

Supplementary

Fig. S1

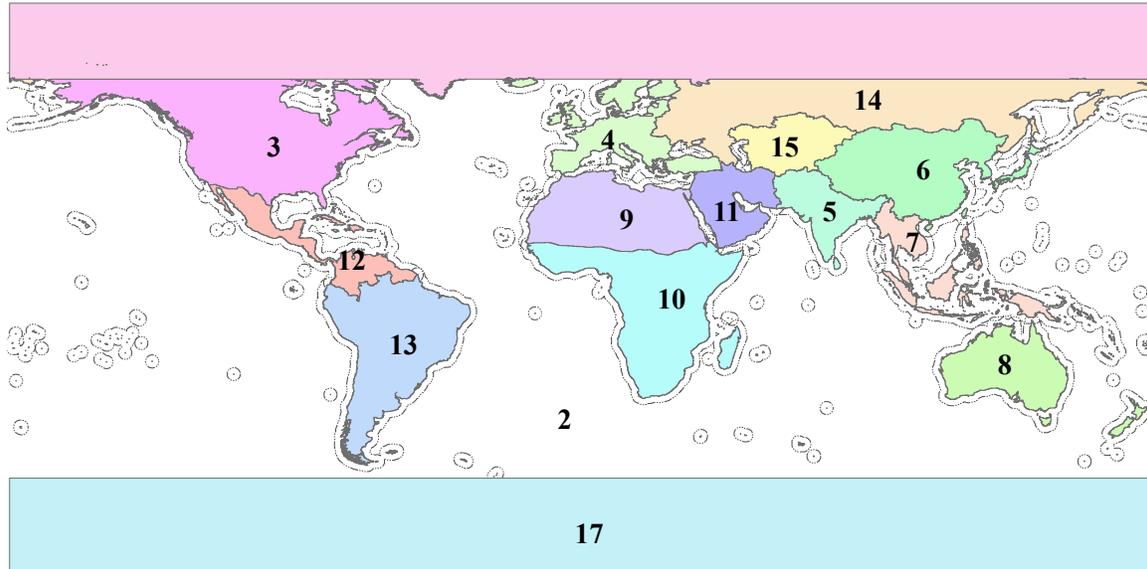
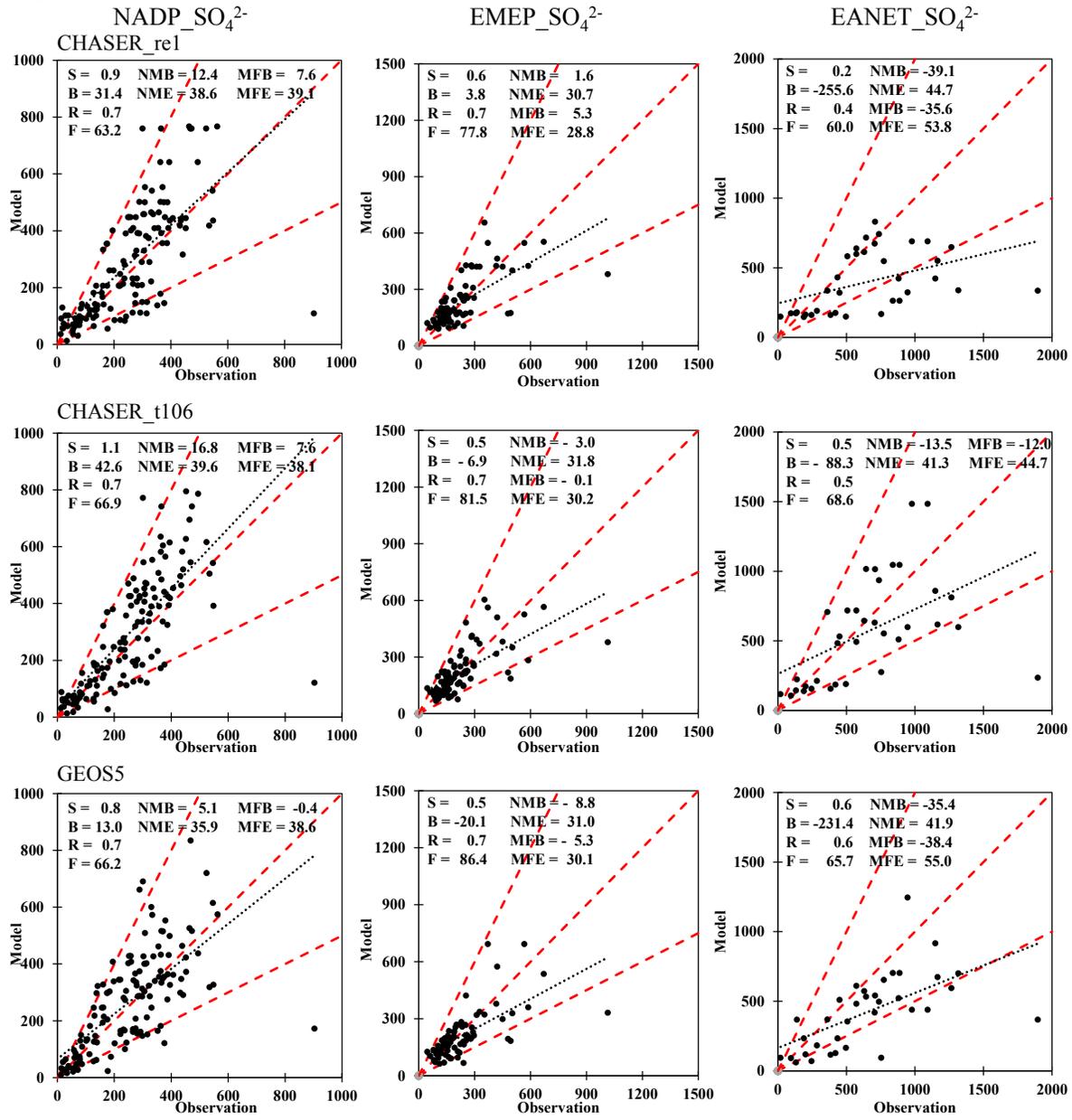


Fig. S1. Regions defined in HTAP phase II and coastal area. Region 1-Global, 2-Ocean (include Arctic), 3-North America, 4-Europe, 5-South Asia, 6-East Asia, 7-Southeast Asia, 8-Australia, 9-North Africa, 10- Sub Saharan Africa, 11-Middle East, 12- Mexico, Central America, Caribbean, Guyanas, Venezuela, Columbia (Central America), 13-South America, 14-Russia, Belarussia, Ukraine (RBU), 15-Central Asia, 17-Antarctic.

Fig. S2



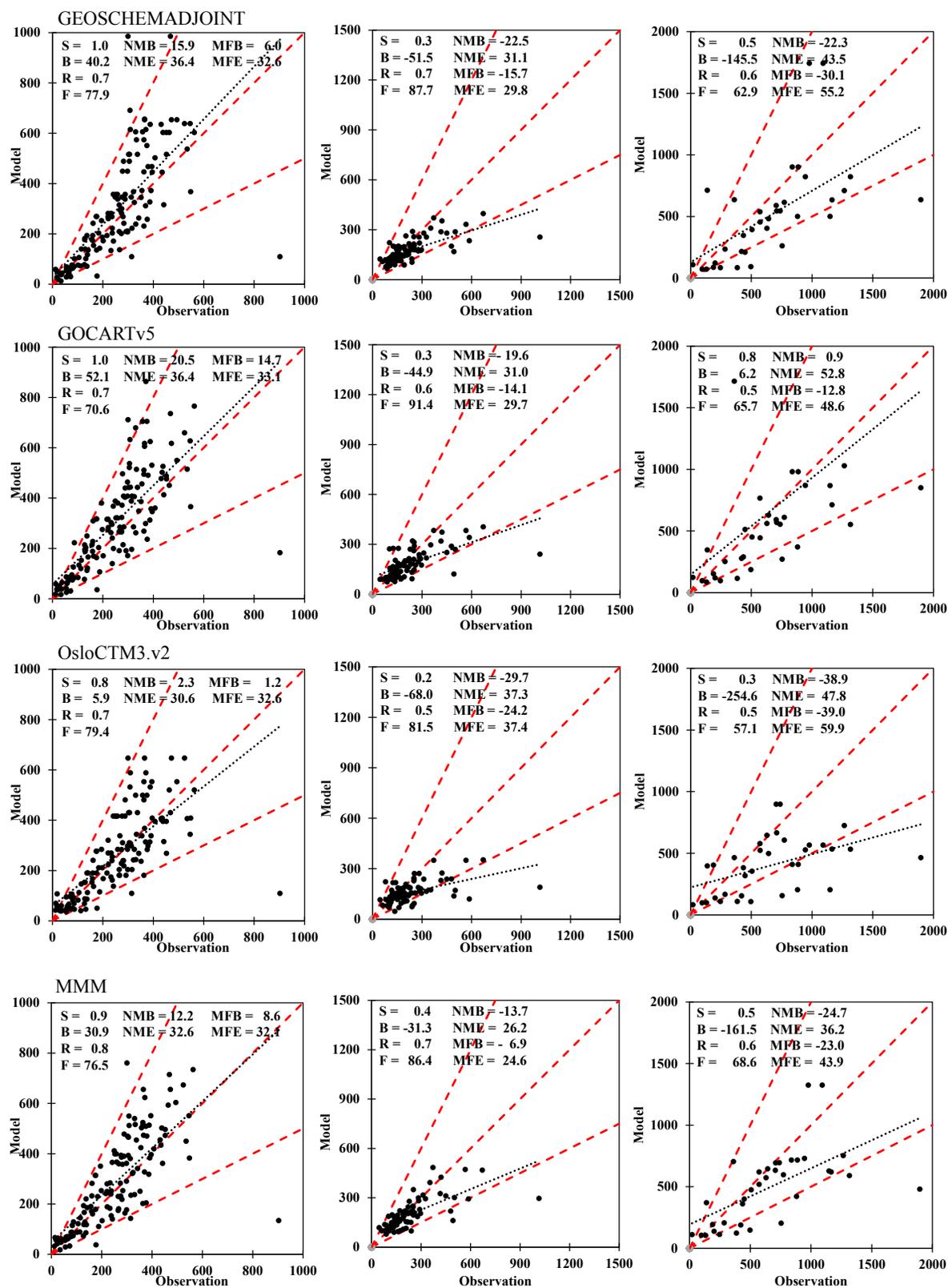
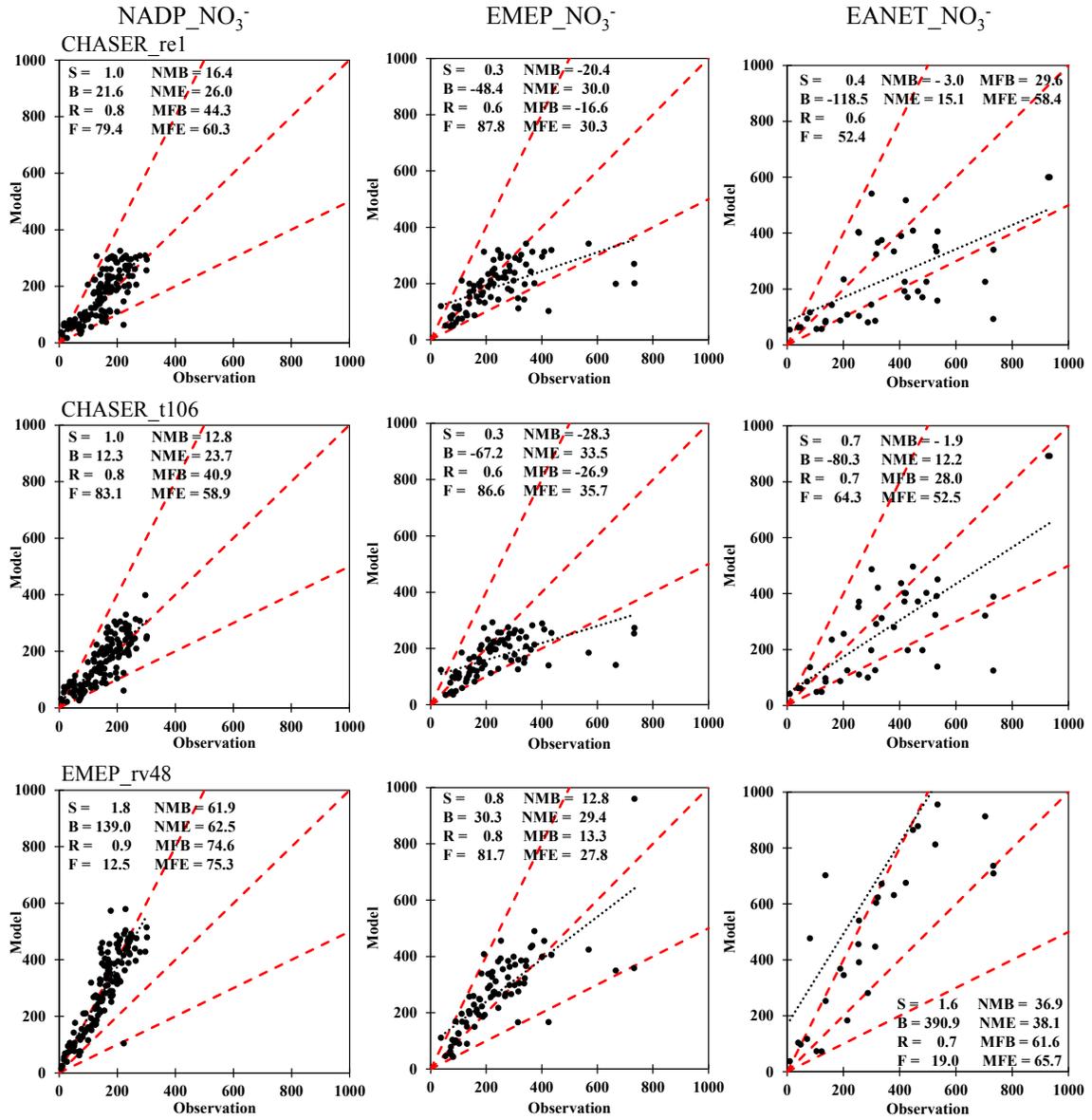


Fig. S2. Individual model performances on SO_4^{2-} wet deposition ($\text{mg (S) m}^{-2}\text{yr}^{-1}$). The model result is the annual deposition in 2010 and the observation is 3-year average annual data of 2009-2011.

Fig. S3



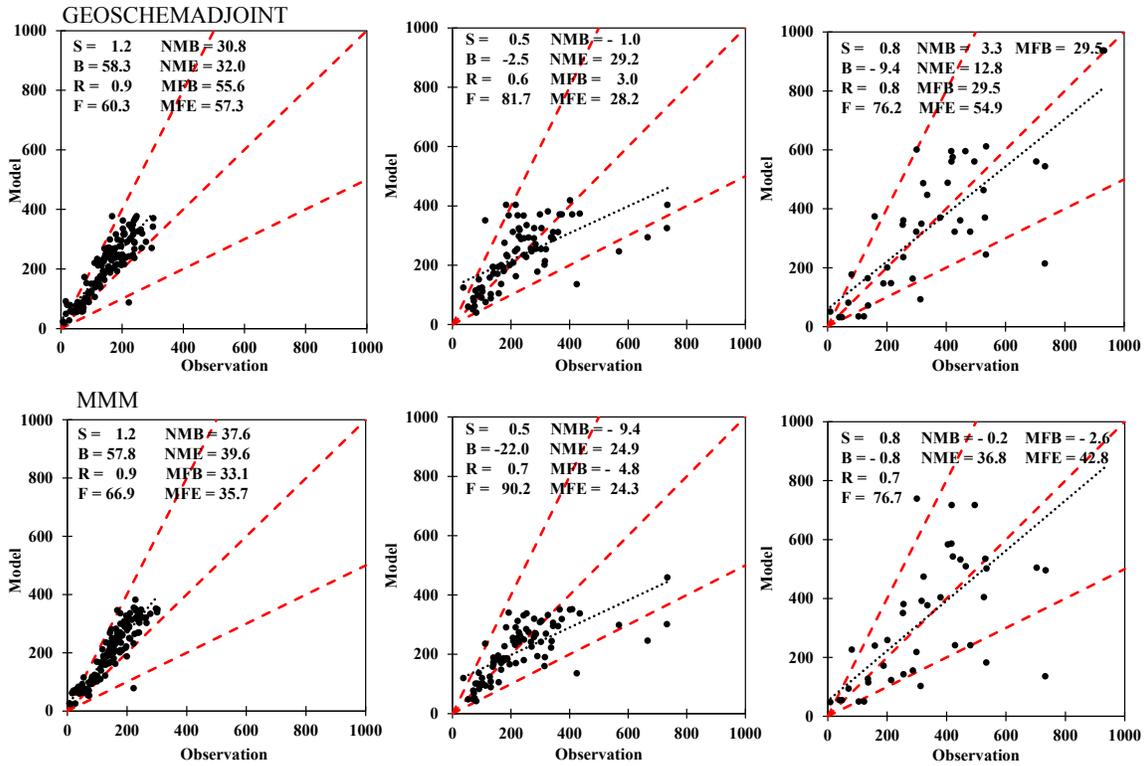


Fig. S3. Same as Fig. S2 but for NO_3^- wet deposition ($\text{mg (N) m}^{-2}\text{yr}^{-1}$)

Fig. S4

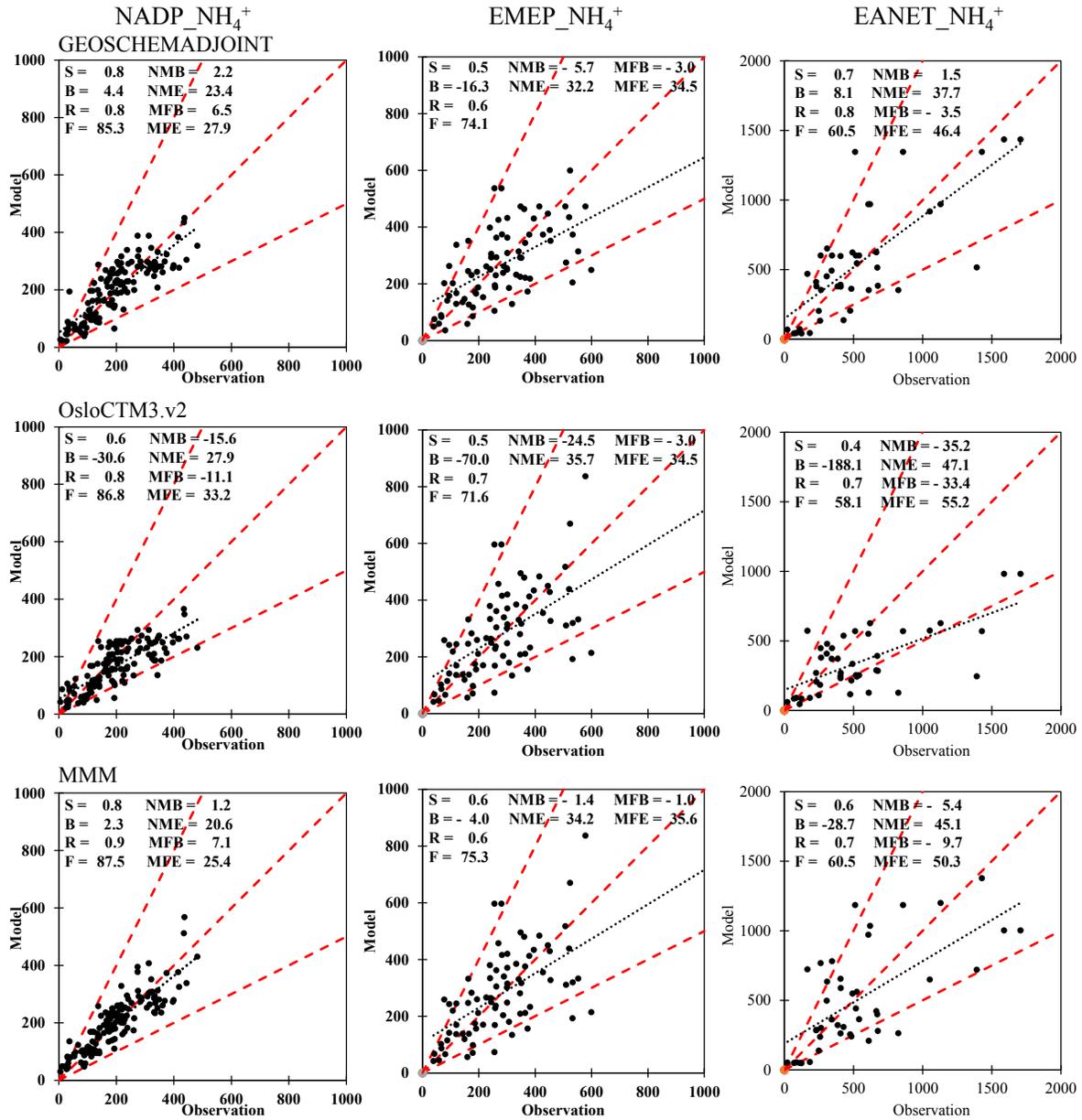
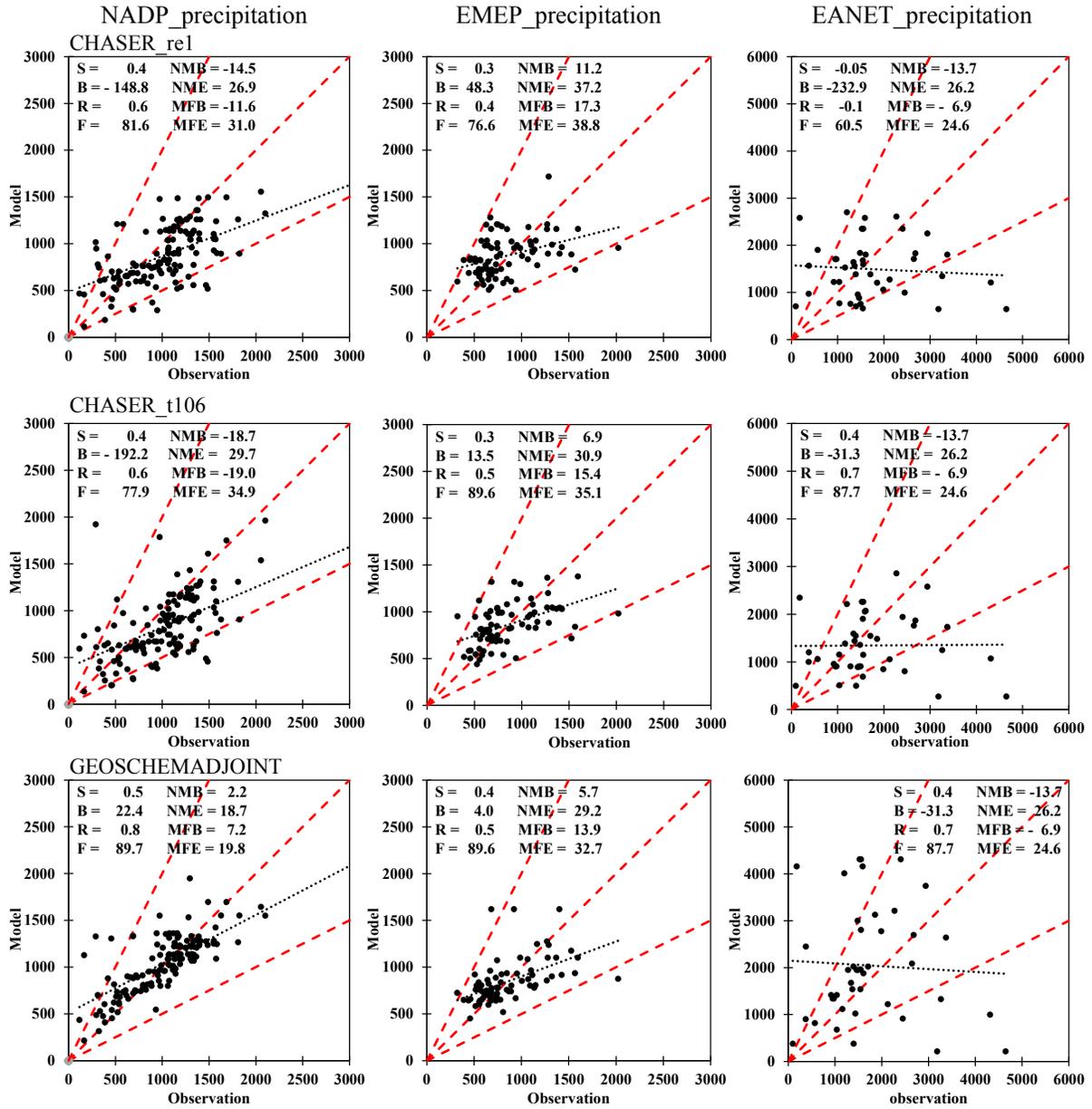


Fig. S4 Same as Fig. S2 but for NH_4^+ wet deposition ($\text{mg (N) m}^{-2}\text{yr}^{-1}$)

Fig. S5



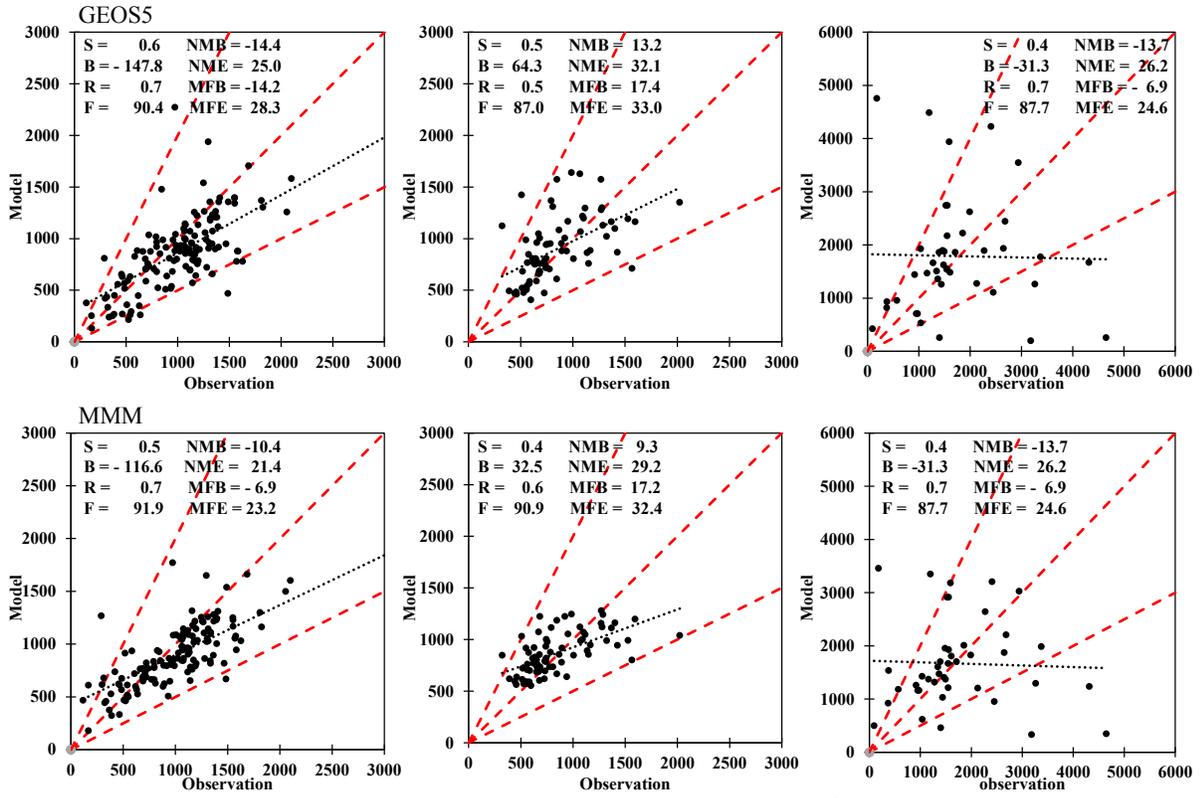


Fig. S5. Individual model performances on precipitation (mm yr^{-1}). The model result is the annual precipitation in 2010 and the observation is 3-year average annual data of 2009-2011.

Fig. S6

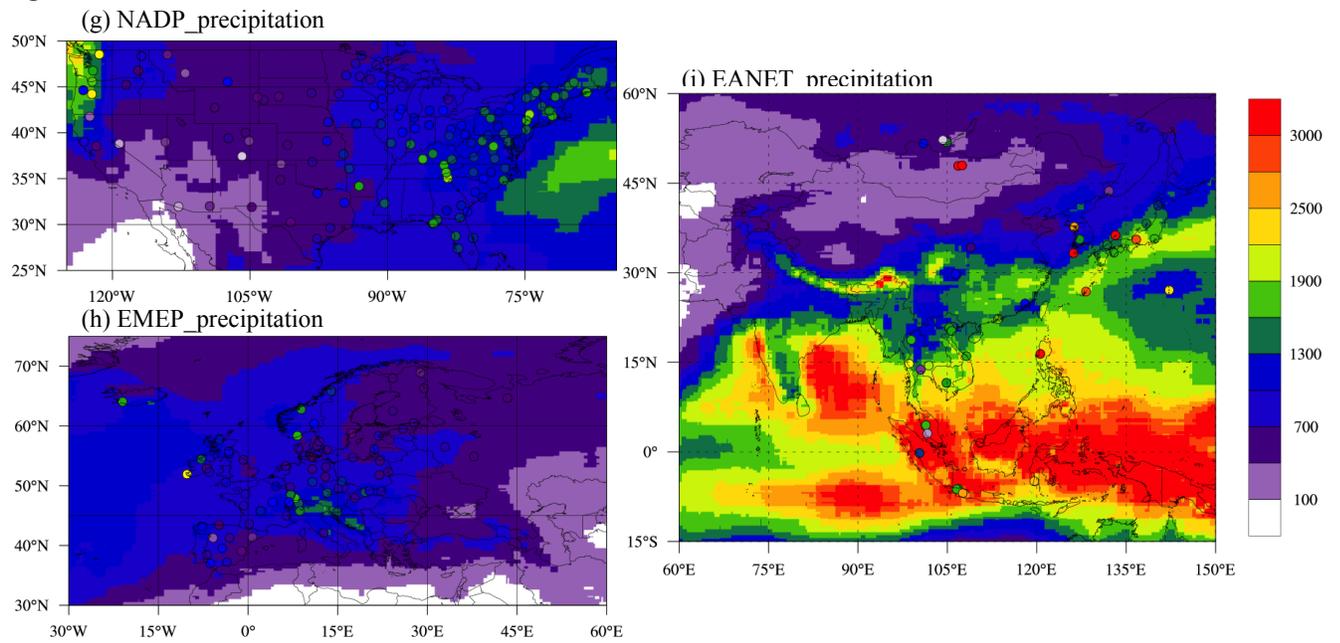


Fig. S6. Distribution of precipitation (mm yr^{-1}) of MMM and observation. The MMM is the annual total precipitation in 2010 and the observation is 3-year average annual data of 2009-2011. Contours are MMM results and filled circles are observation.

Fig. S7

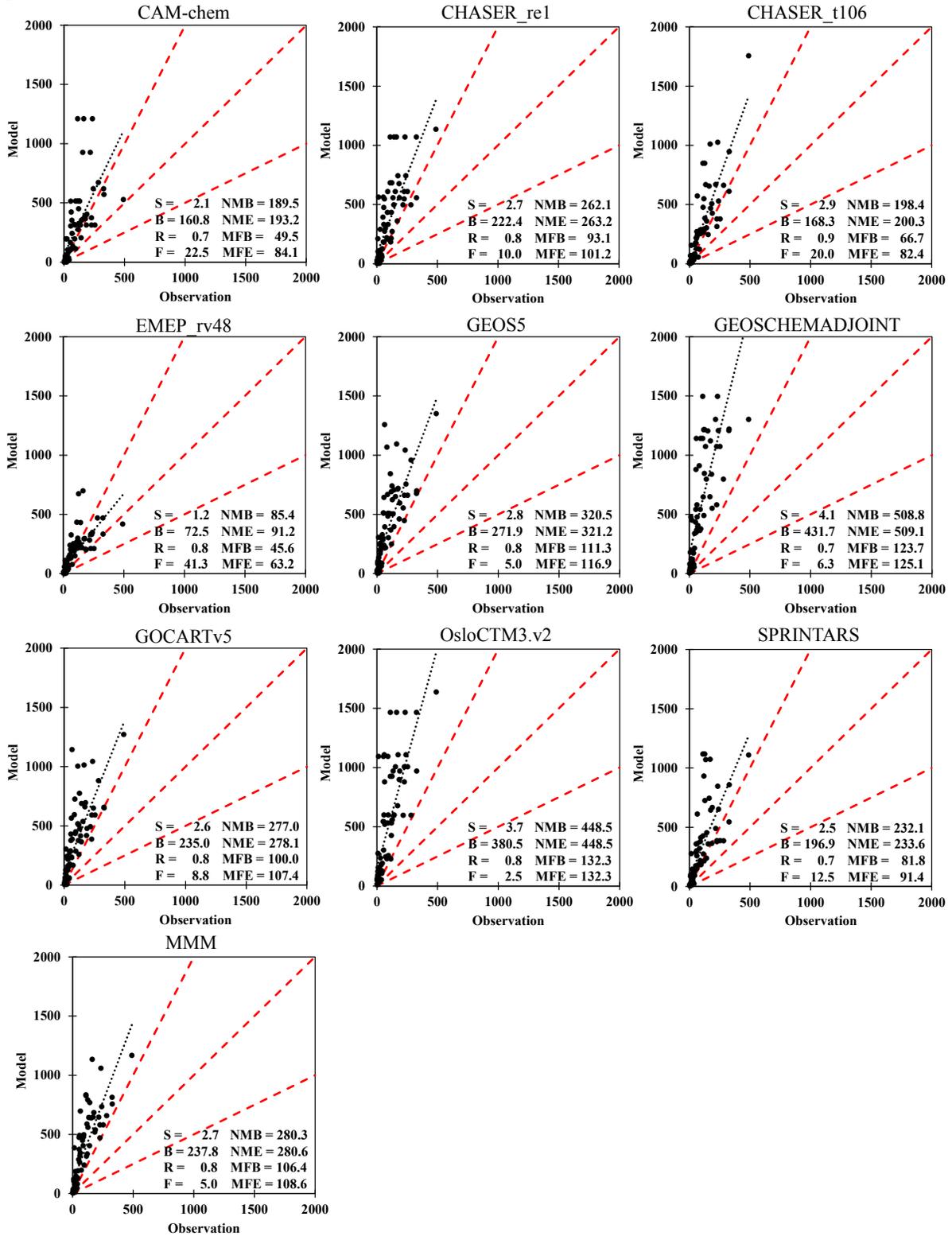


Fig. S7. Individual model performances on SO₂ dry deposition (mg (S) m⁻²yr⁻¹). The model result is the annual deposition in 2010 and the observation is 3-year average annual data of 2009-2011.

Fig. S8

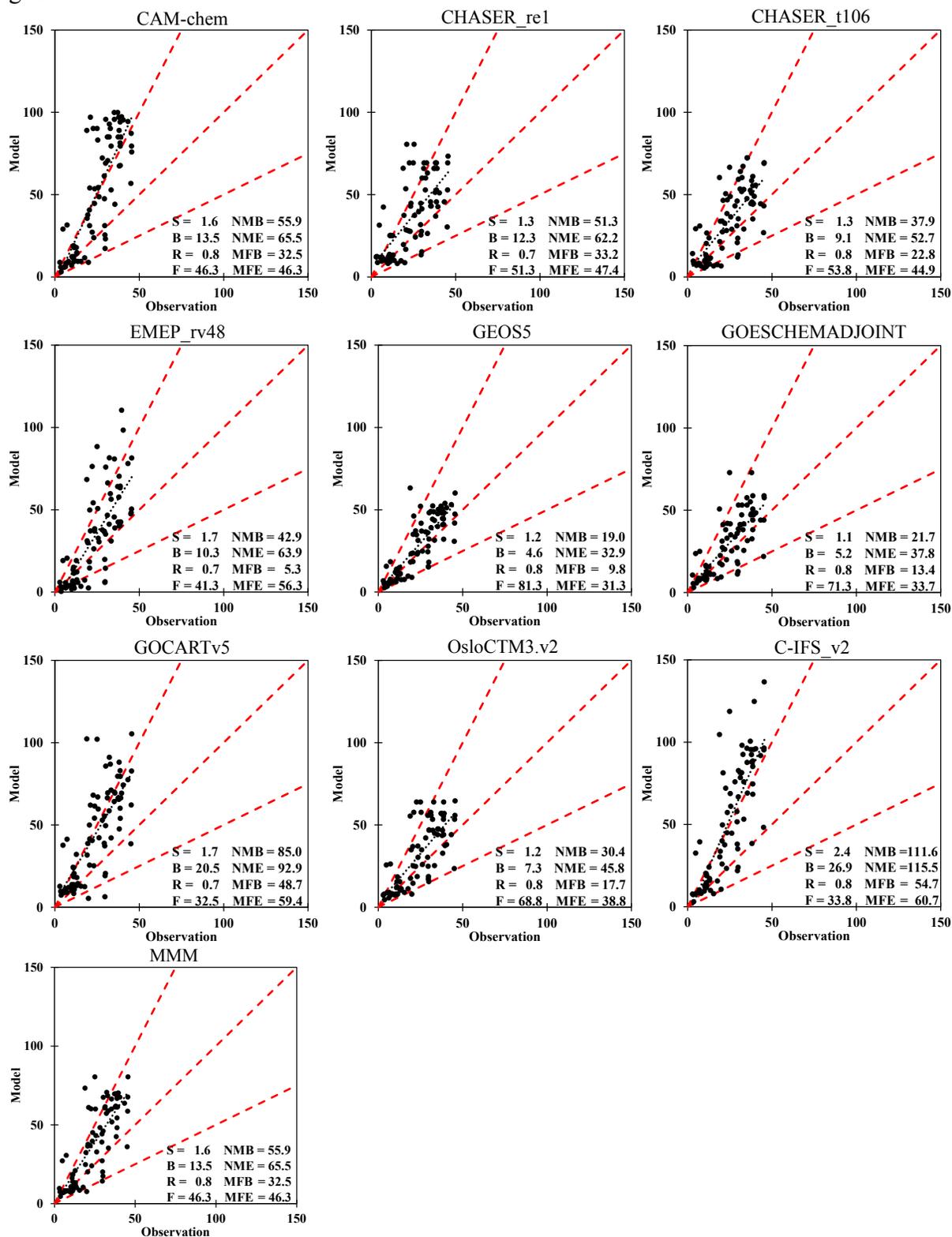


Fig. S8. Same as Fig. S7 but for SO_4^{2-} dry deposition ($\text{mg (S) m}^{-2}\text{yr}^{-1}$)

Fig. S9

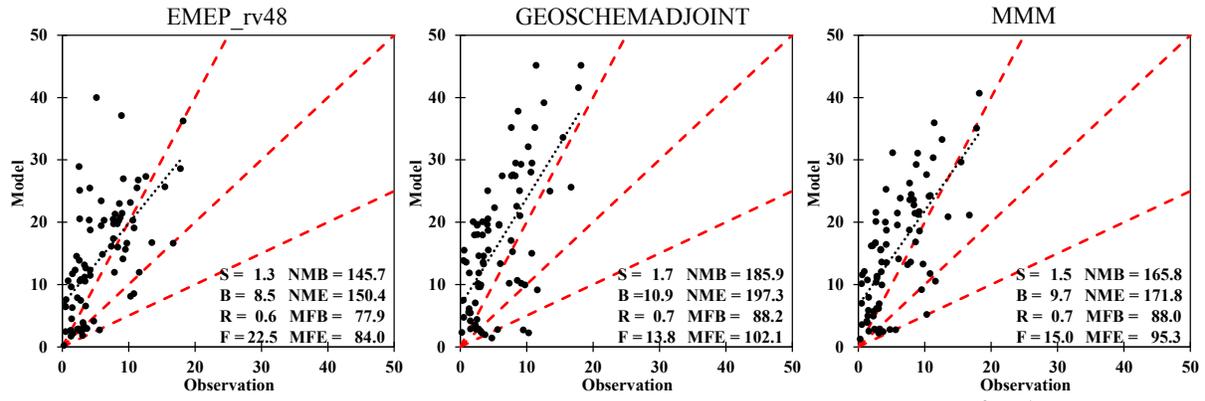


Fig. S9. Same as Fig. S7 but for NO_3^- dry deposition ($\text{mg (N) m}^{-2}\text{yr}^{-1}$)

Fig. S10

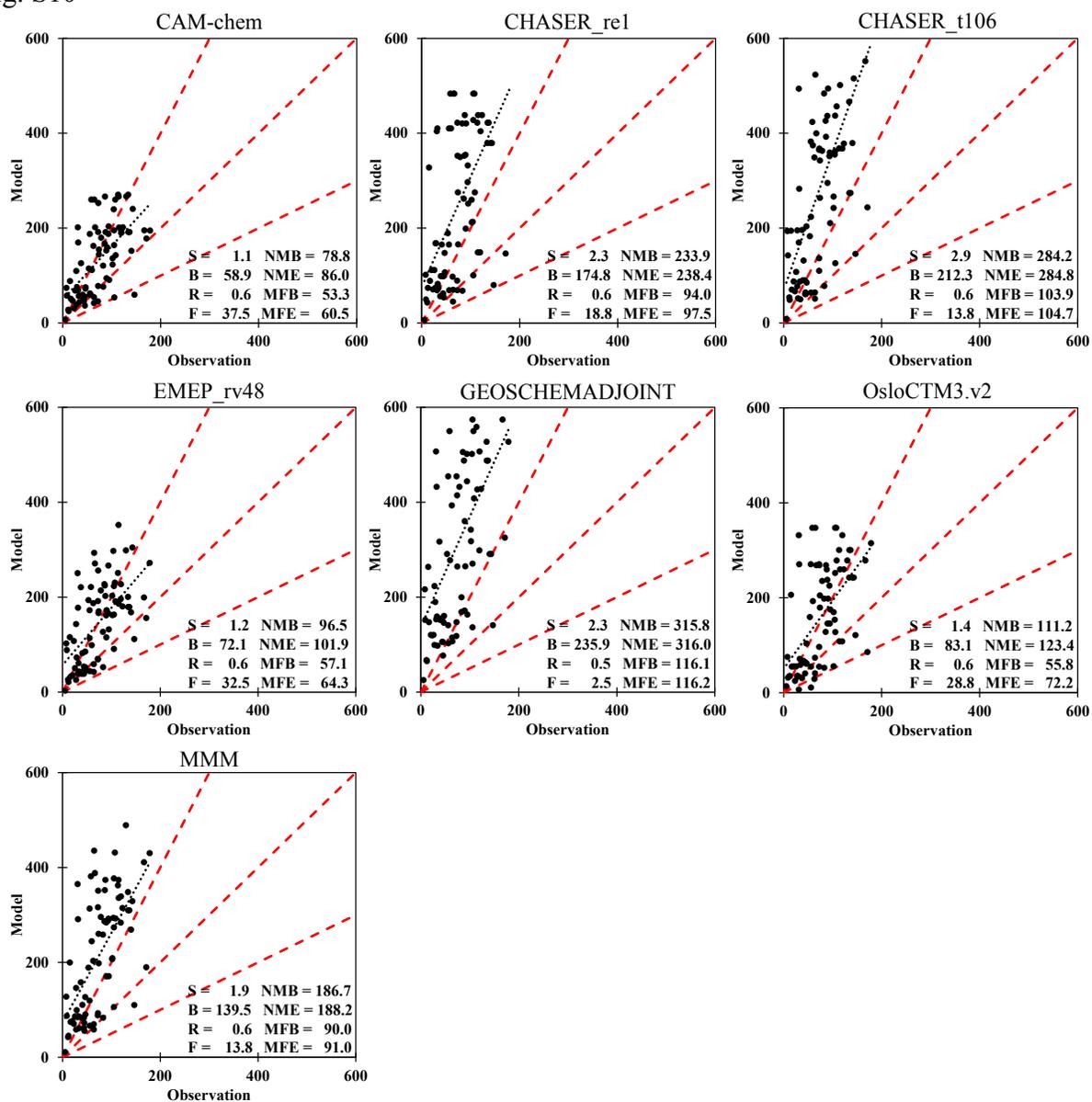


Fig. S10. Same as Fig. S7 but for HNO_3 dry deposition ($\text{mg (N) m}^{-2}\text{yr}^{-1}$)

Fig. S11

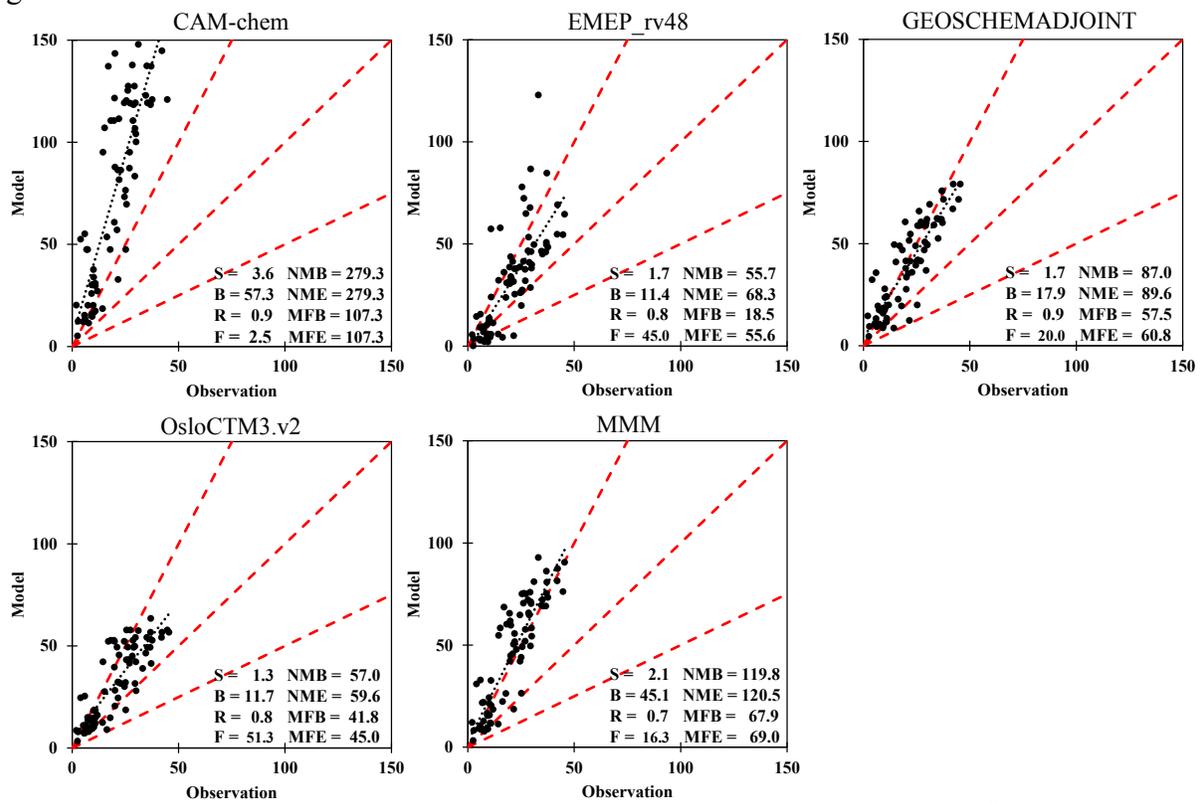


Fig. S11. Same as Fig. S7 but for NH_4^+ dry deposition ($\text{mg (N) m}^{-2}\text{yr}^{-1}$)

Fig. S12

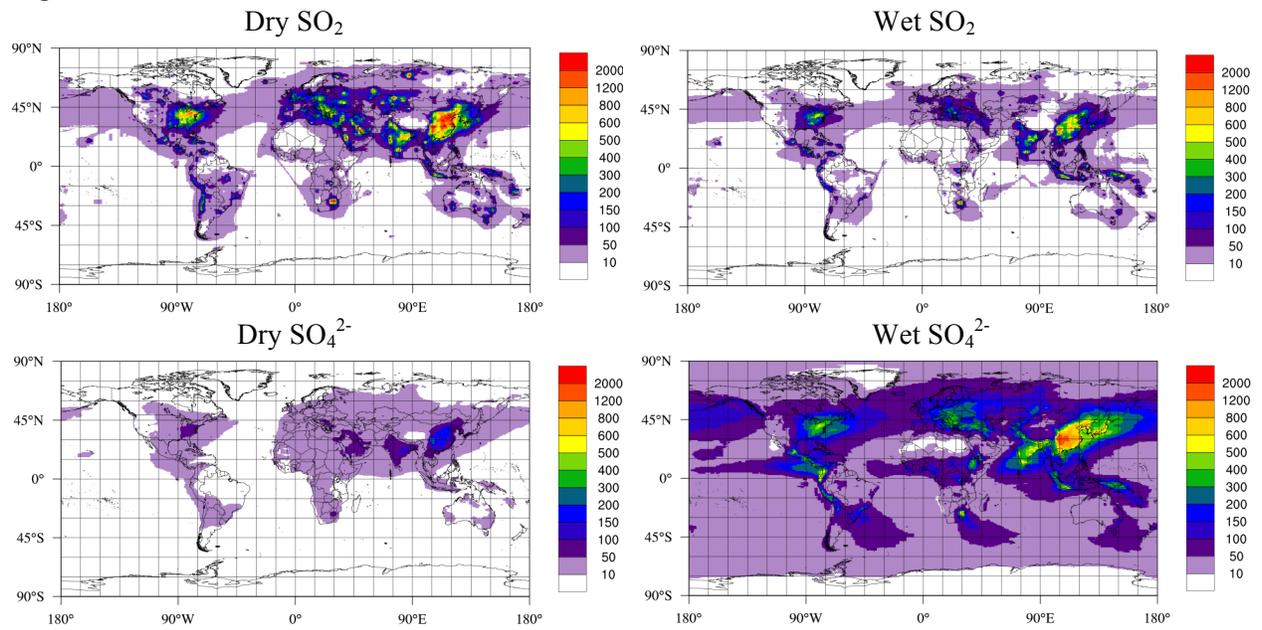


Fig. S12 Spatial distribution of SO_2 and SO_4^{2-} deposition in 2010 from MMM results ($\text{mg(S)} \text{m}^{-2} \text{yr}^{-1}$).

Fig. S13

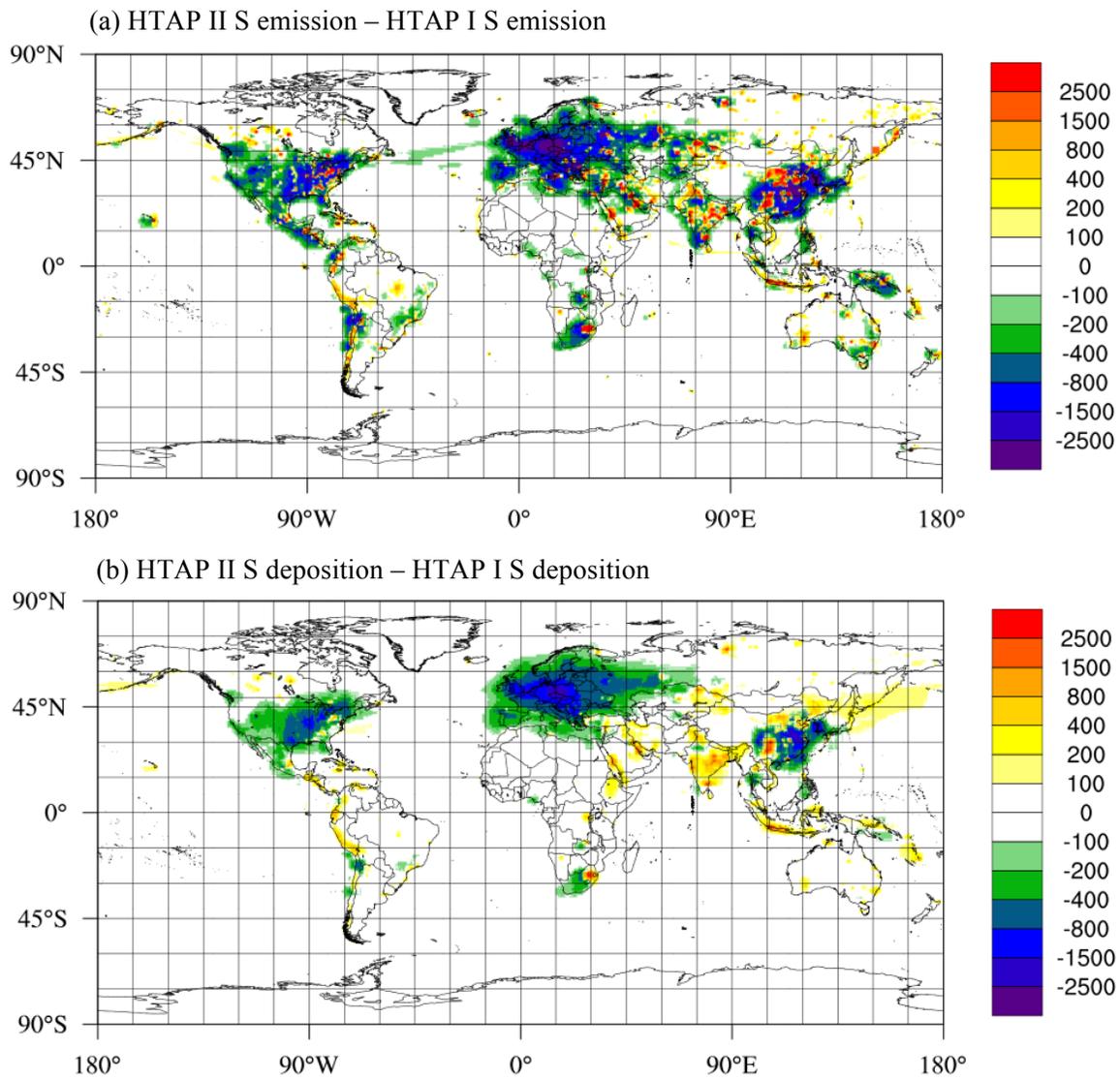


Fig. S13 Difference between MMM of HTAP II and HTAP I for (a) S emission (b) S Deposition. Unit: $\text{mg(S)} \text{ m}^{-2} \text{ yr}^{-1}$.

Fig. S14

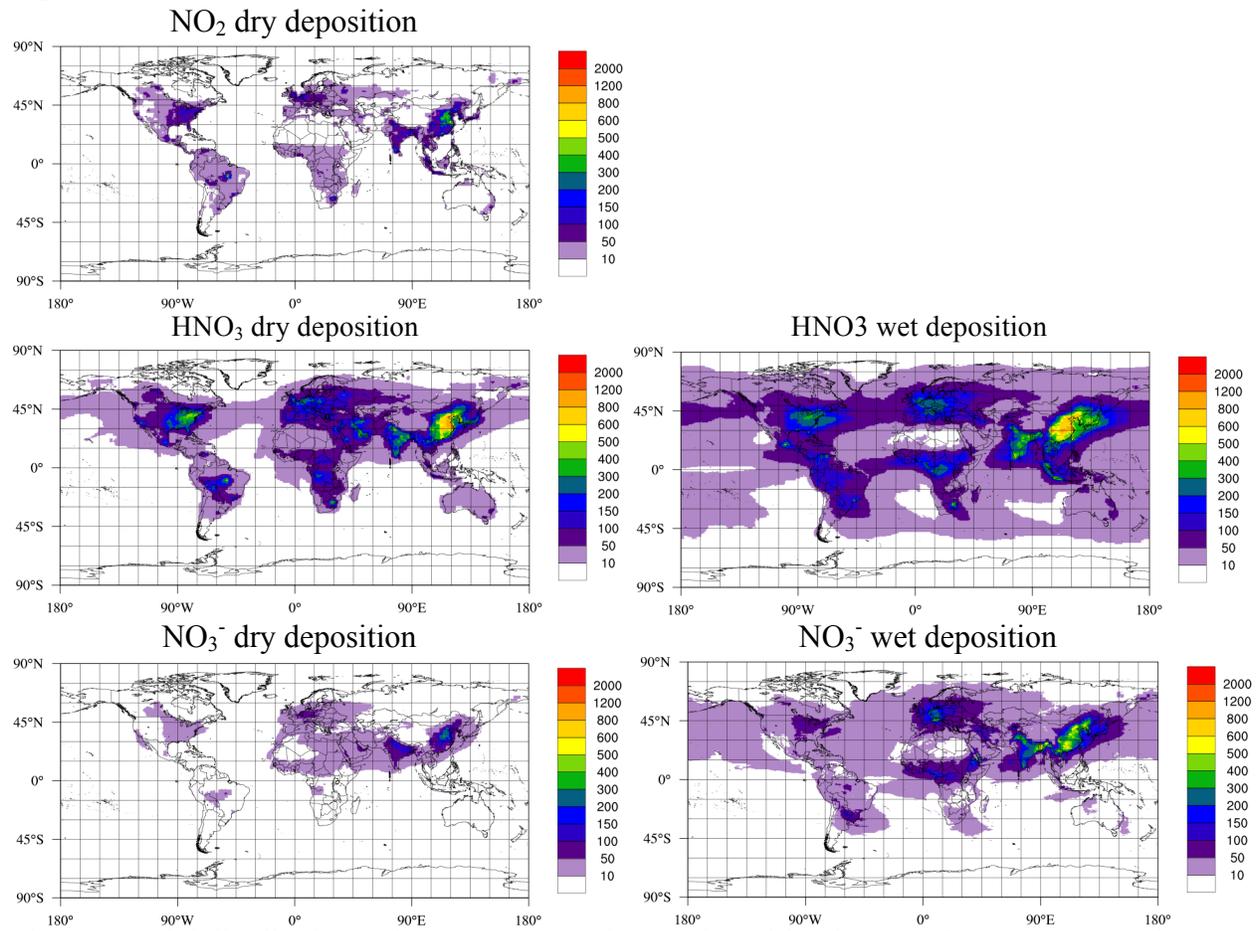


Fig. 14 Spatial distributions of NO₂, HNO₃ and NO₃⁻ deposition in 2010 from MMM results ($\text{mg(N) m}^{-2}\text{yr}^{-1}$).

Fig. S15

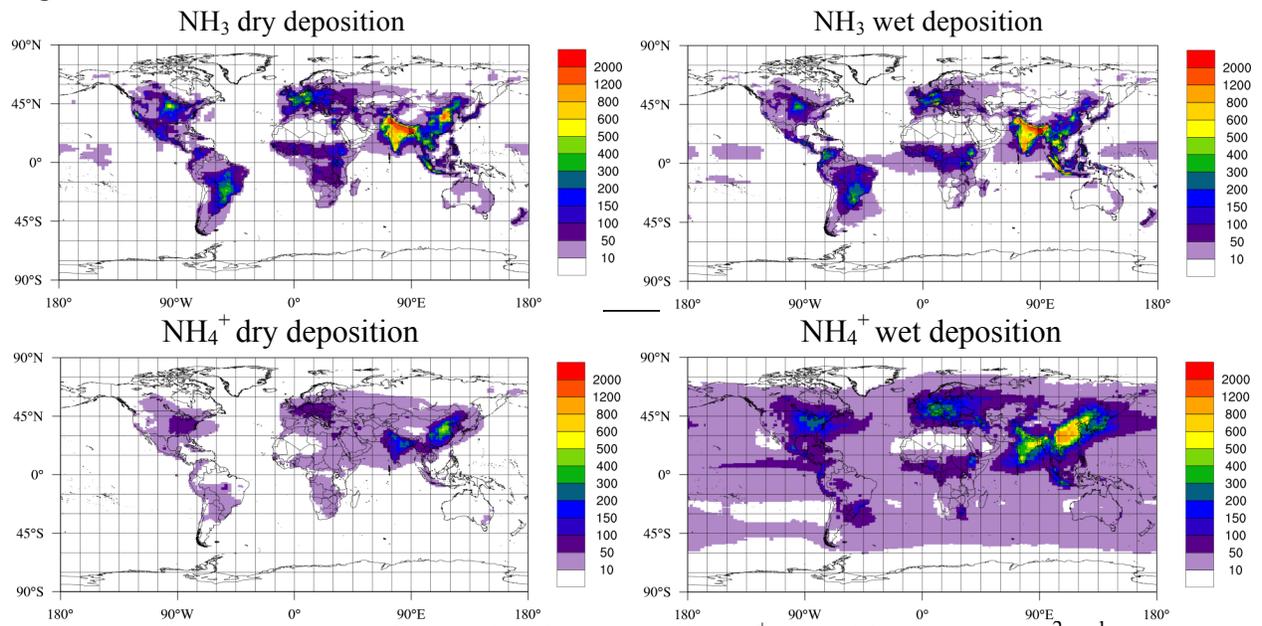
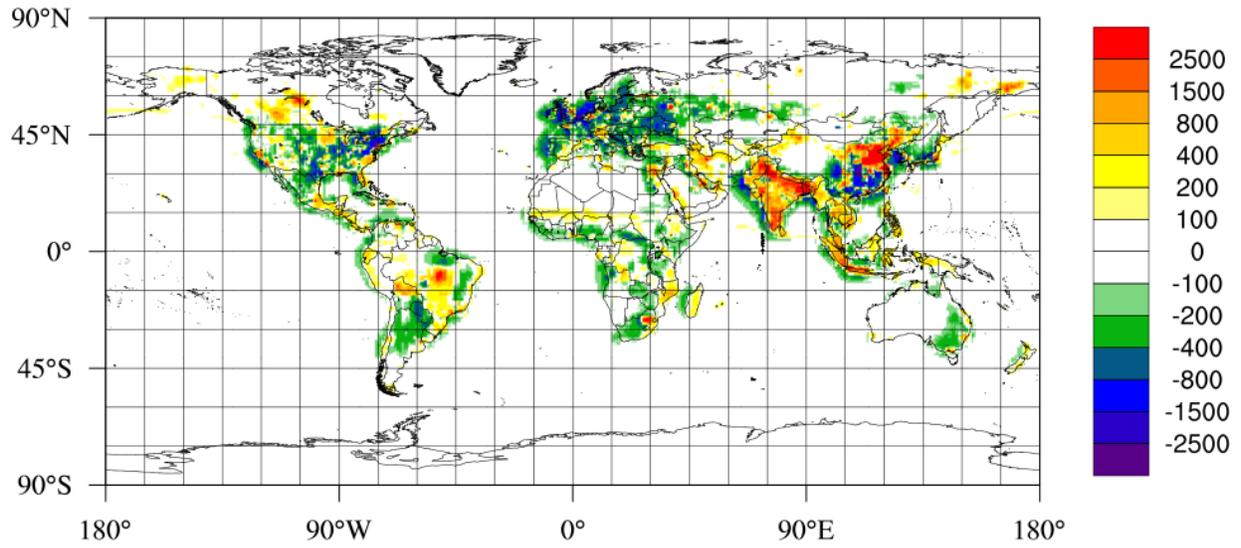


Fig. S15 Same as Fig. S14 but for NH_3 and NH_4^+ deposition ($\text{mg(N) m}^{-2}\text{yr}^{-1}$)

Fig. S16

(a) HTAP II N emission – HTAP I N emission



(b) HTAP II N deposition – HTAP I N deposition

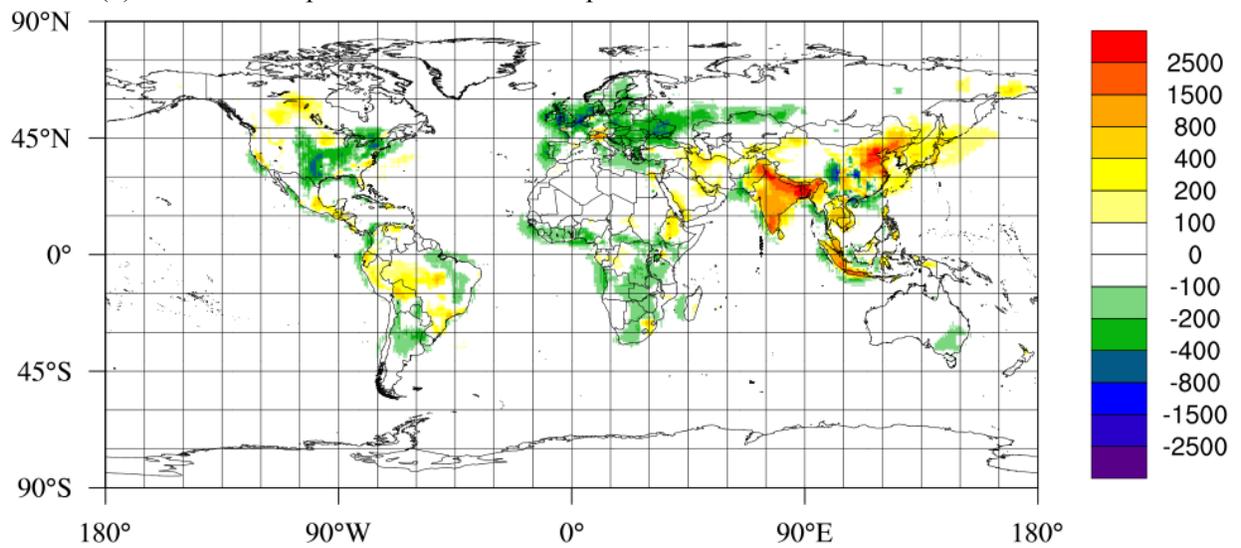


Fig. S16 Difference between MMM of HTAP II and HTAP I for (a) N emission (b) N Deposition. Unit: mg(N) m⁻² yr⁻¹

Fig. S17

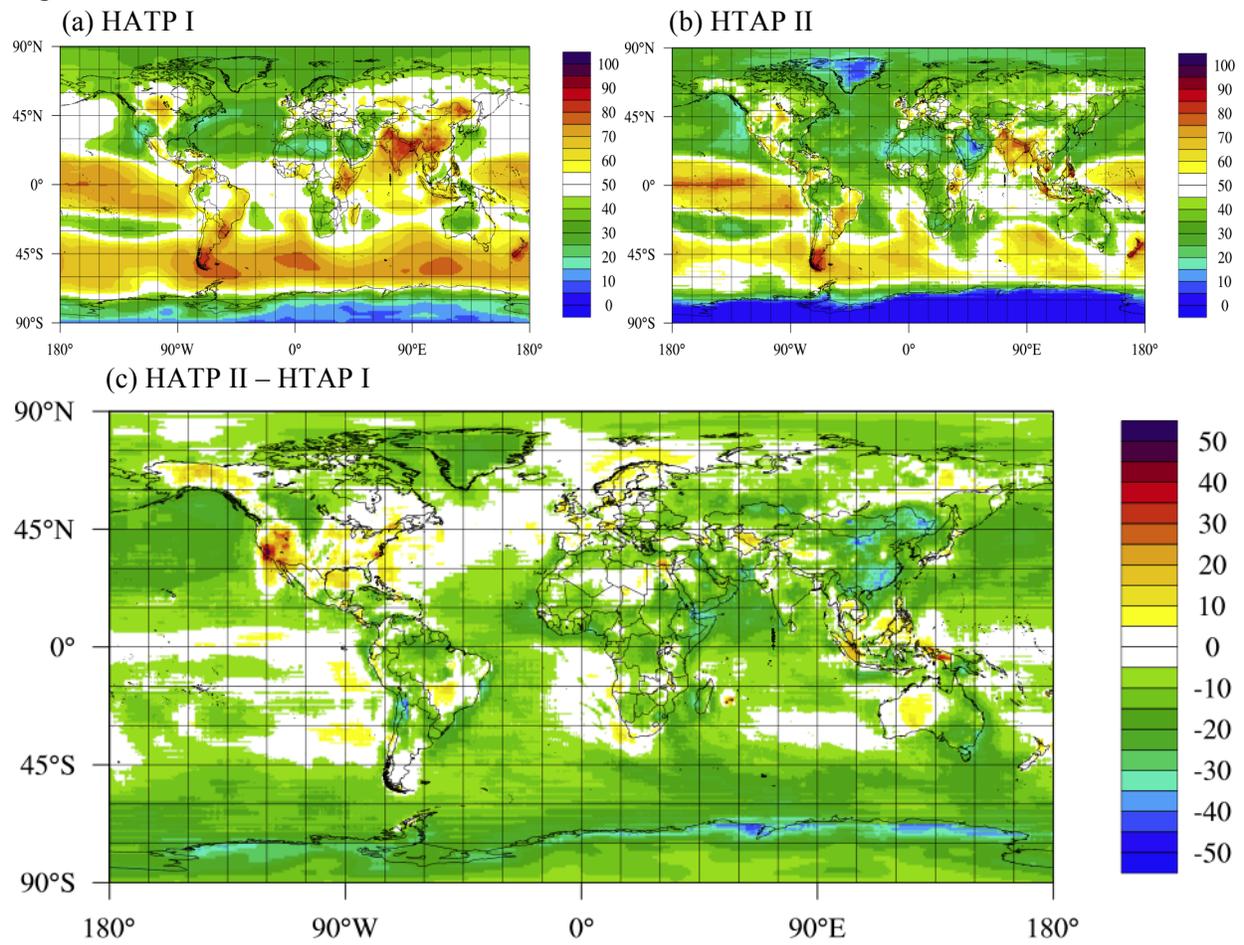


Fig. S17 Ratio of NH_x deposition in N ($\text{NO}_y + \text{NH}_x$) deposition from MMM results of (a) HTAP I (b) HTAP II. (c) HTAP II – HTAP I. Unit: %.

Table S1

Table S1. Summary of Global Total Deposition and Emission of S in 2010 (Tg(S) yr⁻¹)

Model/Species	Dry deposition	Wet deposition	Total Deposition	Emission surface SO ₂	Emission surface SO ₄ ²⁻	Emission DMS	Total Emission
CAM-chem	18	-	-	55	-	28	83
CHASER_re1	25	54	79	55	-	25	80
CHASER_t106	23	53	77	55	-	23	78
EMEP_rv48	16	42	58	-	-	-	-
GEMMACH	-	-	-	66	-	-	-
GEOS5	34	43	77	53	2	31	85
GEOSCHEMADJOINT	32	52	85	62	1	-	-
OsloCTM3.v2	40	63	103	77	2	-	-
GOCARTv5	29	47	76	66	2	-	-
SPRINTARS	26	-	-	60	1	22	84
C-IFS_v2	-	-	-	77	-	-	77
Multimodel mean	28	56	84	62	1	27	91

Note: S deposition is the sum of SO₂, SO₄²⁻, Methanesulfonic acid (MSA) and Dimethyl sulfide (DMS) deposition. S emission is the sum of SO₂, SO₄²⁻ (sulfate dry aerosol particles due to emission) and Dimethyl sulfide (DMS) emission.

Table S2

Table S2. Summary of Global Total Deposition and Emission of NO_y in 2010 (Tg(N) yr⁻¹)

Model/Species	Dry deposition	Wet deposition	Total Deposition	Emis NO _x	Emis INO _x	Total Emission
CAM-chem	16	-	-	-	4	-
CHASER_re1	23	28	51	57	4	60
CHASER_t106	25	27	52	58	5	63
EMEP_rv48	15	45	59	-	-	-
GEMMACH	-	-	-	44	-	44
GEOSCHEMADJOINT	26	28	54	54	-	54
OsloCTM3.v2	25	-	-	51	-	51
Multimodel mean	22	38	59	57	4	60

Note: NO_y deposition is the sum of all simulated oxidized nitrogen species (expressed as nitrogen) including NO, NO₂, HNO₃, HNO₄, NO₃aerosol, NO₃(radical), N₂O₅, Peroxyacyl nitrates (PAN), other organic nitrates other than PAN, but not N₂O (Orgn) deposition. NO_x emission is sum of anthropogenic NO_x emission, aircraft NO emission, soil NO emission and lightening NO_x emission (INO_x).

Table S3

Table S3. Summary of Global Total Deposition and Emission of NH_x in 2010 (Tg(N) yr⁻¹)

Model/Species	Dry deposition		Wet deposition		Total Deposition	Emission
	NH ₃	NH ₄ ⁺	NH ₃	NH ₄ ⁺		
CAM-chem	12	8	-	-	-	54
EMEP_rv48	11	3	13	-	-	-
GEOSCHEMADJOINT	14	4	13	24	55	55
OsloCTM3.v2	19	4	-	21	-	54
Multimodel mean	14	5	13	22	54	54

Note: NH_x deposition is the sum of NH₃ and NH₄⁺ deposition. NH_x emission is NH₃ emission.

Table S4

Table S4. Multi-model performance on simulating SO₂ concentration at CASTNET sites. The unit is µg (S) m⁻³.

Species	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMAD JOINT	GOCART	OsloCTM3 .v2	SPRINTARS	MMM
mean obs	0.82	0.82	0.82	0.82	0.82	0.82	0.82	0.82	0.82
mean model	5.31	5.79	5.51	1.71	5.36	2.49	4.61	1.72	4.06
Linear Fit Slope	6.77	6.65	7.90	2.22	6.33	2.90	5.43	2.03	5.03
mean bias	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Bias ¹	546.66	604.77	570.95	107.92	553.04	203.68	460.86	109.94	394.73
R	0.79	0.81	0.84	0.89	0.76	0.88	0.84	0.76	0.90
F	12.50	6.25	12.50	31.25	16.25	21.25	15.00	43.75	11.25
NMB	546.66	604.77	570.95	107.92	553.04	203.68	460.86	109.94	394.73
NME	548.91	606.20	573.30	116.37	554.09	208.21	462.33	117.67	396.47
MFB	104.46	119.52	101.46	21.00	110.67	65.28	105.73	25.09	99.26
MFE	116.11	125.36	110.97	69.95	113.81	88.82	111.67	61.69	106.31
no of station	80	80	80	80	80	80	80	80	80
Spatial resolution	1.9° × 2.5°	2.8° × 2.8°	1.1° × 1.1°	0.5° × 0.5°	2.0° × 2.5°	1.3° × 1.0°	2.8° × 2.8°	1.1° × 1.1°	

¹ Bias is calculated as dividing Mean Bias with Mean Observation. The unit is %.

Table S5

Table S5. Same as Table S4 but for SO_4^{2-} concentration. The unit is $\mu\text{g (S) m}^{-3}$.

Species	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMADJOINT	OsloCTM3.v2	MMM
mean obs	0.64	0.64	0.64	0.64	0.64	0.64	0.64
mean model	1.06	1.24	1.09	0.74	0.70	0.52	0.89
Linear Fit Slope	1.97	1.90	1.80	1.62	1.17	0.96	1.57
mean bias	0.00	0.00	0.00	0.00	0.00	0.00	0.00
bias%	66.99	94.61	71.61	16.30	10.81	-18.18	40.36
R	0.93	0.84	0.88	0.92	0.92	0.87	0.91
F	41.25	26.25	40.00	57.50	92.50	82.50	63.75
NMB	66.99	94.61	71.61	16.30	10.81	-18.18	40.36
NME	72.20	95.50	73.78	40.59	23.01	25.30	46.73
MFB	34.17	59.16	45.63	-18.17	6.00	-32.63	23.84
MFE	46.67	60.54	49.32	50.23	23.21	37.75	33.98
no of station	80	80	80	80	80	80	80
Spatial resolution	$1.9^\circ \times 2.5^\circ$	$2.8^\circ \times 2.8^\circ$	$1.1^\circ \times 1.1^\circ$	$0.5^\circ \times 0.5^\circ$	$2.0^\circ \times 2.5^\circ$	$2.8^\circ \times 2.8^\circ$	

Table S6

Table S6. Same as Table S4 but for HNO₃ concentration. The unit is µg (N) m⁻³.

Species	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMADJOINT	OsloCTM3.v2	MMM
mean obs	0.17	0.17	0.17	0.17	0.17	0.17	0.17
mean model	0.64	0.83	0.71	0.36	0.64	0.14	0.55
Linear Fit Slope	3.34	4.40	5.10	2.00	2.48	0.72	3.01
mean bias	0.00	0.00	0.00	0.00	0.00	0.00	0.00
bias%	264.73	376.04	309.11	106.39	264.54	-19.82	216.83
R	0.78	0.65	0.82	0.85	0.74	0.76	0.84
F	3.75	2.50	1.25	28.75	1.25	76.25	3.75
NMB	264.73	376.04	309.11	106.39	264.54	-19.82	216.83
NME	265.39	376.18	309.11	106.84	264.54	30.38	216.83
MFB	107.09	117.52	105.97	60.13	113.19	-27.44	98.66
MFE	107.50	117.59	105.97	61.55	113.19	42.18	98.66
no of station	80	80	80	80	80	80	80
Spatial resolution	1.9° × 2.5°	2.8° × 2.8°	1.1° × 1.1°	0.5° × 0.5°	2.0° × 2.5°	2.8° × 2.8°	

Table S7

Table S7. Same as Table S4 but for NO_3^- concentration. The unit is $\mu\text{g (N) m}^{-3}$.

Species	EMEP_rv48	GEOSCHEMADJOINT	MMM
mean obs	0.17	0.17	0.17
mean model	0.17	0.63	0.40
Linear Fit Slope	0.67	2.58	1.63
mean bias	0.00	0.00	0.00
bias%	0.05	270.05	135.05
R	0.80	0.74	0.77
F	65.00	12.50	17.50
NMB	0.05	270.05	135.05
NME	35.60	279.48	144.21
MFB	2.19	103.34	76.92
MFE	41.98	113.85	87.53
no of station	80.0	80	80
Spatial resolution	$0.5^\circ \times 0.5^\circ$	$2.0^\circ \times 2.5^\circ$	

Table S8

Table S8. Same as Table 4 but for NH_4^+ concentration. The unit is $\mu\text{g (N) m}^{-3}$.

Species	EMEP_rv48	GEOSCHEMADJOINT	MMM
mean obs	0.56	0.56	0.56
mean model	1.13	1.94	1.54
Linear Fit Slope	2.00	3.47	2.74
mean bias	0.00	0.00	0.00
bias%	101.13	244.46	172.79
R	0.91	0.94	0.95
F	26.25	1.25	5.00
NMB	101.13	244.46	172.79
NME	101.89	244.46	172.79
MFB	58.09	106.87	88.42
MFE	59.91	106.87	88.42
no of station	80	80	80
Spatial resolution	$0.5^\circ \times 0.5^\circ$	$2.0^\circ \times 2.5^\circ$	

Table S9

Table S9. Comparison of dry deposition velocity of SO_2 between models and CASTNET. The unit is cm s^{-1} .

	CAM-chem	CHASER_r e1	CHASER_t 106	EMEP_rv4 8	GEOSCHE MADJOIN T	GOCART	OsloCTM3. v2	SPRINTAR S	MMM
mean obs	0.27	0.27	0.27	0.27	0.27	0.27	0.27	0.27	0.27
mean model	0.14	0.17	0.16	0.36	0.33	0.41	0.39	0.50	0.24
Linear Fit Slope	0.16	0.02	-0.01	-0.35	0.20	0.03	-0.62	0.15	0.04
mean bias	-0.13	-0.09	-0.11	0.10	0.07	0.15	0.12	0.23	-0.02
bias%	-47.79	-34.88	-41.44	35.54	25.05	54.32	46.08	87.86	-8.39
R	0.18	0.03	-0.02	-0.23	0.09	0.03	-0.44	0.23	0.06
F	36.25	60.00	53.75	56.25	70.00	42.50	50.00	30.00	77.50
NMB	-47.79	-34.88	-41.44	35.54	25.05	54.32	46.08	87.86	-9.03
NME	56.70	44.51	49.62	62.93	55.54	60.74	68.83	88.07	34.88
MFB	-67.47	-39.46	-48.79	27.96	16.61	45.18	36.33	65.08	-3.83
MFE	75.10	52.75	60.26	50.44	42.03	49.40	53.58	65.21	35.00
no of station	80.00	80.00	80.00	80.00	80.00	80.00	80.00	80.00	80

Table S10

Table S10. Same as Table S9 but for dry deposition velocity of SO_4^{2-}

	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMADJOINT	OsloCTM3.v2	MMM
mean obs	0.13	0.13	0.13	0.13	0.13	0.13	0.13
mean model	0.15	0.09	0.10	0.17	0.13	0.21	0.13
Linear Fit Slope	0.29	0.01	-0.01	0.30	0.11	0.21	0.10
mean bias	0.02	-0.03	-0.03	0.04	0.01	0.08	0.00
Bias %	16.15	-26.92	-24.39	31.34	4.06	63.18	0.30
R	0.42	0.07	-0.16	0.17	0.28	0.27	0.34
F	83.75	82.50	85.00	67.50	90.00	36.25	88.75
NMB	16.15	-26.92	-24.39	31.34	4.06	63.18	0.30
NME	26.38	32.21	30.95	45.91	22.36	63.67	21.12
MFB	17.60	-26.05	-22.58	23.00	7.78	50.30	4.56
MFE	26.19	34.91	33.14	37.74	22.75	50.56	21.73
No of stations	80	80	80	80	80	80	80

Table S11

Table S11. Same as Table S9 but for dry deposition velocity of HNO₃

	CAM-chem	CHASER_re1	CHASER_t10 6	EMEP_rv48	GEOSCHEM ADJOINT	OsloCTM3.v2	MMM
mean obs	1.34	1.34	1.34	1.34	1.34	1.34	1.34
mean model	0.68	1.07	1.41	1.35	1.51	3.55	1.25
Linear Fit Slope	0.03	0.06	-0.37	-0.03	-0.40	-1.24	-0.10
mean bias	-0.66	-0.28	0.06	0.01	0.17	2.21	-0.10
Bias %	-49.12	-20.70	4.60	0.74	12.44	164.52	-7.27
R	0.10	0.05	-0.28	-0.03	-0.29	-0.38	-0.12
F	50.00	72.50	77.50	85.00	76.25	16.25	85.00
NMB	-49.12	-20.70	4.60	0.74	12.44	164.52	-7.27
NME	49.92	34.65	36.69	29.96	38.75	165.99	27.47
MFB	-62.09	-27.68	2.14	-0.16	9.65	84.58	-7.01
MFE	63.78	42.88	37.23	30.98	36.88	85.84	29.52
No of stations	80	80	80	80	80	80	80

Table S12

Table S12. Same as Table S9 but for dry deposition velocity of NO₃⁻

	EMEP_rv48	GEOSCHEMADJOINT	MMM
mean obs	0.12	0.12	0.12
mean model	0.29	0.10	0.14
Linear Fit Slope	0.65	0.00	0.06
mean bias	0.17	-0.02	0.02
Bias %	146.99	-16.50	18.69
R	0.26	0.00	0.05
F	7.50	77.50	73.75
NMB	146.99	-16.50	18.69
NME	147.73	40.11	37.35
MFB	81.93	-17.07	17.29
MFE	82.58	38.99	32.05
No of stations	80	80	80

Table S13

Table S13. Same as Table S9 but for dry deposition velocity of NH₄⁺

	CAM-chem	GEOSCHEMADJOINT	MMM
mean obs	0.12	0.12	0.12
mean model	0.22	0.06	0.12
Linear Fit Slope	0.38	0.06	0.13
mean bias	0.10	-0.06	0.00
Bias	81.91	-47.39	-1.72
R	0.40	0.23	0.27
F	15.00	60.00	87.50
NMB	81.91	-47.39	-1.72
NME	82.81	48.76	22.13
MFB	60.11	-57.09	2.01
MFE	60.62	59.99	22.67
No of stations	80	80	80