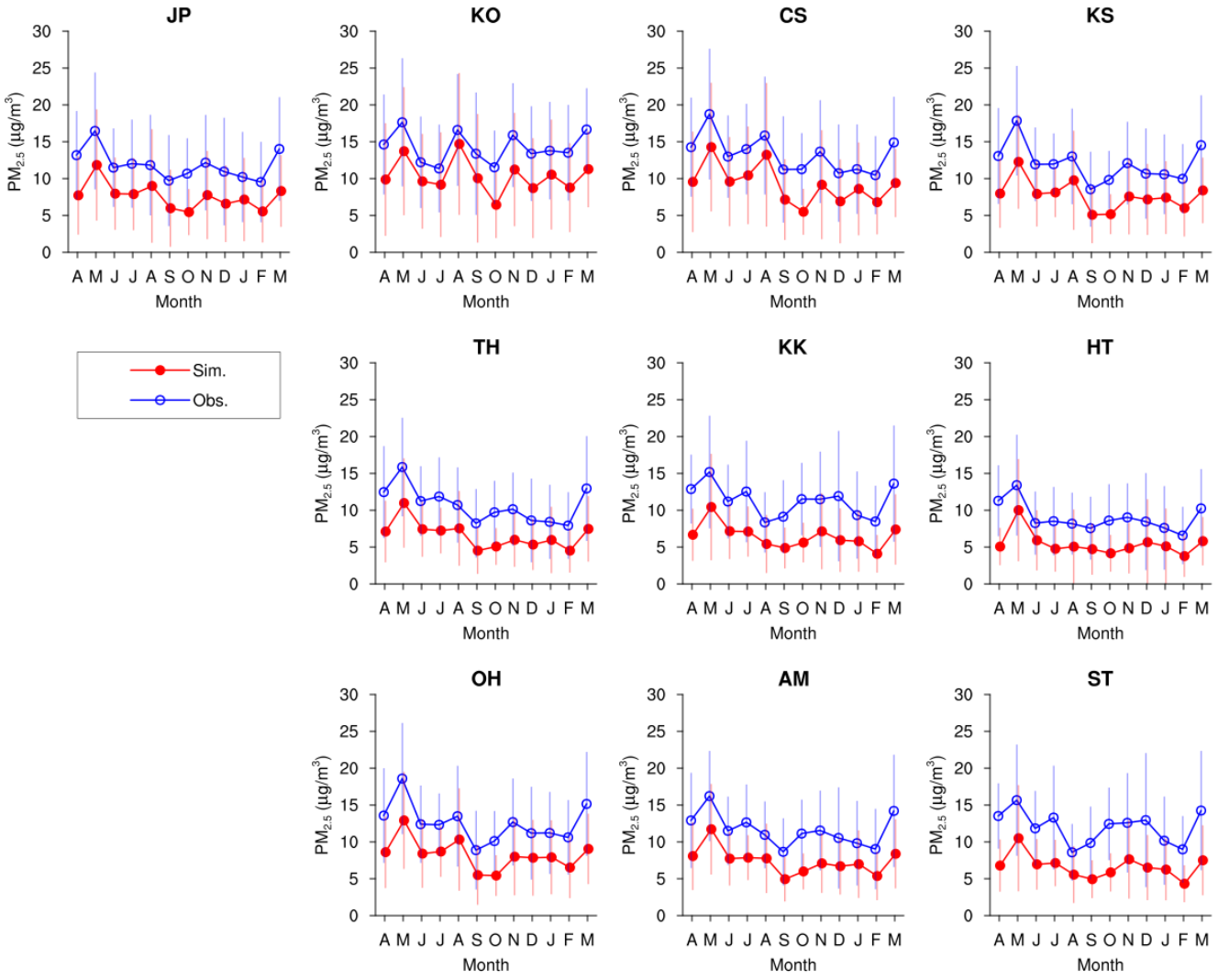
(b2) $PM_{2.5}$ (normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	4.0	3.4	2.8	4.9	5.4	7.1	2.8	8.0	8.8	10.2
s02	5.2	5.2	6.1	6.9	6.6	5.2	3.6	8.8	8.9	7.6
s03	0.6	0.4	0.3	0.7	0.9	1.3	0.4	1.3	1.5	2.1
s04	7.3	5.2	7.0	8.3	9.7	10.4	6.0	9.1	11.6	11.5
s05	0.8	0.5	0.4	0.9	0.9	1.4	0.9	1.7	1.7	3.1
s06	0.3	0.1	0.1	0.2	0.3	0.5	0.4	0.2	0.4	0.7
s07	0.3	0.1	0.2	0.4	0.5	0.8	0.1	0.6	0.8	1.6
s08	9.5	11.0	9.5	9.2	9.4	11.6	7.7	8.8	11.1	10.9
s09	6.7	10.2	8.0	7.4	7.0	6.4	3.6	6.2	5.1	4.4
s10	10.3	10.2	11.7	10.5	9.7	8.1	10.7	9.4	8.1	5.9
s11	58.5	57.8	57.7	54.5	53.4	50.8	66.3	49.7	46.5	45.7
s12	-3.5	-4.1	-3.9	-3.8	-3.8	-3.6	-2.5	-3.7	-4.5	-3.6



(a) MDA8O3

Figure S1: Comparisons of the observed and simulated monthly mean MDA8O3 and $PM_{2.5}$ concentrations at all stations in the regions. Markers and error bars represent mean values and standard deviations, respectively, of the daily concentrations at all monitoring stations for each month.



(b) $PM_{2.5}$

Figure S1: ***Cont'd***

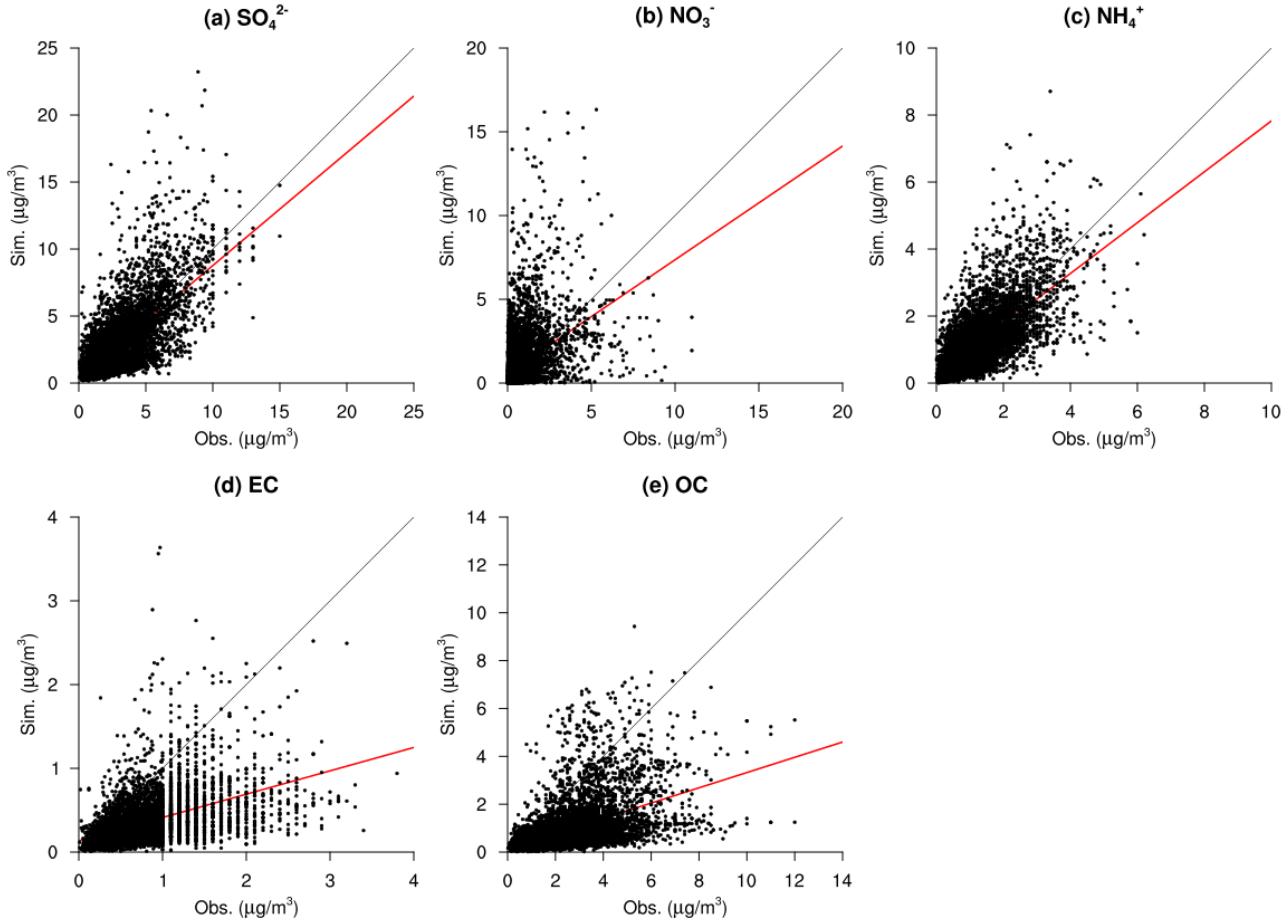


Figure S2: Scatter plots of the observed and simulated concentrations of $PM_{2.5}$ components during the monitoring campaigns of ambient concentrations of $PM_{2.5}$ components at all the locations throughout Japan in all four seasons. Regression lines are represented by red lines.

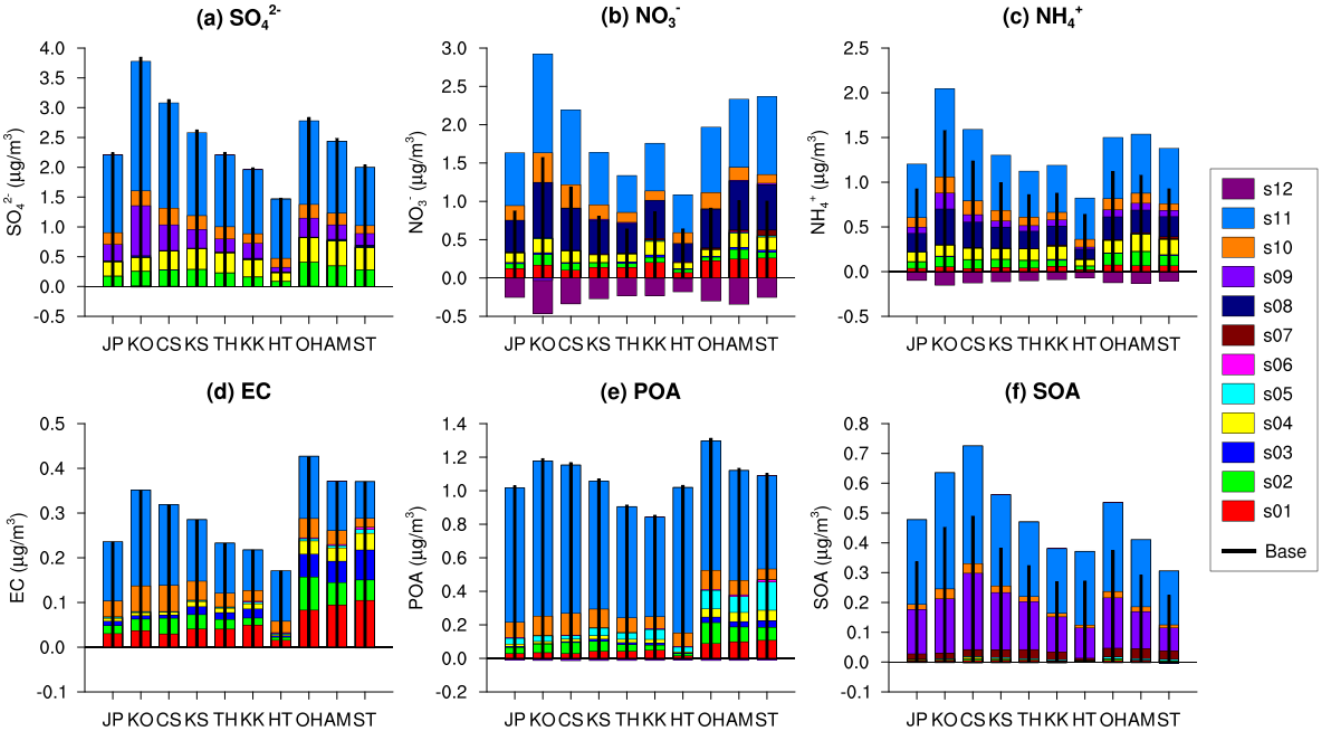


Figure S3: Source sensitivities of the annual mean concentrations of $PM_{2.5}$ components derived by BFM in the regions. Thick black lines represent the simulated concentrations.

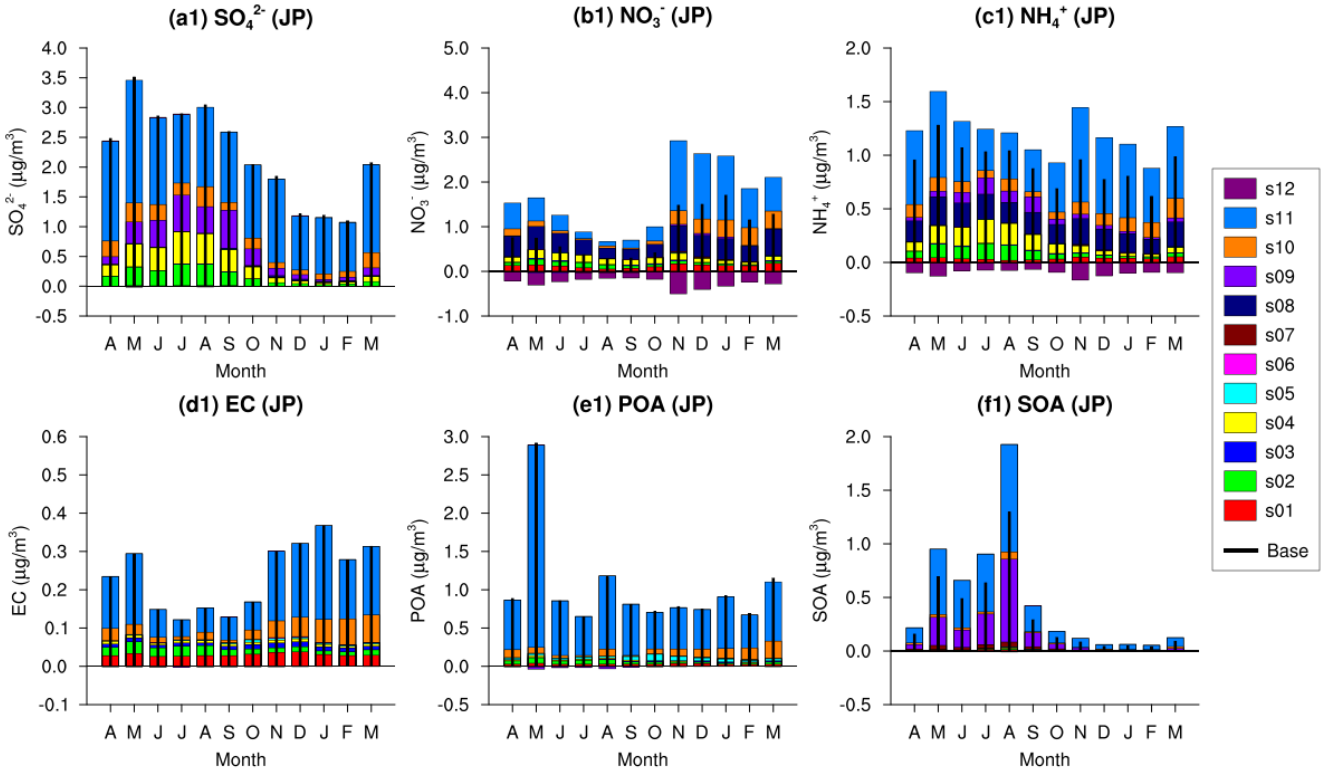


Figure S4: Source sensitivities of the monthly mean concentrations of $PM_{2.5}$ components derived by BFM in entire Japan (JP) and ST. Thick black lines represent the simulated concentrations.

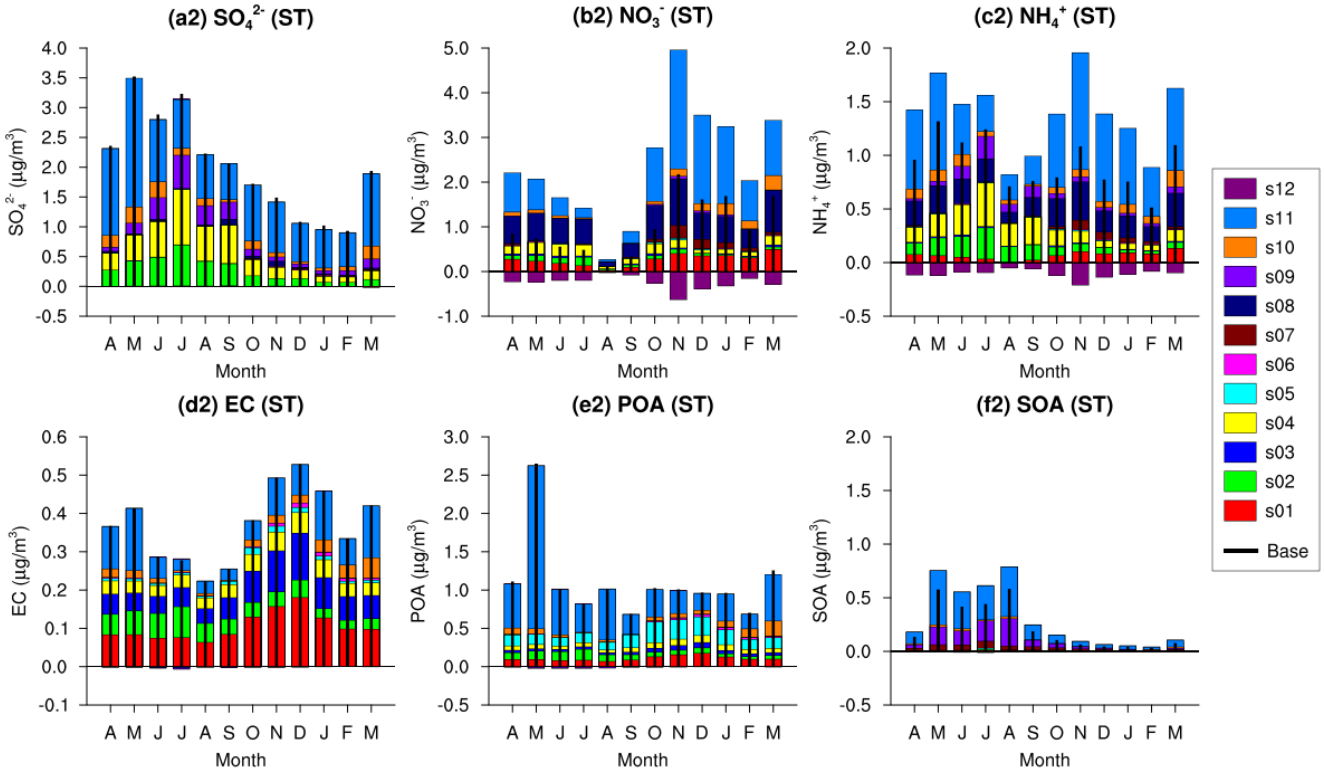


Figure S4: ***Cont'd***

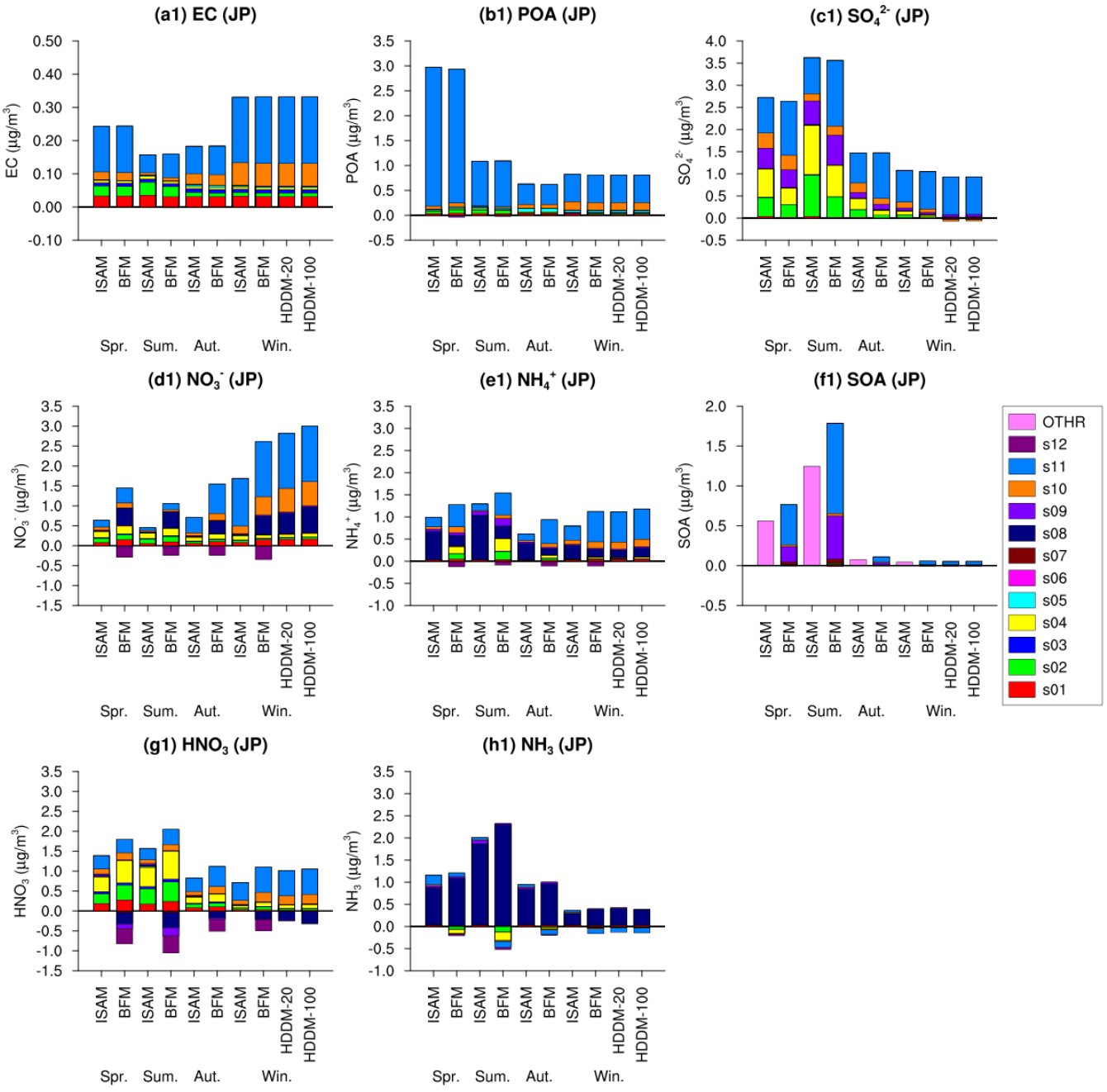


Figure S5: Apportionments derived by ISAM and sensitivities derived by BFM and HDDM of the simulated concentrations of $PM_{2.5}$ components to all source groups in JP and ST for the two target weeks in the four seasons.

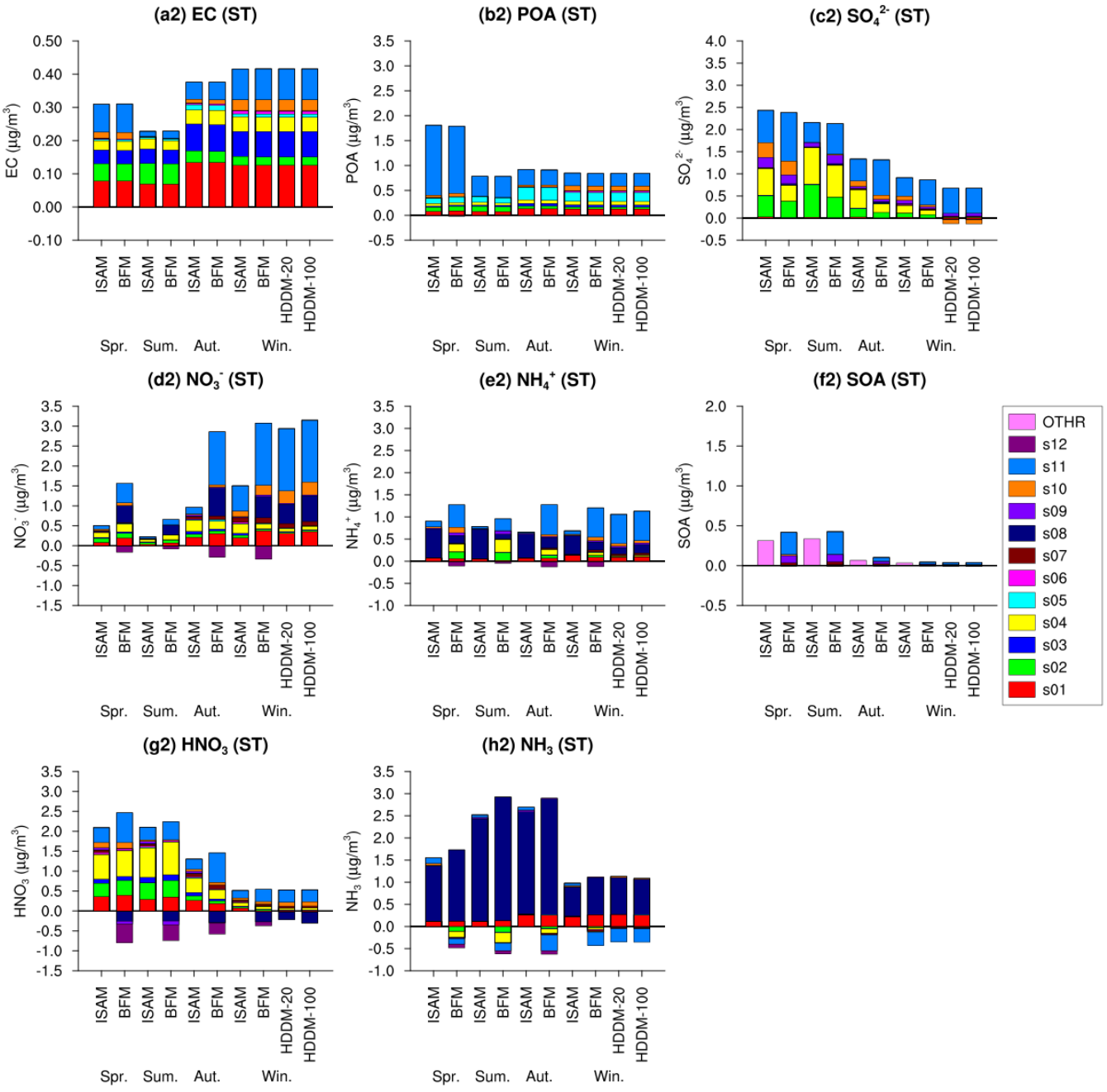


Figure S5: ***Cont'd***

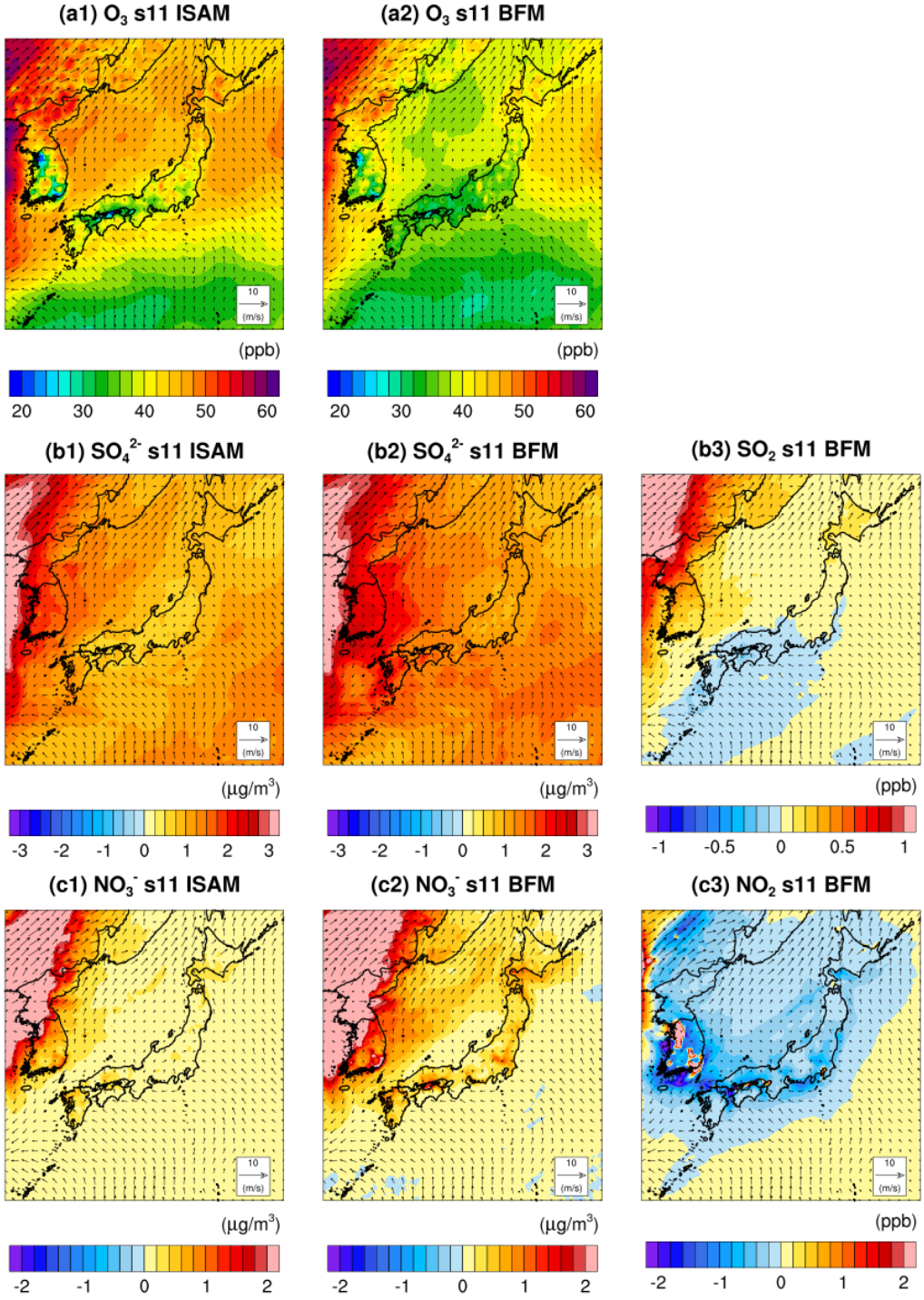


Figure S6: Horizontal distributions of the apportionments and the sensitivities to s11 for the target two weeks of the spring.

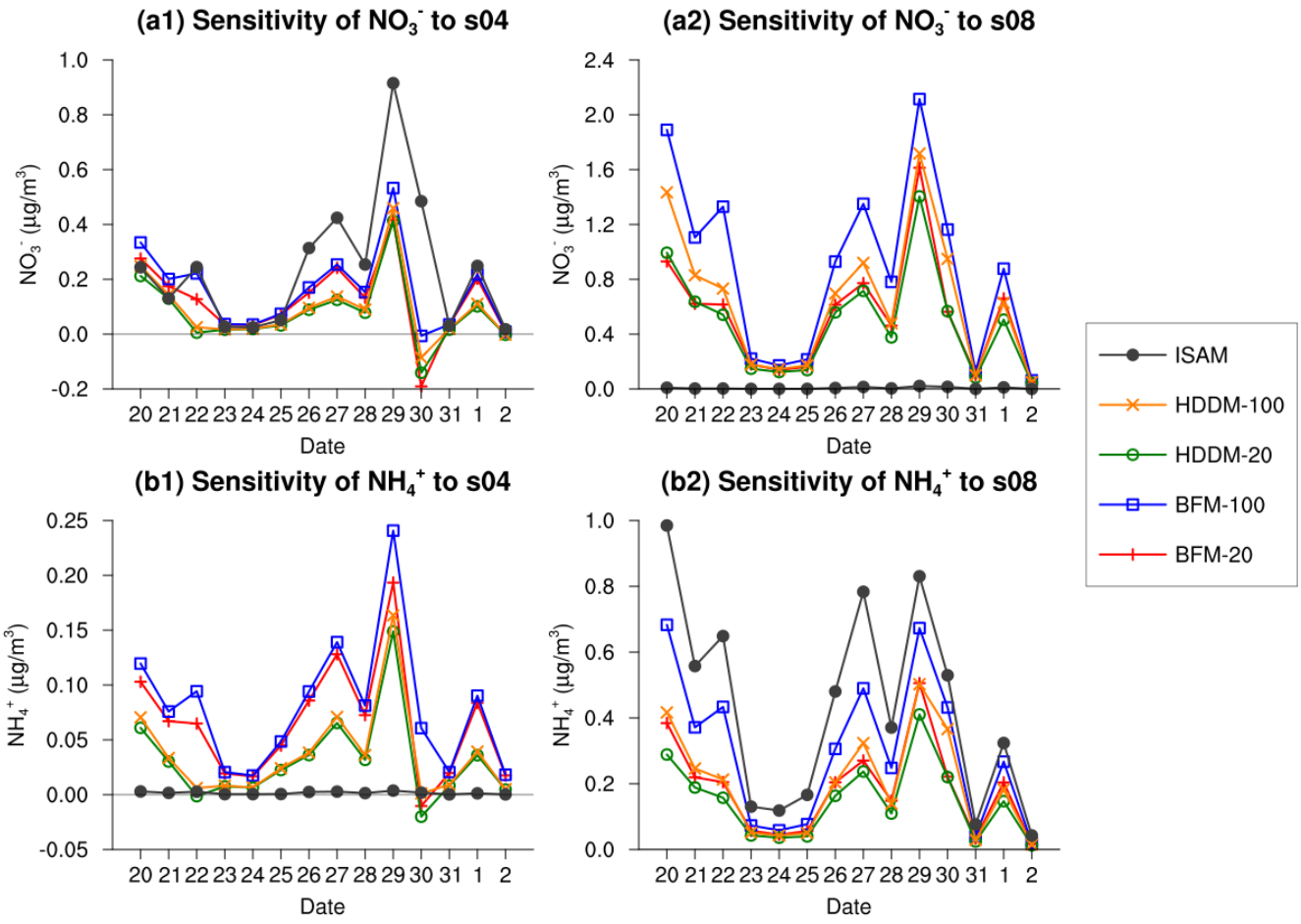


Figure S7: Sensitivities derived by BFM-20, BFM-100, HDDM-20, and HDDM-100, and apportionments derived by ISAM of the daily NO_3^- and NH_4^+ concentrations to s04 and s08 for the two target weeks in winter in ST.

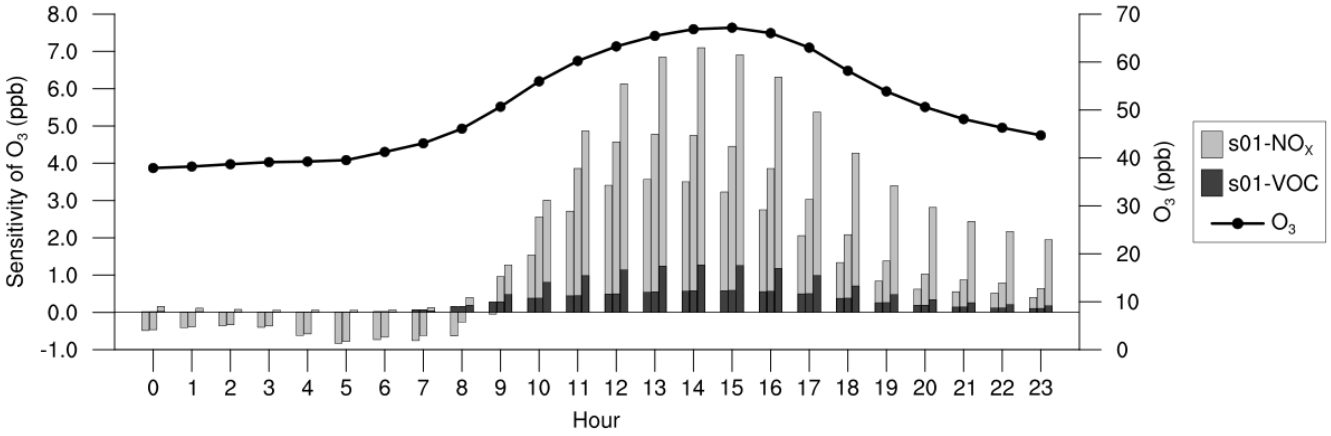


Figure S8: Sensitivities derived by BFM-20 (left) and BFM-100 (middle), and apportionments derived by ISAM (right) of the hourly ozone concentrations (shown by a line with markers) to NO_X and VOC emissions of s01on July 25th in ST.

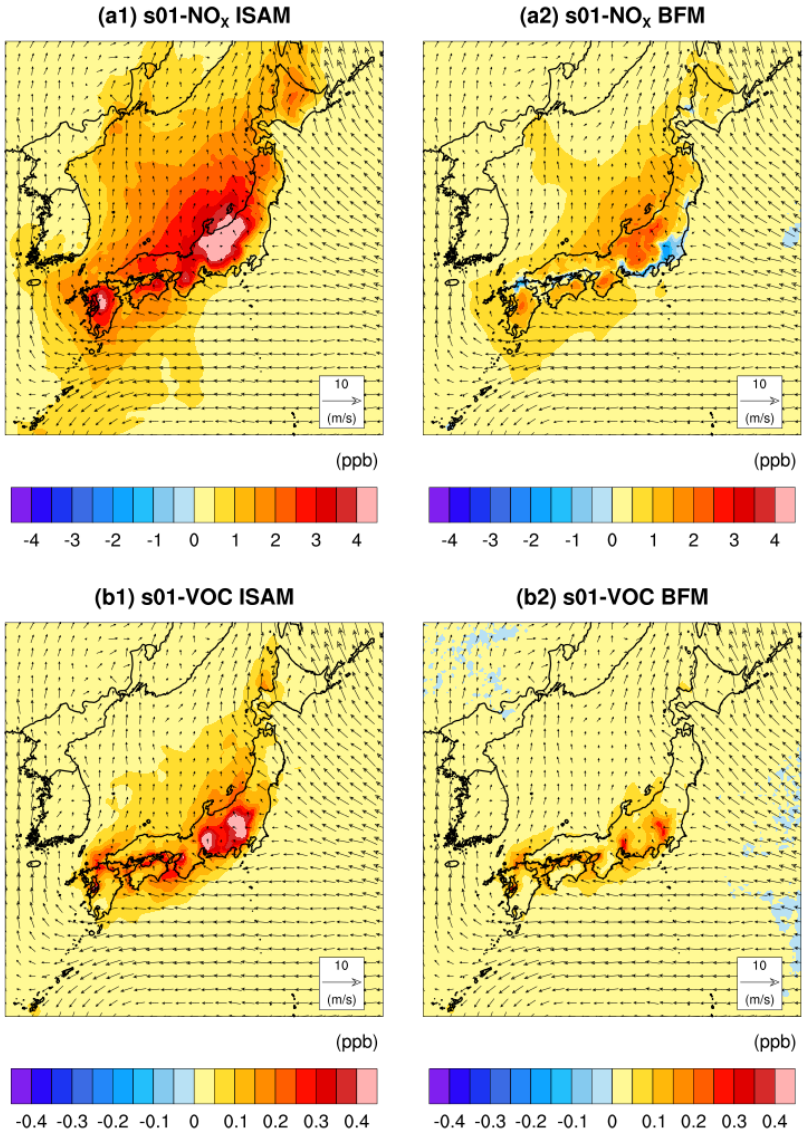


Figure S9: Horizontal distributions of the apportionments and sensitivities of the ozone concentrations to the s01 NO_X and VOC emissions averaged for the two target weeks in summer.

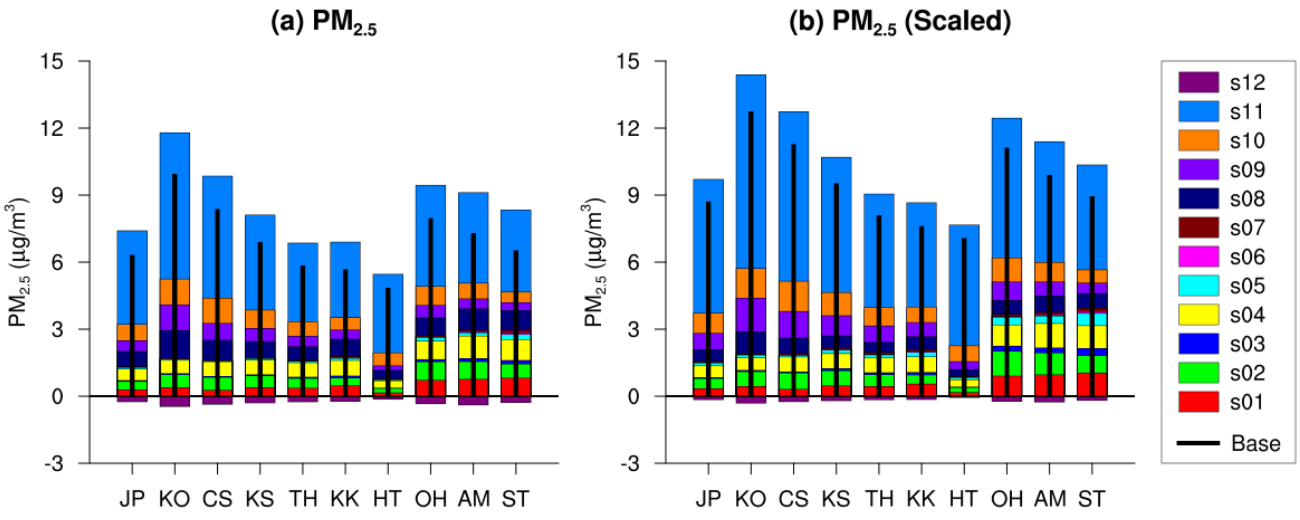


Figure s10. Source sensitivities of the annual mean $PM_{2.5}$ concentrations derived by BFM in the regions. Thick black lines represent the simulated concentrations. The values shown in (b) were scaled by ratios of observed and simulated concentrations of $PM_{2.5}$ components.