Anonymous Referee #1

General Comments

This paper describes the global S and N emissions and deposition in a set of global models run for 2010 under a model intercomparison project (HTAP II) and compares the results with regional monitoring data and previous global modelling. The subject matter is of interest to air quality and ecosystem scientists, particularly those concerned with deposition to oceans, where there is little information from measurements or regional models. I recommend publication with minor revisions, with the most important being an expansion of the dry deposition discussion.

Response: We would like to thank the reviewer for the suggestions to improve this manuscript. Following are the responses to comments.

Specific comments:

Comment: Section 2.1: It isn't crucial, but it would be helpful to include a brief mention of any other papers (published or forthcoming) that describe additional results from this intercomparison project (e.g. ambient concentrations of particulate matter or O3). With virtual special issues, it's not always obvious where to find related papers.

Response: We added the following paragraph in the manuscript to introduce the related publication from this project.

Line 137-145: Following are some highlight findings in HTAP II. (Stjern et al., 2016) estimated the impact of domestic and foreign emission change of BC, OC and SO_4 on regional radiative forcing. (Huang et al., 2017) studied the impact of intercontinental outflow from East Asia to North America on O_3 pollution by simulating the regional-scale Sulfur Transport and dEposition Model (STEM) with boundary conditions provided by 3 global transport models. (Jonson et al., 2018) conducted a source apportionment for O_3 pollution in Europe and calculated the contributions of emission from global wide. (Tan et al., 2018) investigated the intercontinental export of sulfur and nitrogen emission and its impact on local deposition.

Comment: Section 2.2: Can you explain why some values in Tables S1-S3 seem inconsistent? E.g., total S emission from the MMM is 91 Tg in Table S1, but from equation (2) it seems that it should be 55+1+27=83 Tg. Is this an error or am I missing something? Also, the same table seems to show that OsloCTM3 should be excluded for S based on the mass balance criteria described. It's unclear why it is kept. I'm assuming model values that did not meet the criteria are not listed in the tables, though I don't think that was explicitly stated.

Response: Thank you for pointing out this problem. The Multi-model mean of "Emission surface SO_2" should be 62 instead of 55. Therefore the "total S emission" is 62+1+27=91 Tg. We have changed it in the manuscript.

The tables only list the model values that meet both criteria described in Section 2.2, except the condition that if a model hasn't submitted some important components, which make it impossible to check the criteria. For instance, the 1st criteria compares the global emission with deposition for each model. This criterial is used to check the models that submits the major components of both emission and deposition. The OsloCTM3 model submits the major components of dry and wet deposition. The total value is 40+63=103 Tg. But it hasn't submitted the emission of DMS, and we can't calculate its total S emission. Therefore we can't compare its S deposition with emission. According to the 2nd criteria, we check if the model value is within the range of (median of models ± 1.5 × interquartile) for each component. The components of OsloCTM3 model all pass this quality check. Since the model passes the 2nd criteria and it is unable to check the 1st criteria, we still keep this model.

Comment: l. 265: the 81% value in the text does not match Table 1, which says 61%. Which is correct?

Response: 81% is the average percentage of North America, Europe and Asia. Table 1 gives the separate values for these 3 regions, which are 88%, 75% and 61%, respectively. North America has the 136 stations used for evaluation, more than Europe (82) and East Asia (43), thus the 3 area averaged value is closer to its value.

We have added word in red in the following sentences in the manuscript for clarity:
Line243: Overall, 76% of the stations of all networks predicted quantities within ±50% of observations.
Line 269: Overall, 83% of the MMM results are within ±50% of observations at stations of all networks.
Line 286: Overall, 81% of the MMM predictions are within ±50% of observations at stations of all networks.

Comment: ll. 267-291 and Table 1: If possible, I suggest adding the number of stations for each comparison to Table 1 since that is likely different as well; where N is relatively low, the number and location of stations used could have a significant impact on the statistics.

Response: We have added the number of stations in Table 1. Because we check the quality and completeness of observation data, the numbers of stations used for evaluation are less than those that are available. We use 136 out of 267 available stations in North America, 82 out of 102 available stations in Europe and 43 out of 52 available stations in Asia.

Table 1. Intercomparison of HTAP II MMM performance with previous projects on wet deposition. The unit is mg (N or S) m^{-2} yr^{-1}.

Wet SO_4^{2-} Deposition	North America				Europe				Asia			
	PhotoComp	HTAP I	ACCMIP	HTAP II	PhotoComp	HTAP I	ACCMIP	HTAP II	PhotoComp	HTAP I	ACCMIP	HTAP II
Linear Fit Slope	0.9	1	0.6	0.9	0.4	0.6	0.3	0.4	0.4	0.5	0.3	0.5
Mean Bias	46.3	50	-18.8	30.9	-67.1	51.5	-125.3	-31.3	-218.6	-182.1	-292.4	-161.5
Mean Observation	309.8	309.8	309.8	253.7	404.5	404.5	404.5	228.7	686.1	686.1	686.1	653.7
Mean Model	356.1	359.8	291	284.6	337.3	456.1	279.3	197.4	467.5	504.1	393.7	492.2
R	0.9	0.9	0.9	0.8	0.6	0.6	0.6	0.7	0.9	0.9	0.8	0.6
Fraction within ±50%	70.4	70	72.2	76.5	78.7	52.8	78.7	86.4	80	88	72	68.6
Number of stations	346	346	346	136	126	126	126	82	49	49	49	43

Wet NO_3^- Deposition	North America				Europe				Asia			
	PhotoComp	HTAP I	ACCMIP	HTAP II	PhotoComp	HTAP I	ACCMIP	HTAP II	PhotoComp	HTAP I	ACCMIP	HTAP II
Linear Fit Slope	1	1	0.9	1.2	0.3	0.3	0.3	0.5	0.5	0.5	0.4	0.8
Mean Bias	34.8	21.9	44.3	57.8	-41.4	-60	-75.2	-22.0	-47.8	-49.3	-46.4	-0.8
Mean Observation	191.3	191.3	191.3	153.7	300.5	300.5	300.5	237.3	263	263	263	356.4
Mean Model	226.1	213.3	235.6	211.5	259.1	240.5	225.3	215.4	215.2	213.7	216.7	355.7
R	0.8	0.9	0.9	0.9	0.6	0.6	0.6	0.7	0.8	0.8	0.8	0.7
Fraction within ±50%	77	84.3	68.7	66.9	75	85.2	85.2	90.2	84	84	88	76.7
Number of stations	346	346	346	136	126	126	126	82	49	49	49	43

Wet NH_4^+ Deposition	North America				Europe				Asia			
	PhotoComp	HTAP I	ACCMIP	HTAP II	PhotoComp	HTAP I	ACCMIP	HTAP II	PhotoComp	HTAP I	ACCMIP	HTAP II
Linear Fit Slope	0.8	0.9	0.5	0.8	0.4	0.4	0.3	0.6	0.8	0.7	0.1	0.6
Mean Bias	5.5	10.9	-12.1	2.3	-23.9	-49.7	-94.7	-4.0	-69.7	-63.4	-136.2	-28.7
Mean Observation	161.3	161.3	161.3	195.5	336	336	336	286.1	400.5	400.5	400.5	534.5
Mean Model	166.8	172.2	149.2	197.9	312.1	286.4	241.3	282.2	330.8	337.1	264.4	505.8
R	0.9	0.9	0.8	0.9	0.8	0.6	0.6	0.6	0.8	0.8	0.2	0.7
Fraction within ±50%	82.2	84.8	75.7	87.5	73.9	79.5	78.4	75.3	76	68	56	60.5
Number of stations	346	346	346	136	126	126	126	82	49	49	49	43

Comment: Section 3.1.2: Additional discussion of dry deposition is warranted, given the large differences with the CASTNET inferential values. The difference between the CASTNET dry deposition calculations and those using the CAPMoN method are touched on (ll. 300-303) but the implications for the models is not fleshed out. I recommend moving this discussion to the end of the section 3.1.2 and discussing the relevance to the ensemble-measurement comparison. How does the CASTNET dry deposition velocity parametrization compare with those used in the various models? How do the modelled air concentrations of SO2, HNO3, etc. compare with the CASTNET observations?

Response: Thank you for this useful suggestion. We have moved the discussion of uncertainty of CASNET to the end of section 3.1.2 as an explanation of the model bias. We also compare the air concentrations and dry deposition velocities between the models and CASTNET dataset in the manuscript as follows:

Line 349-381: Since the CASTNET dry deposition is not actually measured data but instead a combination of measured concentration of species and modelled dry deposition velocities, it is necessary to investigate which factor of these two contributes to the model bias. We compare the modelled air pollutant concentrations with CASENET measurements as shown in Table S4-S8. The MMM overestimates the SO_2, SO_4^{2-}, HNO_3, NO_3^- and NH_4^+ concentrations by 394%, 40%, 217%, 135% and 173%, respectively. It should be noted that the CASTNET sites are generally located in rural regions that are away from emission sources (Sickles and Shadwick, 2008), thus the measured concentrations of air pollutants are relatively low compared with those of urban sites. While the resolutions of the HTAP II models range from 0.5° to 3°, and are not fine enough to reproduce the characteristic of some rural sites. The models with finer resolutions except CHASER_t106 model (i.e. EMEP_rv48 (0.5 × 0.5) and SPRINTARS (1.1 × 1.1)) generally perform better than the others, while models with coarse resolutions (i.e. CHASER_re1 (2.8 × 2.8) and OsloCTM3.v2) are generally not performing well for all species. This could explain the overestimation of air pollutant concentrations at the CASTNET sites.

In order to check the differences of modelled dry deposition velocity between CASNET and HTAP II models, we adopt the general approach for calculating dry deposition velocity from (Wesely, 1989).

$$V_d = - F_c / C_a \qquad (7)$$

Where V_d is the deposition velocity, F_c is the dry deposition flux and C_a is the concentration of species. The negative mark indicates the direction of the dry deposition velocity. This scheme has been widely adopted in global models (Wesely and Hicks, 2000) with modifications. We compare the calculated dry deposition velocity of models and CASTNET (Table S9-S13). The mean bias of dry deposition velocities for MMM are -8%, 0.3%, 7%, 19% and 2% for SO_2, SO_4^{2-}, HNO_3, NO_3^- and NH_4^+, respectively, which are much lower than those of air pollutants. The model bias for dry deposition at the CASTNET sites mainly comes from the model over prediction of air pollutant concentration.

Table S4. Multi-model performance on simulating SO_2 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 Observation	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
Number of stations	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 by dividing Mean Bias with Mean Observation. The unit is %.

Table S5. Same as Table S4 but for SO_4^{2-} concentration. The unit is μg (S) m^{-3}.

Species	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMADJOINT	OsloCTM3.v2	MMM
Mean Observation	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%[1]	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
Number of stations	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 S6. Same as Table S4 but for HNO_3 concentration. The unit is μg (N) m^{-3}.

Species	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMADJOINT	OsloCTM3.v2	MMM
Mean Observation	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%[1]	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
Number of stations	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. Same as Table S4 but for NO_3^- concentration. The unit is μg (N) m^{-3}.

Species	EMEP_rv48	GEOSCHEMADJOINT	MMM
Mean Observation	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%[1]	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
Number of stations	80.0	80	80
Spatial resolution	0.5° × 0.5°	2.0° × 2.5°	

Table S8. Same as Table 4 but for NH_4^+ concentration. The unit is μg (N) m^{-3}.

Species	EMEP_rv48	GEOSCHEMADJOINT	MMM
Mean Observation	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%[1]	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
Number of stations	80	80	80
Spatial resolution	0.5° × 0.5°	2.0° × 2.5°	

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

	CAM-chem	CHASER_re1	CHASER_t106	EMEP_rv48	GEOSCHEMADJOINT	GOCART	OsloCTM3.v2	SPRINTARS	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
Number of station	80.00	80.00	80.00	80.00	80.00	80.00	80.00	80.00	80

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
Number of stations	80	80	80	80	80	80	80

Table S11. Same as Table S9 but for dry deposition velocity of HNO_3

	CAM-chem	CHASER_re1	CHASER_t106	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
Number of stations	80	80	80	80	80	80	80

Table S12. Same as Table S9 but for dry deposition velocity of NO_3^-

	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
Number of stations	80	80	80

Table S13. Same as Table S9 but for dry deposition velocity of NH_4^+

	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
Number of stations	80	80	80

Comment: l. 347: Australia appears to receive higher coastal S deposition than E. Asia, so should be listed here as well.

Response: Coastal Australia emitted 2.0 Tg(S) yr^{-1} of S emission, higher than that of coastal East Asia (1.8 Tg(S) yr^{-1}). But coastal Australia received 1.5 Tg(S) yr^{-1} of S deposition, lower than that of coastal East Asia (2.9 Tg(S) yr^{-1}). This is because the high S emission emitted in non-coastal East Asia (15.0 Tg(S) yr^{-1}) brings deposition to its coastal region via long-range transport. While lower S emission in non-coastal Australia (1.5 Tg(S) yr^{-1}) has less impact on its coastal region.

Comment: l. 374: Why the 32% increase in ocean S emissions? Is that real or the result of improved emission budgets?

Response: The total S emission in HTAP I is 91 Tg(S) in 2001, of which 66.4 Tg(S) is SO_2 emission, 6.3 Tg(S) is SO_4^{2-} emission and 18.2 Tg(S) is DMS emission. The total S emission in HTAP II is 91 Tg(S) in 2010, of which 63 Tg(S) is SO_2 emission, 1 Tg(S) is SO_4^{2-} emission and 27 Tg(S) is DMS emission.

The amount of total S emission and SO_2 emission are similar between HTAP I and HTAP II. While the SO_4^{2-} emission is decreased by 5 Tg(S) and DMS emission is increased by 9 Tg(S). Since the DMS emission is generally from coastal and ocean sources, the large difference of oceanic S emissions comes from the DMS emission.

We compare the emission of DMS with literatures. The range of DMS is estimated to be 23-35 Tg(S) by (Simo and Dachs, 2002) from remote sensing of biogeophysical data and to be about 28 Tg(S) estimated by (Kloster et al., 2006). The 27 Tg(s) of HTAP II is closer to the abovementioned range, and the 18 Tg(s) of HTAP I could be slightly underestimated.

Another possible reason is the calculation of multi-model mean of DMS emission. The following table listed the S emission by different models. Although all models except EMEP_rv48 and GEMMACH are confirmed to include DMS emission in simulations, but only 5 out of 10 models have submitted the DMS emission. The relative low number of submission could cause uncertainty in calculating the multi-model ensemble of DMS emission.

Table 1. Summary of Global Emission of S in 2010 (Tg(S) yr^{-1}

Model/Species	DMS
CAMChem	28
CHASER_re1	25
CHASER_t106	23
EMEP_rv48	Not used
GEMMACH	Not given
GEOS5	31
GEOSCHEMADJOINT	Include
OsloCTM3.v2	Include
GOCARTv5	Include
SPRINTARS	22
C-IFS_v2	Include
Multimodel mean*	27

Comment: ll. 520-536: There is discussion of the areas of increasing NHx ratio, but globally there appears to be a general decrease (e.g. over the oceans). Maybe add a comment on this.

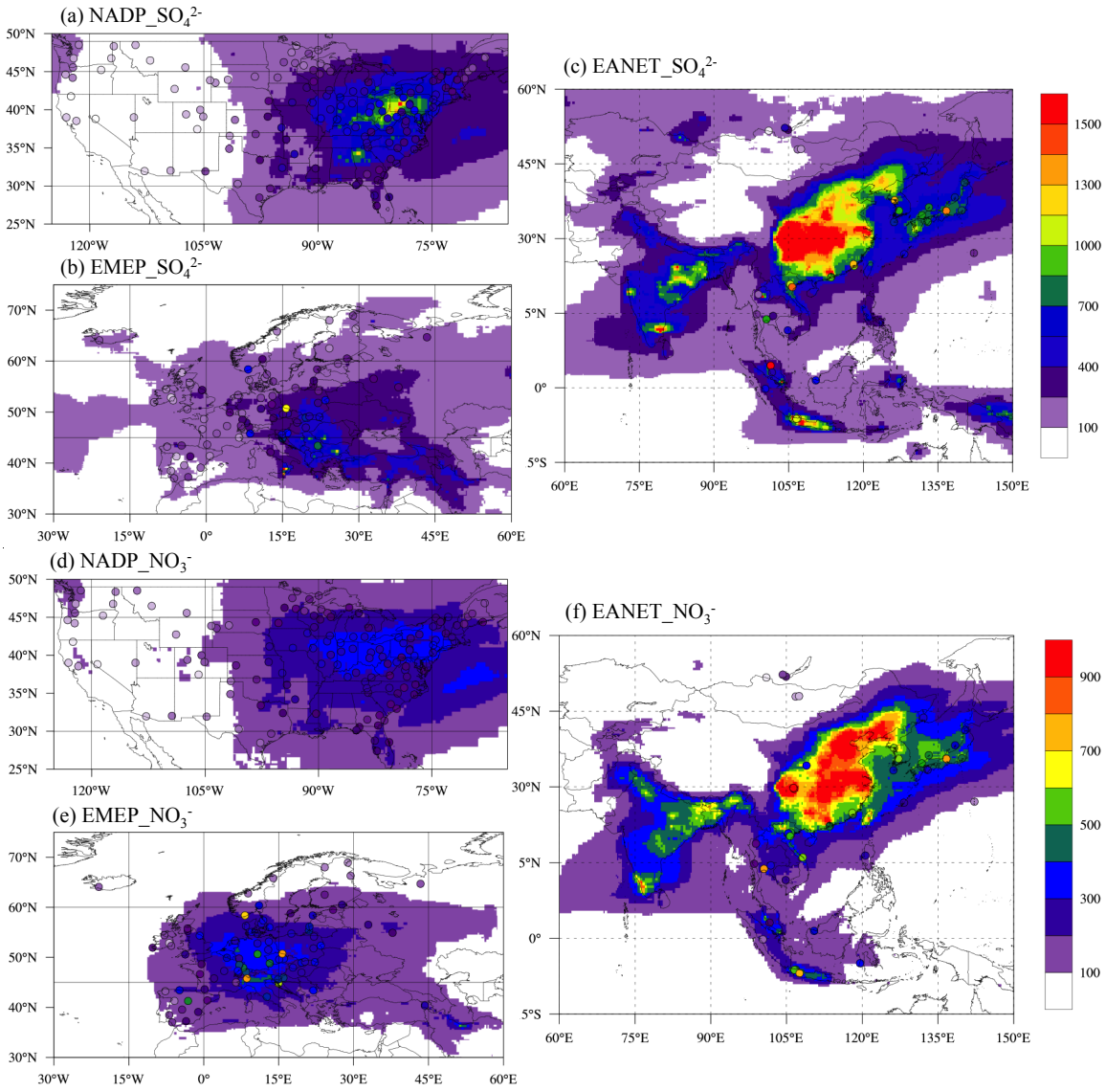
Response: Thank you for your suggestion. We add the following sentence in the manuscript.

Line 570-572: Generally, we found a 10% decrease in the ratio of NHx deposition from 2001 to 2010. In particular, a 30% decrease in the ratio of NHx is found in southeastern China, mainly due to the large increase in NOx emission during the last decade.

Comment: Fig. 2: Observation (point) values are very difficult to see on these small plots. Can they be enlarged, since the discussion in 3.1.1 hinges on the regional comparison? Fig. 4 is better; I would suggest that size is the minimum needed.

Response: we have enlarged the circles in the figure. Following are the new figures.

Fig. 2



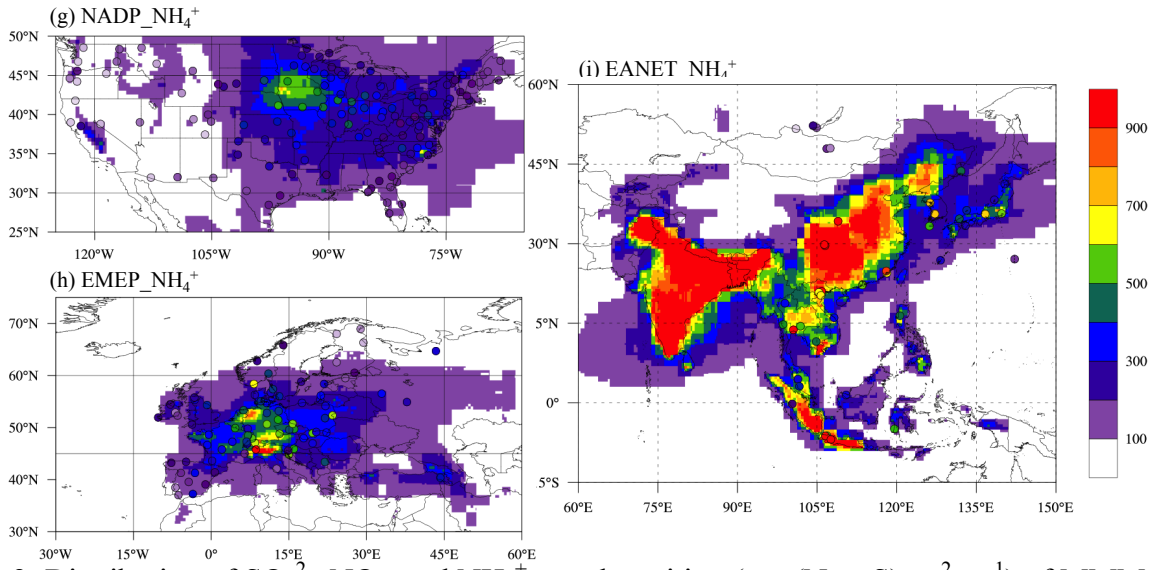


Fig. 2. Distribution of SO_4^{2-}, NO_3^- and NH_4^+ wet deposition (mg (N or S) m^{-2} yr^{-1}) of MMM and observation. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011.Contours are MMM results and filled circles are observation.

Technical comments: The manuscript would generally benefit from careful copyediting to correct minor issues with non-standard English usage. I've only highlighted errors where the meaning was somewhat unclear:

Response: We want to thank review#1 for the careful review of the manuscript. Following are the point-to-point replies to the comments. We have made all the corrections in the manuscript.

l. 53: change "shows that. . . increases" to "predicts that. . . will increase"

Response: we have changed the sentence in the manuscript.

ll. 67-90: previous results should all be in past tense

Response: We have checked this paragraph and changed present tense to past tense. But we use present tense for sentence after "that" in that-clauses.

For instance, Model evaluation showed that 60-70% of modelled wet deposition is within ±50% of measurements in Europe and North America.

l. 123: The HTAP project? Task Force?

Response: Thank you for pointing out this mistake. The full name is Task Force Hemispheric Transport of Air Pollution. It is firstly mentioned it in Line 76: Sanderson et al. (2008) used the ensemble results of the 1st phase of the Task Force Hemispheric Transport of Air Pollution (HTAP I) to estimate the long-range transport of oxidized nitrogen.

In the after content, we use HTAP to refer to Task Force Hemispheric Transport of Air Pollution.

l. 130 and 146: update the Galmarini reference to the final ACP paper (2017)

Response: we have updated the citation.

l. 228: keeping with your sign convention in Table 1, the bias increases (or changes) from -160 to -300

Response: we have rephrased the sentence.

"the mean bias for East Asia will change from -160 mg (S) m^{-2} yr^{-1} to -300 mg (S) m^{-2} yr^{-1}".

l. 230 change "highest deposition" to "highest modelled deposition" if that is what is meant

Response: we have changed it in the manuscript.

ll. 240-243: Reword; the stations do not underestimate/under-predict the deposition, the MMM underestimates deposition at those stations.

Response: We have rephrased the sentence in the manuscript.

"According to Fig. 2(e), wet deposition at 3 stations in Poland, Norway and Spain were underestimated by 430 (59%), 420 (63%) and 290 (67%) mg N m^{-2} yr^{-1}, respectively."

l. 300: Reword to "Schwede et al. (2011) compared CASTNET dry deposition estimates with those of the Canadian. . ."

Response: we have changed it in the manuscript.

l. 312: suggest changing "0.5-1 times" to "50-100%" for consistency

Response: we have changed it in the manuscript.

ll. 321-22: change end of sentence to ". . .but this gradient is much weaker in the inferential data."

Response: we have changed it in the manuscript.

Suggest changing title of 3.2 to "Total S deposition" and similar for 3.3

Response: we have changed it in the manuscript.

l. 351-352: change to ". . .S deposition to the ocean and coastal areas in 2010." Remove text in parentheses.

Response: we have changed it in the manuscript.

l. 449: remove "and Mexico" since it's part of N. America

Response: In HTAP II, Mexico is separated from North America as shown in Fig. S1.

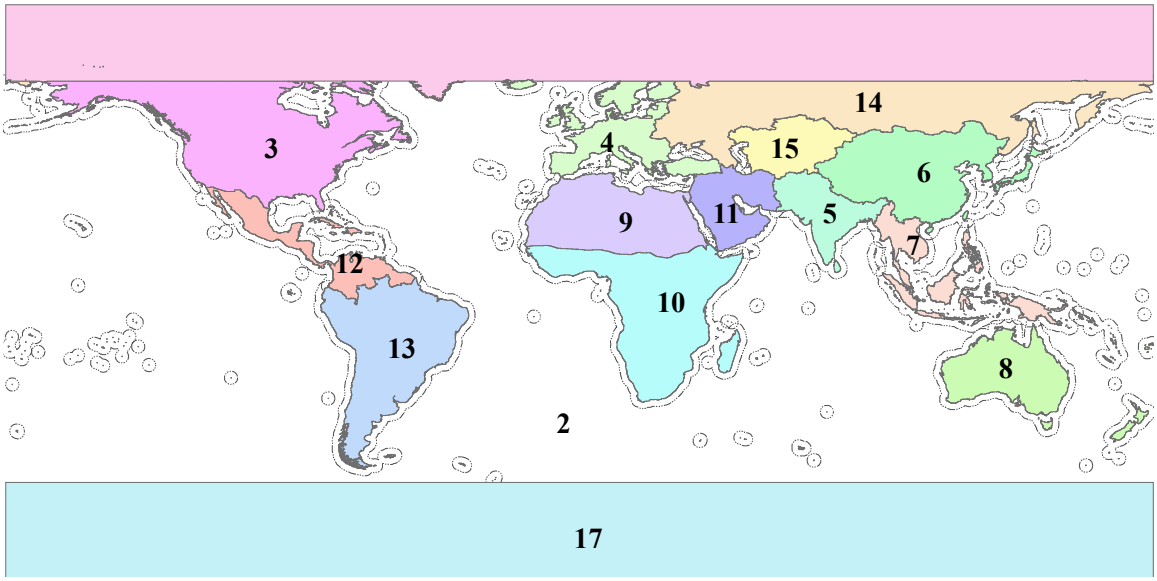


Fig. S1. Regions defined in HTAP phase II and coastal area. Region 1-Global, 2-Ocean (include Arctic), 3-North America, 4-Europe, 5-South Asia, 6-East Asia, 7-Southeast Asia, 8-Australia, 9-North Africa, 10- Sub Saharan Africa, 11-Middle East, 12- Mexico, Central America, Caribbean, Guyanas, Venezuela,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

l. 485: replace "positive changes" with "increases" to avoid the message that this is a desirable change

Response: we have changed it in the manuscript.

l. 571: "large net changes" could replace "large changes" for clarity

Response: we have changed it in the manuscript.

Tables 3-5: Merge the coastal numbers into a single cell. Add text to the caption to remind the reader that the values in parentheses are percentages (Tables 3 and 4).

Response: we have changed it in the manuscript.

Reference:

Huang, M., Carmichael, G. R., Pierce, R. B., Jo, D. S., Park, R. J., Flemming, J., Emmons, L. K., Bowman, K. W., Henze, D. K., Davila, Y., Sudo, K., Jonson, J. E., Tronstad Lund, M., Janssens-Maenhout, G., Dentener, F. J., Keating, T. J., Oetjen, H., and Payne, V. H.: Impact of intercontinental pollution transport on North American ozone air pollution: an HTAP phase 2 multi-model study, Atmos. Chem. Phys., 17, 5721-5750, 10.5194/acp-17-5721-2017, 2017.

Jonson, J. E., Schulz, M., Emmons, L., Flemming, J., Henze, D., Sudo, K., Tronstad Lund, M., Lin, M., Benedictow, A., Koffi, B., Dentener, F., Keating, T., and Kivi, R.: The effects of intercontinental emission sources on European air pollution levels, Atmos. Chem. Phys. Discuss., 2018, 1-26, 10.5194/acp-2018-79, 2018.

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Sanderson, M. G., Dentener, F. J., Fiore, A. M., Cuvelier, C., Keating, T. J., Zuber, A., Atherton, C. S., Bergmann, D. J., Diehl, T., Doherty, R. M., Duncan, B. N., Hess, P., Horowitz, L. W., Jacob, D. J., Jonson, J. E., Kaminski, J. W., Lupu, A., MacKenzie, I. A., Mancini, E., Marmer, E., Park, R., Pitari, G., Prather, M. J., Pringle, K. J., Schroeder, S., Schultz, M. G., Shindell, D. T., Szopa, S., Wild, O., and Wind, P.: A multi-model study of the hemispheric transport and deposition of oxidised nitrogen, Geophys Res Lett, 35, 10.1029/2008gl035389, 2008.

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Stjern, C. W., Samset, B. H., Myhre, G., Bian, H., Chin, M., Davila, Y., Dentener, F., Emmons, L., Flemming, J., Haslerud, A. S., Henze, D., Jonson, J. E., Kucsera, T., Lund, M. T., Schulz, M., Sudo, K., Takemura, T., and Tilmes, S.: Global and regional radiative forcing from 20 % reductions in BC, OC and SO4 – an HTAP2 multi-model study, Atmos. Chem. Phys., 16, 13579-13599, 10.5194/acp-16-13579-2016, 2016.

Tan, J., Fu, J. S., Dentener, F., Sun, J., Emmons, L., Tilmes, S., Flemming, J., Takemura, T., Bian, H., Zhu, Q., Yang, C. E., and Keating, T.: Source contributions of sulfur and nitrogen deposition - an HTAP II multi model study on hemispheric transport, Atmos. Chem. Phys. Discuss., 2018, 1-31, 10.5194/acp-2018-109, 2018.

Wesely, M. L.: Parameterization of Surface Resistances to Gaseous Dry Deposition in Regional-Scale Numerical-Models, Atmospheric Environment, 23, 1293-1304, 1989.

Wesely, M. L., and Hicks, B. B.: A review of the current status of knowledge on dry deposition, Atmospheric Environment, 34, 2261-2282, 2000.

Anonymous Referee #2

General Comments

This study gives a comprehensive overview of the global atmospheric deposition of sulfur and nitrogen using a range of global atmospheric transport model, compared to observations for 2010. The manuscript is well written. I have a few questions and remarks:

Response: We would like to thank the reviewer for the suggestions to improve the manuscripts. Following are the responses to comments.

Comment: Line 157. What does it mean that models are excluded if they fall outside their emission values? Several of the models given in table S1 and S2 don't calculate wet/total deposition? Are these models not used or have you deleted part of the calculations (i.e only used aerosol and not wet deposition)? Maybe indicate which models used for each ensemble mean. If that is S1 and S2, maybe indicate what has been deleted? Are you looking at the surface emissions or total emissions, several models do not include emissions of DMS? A follow up question on this topic, why don't the total emissions and deposition match up (i.e.7 Tg S and 1 Tg N differences in table 3 and 4)? Where does the left offers go, Have the models included organic N and S species?

Response: We compare the global total amount of emission with deposition for each model. If the global deposition is outside the ranges of ±20% of global emission, we don't show its value in table S1 and S2 and don't include this model in calculating the multi-model mean. This criteria is used to check the models that submits the major components of both emission and deposition. For models that submit part of the major components, we only check the 2nd criteria. The 2nd criteria checks if the model value is within the range of (median of models ± 1.5 × interquartile). If one model passes criteria 2, we still use it in calculating the multi-model mean of this components.

Following are 3 tables that give details about model values that used/not used to calculate ensemble mean. Dash ("-") symbols mean the models haven't submitted these components. Red-color values mean the models have submitted these components, but failed to pass the quality check. These values are not used for calculating multi-model mean. Since this study doesn't include inter-model comparison, we only give what models we have used in table S1 and S2.

Table 1. Summary of Global Total Deposition and Emission of S in 2010 (Tg(S) yr^{-1}

Model/Species	Dry deposition					Wet deposition					Total deposition	Emission			
	Total	SO_4^{2-}	SO_2	MSA	dms (C_2H_6S)	Total	SO_4^{2-}	SO_2	MSA	dms (C_2H_6S)		Total	SO_2	SO_4^{2-}	dms
CAMChem	18	6	12	-	-	-	45	-	-	-	-	83	55	-	28
CHASER_re1	25	7	17	-	1	54	45	9	-	0	79	80	55	-	25
CHASER_t106	23	7	16	-	1	53	43	10	-	0	77	78	55	-	23
EMEP_rv48	16	3	13	-	-	42	32	11	-	-	58	-	-	-	Not used
GEMMACH	-	-	43	-	-	-	-	-	-	-	-	-	66	-	Not given
GEOS5	34	4	30	0.2	0	43	33	8	2	0	77	85	53	2	31
GEOSCHEMADJOINT	32	4	28	0.4	-	52	33	15	5	-	85	-	62	1	Include
OsloCTM3.v2	40	6	33	1	0	63	55	0	8	0	103	-	77	2	Include
GOCARTv5	29	6	23	-	0	47	35	12	-	0	76	-	66	2	Include
SPRINTARS	26	9	16	-	1	70	67	3	-	-	-	84	60	1	22
C-IFS_v2	-	5	43	-	-	-	-	-	-	-	-	77	-	-	Include
Multimodel mean*	28	5	21	1	1	56	40	11	5	0	84	91	-	1	27

* *The multi-model mean values of components are calculated by averaging the values of all available model outputs. The multi-model mean values of "Total" columns are calculated by summing up the multi-model mean of related components instead of averaging the values of model's "Total" results.*

Table 2. Summary of Global Total Deposition and Emission of NOy in 2010 (Tg(N) yr^{-1})

	Dry deposition						Wet deposition				Emission				
	Total NO_y	NO_2	HNO_3	NO_3	Pan	Orgn	Total NO_y	HNO_3	NO_3	Orgn	Total	Surface	Lightening	No aircraft	No soil
CAMChem	16	3	10	-	-	-	-	-	-	-	-	94	4	-	-
CHASER_re1	23	4	17	-	0.3	2	28	28	-	1	60	50	4	1	6
CHASER_t106	25	4	19	-	0.3	2	27	26	-	1	63	51	5	1	6
EMEP_rv48	15	3	8	4	1	0.2	44	27	18	-	-	-	-	-	-
GEMMACH	-	-	-	-	-	-	-	-	-	-	44	44	-	-	-
GEOSCHEMADJOINT	26	2	21	2	1	0.4	28	24	4	-	54	54	-	-	-
OsoloCTM3.v2	25	5	9	11	-	-	-	10	-	-	51	51	-	-	-
Multi-model mean	22	4	14	3	1	1	38	26	11	1	60	50	4	1	6

Table 3. Summary of Global Total Deposition and Emission of NHx in 2010 (Tg(N) yr-1)

	Dry deposition		Wet deposition		Emis_NH_3
Model/Species	NH_3	NH_4^+	NH_3	NH_4^+	
CAMChem	12	8	-	-	54
CHASER_re1	15	15	5	5	45
CHASER_t106	15	15	6	6	45
EMEP_rv48	11	3	13	-	-
GEOSCHEMADJOINT	14	4	13	24	55
OsoloCTM3.v2	19	4	6	21	54
Multi-model mean	14	5	13	22	54

Are you looking at the surface emissions or total emissions, several models do not include emissions of DMS? A follow up question on this topic, why don't the total emissions and deposition match up (i.e.7 Tg S and 1 Tg N differences in table 3 and 4)? Where does the left offers go, Have the models included organic N and S species?

The emission values listed in Table S1-S3 are total emissions, including surface and aloft. We have checked the emission of DMS with modelers (Table S1). The EMEP_rv48 model does not include DMS emission in simulation. This is consistent with the factor that the EMEP_rv48 model has lower dry and wet deposition than the other models. As a result, the mmm results, which includes EMEP_rv48 model, could underestimated the S deposition. The GEMMACH model has not given whether used DMS emission or not, but its SO_2 dry deposition is not included in the mmm, so the impact on mmm results is negligible. The GEOSCHEMADJOINT, OsloCTM3.v2, GOCARTv5 and C-IFS_v2 model used DMS emission in simulation, but they haven't submitted the emission of DMS to HTAP II.

The models include organic S and N speics. As shown in Table 1 and Table2, the models submit Methanesulfonic acid (MSA), dimethyl sulfate (dms), peroxyacyl nitrate (PAN) and other

organic nitrates than PAN (Orgn). Following are our explanation for the discrepancy between global amounts of emission and deposition.

1) There is high uncertainty in the formation ratio of DMS emission to SO_4^{2-}. The DMS emission can be oxidized to SO_2 as well as DMSO by different pathways. The DMSO is an intermediate product, which prevents the formation of aerosol. The transformation rate of DMS is about 86% to SO_2, while the rest 14% is oxidized to DMSO (Boucher et al., 2003). If we use 86% as the rate, the actual S emission should be 53+2+31*0.86=81 Tg, closer to the 77 Tg of S emission. The DMSO is further oxidized to dimethyl sulfone ($DMSO_2$), methyl sulfinic acid (MSIA) and MSA. Although the last one (MSA) is also listed as a deposition component in the table, but MSIA is the main product (Hoffmann et al., 2016). Therefore, there could be missing a large part in the S budget. In addition, the DMS and DMSO are found to be inter-changeable (Bardouki et al., 2003), which could be considered as another a reason for difference between S emission and deposition.

2) Different models are used to form multi-model mean value for emission and deposition. For instance, the GEMMACH model contributes to the S emission, but not S deposition. We adopt this way to form the multi-model mean in order to include all available model outputs in the ensemble results, but could cause inconsistency between emission and deposition.

3) The NO_y deposition is about 1 Tg N higher than NOx emission. This result is consistent to Lamarque et al. (2013), who considered this 1 Tg N from the stratosphere, which agrees well with observation data.

Comment: Line 201. Data from 43 stations of the 52 available EANET stations are used. It seems like you have included all station times, urban as well as remote, which surely have different representativity for the region. Later you state that you delete sites with high Ca values (line 219), which can be an indication of urban dust, but these may also be from also from regional dust. Not sure if I understand the reasoning behind this way of selecting the sites.

Response: In line 236 (previous line 219), we showed the evaluation results excluding the stations with high Ca^{2+} values. This is the way of screening SO_4^{2-} wet deposition observation in EANET stations adopted by the 3 previous projects of PhotoComp (Dentener et al., 2006), HTAP I (Vet et al., 2014) and ACCMIP (Lamarque et al.,2013). We adopt this method to 1) facilitate a comparison with these 3 projects to investigate improvement and remaining problem in model accuracy, which is one of the main purpose of this study. 2) as mentioned by Dentener et al. (2006), these stations with both high SO_4^{2-} and CA^{2+} measurements are likely influenced by dust emission, which is not yet included as emission in model inputs.

In line 247, we also illustrated the evaluation results if include the dust stations. "It should be noted that for the 3 excluded stations (located in China) with high Ca^{2+} deposition, the SO_4^{2-} wet deposition is largely underestimated by more than 1000 mg (S) m^{-2} yr^{-1}(not shown in figures). If we include these stations in the model evaluation, the mean bias for East Asia increases from 160 mg (S) m^{-2} yr^{-1} to 300 mg (S) m^{-2} yr^{-1}."

Comment: Line215. The outliers in Norway and Poland are probably due these specific location with high precipitation amount (Norway) and high altitude (Polish site PL03 is at 1600moh.). Have you checked how well the models compare with precipitation amount contra concentration levels in precipitation?

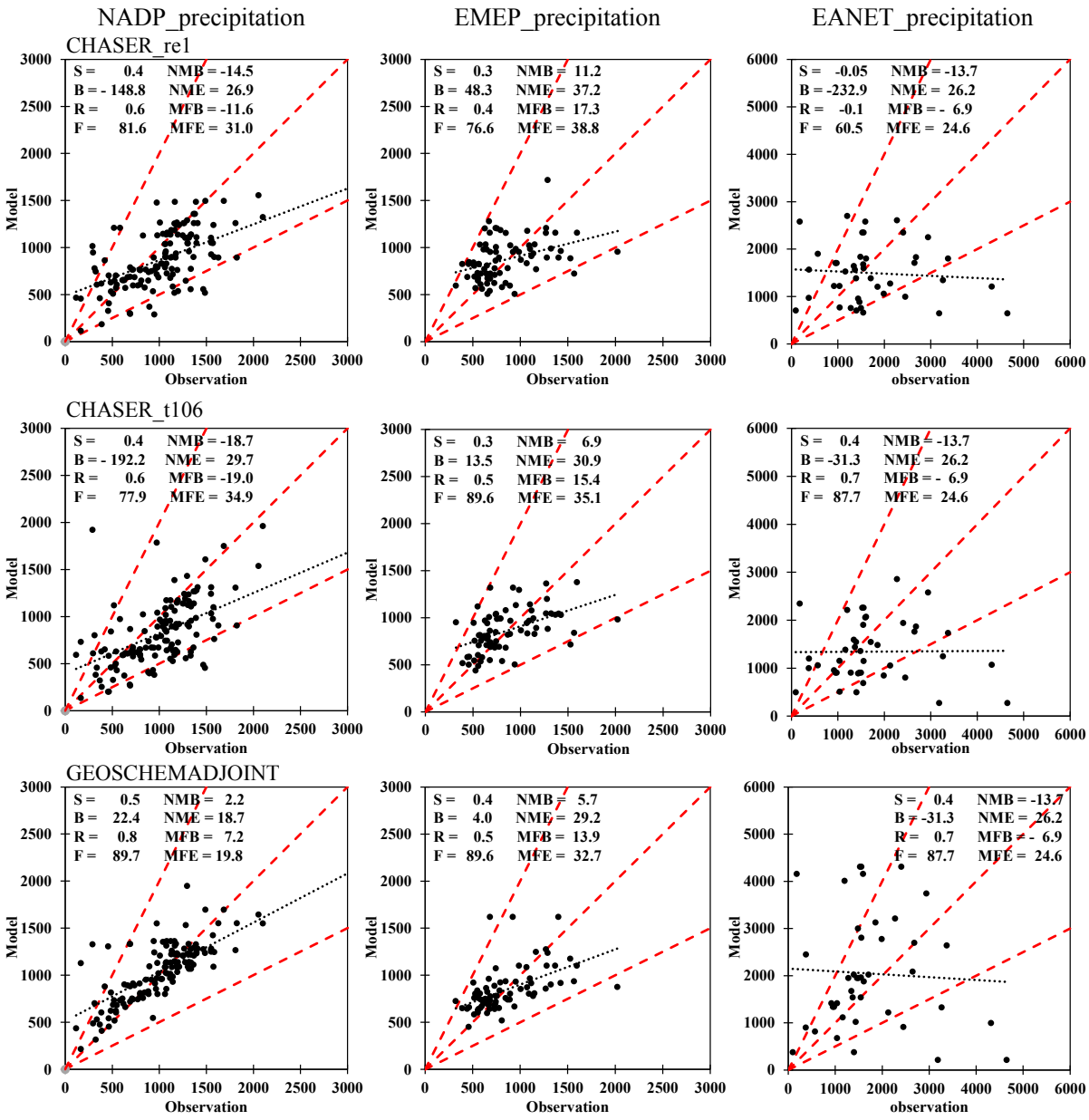
Response: we evaluate the precipitation with observation as shown in Fig. S16 and Fig. S17. For the Norway (NO01) site, the observed precipitation is 1566 mm yr^{-1} and the mmm underestimated the precipitation by 49%. It's SO_4^{2-}, NH_4^+ and NO_3^- wet deposition is underestimated by 332 mg(S) m^{-2} yr^{-1} (50%), 385 mg(N) m^{-2} yr^{-1} (64%) and 248 mg(N) m^{-2} yr^{-1} (63%), respectively. The 49% underestimation of precipitation in the Norway site fits well to the underestimation of SO_4^{2-} wet deposition.

For the Polish (PL03) site, the observed precipitation is 1137 mm yr^{-1} and the mmm underestimated the precipitation by 21%. It's SO_4^{2-}, NH_4^+ and NO_3^- wet deposition is underestimated by 718 mg(S) m^{-2} yr^{-1} (71%), 213 mg(N) m^{-2} yr^{-1} (40%) and 301 mg(N) m^{-2} yr^{-1} (60%), respectively. As mentioned in the comment, one possible reason could be the complicated topography of the sites. The height of the Polish site is 1603 meters above sea, which is one of the highest sites among all EMEP sites. Similar to the PL03 site, the ES09R sites in Spain, which is 1360 meters high, is underestimated by 142 mg(S) m^{-2} yr^{-1} (59%), 184 mg(N) m^{-2} yr^{-1} (57%) and 135 mg(N) m^{-2} yr^{-1} (67%) for its SO_4^{2-}, NH_4^+ and NO_3^- wet deposition, while its precipitation is well simulated with a positive model bias of 5%.

We have added the following paragraph in the manuscript to explain the reason for model bias in the manuscript.

Line 226: We evaluated the model performance on simulating precipitation (Fig. S5 and Fig. S6). For the Norway site, the observed precipitation is 1566 mm yr^{-1} and the mmm underestimated the precipitation by 49%, which fits well for the 50% underestimation of SO_4^{2-} wet deposition at this site. For the Polish site, the observed precipitation is 1137 mm yr^{-1} and the mmm underestimated the precipitation by 21%. The underestimation in precipitation could partly explain the negative model bias in simulating SO_4^{2-} wet deposition. Another possible reason is the high topography of the sites. The Polish site is 1603 meters above sea, which is one of the highest sites among the European sites. Similar to the Polish sites, one site in Spain, which is 1360 meters height, is underestimated by 142 mg (S) m^{-2} yr^{-1} (59%) for SO_4^{2-} wet deposition, while its precipitation is well simulated with a slight positive model bias of 5%.

Fig. S5


NADP_precipitation
EMEP_precipitation
EANET_precipitation
CHASER_re1
S = 0.4 NMB = -14.5
B = - 148.8 NME = 26.9
R = 0.6 MFB = -11.6
F = 81.6 MFE = 31.0
S = 0.3 NMB = 11.2
B = 48.3 NME = 37.2
R = 0.4 MFB = 17.3
F = 76.6 MFE = 38.8
S = -0.05 NMB = -13.7
B = -232.9 NME = 26.2
R = -0.1 MFB = - 6.9
F = 60.5 MFE = 24.6
CHASER_t106
S = 0.4 NMB = -18.7
B = - 192.2 NME = 29.7
R = 0.6 MFB = -19.0
F = 77.9 MFE = 34.9
S = 0.3 NMB = 6.9
B = 13.5 NME = 30.9
R = 0.5 MFB = 15.4
F = 89.6 MFE = 35.1
S = 0.4 NMB = -13.7
B = -31.3 NME = 26.2
R = 0.7 MFB = - 6.9
F = 87.7 MFE = 24.6
GEOSCHEMADJOINT
S = 0.5 NMB = 2.2
B = 22.4 NME = 18.7
R = 0.8 MFB = 7.2
F = 89.7 MFE = 19.8
S = 0.4 NMB = 5.7
B = 4.0 NME = 29.2
R = 0.5 MFB = 13.9
F = 89.6 MFE = 32.7
S = 0.4 NMB = -13.7
B = -31.3 NME = 26.2
R = 0.7 MFB = - 6.9
F = 87.7 MFE = 24.6
Model
Observation
observation

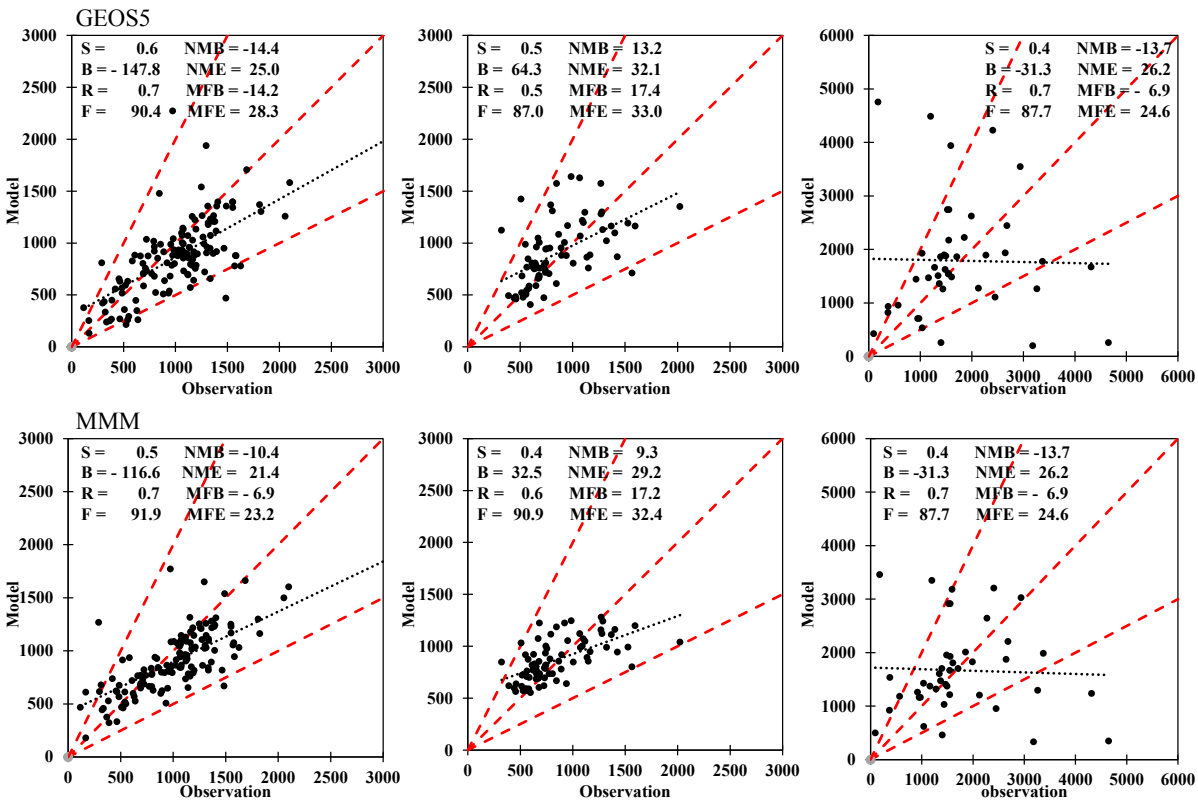


Fig. S5. Individual model performances on precipitation (mm yr^{-1}). The model result is the annual precipitation in 2010 and the observation is 3-year average annual data of 2009-2011.

Fig. S6

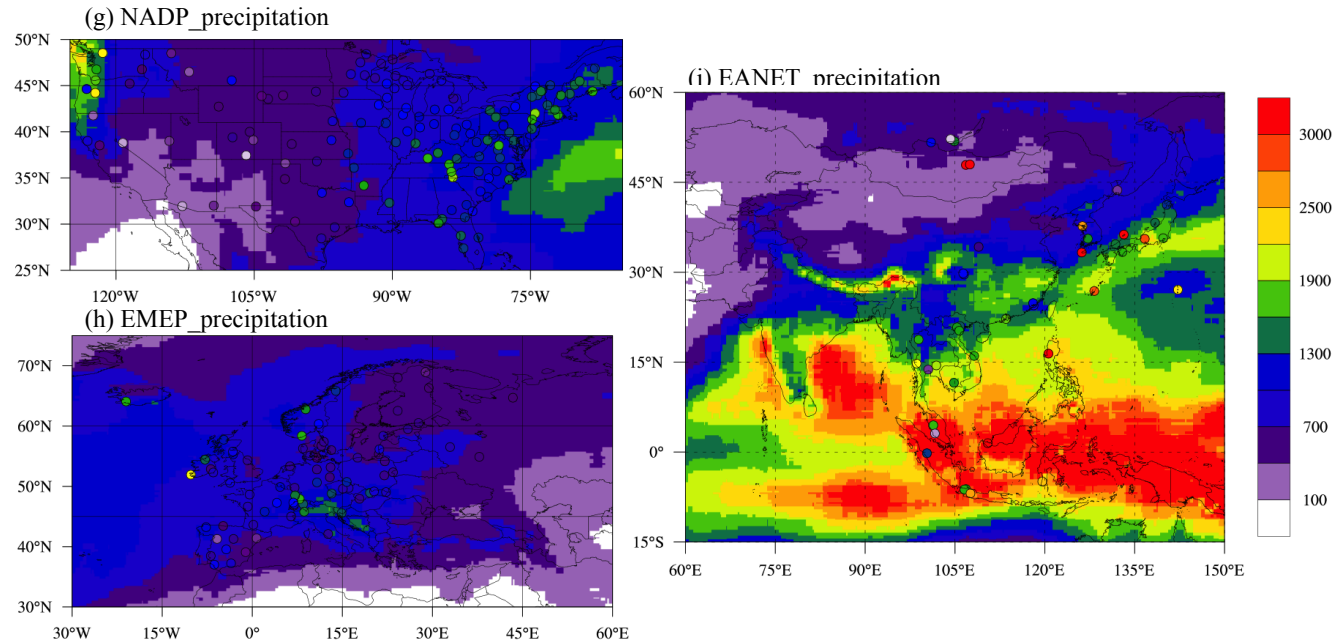


Fig. S6. Distribution of precipitation (mm yr^{-1}) of MMM and observation. The MMM is the annual total precipitation in 2010 and the observation is 3-year average annual data of 2009-2011.Contours are MMM results and filled circles are observation.

Comment: Line 235 “According to Fig. 2(d), the over-predicted stations are mainly located in Midwestern and Southeast United”. For me it seems like a general tendency (fig 1d). Maybe include information that 67 % of the station are within 50%.

Response: Thank you for pointing this out. We have revised the sentence in the manuscript. “According to Fig. 2(d), there is a general tendency of overestimation throughout the stations in United States, especially the stations located in Midwest and Southeast.

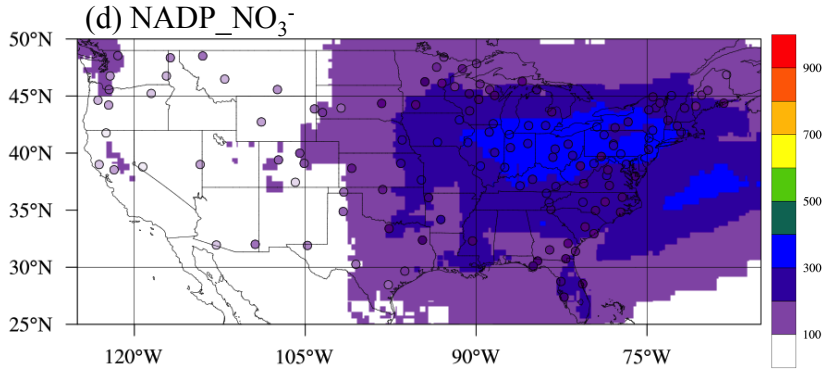


Fig. 2(g) Distribution of NH_4^+ wet deposition (mg (N) m^{-2} yr^{-1}) of MMM and observation. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011.Contours are MMM results and filled circles are observation.

Comment: Line 265. "The NH_4^+ wet deposition is somewhat underestimated in all 3 regions". This is not the case for US (NADP) if one look at table 1 where the HTAPII is higher than observations.

Response: According to table 1, the average value of modelled wet NH_4^+ deposition is 2.3 mg(N) m^{-2} y^{-1} higher than that of observation. This is mainly due to that the model over-estimated two sites (indicated by red circle in Fig. 1(g) in following figure) by about 100 mg(N) m^{-2} y^{-1}. But the linear fit slope between model and observation is 0.8 (<1) (as shown in Table 1 and Fig. 1(g) in the following figure), which indicates a generally trend of slight underestimation, due to the generally underestimation found in Southeast (Fig. 2(g)).

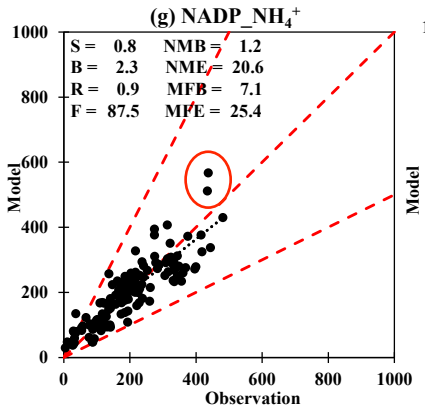


Fig. 1(g) Evaluation of MMM performance of NH_4^+ wet deposition (mg (N) m^{-2} yr^{-1}) at NADP stations. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011.

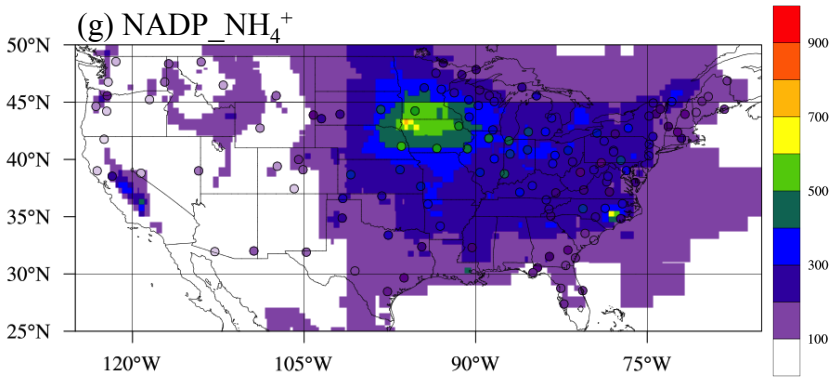


Fig. 2(g) Distribution of NH_4^+ wet deposition (mg (N) m^{-2} yr^{-1}) of MMM and observation. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011.Contours are MMM results and filled circles are observation.

Comment: Line 350. “The ocean serves as an important sink of S deposition”. But it is also a very important source. The net effect is only 3 TgS.

Response: The ocean (including coastal region) emits 39.2 TgS of S emission in 2010, accounts for 43% of global total S emission, while it receives 43 TgS of S deposition, accounts for 51% of global total S deposition. The difference of 3 TgS is considerably small if compared to the 43 TgS of deposition. In order to avoid misleading the readers, we have deleted that sentence in the manuscript.

Comment: Table 3,4,5. It is a bit confusing for the reader when you have defined two different categories continental coastal and ocean coastal which are the same thing. Would be more readable and less confusion if these cells are merges so it is clear that there are three categories (Ocean, Continent and Coast)

Response: We have merged them into one row in the manuscript.

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