

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 $\pm 20\%$ 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 $\pm 1.5 \times$ 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 ₂ H ₆ S)	Total	SO ₄ ²⁻	SO ₂	MSA	dms (C ₂ H ₆ S)		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 NO_y 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 NH_x in 2010 (Tg(N) yr⁻¹)

Model/Species	Dry deposition		Wet deposition		Emis_NH ₃
	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 SO₂ 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 NO_x 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 $1000 \text{ mg (S) m}^{-2} \text{ yr}^{-1}$ (not shown in figures). If we include these stations in the model evaluation, the mean bias for East Asia increases from $160 \text{ mg (S) m}^{-2} \text{ yr}^{-1}$ to $300 \text{ mg (S) m}^{-2} \text{ yr}^{-1}$.”

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?

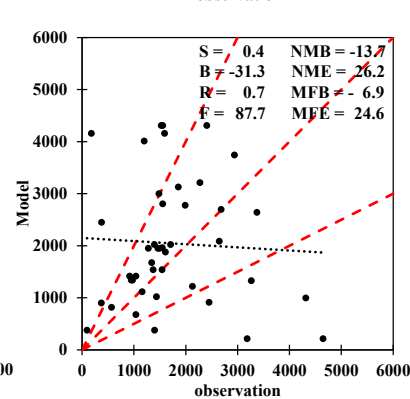
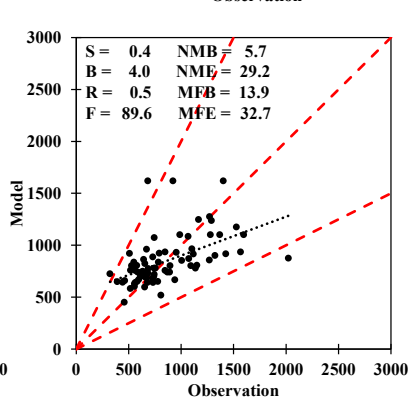
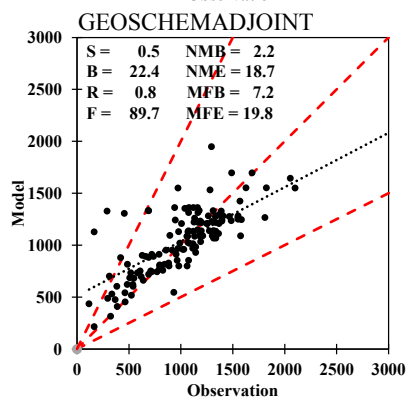
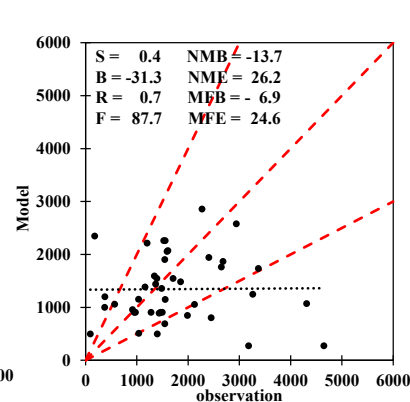
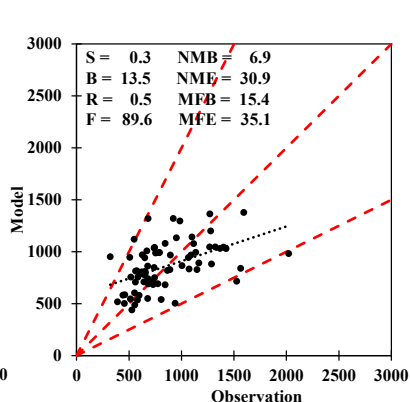
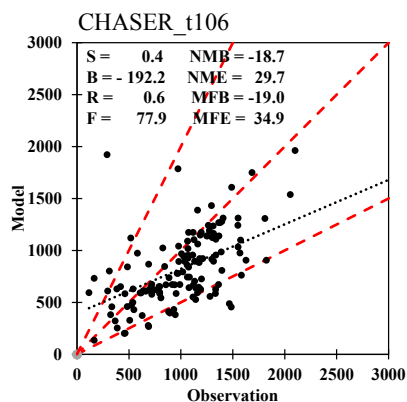
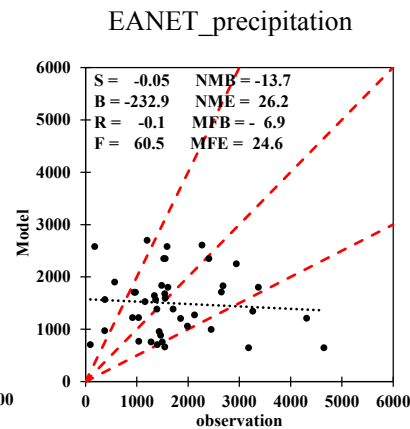
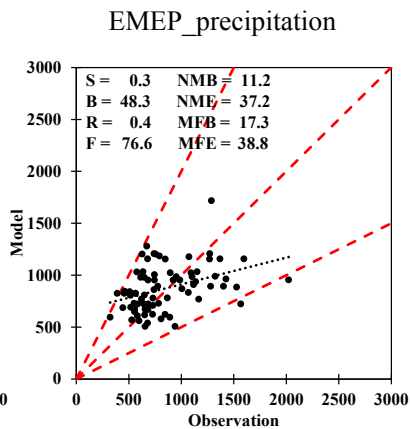
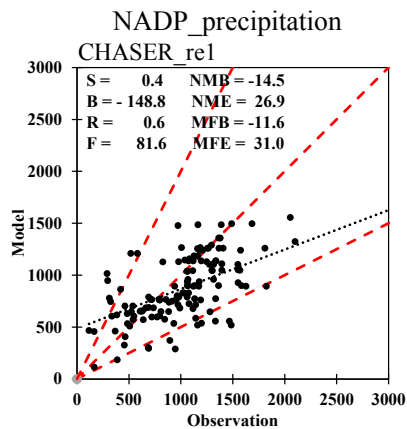
Response: we evaluate the precipitation with observation as shown in Fig. S16 and Fig. S17. For the Norway (NO01) site, the observed precipitation is 1566 mm yr^{-1} and the mmm underestimated the precipitation by 49%. It's SO_4^{2-} , NH_4^+ and NO_3^- wet deposition is underestimated by $332 \text{ mg(S) m}^{-2} \text{ yr}^{-1}$ (50%), $385 \text{ mg(N) m}^{-2} \text{ yr}^{-1}$ (64%) and $248 \text{ mg(N) m}^{-2} \text{ yr}^{-1}$ (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 $718 \text{ mg(S) m}^{-2} \text{ yr}^{-1}$ (71%), $213 \text{ mg(N) m}^{-2} \text{ yr}^{-1}$ (40%) and $301 \text{ mg(N) m}^{-2} \text{ yr}^{-1}$ (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 $142 \text{ mg(S) m}^{-2} \text{ yr}^{-1}$ (59%), $184 \text{ mg(N) m}^{-2} \text{ yr}^{-1}$ (57%) and $135 \text{ mg(N) m}^{-2} \text{ yr}^{-1}$ (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 $142 \text{ mg (S) m}^{-2} \text{ yr}^{-1}$ (59%) for SO_4^{2-} wet deposition, while its precipitation is well simulated with a slight positive model bias of 5%.

Fig. S5



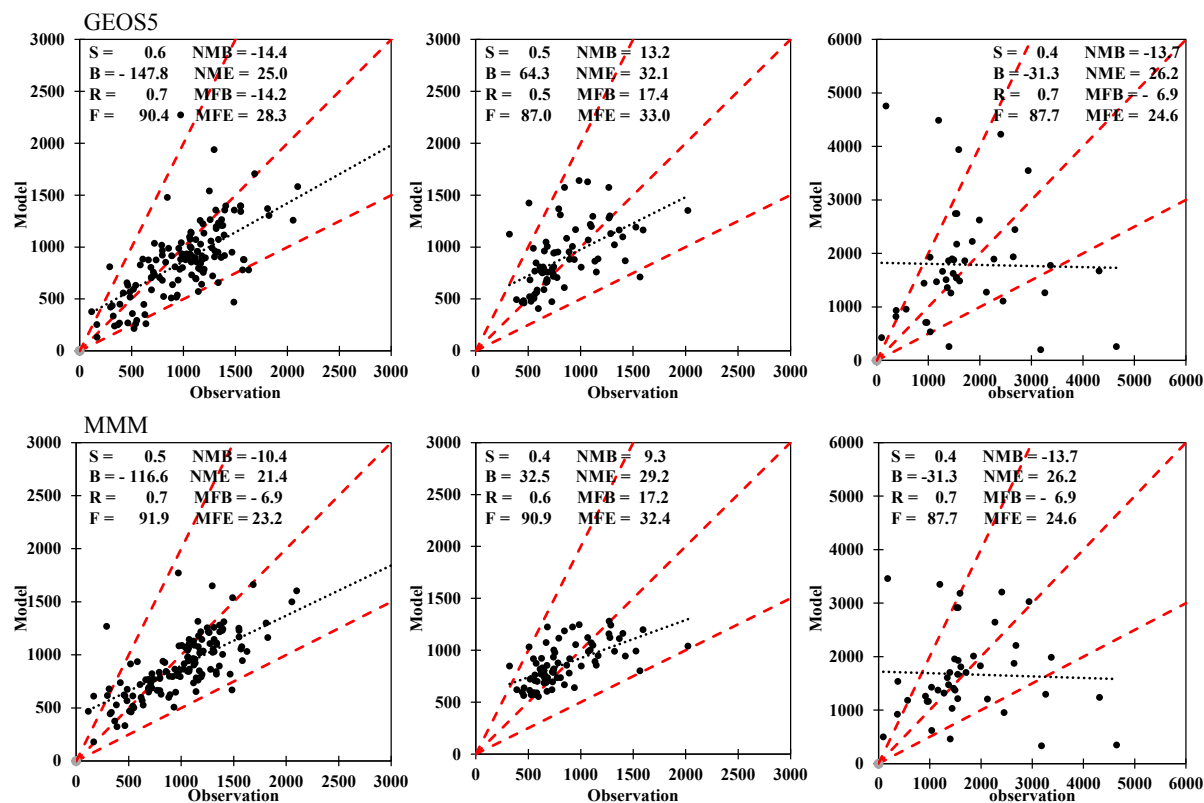


Fig. S5. Individual model performances on precipitation (mm yr^{-1}). The model result is the annual precipitation in 2010 and the observation is 3-year average annual data of 2009-2011.

Fig. S6

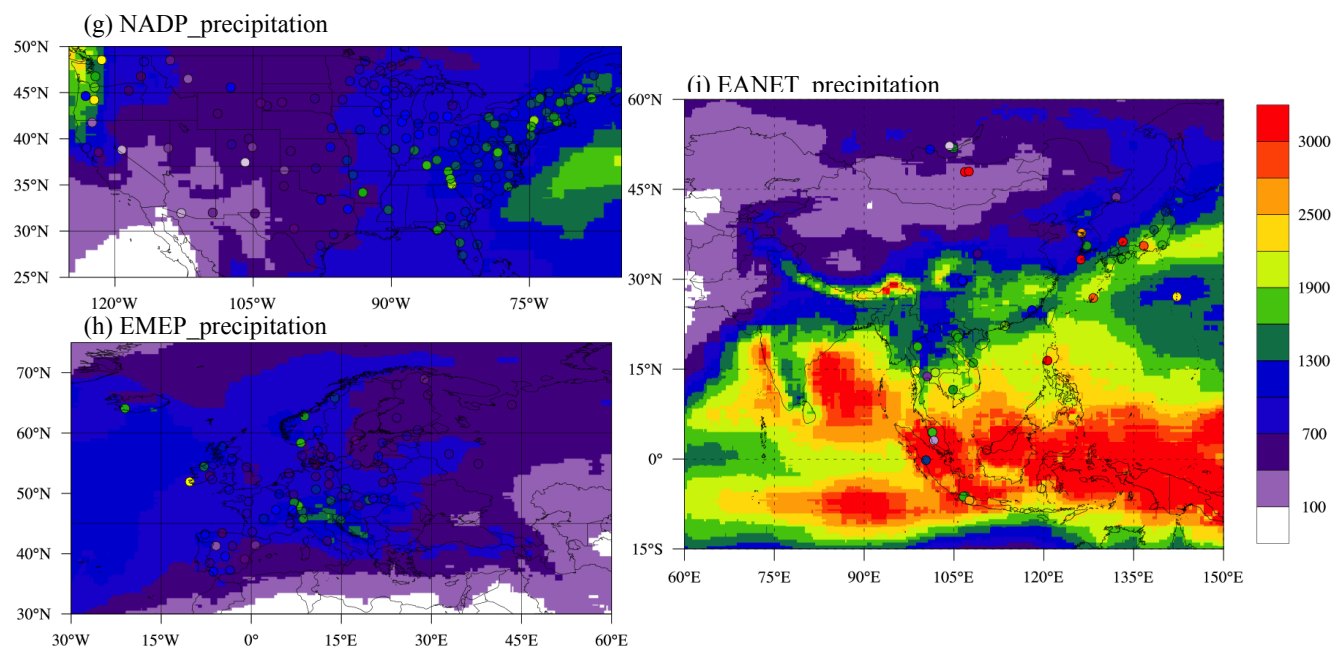


Fig. S6. Distribution of precipitation (mm yr^{-1}) of MMM and observation. The MMM is the annual total precipitation in 2010 and the observation is 3-year average annual data of 2009-2011. Contours are MMM results and filled circles are observation.

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.

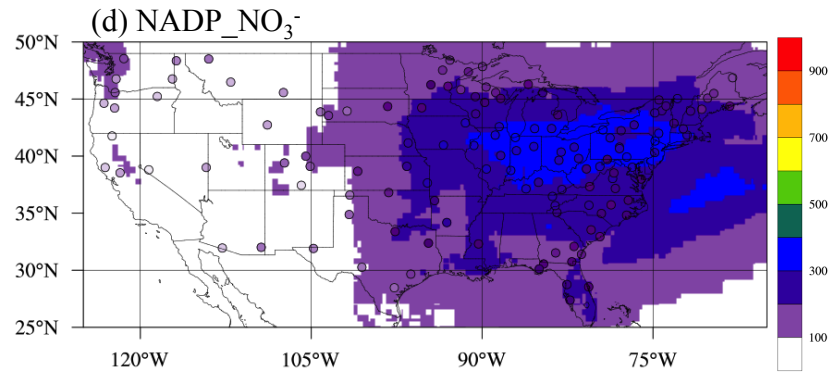


Fig. 2(g) Distribution of NH_4^+ wet deposition ($\text{mg (N) m}^{-2} \text{ yr}^{-1}$) of MMM and observation. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011. Contours are MMM results and filled circles are observation.

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 $2.3 \text{ mg(N) m}^{-2} \text{ y}^{-1}$ 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 $100 \text{ mg(N) m}^{-2} \text{ y}^{-1}$. 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)).

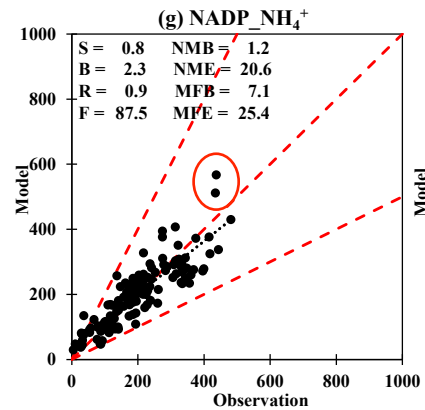


Fig. 1(g) Evaluation of MMM performance of NH_4^+ wet deposition ($\text{mg (N) m}^{-2} \text{ yr}^{-1}$) at NADP stations. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011.

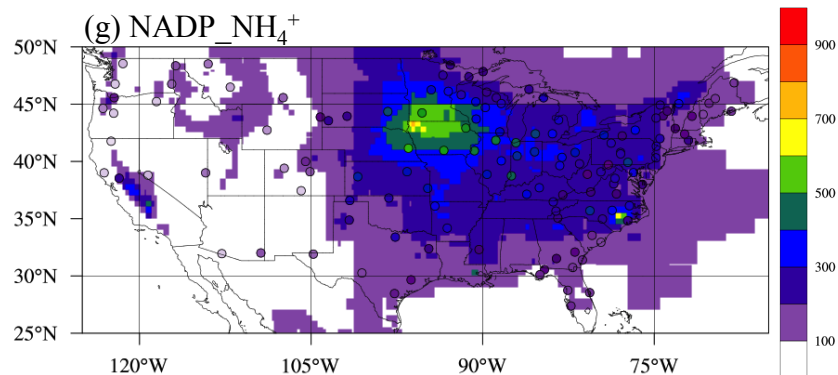


Fig. 2(g) Distribution of NH_4^+ wet deposition ($\text{mg (N) m}^{-2} \text{ yr}^{-1}$) of MMM and observation. The MMM is the annual wet deposition in 2010 and the observation is 3-year average annual data of 2009-2011. Contours are MMM results and filled circles are observation.

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 merged so it is clear that there are three categories (Ocean, Continent and Coast)

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

Reference

Bardouki, H., Berresheim, H., Vrekoussis, M., Sciare, J., Kouvarakis, G., Oikonomou, K., Schneider, J., and Mihalopoulos, N.: Gaseous (DMS, MSA, SO₂, H₂SO₄ and DMSO) and particulate (sulfate and methanesulfonate) sulfur species over the northeastern coast of Crete, *Atmospheric Chemistry and Physics*, 3, 1871-1886, 2003.

Boucher, O., Moulin, C., Belviso, S., Aumont, O., Bopp, L., Cosme, E., von Kuhlmann, R., Lawrence, M. G., Pham, M., Reddy, M. S., Sciare, J., and Venkataraman, C.: DMS atmospheric concentrations and sulphate aerosol indirect radiative forcing: a sensitivity study to the DMS source representation and oxidation, *Atmospheric Chemistry and Physics*, 3, 49-65, 2003.

Hoffmann, E. H., Tilgner, A., Schrodner, R., Brauera, P., Wolke, R., and Herrmann, H.: An advanced modeling study on the impacts and atmospheric implications of multiphase dimethyl sulfide chemistry, *Proceedings of the National Academy of Sciences of the United States of America*, 113, 11776-11781, 2016.