1 **Source contributions of sulfur and nitrogen deposition – an HTAP II multi-**
2 **model study on hemispheric transport**

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21 **Abstract.** With rising emissions by human activities, enhanced concentrations of air pollutants

22 have been detected in hemispheric air flows in recent years, aggravating the regional air

23 pollution and deposition burden. However, contributions of hemispheric air pollution to

24 deposition at global scale have been given little attention in the literature. In this light, we assess

25 the impact of hemispheric transport on sulfur (S) and nitrogen (N) deposition for 6 world

26 regions: North America, Europe, South Asia, East Asia, Middle East and Russia in 2010, by

27 using the multi-model ensemble results from the 2nd phase of Task Force Hemispheric Transport

28 of Air Pollution (HTAP II) with and without 20% emission perturbation experiments. About 27-

29 58%, 26-46% and 12-23% of local S, NO_x and NH_3 emissions are transported and removed by

30 deposition outside of the source regions annually, with 5% higher fraction of export in winter

31 and 5% lower in summer. For receptor regions, 20% emission reduction in source regions affects

32 the deposition in receptor regions by 1-10% for continental non-coastal regions and 1-15% for

33 coastal regions and open oceans. Significant influences are found from North America to the

34 North Atlantic Ocean (5-15%), from South Asia to western East Asia (2-10%) and from East

35 Asia to the North Pacific Ocean (5-10%) and western North America (5-8%). The impact on

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36   deposition caused by transport between neighbouring regions (i.e. Europe and Russia) occurs

37   throughout the whole year (slightly stronger in winter), while that by transport over long

38   distances (i.e. from East Asia to North America) mainly takes place in spring and fall, which is

39   consistent with the seasonality found for hemispheric transport of air pollutants. Deposition in

40   emission intense regions such as East Asia is dominated (~80%) by own region emission, while

41   deposition in low emission regions such as Russia is almost equally affected by own region

42   emission (~40%) and foreign impact (~23-45%). We also find that deposition on the coastal

43   regions or near coastal open ocean is twice more sensitive to hemispheric transport than non-

44   coastal continental regions, especially for regions (i.e. west coast of North America) in the

45   downwind location of major emission source regions. This study highlights the significant

46   impact of hemispheric transport on deposition in coastal regions, open ocean and low emission

47   regions. Further research is proposed for improving ecosystem and human health in these

48   regions, with regards to the enhanced hemispheric transport.

49   **1 Introduction**

50   The increasing consumption of energy by human activities has largely increased the deposition

51   of sulfur (S) and nitrogen (N) over the terrestrial and marine ecosystem in recent years (Kim et

52   al., 2011; Galloway et al., 2008; Duce et al., 2008) and the amount of atmospheric deposition is

53   estimated to continue increasing in the near future (Bleeker et al., 2011; Lamarque et al., 2013;

54   Kanakidou et al., 2016; Paulot et al., 2013; Lamarque et al., 2005). While deposition supplies

55   ecosystem with nutrients, too much deposition could cause various adverse impacts on the

56   environment, including acidification and eutrophication of the forest and waterbody (Bouwman

57   et al., 2002; Bergstrom and Jansson, 2006; Dentener et al., 2006; Phoenix et al., 2006), soil

58   acidification that slows down the crop production (Guo et al., 2010; Janssens et al., 2010) and

59   even decrease plant biodiversity (Bobbink et al., 2010; Clark and Tilman, 2008). The control of

60   deposition has become a growing worldwide concern.

61   Hemispheric transport of air pollutants is found to aggravate the regional air pollution

62   (Wild and Akimoto, 2001; Sudo and Akimoto, 2007; Fu et al., 2012; Fiore et al., 2009) as well as

63   enlarge the local deposition burden (Glotfelty et al., 2014; Sanderson et al., 2008). Air pollution

64   from Asia contributes to the concentration of $PM_{2.5}$ in western United States by 1.5 µg m^{-3} (Tao

65   et al., 2016), the O_3 concentration by 3-10 ppbv (Zhang et al., 2009; Zhang et al., 2008; Yienger

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66   et al., 2000; Reidmiller et al., 2009; Jacob et al., 1999; Brown-Steiner and Hess, 2011) and the

67   peroxyacyl nitrate (PAN) concentration by 26 ppbv (Berntsen et al., 1999) in spring. The long-

68   range transport of air pollution from North America is estimated to contribute by 3-5 ppb to (7-

69   11%) to the O_3 concentration in Europe annually (Auvray and Bey, 2005; Guerova et al., 2006;

70   Derwent et al., 2004; Li et al., 2002) and the increment can reach 25-28 ppbv during particular

71   events (Guerova et al., 2006). European outflow affects the surface O_3 concentration in western

72   China by 2-6 ppbv in spring and summer (Li et al., 2014) and North Africa by 5-20 ppbv in

73   summer (Duncan et al., 2008; Duncan and Bey, 2004). The study by Yu et al. (2013) found that

74   the long-range transport contributes by 6-16% and 22-40% to aerosol optional depth and direct

75   radiative forcing in 4 regions including North America, Europe, East Asia and South Asia.

76   Recent studies have reported an increasing trend in the hemispheric transport of air pollution. In

77   particular, the air pollution exported from Asia to North America has increased significantly in

78   recent years (Jaffe et al., 2003; Parrish et al., 2009; Parrish et al., 2004; Verstraeten et al., 2015;

79   Zhang et al., 2008) due to the rapid growth of Asian emission (Richter et al., 2005; Lu et al.,

80   2010; Zhang et al., 2007; van der A et al., 2006; van der A et al., 2008).

81   Compared to the impact on air pollution, the impact of hemispheric transport on

82   deposition hasn't been fully studied. Arndt and Carmichael (1995) developed a source-receptor

83   (S-R) relationship for S deposition among the Asia regions in early 1900s. Zhang et al. (2012)

84   found foreign anthropogenic emission contributes to 6% and 8% of the NO_y and NH_x deposition

85   in contiguous United States, respectively. A systematic study by Sanderson et al. (2008) shed

86   light on the impact of long-range transport on deposition of oxidized nitrogen at global scale.

87   That study uses the model ensemble results from phase I of Task Force Hemispheric Transport

88   of Air Pollution (HTAP I) to calculate the S-R relationship for NO_y deposition in 2001 among 4

89   regions: Europe, North America, South Asia and East Asia. Results showed that about 12-24%

90   of the NO_x emission is deposited out of source regions. About 3-10% of the emission is

91   deposited on the other 3 regions and affects their deposition by about 1-3%. However, these

92   studies focused on emission intense regions, where the foreign disturbance is relatively small

93   compared to huge own region emission. The foreign impact on low emission regions was not

94   evaluated in the same detail. Furthermore, both the magnitude and spatial distribution of S and N

95   emission and deposition have been changed considerably during the last 10 years (2001-2010)

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96   (Tan et al., 2018). It is necessary to update the S-R relationship for more recent years with

97   regards to these changes.

98   To explore these questions, this study assesses the impact of hemispheric transport of S,

99   NO_x and NH_3 emissions on S and N deposition, with multi-model ensemble results from 2nd

100   phase of HTAP (HTAP II). Additional to the 4 regions: North America (NA), Europe (EU),

101   South Asia (SA), East Asia (EA) used in Sanderson's (2008) study for HTAP I, we include 2

102   regions: Middle East (ME) and Russia, Belarussia, Ukraine (RU) in this study. These two

103   regions have low S and N emissions relative to their areal extent, but are located close to high

104   emission regions such as EU, SA and EA. We calculate the amount of deposition brought by

105   hemispheric transport by comparing model results for base case and for 20% emission

106   perturbation cases. The experimental design is described in Section 2. Section 3 is the result part

107   and has 3 subsections. We explore the following questions:

108   1) Which fraction (percentage) of the S or N emissions is transported and deposited outside of

109   this region? Which fraction is finally deposited on the other 5 receptor regions, other regions

110   and oceans? What is the seasonality of the exported fraction?

111   2) As receptor regions, what is the amount of deposition brought by hemispheric transport?

112   How much will it affect the local deposition? Is there any seasonality for this impact?

113   3) For each region, what are the contributions of hemispheric transport from foreign regions and

114   of control of own region emission on deposition? In line with the analysis for other pollutants,

115   to this purpose we evaluate the so-called response to extra-regional emission reduction

116   (RERER) metric. We also discuss the own region and foreign impact specifically on the

117   coastal regions. The inter-model variations are also illustrated in this section.

118   Section 4 is a summary of the findings in this study and some suggestions for future study.

119   **2 Methodology**

120   **2.1 HTAP II and experiment set-up**

121   The HTAP was created in 2004 under the Convention on Long-range Transboundary Air

122   Pollution (CLRTAP). The project involves efforts from international scientists aiming at

123   understanding the hemispheric transport of air pollutants and its impact on regional and global

124   air quality, public health and near-term climate change. Until now, two phases of HTAP

125   experiments have been conducted successfully. The HTAP I involved more than 20 models from

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126   international modelling groups, with 2001 as the base year for modeling studies. A

127   comprehensive report of the major findings of HTAP was released in 2010 and could be

128   downloaded from http://www.htap.org/. The HTPA II was launched in 2012, with 2010 as the

129   base simulation year. HTAP II required all models to use the same prescribed anthropogenic

130   emission inventory and boundary conditions instead of using the best estimates of emission by

131   each model group as HTAP I, which facilitated an inter-model comparison between models. In

132   addition, HTAP II had a refined definition for the boundaries of regions, which enabled an

133   update in the S-R relationships for air pollutants and deposition among regions.

134   This study uses the ensemble of 11 global models from HTAP II (including CAM-Chem,

135   CHASER_re1, CHASER_t106, EMEP_rv48, GEMMACH, GEOS5, GEOSCHEMAJOINT,

136   OsloCTM3v.2, GOCARTv5, SPRINTARS and C-IFS_v2). A detailed description of the

137   experiment set-up could be found in Galmarini et al. (2017). The S deposition includes SO_2

138   deposition and SO_4^{2-} deposition. N deposition is categorized to oxidized nitrogen (NO_y) and

139   reduced nitrogen (NH_x) deposition. NO_y deposition is a sum of all oxidized N except N_2O,

140   including NO_2, HNO_3, NO_3^-, PAN and other organic nitrates than PAN (Orgn). NH_x deposition

141   includes NH_3 deposition and NH_4^+ deposition. To form the multi-model ensemble, we re-grid all

142   models to a uniformed horizontal resolution of $0.1° \times 0.1°$. We use the multi-model mean value

143   (MMM) of all models to present the ensemble results, a procedure which has previously been

144   proven to have a better agreement with observations than single model results (Dentener et al.,

145   2006; Tan et al., 2018). The mean values of the compositions of S or N deposition are calculated

146   separately and then are combined to compute the total S or N deposition. More details can be

147   found in Tan et al. (2018). The inter-model variations are discussed in section 3.3.

148   **2.2 Simulation scenarios**

149   The base simulation uses anthropogenic emissions in 2010 (Janssens-Maenhout et al.,

150   2015), which is called "base case" in this study. The MMM performance on wet deposition has

151   been evaluated with observations from National Atmospheric Deposition Program (NADP)

152   (http://nadp.sws.uiuc.edu/) for NA, European Monitoring and Evaluation Programme (EMEP)

153   CCC reports (http://www.nilu.no/projects/ccc/reports.html) for EU and Acid Deposition

154   Monitoring Network in East Asia (EANET) (http://www.eanet.asia/) for EA in Tan et al. (2018).

155   In terms of wet deposition, MMM results are evaluated with site observation of deposition. SO_4^{2-}

156   wet deposition is generally well simulated with 76% of the stations of all networks predicted

157   within ±50% of observation. Negative model biases (-20%) are found at some East Asian

158   stations. Modeled NO_3^- wet deposition is within ±50% of observation for 83% of the stations of

159   all networks. The European and Southeast Asian stations with high observed NO_3^- wet deposition

160   are found to be somewhat underestimated. 81% of modeled NH_4^+ wet deposition at stations of all

161   networks are within ±50% of observation. A general underestimation is found in modelled NH_4^+

162   wet deposition, especially at East Asian stations. In terms of dry deposition, due to the lack of

163   directly measured dry deposition, we compare the modeled dry deposition with inferential data

164   from the Clean Air Status and Trends Network (CASTNET) over United States. The CASTNET

165   inferential data is calculated with observed aerosol concentration and modelled dry deposition

166   velocity, therefore it has high uncertainty in data quality. Results show that the modelled dry

167   deposition is generally a factor of 1-2 higher than the CASTNET inferential data. This is a

168   common feature of many global and regional models (WMO, 2017) and subject to further

169   research.

170   In addition to the base case simulations, emission perturbation experiments are conducted

171   for certain regions. The boundaries of 17 regions in HTAP II are defined in Fig. 1. In the

172   perturbation experiments, the anthropogenic emissions (including NO_X, SO_2, NH_3, VOC, CO

173   and PM) of specific regions (i.e. NA) are reduced by 20% from the amounts in the base case

174   simulation, while the other regions keep the same emissions. This study uses the perturbation

175   experiments of 6 regions (with color in Fig.1) with high priority: NA, EU, SA, EA, ME and RU.

176   In addition, a global perturbation experiment referred as "GLO" is conducted with 20% decrease

177   in global anthropogenic emissions. We estimate the impact of hemispheric transport on

178   deposition by comparing the model results under perturbation experiments with those under base

179   case simulation. In order to validate the quality of model outputs, we check the mass balance

180   between emission and deposition at global scale. The mass balance for base case simulation has

181   been checked in Tan et al. (2018), therefore we show the mass balance for perturbation

182   experiments in this study. We compare the global total amounts of changes of deposition (Δ

183   Depo) with changes of emissions (Δ Emis) for all perturbation cases (Table S1). According to

184   our results, the amounts of Δ Depo are almost identical to Δ Emis for all perturbation cases. The

185   Δ Depo of NH_x deposition under EA perturbation case is not available due to that no model

186   meets the mass balance requirements mentioned above.

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3 Results

3.1 Export of S and N emissions from source regions

This section studies the export of S and N emissions and oxidation products from source regions. Table 1 shows the S-R relationship of S and N deposition among the 6 regions. The numbers are the sensitivity ($SEN_{r \to s}$) of deposition in the receptor/source regions to emission changes in the source regions (Sanderson et al., 2008). The metric is calculated as Δ Depo in the receptor/source regions divided by Δ Emis in the source regions following equation (1).

$$SEN_{r \to s} = \frac{\Delta\,Depo\,(r/s)}{\Delta\,Emis\,(s)} \times 100\% \qquad (1)$$

where s is the source region and r is the receptor region. Δ Depo (r/s) is the deposition change in the receptor/source regions, Δ Emis (S) is the emission change in the source regions. This value indicates the fraction of emission from source regions that is deposited locally or exported to foreign regions.

The numbers in Table1 are for coastal and non-coastal regions together and the numbers in the parenthesis are specifically for coastal regions (defined in Fig. 1). "Others" means the other regions in the world than the 6 regions (white color in Fig.1). The NA region has 69% of its S emission deposited within itself, including 9% deposited on its coastal region. The remaining 31% is exported to the other regions, mostly to the "Others" and less than 1% is deposited on the other 5 regions (EU, SA, EA, ME and RU). A relatively large fraction (14 %) of European S emissions are exported to RU region. Other major pathways of export of sulphur emissions/reaction products are from SA to EA (9%), from EA to RU (5%) and from RU to EU (7%) and EA (5%). ME has considerable percentages of S emission exported to its nearby regions such as SA (8%), EA (5%) and RU (5%). The S-R relationship of NO_y deposition is similar to that of S deposition, except that EU and ME have 66% and 54% of NO_x emissions deposited within the source region, which are 6% and 12% higher than those of S emissions, likely due to the large average emission altitude of S emissions and somewhat longer lifetimes compared to NOx emissions. In terms of NH_x deposition, about 20% more NH_3 emission is deposited within the source region due to its short lifetime in the atmosphere.

The seasonal variation of the export of S and N emissions by source regions is shown in Fig. S1. In terms of S emission, there is 5-10% of seasonal variation around the annual average

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216   of 25% in the export fractions for all regions except SA. SA exports almost half of its annual S

217   emission in spring, which is twice the numbers in summer (20%) and fall (25%), related to the

218   specific dry period and monsoon circulation. The seasonal export fractions of NO_x and NH_3

219   emission are similar to that of S emission, but generally lower in all seasons. Generally, the

220   source regions export highest percentage of their emissions in winter and spring and lowest in

221   summer. More proficient oxidation chemistry in summer resulting in more soluble component,

222   and local weather systems, especially the episodic of precipitation has large influence on this

223   seasonality. For most continental regions, the wet deposition accounts for 50-70% of total

224   deposition (Tan et al., 2018; Vet et al., 2014; Dentener et al., 2006). Therefore, local

225   precipitation washes out large proportion of the air pollutants in the atmosphere during the

226   rainfall seasons (i.e. summer). In addition, the strong westerly winds in winter and spring favor

227   the hemispheric transport, while the rapid vertical convection in summer slows down the zonal

228   transport of air flows and accelerates the removal process.

229   We compare our results with previous studies. Bey et al. (2001) estimated that 70% of

230   emitted NO_x from Asia is lost within its boundary by deposition of HNO_3 in spring. The

231   estimation is our study is 70% for EA and 61% for SA, close to Bey's result. Li et al. (2004)

232   reported that about 20% of anthropogenic NO_x emitted by NA is deposited out of its boundary

233   (about 1000 km offshore). Stohl et al. (2002) calculated that 9-22% of surface NO_y emissions is

234   exported out of the boundary layer of NA. Our result is about 30%, higher than Li's and Stohl's

235   results. HTAP I study by Sanderson et al. (2008) developed a SR relationship for NO_y deposition

236   among NA, EU, SA and EA. About 12-24% of the emitted NO_x is deposited out of source

237   regions and the corresponding percentage in this study is 26-34%. It should be noted that the

238   estimations of different studies are influenced by several factors and the results are not fully

239   comparable. (1) Definition of boundaries of source and receptor regions. For instance, Li et al.

240   (2004) defined the boundary of NA by a squared domain: 65-130°W, 25-55°N, while we use the

241   continental boundaries defined by HTAP II. There are also changes in the definition of

242   boundaries between HTAP II and HTAP I. For instance, HTAP I includes Mexico and Central

243   America in NA, but they are defined as a separate region in HTAP II (region 12 in Fig. 1). The

244   boundary of EU is also changed in HTAP II. (2) HTAP I used the perturbation simulation that

245   only reduce the NO_x emission, but HTAP II simulations also reduce other anthropogenic

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246 emissions, including SO_2 and PM. The joint control of multiple emissions may cause more

247 reduction in NO_y deposition and it is hard to estimate this effect in this study.

3.2 Impact of hemispheric transport on deposition

249 This section investigates the impact of hemispheric transport on deposition in the receptor

250 regions. Fig. 2 is annual response of S deposition to 20% emission reduction in source regions,

251 calculated as (Δ Depo under perturbation case) / (deposition in base case) ×100%. The negative

252 values mean that the corresponding deposition decreases with reduction in emission. Table S2

253 summarizes the regional median deposition fluxes under base case and under emission

254 perturbation in source regions. Fig. 2(a) shows the global response of deposition to 20%

255 emission reduction in NA. The largest changes happen in the source region NA, with a 5-20%

256 decrease in S deposition in the non-coastal region and 15-20% decrease at the east coast. The

257 impact on the North Atlantic Ocean deposition declines gradually from near coastal region (12-

258 15%) to open ocean (5-12%) and Eurasia (<1%). Fig. 2(b) shows the global response of

259 deposition to 20% emission reduction in EU. Although the impact on continental non-coastal

260 regions is high (12-20%), the impact on the coastal regions is within 5%, much lower than NA's

261 impact on its east coast (15-20%). The deposition in North Africa, central Asia and western RU

262 is affected by 2-5%. 20% emission reduction in SA (Fig. 2(c)) shows large influence over its

263 south-west coast and the Arabian Sea (5-12%). The SA's outflow affects the deposition in

264 southeastern ME and eastern Sub Saharan Africa by 1-5% and western EA and Southeast Asia

265 (Bangladesh) by 2-10%. Fig. 2(d) shows the impact of 20% emission reduction on deposition in

266 EA. On one hand, the impact is strong on the east coast of China (12-15%) and decreases

267 gradually over the North Pacific Ocean (5-15%). Although the majority of S emission is

268 deposited on the Pacific Ocean, the influence on western NA can still reach 5-8%. On the other

269 hand, the impact on Southeast Asia and South Asia is much lower (2-5% and <1%), due to the

270 block of air flows by the Himalaya Mountains (Fig. S4). 20% emission decrease in ME mainly

271 affects the S deposition in Africa by 2-10% and western EA by 2-5%. Fig. 2(f) shows the S

272 deposition change with 20% emission reduction in RU. The regions of impact are mainly at high

273 latitudes, including northern EU (2-5%) and western Arctic Circle (1-5%).

274   The impact of NO_x emission reduction on NO_y deposition from each source region is

275 shown in Fig 3. The overall impact is qualitatively similar to that of S emission with some

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276   differences. For some regions, the impact of hemispheric transport on NO_y deposition is lower

277   than that on S deposition. For instance, SA's impact on eastern Africa is about 2-5% on S

278   deposition, but is <1% on NO_y deposition. ME's impact on the east coast of Africa and Gulf of

279   Guinea is about 2-5% on S deposition, but is <1% on NO_y deposition. These smaller sensitivities

280   reflect differences in lifetimes, and the lower formation of aerosol nitrate under warm conditions

281   in tropical regions. Under the NA perturbation case (Fig. 3(a)), a 2-12% change of NO_y

282   deposition is found on the west coast of California, due to high NO_x emission in California from

283   mobile source, which is not seen in S deposition. The impact of emission reduction in EU and

284   EA on their coastal regions is generally 2-5% higher for NO_y deposition than S deposition (Fig.

285   3(b) and (d)). The impact on NH_x deposition is similar to NO_y deposition (Fig. S2). It should be

286   noted that this is the result from 20% emission reduction in the source regions, therefore the

287   actual impact (100% emission reduction) could be 5 times higher when assuming a linear

288   relationship between 20 and 100% emission reduction on deposition.

289   We quantify the amount of deposition carried by hemispheric transport and study its

290   seasonality. Fig. 4 shows the monthly changes of S deposition for 20% emission reductions in

291   source regions. The values are meridional sum with a west-east resolution of 0.1 degree, and

292   display well the locations of the source regions. The negative values indicate the amounts of

293   pollutants transported from source regions to receptor regions. According to Fig 4(a), NA has

294   about $(1\text{-}10) \times 10^4$ kg(S) month^{-1} per 0.1° longitude of its S emission transported and deposited

295   over the North Atlantic Ocean (15-75°W) throughout the whole year. We also find about (1-3)

296   $\times 10^4$ kg(S) month^{-1} per 0.1° longitude decrease of S deposition at about 90°E and 120°E in

297   spring and fall, which gives evidence to NA's influence on Eurasia via transatlantic flow,

298   although this amount accounts for less than 1% of local S deposition (white in Eurasia in

299   Fig.2(a)). Fig. 4(b) shows that about $(1\text{-}3) \times 10^4$ kg(S) month^{-1} per 0.1° longitude of EU's

300   emission is transported and deposited at 30-60°E in RU throughout the whole year and at 100-

301   120°E in EA in spring and fall. According to Fig. 4(c), SA exports its S emission to 30-60°E in

302   ME and eastern Africa in early spring and to 90°E-180° in EA and North Pacific Ocean from late

303   spring until fall. In particular, the influence on EA can reach $(5\text{-}10) \times 10^4$ kg(S) month^{-1} per 0.1°

304   longitude in mid-spring. According to Fig. 4(d), EA's S emission is widely transported and

305   deposited over the North Pacific Ocean throughout the whole year. The Asian outflow arrives at

306   the west coast of NA (~130°W) in all seasons except summer, but only reaches far in western

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307   NA (~90°W) in spring and brings about 1×10^4 kg(S) month^{-1} per 0.1° longitude of S deposition.

308   The monthly changes of NO_y deposition with perturbation experiments are shown in Fig. 5.

309   Compared to S deposition, the change in NO_y deposition by hemispheric transport is generally

310   smaller. For instance, the NA's impact on Eurasia is $(1-3) \times 10^4$ kg(S) month^{-1} per 0.1° longitude

311   for S deposition, but is less than 0.5×10^4 kg(N) month^{-1} per 0.1° longitude for NO_y deposition.

312   The SA's impact on EA (90-120°E) can reach $(5-10) \times 10^4$ kg(S) month^{-1} per 0.1° longitude for S

313   deposition, but the amount is 4 times lower for NO_y deposition. This result is in accordance with

314   the S-R results in section 3.1 that more S emission is transported out of the source regions than N

315   emission, due to higher emission altitudes and longer chemical lifetimes. Patterns similar to NO_y

316   are also found in the monthly changes of NH_x deposition (Fig. S3).

317   The deposition change via transport between neighboring regions is found throughout the

318   whole year and is slightly stronger in winter, such as between EU and RU (~30°E) (Fig. 4(b) and

319   (f)) and from EA to the North Pacific Ocean (~130°E) (Fig. 4(d)). This is consistent with the

320   seasonality we found for the export of emission by source regions in section 3.1. In addition,

321   most source regions reduce more S and NO_x emissions in winter than the other seasons (Table

322   S3), thus more emissions are exported abroad in winter. On the contrary, most of the deposition

323   change by transport over long distance occurs in spring and fall, especially for the hemispheric

324   transport from NA to EU, from EU to EA and from EA to NA. This agrees with the seasonality

325   of the transpacific, transatlantic and trans-Eurasia flows of air pollutants (Holzer et al., 2005;Liu

326   et al., 2005;Liang et al., 2004;Brown-Steiner and Hess, 2011;Li et al., 2014;Auvray and Bey,

327   2005;Wild et al., 2004;Liu et al., 2003). The long distance transport of emissions is low in winter,

328   due to that the formation of secondary species like PAN is suppressed due to slow oxidation

329   (Berntsen et al., 1999;Deolal et al., 2013;Moxim et al., 1996), which plays an important role as a

330   reservoir for NO_x in the long-range transport of air pollution (Lin et al., 2010;Hudman et al.,

331   2004).

332   **3.3 Own region and foreign contributions on deposition**

333   This section compares the contributions of hemispheric transport and own region emission

334   control on deposition. The second metric is the response to extra-regional emission reduction

335   (RERER) as shown in Table 2. It is calculated by dividing the Δ Depo due to foreign emission

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336   reduction by Δ Depo due to global (foreign + own region) emission control following equation

337   (2).

338    $$RERER_i = \frac{\Delta\,Depo_i\,(foreign)}{\Delta\,Depo_i\,(global)} \qquad (2)$$

339   where i is the region of focus. Δ Depo$_i$ (foreign) is the Δ Depo in region i due to 20% foreign

340   emission reduction. It is calculated by subtracting the Δ Depo due to 20% own region emission

341   change from Δ Depo due to 20% global emission change. Δ Depo$_i$ (global) is the Δ Depo in

342   region i due to 20% global emission reduction. This metric compares the contributions from

343   foreign emission reduction with own region emission control on local deposition. A low RERER

344   value (close to 0) indicates a predominance impact of own region emission on local deposition,

345   while high RERER value (close to 1) means strong impact of hemispheric transport on local

346   deposition.

347   The total column includes both non-coastal and coastal regions. As we expected, NA

348   (0.07-0.17), SA (0.04-0.18) and EA (0.16) regions have relatively low RERER values, due to

349   large emissions and deposition in those regions compared to the foreign contributions. EU (0.12-

350   0.36) and ME (0.32-0.42) have relatively higher RERER values. RU is the only region with

351   RERER (0.55-0.61) higher than 0.5, which means its deposition is almost equally sensitive to

352   foreign impact and own region control. The RERER values of S deposition and NO$_y$ deposition

353   are of similar magnitudes, while the RERER of NH$_x$ deposition is 0.1 lower, due to the lack of

354   contribution from EA.

355   For non-coastal regions, the own region impact includes control of both its coastal and

356   non-coastal regions. The foreign impact comes from emission reduction of foreign coastal and

357   non-coastal regions. The RERER values of coastal regions are generally 0.1-0.3 higher than

358   those of non-coastal regions. In particular, the values for RU's coast are all higher than 0.84.

359   Even regions with low total RERER such as NA and SA have high RERER on coastal regions.

360   For instance, the RERER of NA reached 0.3-0.4 for its coastal region, more than double of the

361   RERER on its non-coastal regions (0.05-0.12). Coastal regions receive high proportion of

362   deposition from foreign transport. According to table 1, EA exports 5% of its S and N emission

363   to RU, almost half of which is deposited on RU's coastal regions. RU exports 7-12% of S and N

364   emission to EA, of which 30% is deposited on EU's coastal regions. The impact of hemispheric

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365   transport is identical or even larger than the effect of own region emission control for some

366   coastal or near coastal regions. According to Fig. 2, 20% emission reduction in EA can reduce 2-

367   5% of S deposition in the west coast of NA. This effect is even larger than 20% emission

368   reduction in NA (<1%). Similarly, 20% emission reduction in NA can change 2-5% of S

369   deposition in west coast of EU, which is almost identical to the effect of 20% emission control in

370   EU. On one hand, the emissions in western NA and western EU are relatively low, thus the effect

371   of own region control is not significant. On the other hand, these coastal regions are in the

372   downwind location of eastern EA and eastern NA, which are the main source regions of S and N

373   emissions. Coastal regions serve as transit places for air-sea exchange with vulnerable ecosystem

374   (Jickells, 2006;Jickells et al., 2017). The over-richness of deposition in coastal water and

375   ecosystem can evoke a number of environmental issues, of which some are specifically for

376   coastal regions such as threats to coastal benthic and planktonic system and sustainability of

377   fishery (Paerl, 2002;Doney et al., 2007).

378   Figure 6 compares the foreign and own region contributions on own region deposition.

379   Other (OTH, pattern fill in the figure) is calculated as $\Delta \text{Depo}_{(GLO)} - \sum \Delta \text{Depo}_{(case)}$ (case = 6,

380   including NA, EU, SA, EA, ME and RU). It indicates the deposition change in the receptor

381   region due to other reasons than the sum of separate emission reduction in the 6 regions. For

382   regions with low RERER values (NA, SA and EA), the own region emission dominates the

383   deposition by more than 80%. For these regions, determined by vicinity and transport patterns,

384   the foreign impact is somewhat dominated by certain source regions, such as from EA to NA (2-

385   4% out of 4-5%), from ME to SA (5-6% out of 7-11%) and from SA to EA (3-4% out of 4-7%).

386   For EU and ME, there is about 20% contribution from "OTH". It could come from the emission

387   reduction in rest of world, especially nearby regions such as Central Asia and North Africa. It

388   could also come from the joint effects of emission control by multiple source regions. However,

389   the model simulations do not allow to separate these two contributions in this study. Beside this,

390   RU contributes 4-5% to EU's deposition and EU contributes 5% to ME's deposition. For high

391   RERER regions, RU has a different pattern than the other regions. The contributions of

392   hemispheric transport from other 5 regions (23-45%) are almost equivalent to its own region

393   emission control (39-45%). There are significant contributions from EA (20-24%) and EU (13-

394   15%), which is reasonable since RU emits low S and N emissions, but is located close to these

395   two major source regions.

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396   Fig. 7 shows the inter-model variation on simulating the changes of deposition of SO_x,

397   NO_y, and NH_x under emission perturbation cases, separate for wet and dry deposition. The

398   values are global integrated changes in component deposition for perturbation experiments from

399   MMM results with error bars showing the maximum and minimum values of models. The figure

400   only shows main compositions of S and N deposition, which together account for more than 95%

401   of total deposition. In terms of S deposition (Fig. 7(a)), the differences between the maximum

402   and minimum values of models for different perturbation cases range from ±(0.03-0.13) Tg(S)

403   (±29-83% of MMM), ±(0.00-0.11) Tg(S) (±39-78%), ±(0.00-0.01) Tg(S) (±21-57%) and ±(0.01-

404   0.09) Tg(S) (±11-51%) for SO_2 dry and wet deposition and SO_4^{2-} dry and wet deposition,

405   respectively. High uncertainty is found in EA perturbation case, where the model divergence are

406   mainly found on SO_2 wet and dry deposition and SO_4^{2-} wet deposition. In terms of NO_y

407   deposition (Fig. 5(b)), the differences among cases are about ±(0.00-0.05) Tg(N) (±16-39%),

408   ±(0.02-0.28) Tg(N) (±23-70%) and ±(0.02-0.47) Tg(N) (±15-45%) for NO_2 dry deposition and

409   NO_3^- dry and wet deposition, respectively. The EA case also has the largest inter-model

410   variation, with high uncertainty in simulating both the NO_3^- wet and dry deposition. In terms of

411   NH_x deposition (Fig. 5(c)), the differences are about ±(0.00-0.06) Tg(N) (±2-27%), ±(0.00-0.08)

412   Tg(N) (±10-55%) and ±(0.01-0.06) Tg(N) (±5%) for NH_3 dry deposition and NH_4^+ dry and wet

413   deposition, respectively. Both EA and SA have relatively high uncertainties on NH_4^+ dry

414   deposition. Overall, the inter-model variation is considerably high for the deposition change

415   under EA emission perturbation. On one hand, the EA perturbation case assumes the largest

416   amount of emission reductions among all cases (Table S3). On the other hand, model evaluation

417   (Tan et al., 2018) also has reported high model bias in simulating the deposition in this region,

418   and suggest an incomplete knowledge from the combined picture provided by observations and

419   models.

420   **4 Conclusion**

421   This study assesses the impact of hemispheric transport on S and N deposition for 6 regions:

422   North America, Europe, South Asia, East Asia, Russia and Middle East, by using multi-model

423   ensemble results from 11 models of HTAP II project, with simulations under base case and 20%

424   emission reduction scenarios.

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425   We investigate the export of S and N emissions by source regions. Results show that

426   about 27-58%, 26-46% and 12-23% of the emitted S, NO_x and NH_3 emissions are deposited

427   outside of the source regions. The most significant exports of emissions are from Europe to

428   Russia (10-14%), from South Asia to East Asia (4-9%), from East Asia to Russia (5%) and from

429   Russia to Europe (7-12%) and East Asia (4-5%). Most regions export 5-10% more emission to

430   abroad in winter than summer, which is highly influenced by chemistry, precipitation amount

431   and frequency, atmospheric mixing and transport patterns.

432   We explore the impact of hemispheric transport on deposition in receptor regions.

433   Overall, 20% emission reduction in source regions could affect 1-10% of deposition on foreign

434   continental regions and 1-15% on foreign coastal regions and open ocean, especially from North

435   America to the North Atlantic Ocean (5-15%), from South Asia to western East Asia (2-10%)

436   and from East Asia to North Pacific Ocean (5-15%) and western North America (5-8%). The

437   amounts of deposition brought by hemispheric transport range from 10^4-10^5 kg(S or N) month^{-1}

438   per 0.1° longitude (meridional sum). The impact on deposition via transport between

439   neighbouring regions (i.e. Europe and Russia) is generally found throughout the whole year and

440   slightly stronger in winter, while that via transport over long distance (i.e. from East Asia to

441   North America) mainly takes place in spring and fall.

442   We compare the own region and foreign impact on deposition. The deposition in North

443   America, South Asia and East Asia is dominated (~80%) by their own region emission, while

444   Europe, Middle East and Russia receive 40-60% of impact from hemispheric transport. In

445   particular, Russian deposition is even equally sensitive to foreign inputs and own region

446   emission, with high contributions from two neighbouring source regions: East Asia (~20%) and

447   Europe (~15%). Coastal regions receive upmost half of the hemispheric transport from foreign

448   regions. Deposition in coastal regions or near-coastal open ocean is found twice more sensitive

449   to long-range transport than non-coastal regions. For some coastal regions such as west coast of

450   North America and west coast of Europe, the impact of hemispheric transport is identical or even

451   larger than that of own region emission control.

452   This study highlights the impact of hemispheric transport on deposition in coastal regions

453   and open ocean, which hasn't been fully studied in the literature. We therefore suggest further

454   research on this impact on the mitigation of coastal and oceanic ecosystem, with regards to the

455   increasing concentration of air pollutants in hemispheric outflow. We also find significant impact

456 of hemispheric transport on deposition in relatively low emission regions such as Russia. The

457 impact on their ecosystem and human health requires further research. Meanwhile, there is still a

458 portion of foreign impact that hasn't been attributed in this study (aggregated as other regions

459 "OTH" in Fig. 6). Some regions are not included in perturbation experiments, but are found with

460 large impact by/to other regions. For instance, at least 4 regions (North America, Europe, South

461 Asia and Middle East) have shown considerable impact (2-10%) on the S and N deposition in

462 North Africa. The impact from/by Southeast Asia is also unknown, which is regarded as a big

463 contributor of global S and N emissions in Asia. We suggest the future HTAP simulations to

464 include these regions in the perturbation experiments.

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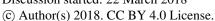and Physics
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742   Caption:

743   Fig. 1. Administration boundaries of regions and coastal areas. 6 Regions with perturbation
744   experiments: 3-North America (NA), 4-Europe (EU), 5-South Asia (SA), 6-East Asia (EA), 11-
745   Middle East (MD) and 14-Russia, Belarussia, Ukraine (RU). Other regions: 1-Global, 2-Ocean
746   (including Arctic), 7-Southeast Asia, 8-Australia, 9-North Africa, 10- Sub Saharan Africa, 12-
747   Mexico, Central America, Caribbean, Guyanas, Venezuela, Columbia (Central America), 13-
748   South America, 15-Central Asia and 17-Antarctic.

749   Fig. 2 The response of S deposition to 20% emission reduction in source regions. The values are
750   the percentage changes (%) in deposition calculated as (changes in deposition with 20%
751   emission reduction) / (base case deposition) ×100%. The unit is %.

752   Fig.3 Same as Fig.2 but for NO_y deposition. The unit is %.

753   Fig. 4 The monthly changes of S deposition with 20% emission reduction in source regions. The
754   values are meridional total values versus time with a west-east resolution of 0.1 degree. The unit
755   is ×10^4 kg(S) month^{-1} per 0.1° longitude. The negative values indicate decline in deposition with
756   reduction in emission.

757   Fig. 5 Same as Fig.4 but for NO_y deposition. The unit is ×10^4 kg(N) month^{-1} per 0.1° longitude.

758   Fig. 6 Own region and foreign contributions on own region deposition. The values are calculated
759   by changes with 20% emission reduction. Other (OTH, pattern fill) is the contribution by other
760   reasons than emission reduction in the 6 regions (see text for details).

761   Fig. 7 Inter-model variations in deposition changes (unit: Tg(S or N) yr^{-1}) under emission
762   perturbation experiments. The values are MMM with error bars showing the max and min values
763   among all models.
764

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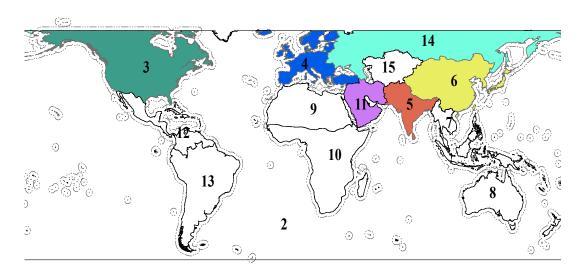Discussions

765 **Fig. 1**

766
767 Fig. 1. Boundaries of regions and coastal areas (dashed). 6 Regions with perturbation
768 experiments: 3-North America (NA), 4-Europe (EU), 5-South Asia (SA), 6-East Asia (EA), 11-
769 Middle East (MD) and 14-Russia, Belarussia, Ukraine (RU). Other regions: 1-Global, 2-Ocean
770 (including Arctic), 7-Southeast Asia, 8-Australia, 9-North Africa, 10- Sub Saharan Africa, 12-
771 Mexico, Central America, Caribbean, Guyanas, Venezuela, Columbia (Central America), 13-
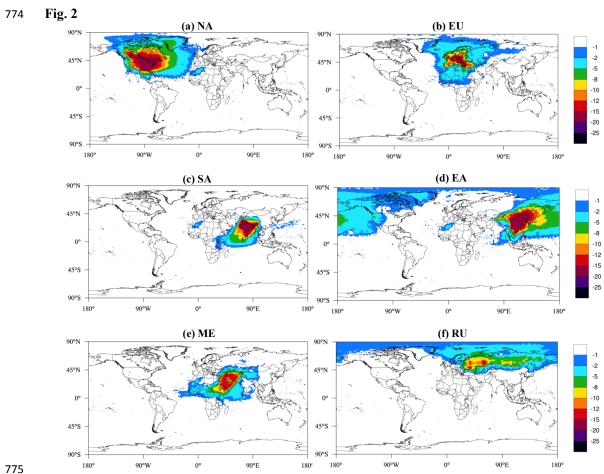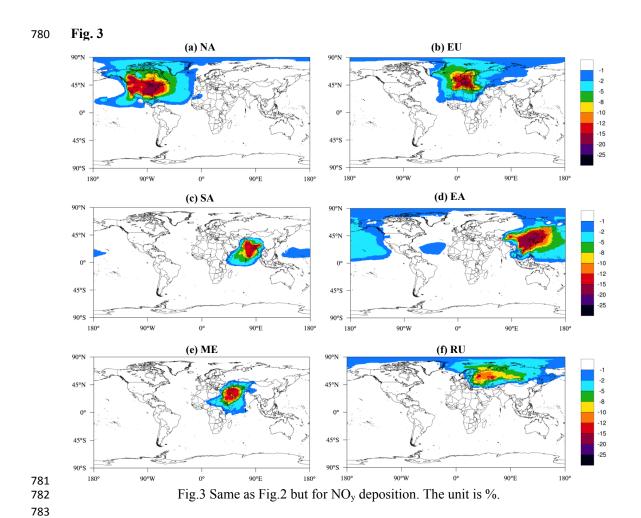772 South America, 15-Central Asia and 17-Antarctic.
773

774 **Fig. 2**

Fig. 2 The response of S deposition to 20% emission reduction in source regions. The values are
the percentage changes (%) in deposition calculated as (changes in deposition with 20%
emission reduction) / (base case deposition) ×100%. The unit is %.

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780   **Fig. 3**

782   Fig.3 Same as Fig.2 but for NO$_y$ deposition. The unit is %.
783

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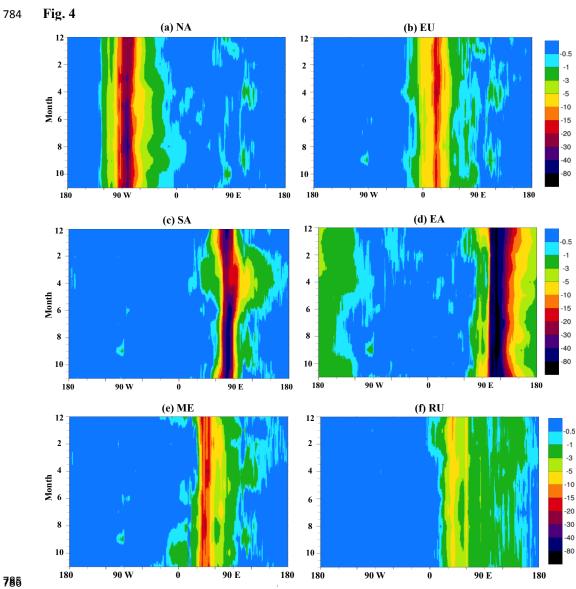Discussions

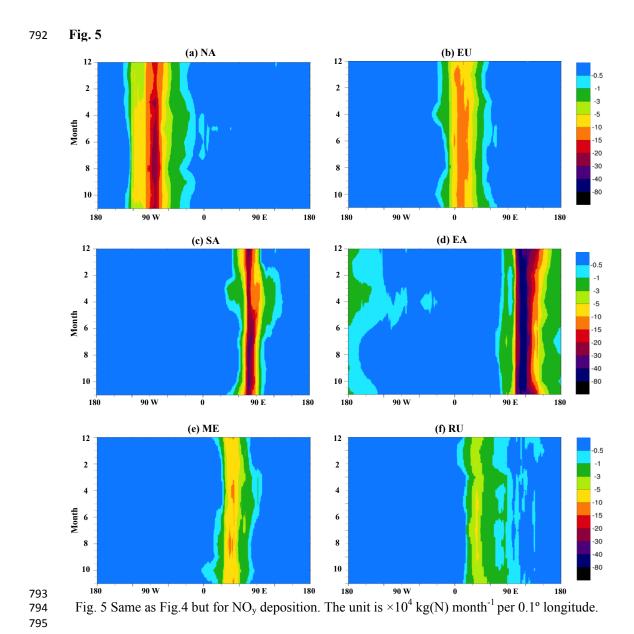784   **Fig. 4**

785
786
787   Fig. 4 The monthly changes of S deposition with 20% emission reduction in source regions. The
788   x-axis values are meridional total values versus time (y-axis) with a west-east resolution of 0.1
789   degree. The unit is $\times 10^4$ kg(S) month^{-1} per 0.1° longitude. Negative values indicate decline in
790   deposition with reduction in emission.
791

792   **Fig. 5**

793
794   Fig. 5 Same as Fig.4 but for NO$_y$ deposition. The unit is $\times 10^4$ kg(N) month^{-1} per 0.1° longitude.
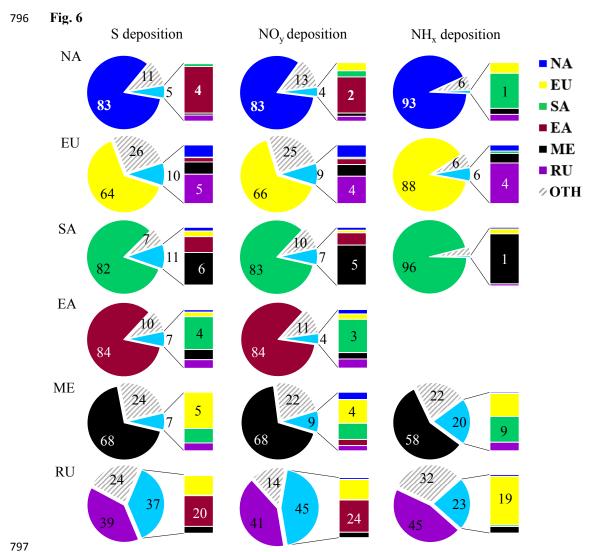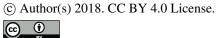795

Fig. 6

Fig. 6 Own region and foreign contributions on own region deposition. The values are calculated by changes with 20% emission reduction. Other (OTH, pattern fill) is the contribution by other reasons than emission reduction in the 6 regions (see text for details).

802   **Fig. 7**

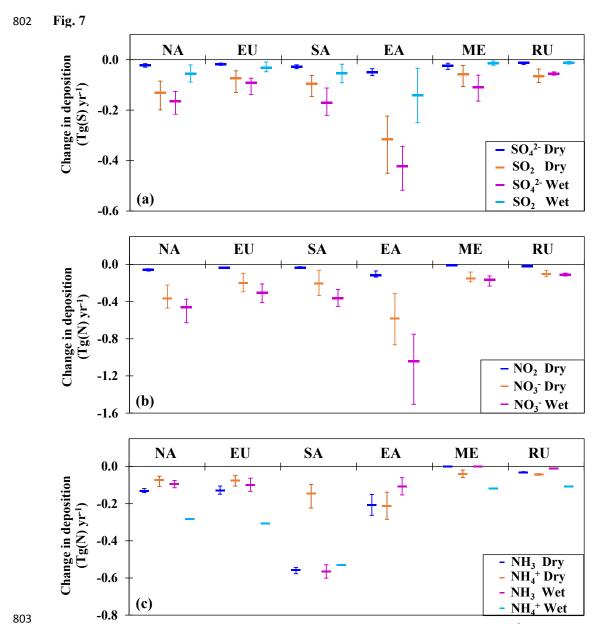

803
804   Fig. 7 Inter-model variations in wet and dry deposition changes (unit: Tg(S or N) yr^{-1}) under
805   emission perturbation experiments. The values are global integrated changes in components of S
806   and N deposition for perturbation experiments from MMM results with error bars showing the
807   max and min values among all models.
808

809   Tables

810   Table 1. Source-receptor relationship of S/NO_y/NH_x deposition (%) for regions (including
811   continental coastal and non-coastal regions). The values in the parentheses are for coastal regions
812   as a subset of the total.

	Receptor Regions	Source Regions					
		NA	EU	SA	EA	ME	RU
S Deposition	NA	68.9 (8.9)	0.2 (0.1)	0.2 (0.0)	1.5 (0.6)	0.3 (0.1)	1.2 (0.6)
	EU	1.1 (0.6)	60.4 (14.4)	0.0 (0.0)	0.2 (0.1)	2.1 (0.2)	6.9 (2.5)
	SA	0.5 (0.1)	1.2 (0.3)	66.4 (10.0)	0.9 (0.4)	7.9 (1.6)	0.3 (0.1)
	EA	0.6 (0.2)	1.8 (0.4)	8.8 (1.3)	73.4 (11.5)	4.6 (0.8)	5.2 (1.4)
	ME	0.0 (0.0)	2.6 (0.6)	0.6 (0.3)	0.0 (0.0)	42.4 (8.2)	0.8 (0.2)
	RU	0.4 (0.1)	13.6 (2.2)	0.1 (0.1)	5.1 (2.2)	5.0 (1.1)	62.2 (4.4)
	Others	28.5	20.1	23.8	19.1	37.7	23.4
NO_y Deposition	NA	71.5 (7.8)	0.8 (0.2)	0.5 (0.1)	1.0 (0.3)	0.5 (0.1)	1.0 (0.3)
	EU	1.3 (0.6)	66.2 (17.5)	0.2 (0.1)	0.3 (0.1)	3.5 (0.9)	9.8 (2.9)
	SA	0.2 (0.0)	0.2 (0.0)	66.2 (8.6)	0.5 (0.2)	7.9 (1.3)	0.2 (0.0)
	EA	0.6 (0.1)	1.2 (0.2)	6.2 (0.7)	74.4 (14.3)	2.4 (0.3)	4.3 (0.9)
	ME	0.4 (0.1)	1.6 (0.3)	0.9 (0.4)	0.1 (0.0)	54.4 (8.0)	0.8 (0.2)
	RU	0.6 (0.1)	10.3 (1.3)	0.1 (0.0)	5.1 (2.2)	4.9 (1.3)	61.4 (3.1)
	Others	25.6	19.7	25.8	18.6	26.4	22.5
NH_x Deposition	NA	88.4 (5.6)	0.2 (0.1)	0.3 (0.1)	-*	0.7 (0.3)	0.4 (0.2)
	EU	0.6 (0.3)	83.2 (17.8)	0.0 (0.0)	-	4.6 (1.2)	11.9 (3.1)
	SA	0.0 (0.0)	0.1 (0.0)	85.1 (7.6)	-	8.6 (2.4)	0.0 (0.0)
	EA	0.0 (0.0)	0.4 (0.1)	4.2 (0.3)	-	2.6 (0.5)	3.8 (1.0)
	ME	0.1 (0.0)	1.3 (0.3)	0.4 (0.2)	-	49.4 (5.9)	1.5 (0.4)
	RU	0.4 (0.1)	10.3 (1.3)	0.1 (0.0)	-	7.3 (1.5)	76.9 (4.1)
	Others	10.5	4.4	9.7	-	26.9	5.7

813   * Lack of NH_4^+ wet deposition under EA emission perturbation experiment from all models.

814

815   Table 2. RERER values of S/NO_y/NH_x deposition for continent non-coastal and coastal regions.
816   Total column gives the RERER for coastal and non-coastal together.

Regions	S deposition			NO_y deposition			NH_x deposition		
	Total	Non-coastal	Coastal	Total	Non-coastal	Coastal	Total	Non-coastal	Coastal
NA	0.17	0.12	0.40	0.17	0.12	0.43	0.07	0.05	0.31
EU	0.36	0.27	0.53	0.34	0.27	0.48	0.12	0.09	0.22
SA	0.18	0.14	0.35	0.17	0.12	0.37	0.04	0.03	0.17
EA	0.16	0.14	0.28	0.16	0.12	0.27	-	-	-
ME	0.32	0.27	0.46	0.32	0.27	0.50	0.42	0.36	0.67
RU	0.61	0.56	0.84	0.59	0.52	0.90	0.55	0.49	0.85

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