

## MIT Open Access Articles

*Intercontinental influence of NO<sub>x</sub> and CO emissions on particulate matter air quality*

The MIT Faculty has made this article openly available. **Please share** how this access benefits you. Your story matters.

**Citation:** Leibensperger, Eric M., Loretta J. Mickley, Daniel J. Jacob, and Steven R.H. Barrett. "Intercontinental Influence of NO<sub>x</sub> and CO Emissions on Particulate Matter Air Quality." *Atmospheric Environment* 45, no. 19 (June 2011): 3318–3324.

**As Published:** <http://dx.doi.org/10.1016/j.atmosenv.2011.02.023>

**Publisher:** Elsevier

**Persistent URL:** <http://hdl.handle.net/1721.1/99449>

**Version:** Author's final manuscript: final author's manuscript post peer review, without publisher's formatting or copy editing

**Terms of use:** Creative Commons Attribution-Noncommercial-NoDerivatives



1 **Intercontinental influence of NO<sub>x</sub> and CO emissions on particulate matter air**  
2 **quality**

3  
4 **Eric M. Leibensperger (eleibens@fas.harvard.edu), Loretta J. Mickley, Daniel J. Jacob**  
5 *School of Engineering and Applied Sciences, Harvard University, Cambridge, MA USA*  
6 **Steven R. H. Barrett**  
7 *Department of Aeronautics and Astronautics, Massachusetts Institute of Technology, Cambridge,*  
8 *MA USA*

9  
10 **Submitted October 8, 2010**

11 **Revised January 31, 2011**

12  
13 **Corresponding Author:**

14 Eric Leibensperger  
15 110J Pierce Hall  
16 29 Oxford St.  
17 Cambridge, MA 02138  
18  
19 Phone: 1-617-384-7835  
20 Fax: 1-617-495-4551  
21 E-mail: eleibens@fas.harvard.edu  
22

23 **Abstract**

24 Anthropogenic emissions of nitrogen oxides ( $\text{NO}_x \equiv \text{NO} + \text{NO}_2$ ) and carbon monoxide (CO)  
25 affect particulate matter (PM) air quality on an intercontinental scale by changing background  
26 concentrations of oxidants (OH, ozone,  $\text{H}_2\text{O}_2$ ) and thus increasing the oxidation rate of sulfur  
27 dioxide ( $\text{SO}_2$ ) to sulfate and  $\text{NO}_x$  to nitrate. We conduct sensitivity simulations with the GEOS-  
28 Chem chemical transport model and find that these intercontinental influences of  $\text{NO}_x$  and CO  
29 emissions on PM can be greater than those from  $\text{SO}_2$  emissions (a direct PM precursor). The  
30 intercontinental impact of oxidant precursors is greatest in receptor regions with high domestic  
31  $\text{SO}_2$ ,  $\text{NO}_x$ , and ammonia emissions and hence already high levels of PM. US  $\text{NO}_x$  and CO  
32 emissions increase annual mean PM in northern Europe and eastern China by up to  $0.25 \mu\text{g m}^{-3}$ .  
33 The increase in Europe is mostly as sulfate, whereas in China it is mostly as nitrate. East Asian  
34  $\text{NO}_x$  and CO emissions have a weaker intercontinental influence ( $\sim 0.2 \mu\text{g m}^{-3}$  in northern Europe,  
35  $\sim 0.1 \mu\text{g m}^{-3}$  in the eastern US). These intercontinental effects of  $\text{NO}_x$  and CO emissions on PM  
36 depend in a complex way on the chemical environment of receptor regions. Intercomparison of  
37 results from different models would be of great interest.

38

39 **1. Introduction**

40 Developed countries worldwide regulate domestic sources of particulate matter (PM) to meet air  
41 quality goals designed to protect public health and visibility. These regulations may involve  
42 neighboring countries to address transboundary transport. However, PM can also be transported  
43 on intercontinental scales with significant implications for air quality in the receptor continent  
44 (Park et al., 2006; Liu et al., 2009a). Here we show that this intercontinental influence on PM  
45 does not only involve emissions of PM and its direct precursors from the source continent, but  
46 also emissions of nitrogen oxides ( $\text{NO}_x \equiv \text{NO} + \text{NO}_2$ ) and CO that affect PM through global  
47 perturbations to oxidant concentrations.

48  
49 There has been much recent interest in quantifying the intercontinental transport of PM pollution  
50 (Jaffe et al., 1999; Park et al., 2004; Heald et al., 2006; Chin et al., 2007; Liu et al., 2009b).

51 Intercontinental transport of desert dust and of plumes from large forest fires has long been  
52 recognized as a contributor to surface PM (Prospero, 1999; Forster et al., 2001; Husar et al.,  
53 2001; Fairlie et al., 2007). Surface, aircraft, and satellite observations have identified episodic  
54 transport of anthropogenic PM across the Pacific (Jaffe et al., 1999; Jaffe et al., 2003; Yu et al.,  
55 2008). Global models indicate annual mean sulfate enhancements in US surface air of up to 0.2  
56  $\mu\text{g m}^{-3}$  from anthropogenic Asian  $\text{SO}_2$  emissions (Park et al., 2004; Heald et al., 2006; Chin et al.,  
57 2007). Direct intercontinental transport of anthropogenic organic and nitrate aerosols appears to  
58 be far less important based on both observational and modeling evidence (Heald et al., 2006;  
59 Peltier et al., 2008; van Donkelaar et al., 2008).

60

61 Sulfate, nitrate, and organic aerosol form in the atmosphere by oxidation of their precursor gases  
62 SO<sub>2</sub>, NO<sub>x</sub>, and volatile organic compounds (VOCs). Oxidant levels affect the rate of aerosol  
63 production. Previous global model studies have found that global changes in anthropogenic  
64 emissions of oxidant precursors (NO<sub>x</sub>, CO, VOCs) affect surface PM concentrations and  
65 radiative forcing by perturbing background concentrations of the oxidants OH, H<sub>2</sub>O<sub>2</sub>, and ozone  
66 (Unger et al., 2006; Rae et al., 2007; Kloster et al., 2008; Shindell et al., 2008; Shindell et al.,  
67 2009). A recent study by Barrett et al. (2010) found that NO<sub>x</sub> emissions from aircraft at cruising  
68 altitudes enhance surface sulfate PM production by increasing background oxidant levels such  
69 that surface sulfate concentrations increase linearly with aircraft NO<sub>x</sub> emissions.

70

71 Current understanding of intercontinental pollution influence on PM air quality has been  
72 assessed by the Task Force on Hemispheric Transport of Air Pollutants (TF-HTAP, 2007) of the  
73 United Nations Environmental Program (UNEP). The assessment presents global multi-model  
74 estimates of intercontinental source-receptor relationships for PM and its precursors. These  
75 estimates can be made with either of two methods. In the first, PM or precursors from a given  
76 region are “tagged” and tracked as they undergo transport and chemical evolution. This method  
77 is applicable only for linear problems. In the second, more general method, sensitivity  
78 simulations with perturbed emissions in the source continent are compared to a control  
79 simulation. The TF-HTAP (2007) multi-model assessment used the second method but did not  
80 investigate the role of oxidant precursor emissions in contributing to intercontinental PM  
81 influences. We do so here.

82

83 **2. Model Simulations**

84 We conducted detailed simulations of coupled tropospheric ozone-NO<sub>x</sub>-VOC-aerosol chemistry  
85 with the GEOS-Chem chemical transport model (version 8-01-01; <http://geos-chem.org>) driven  
86 by assimilated meteorological data from the Goddard Earth Observing System (GEOS)-4. The  
87 model has a horizontal resolution of 2° latitude x 2.5° longitude and 30 vertical levels. GEOS-  
88 Chem simulates gas-phase oxidant chemistry together with the mass concentrations of the major  
89 aerosol types including sulfate-nitrate-ammonium (SNA), black carbon, primary organic carbon,  
90 secondary organic carbon, fine and coarse mode sea salt, and dust in four size classes (Park et al.,  
91 2003; Alexander et al., 2005; Park et al., 2006; Fairlie et al., 2007; Liao et al., 2007). Gas-phase  
92 and aerosol chemistry are coupled by in-cloud SO<sub>2</sub> oxidation, gas-aerosol thermodynamic  
93 partitioning of SNA and secondary organic aerosol (SOA), aerosol effects on photolysis rates  
94 (Martin et al., 2003), and heterogeneous chemistry (Jacob, 2000).

95  
96 The model forms sulfate aerosol from SO<sub>2</sub> through gas-phase oxidation by OH and in-cloud  
97 oxidation by H<sub>2</sub>O<sub>2</sub> and ozone (Park et al., 2004). The MARS-A aerosol thermodynamic  
98 equilibrium model is used to calculate SNA aerosol formation (Binkowski and Roselle, 2003).  
99 Nitric acid is formed by the gas phase reaction of NO<sub>2</sub> with OH and ozone, the latter leading to  
100 formation of N<sub>2</sub>O<sub>5</sub> which hydrolyzes to HNO<sub>3</sub> in aqueous aerosol (Evans and Jacob, 2005). SOA  
101 is formed by oxidation of VOCs by ozone and OH, following the work of Chung and Seinfeld  
102 (2002), as implemented in GEOS-Chem by Liao et al. (2007).

103  
104 Successive versions of GEOS-Chem have been extensively evaluated with surface, aircraft, and  
105 satellite observations of tropospheric oxidants, aerosols, and related species. Recent worldwide  
106 evaluations with satellite data are presented by Zhang et al. (2010) for ozone, Kopacz et al.

107 (2010) for CO, and van Donkelaar et al. (2010) for aerosol. Comparisons to OH and H<sub>2</sub>O<sub>2</sub>  
108 vertical profiles measured on aircraft missions have been presented by Hudman et al. (2007) and  
109 Mao et al. (2010). Air quality relevant evaluations with surface data for ozone and aerosols have  
110 been presented for North America and China (Choi et al., 2009; Wang et al., 2009). In addition, a  
111 number of GEOS-Chem studies have evaluated the model with observations specifically in the  
112 context of intercontinental influences on surface PM (Park et al., 2004; Heald et al., 2006; van  
113 Donkelaar et al., 2008) and surface ozone (Fiore et al., 2009; Zhang et al., 2009a). GEOS-Chem  
114 results contributed to the TF HTAP (2007) multi-model assessment were within the ranges of  
115 results from other models.

116  
117 We conducted a control simulation for year 2000 and six sensitivity simulations where we  
118 removed individually anthropogenic emissions of CO, NO<sub>x</sub>, and SO<sub>2</sub> from the contiguous US  
119 and East Asia. Anthropogenic emissions include fuel and industry, not open fires. We define  
120 East Asia as the emission inventory domain of Streets et al. (2003), which extends from Pakistan  
121 to Japan in the west-east direction and from Indonesia to Mongolia in the south-north direction.  
122 Each simulation used meteorological data for 2000-2001. The first year (2000) was used for  
123 model initialization and the second year (2001) for analysis. The model does not allow for  
124 aerosols or ozone to affect meteorology. Changes in PM concentrations are thus solely due to  
125 atmospheric chemistry.

126  
127 Anthropogenic emissions of NO<sub>x</sub> and SO<sub>2</sub> are from the EDGAR 3.2 FT inventory for 2000  
128 (Olivier and Berdowski, 2001). These include 4.8 Tg N a<sup>-1</sup> and 7.5 Tg S a<sup>-1</sup> from the contiguous  
129 US and 10 Tg N a<sup>-1</sup> and 27 Tg S a<sup>-1</sup> from East Asia. US anthropogenic emissions of CO are from

130 the EPA National Emissions Inventory 1999 (NEI99;  
131 <http://www.epa.gov/ttn/chief/net/1999inventory.html>) and amount to 82 Tg CO a<sup>-1</sup>. East Asian  
132 fossil anthropogenic emissions of CO are from Streets et al. (2006), and amount to 250 Tg CO a<sup>-1</sup>.  
133 <sup>1</sup>. Ammonia emissions are from Bouwman et al. (1997). Additional source information for the  
134 model is available from van Donkelaar et al. (2008).

135

### 136 **3. Intercontinental PM enhancements**

#### 137 **3.1 Effect of US emissions**

138 Figure 1 shows the annual mean PM enhancements in Europe and Asia from US anthropogenic  
139 sources of SO<sub>2</sub>, NO<sub>x</sub>, and CO as calculated by GEOS-Chem. PM enhancements are diagnosed as  
140 the differences in surface air concentrations of dry sulfate, nitrate, ammonium, and organic  
141 aerosol between the control simulation and a simulation with the corresponding emissions shut  
142 off. Figure 2 shows the SNA speciation and seasonality of this enhancement for receptor regions  
143 (outlined as boxes in Figure 1). Intercontinental enhancements in organic aerosol concentrations  
144 are small (< 5 ng m<sup>-3</sup>) and will not be discussed further.

145

146 The intercontinental effects of SO<sub>2</sub> emissions arise from the direct transport of SO<sub>2</sub> and sulfate,  
147 and decrease rapidly with distance downwind due to wet and dry removal. US SO<sub>2</sub> emissions  
148 thus mainly influence western Europe, by up to 0.2 μg m<sup>-3</sup> on an annual mean basis. This  
149 enhancement is comparable to the results of Park et al. (2004), but larger than those of Chin et al.  
150 (2007) and Liu et al. (2009b). Increases in sulfate are partly offset by decreases in nitrate due to  
151 competition for ammonium (West et al., 1999; Park et al., 2004). US SO<sub>2</sub> emissions actually



152 induce a net decrease in PM over eastern China in winter because the replacement of  $2\text{NH}_4\text{NO}_3$   
153 by  $(\text{NH}_4)_2\text{SO}_4$  results in net loss of PM mass.

154

155 We find that US  $\text{NO}_x$  emissions enhance European and Asian PM concentrations by up to  $0.25$   
156  $\mu\text{g m}^{-3}$  on an annual basis. The patterns in Figure 1 show that this is not due to direct  
157 intercontinental transport of nitrate and its precursors, but to an increase in background oxidant  
158 levels that promotes formation of sulfate and nitrate from local sources of  $\text{SO}_2$  and  $\text{NO}_x$  in the  
159 receptor regions. US  $\text{NO}_x$  emissions increase annual mean tropospheric ozone by 3.3%, OH by  
160 3.3%, and  $\text{H}_2\text{O}_2$  by 0.2% in the Northern Hemisphere, with larger effects in Europe, which is  
161 directly downwind.

162

163 Figure 2 shows that the intercontinental PM enhancement from US  $\text{NO}_x$  emissions is mostly  
164 driven by sulfate in northern Europe but by nitrate in eastern China. Conversion of  $\text{SO}_2$  to sulfate  
165 in the model is mostly by in-cloud oxidation by  $\text{H}_2\text{O}_2$  and ozone. The process by  $\text{H}_2\text{O}_2$  is faster  
166 than that by ozone, and in the presence of excess  $\text{H}_2\text{O}_2$ ,  $\text{SO}_2$  oxidation is insensitive to increases  
167 in oxidants. At the high latitudes of northern Europe,  $\text{H}_2\text{O}_2$  production is slow so that  $\text{SO}_2$  in-  
168 cloud oxidation is  $\text{H}_2\text{O}_2$ -limited over  $\text{SO}_2$  source regions for most of the year; thus increases of  
169 ozone and  $\text{H}_2\text{O}_2$  result in increased sulfate. The sulfate increase is largest in fall due to a  
170 combination of extensive cloud cover and significant enhancement of oxidants. Cloud cover is  
171 also extensive in winter but the increase in oxidants is then less. In eastern China, by contrast to  
172 northern Europe,  $\text{H}_2\text{O}_2$ -limited conditions prevail only in winter, and cloud cover is then  
173 infrequent due to the winter monsoon.

174

175 The large enhancement of nitrate PM in eastern China compared to northern Europe in Figure 2  
176 reflects the regional presence of excess ammonia for  $\text{NH}_4\text{NO}_3$  formation. We find that increases  
177 in total inorganic nitrate (gas-phase  $\text{HNO}_3$  plus aerosol nitrate) are comparable in the two  
178 regions, but northern Europe has less excess ammonia so that this inorganic nitrate remains in the  
179 gas phase as  $\text{HNO}_3$ . The effect of increased oxidants on nitrate PM in eastern China is largest in  
180 winter, when cold temperatures promote nitrate partitioning into the aerosol and when oxidation  
181 of  $\text{NO}_x$  is relatively slow. In summer, oxidation of  $\text{NO}_x$  by OH in the region is sufficiently fast  
182 that the effect of added oxidants is small. This is not the case for northern Europe where OH  
183 concentrations are much lower because of the higher latitude.

184

185 CO emissions in the US decrease annual mean tropospheric OH by 1.9% in the Northern  
186 Hemisphere, but increase ozone by 1.3% and  $\text{H}_2\text{O}_2$  by 3.4%. The effect on PM is qualitatively  
187 similar to that of US  $\text{NO}_x$  emissions, but not as large due to the decrease in OH concentrations.  
188 Speciation and seasonal patterns in Figure 2 are similar to those for the effect of US  $\text{NO}_x$   
189 emissions. The decrease in OH, affecting  $\text{NO}_x$  oxidation, results in some negative effects on  
190 nitrate in northern Europe. Nitrate concentrations still increase over China in seasons outside of  
191 summer, when oxidation of  $\text{NO}_x$  by ozone is a significant nitrate formation pathway (Dentener  
192 and Crutzen, 1993).

193

### 194 **3.2 Effect of Asian emissions**

195 Figure 3 shows the intercontinental increases in PM from Asian emissions. The intercontinental  
196 influence of Asian  $\text{SO}_2$  emissions are strongest in the western US where subsidence from the free  
197 troposphere brings Asian outflow to the surface. The enhancement of  $0.1\text{-}0.25 \mu\text{g m}^{-3}$  in this

198 region is comparable to the results of Park et al. (2004), Heald et al. (2006), and Chin et al.  
199 (2007). The effect on Europe is weaker and more uniform than that of US SO<sub>2</sub> emissions, which  
200 are closer upwind (Figure 1). Increases in sulfate cause lower levels of nitrate due to competition  
201 for ammonium, similar to the effects of US SO<sub>2</sub> emissions previously discussed.

202

203 The intercontinental influence of Asian NO<sub>x</sub> on PM shown in Figure 3 is weaker than that of US  
204 emissions (Figure 1). Asian NO<sub>x</sub> emissions increase annual mean tropospheric ozone by 8.8%,  
205 OH by 12.7%, and H<sub>2</sub>O<sub>2</sub> by 1.1%. These increases are much larger than those from US emissions  
206 (section 3.1), but the effect on PM is also contingent on emissions in the receptor continent. Thus  
207 the effect on the US is relatively weak, up to 0.1 μg m<sup>-3</sup> over the central US where excess  
208 ammonia from agricultural emissions promotes ammonium nitrate formation. The effect on  
209 northern Europe (up to 0.2 μg m<sup>-3</sup>) is also weaker than for US emissions, which are closer  
210 upwind (Figure 1). Figure 4 shows that sulfate accounts for most of the overall increase in PM  
211 over the eastern US and northern Europe. The increase is weakest in summer when Asian NO<sub>x</sub>  
212 emissions actually cause H<sub>2</sub>O<sub>2</sub> concentrations to decrease over the US and Europe. This reflects  
213 the large Asian source of NO<sub>x</sub> and complicated effects of NO<sub>x</sub> emissions on H<sub>2</sub>O<sub>2</sub>. On the one  
214 hand, NO<sub>x</sub> leads to ozone production and from there to H<sub>2</sub>O<sub>2</sub> production. On the other hand, NO<sub>x</sub>  
215 increases OH, which is a major H<sub>2</sub>O<sub>2</sub> sink.

216

217 Asian CO emissions decrease annual mean tropospheric OH by 5.6% and increase ozone by  
218 3.1% and H<sub>2</sub>O<sub>2</sub> by 9.4%. Figures 3 and 4 show that Asian CO affects northern European PM in a  
219 very similar manner to US CO emissions (Figures 1 and 2) and with a similar magnitude as  
220 Asian NO<sub>x</sub>. Similar to the effect of US CO emissions, Asian CO emissions increase in-cloud

221 sulfate production by H<sub>2</sub>O<sub>2</sub> over Europe causing sulfate enhancements in all seasons. Changes in  
222 sulfate in the US are largely driven by increased production by H<sub>2</sub>O<sub>2</sub>. A reduction in gas-phase  
223 production of sulfate by OH detracts from increased aqueous production. Nitrate concentrations  
224 decrease in both northern Europe and the US because of the lower OH concentrations.

225

#### 226 **4. Discussion**

227 Anthropogenic emissions of NO<sub>x</sub> and CO perturb sulfate and nitrate PM concentrations on  
228 intercontinental scales by affecting the background concentrations of oxidants and hence the  
229 oxidation rates of SO<sub>2</sub> and NO<sub>x</sub> emitted in receptor regions. NO<sub>x</sub> emissions cause OH, ozone,  
230 and H<sub>2</sub>O<sub>2</sub> to increase on the hemispheric scale although H<sub>2</sub>O<sub>2</sub> can decrease in some regions and  
231 seasons. CO emissions cause OH to decrease, but ozone and H<sub>2</sub>O<sub>2</sub> concentrations to increase.  
232 CO increases H<sub>2</sub>O<sub>2</sub> formation in part by decreasing OH, a major sink of H<sub>2</sub>O<sub>2</sub> and in part by  
233 decreasing the OH/HO<sub>2</sub> ratio and thus promoting H<sub>2</sub>O<sub>2</sub> formation. Most of the intercontinental  
234 effect on sulfate is through increases in ozone and H<sub>2</sub>O<sub>2</sub>, which drive faster SO<sub>2</sub> in-cloud  
235 oxidation under H<sub>2</sub>O<sub>2</sub>-limited conditions. Most of the effect of nitrate is through the increase in  
236 ozone in winter and the shoulder seasons, when NO<sub>x</sub> oxidation is otherwise slow and the cold  
237 temperatures promote nitrate fractionation into the aerosol.

238

239 The intercontinental PM enhancement from NO<sub>x</sub> and CO emissions tends to peak in receptor  
240 regions with the highest domestic sources of SO<sub>2</sub>, NO<sub>x</sub>, and NH<sub>3</sub>, and hence with the highest PM  
241 concentrations. It is thus of particular policy relevance. US emissions of NO<sub>x</sub> and CO increase  
242 PM by over 0.25 μg m<sup>-3</sup> (annual mean) in polluted regions of northern Europe and eastern China,  
243 a much larger effect than that of US SO<sub>2</sub> emissions. PM concentrations in the US are less

244 sensitive to intercontinental NO<sub>x</sub> and CO emissions because domestic PM sources are lower. The  
245 effect of Asian NO<sub>x</sub> and CO on PM in the eastern US is still ~ 0.1 μg m<sup>-3</sup>, comparable to the  
246 effect of Asian SO<sub>2</sub> emissions. Northern Europe is more sensitive to US than to Asian emissions  
247 of NO<sub>x</sub> and CO because of the greater upwind proximity.

248  
249 Our study used emission inventories for 2000. Emissions have changed over the past decade and  
250 also have some uncertainty. The more recent EPA NEI 2005 inventory for the US  
251 (<http://www.epa.gov/ttnchie1/net/2005inventory.html>) includes 2% more NO<sub>x</sub>, 20% less CO, and  
252 57% less SO<sub>2</sub> than used in this work, while the East Asian 2006 inventory of Zhang et al.  
253 (2009b) include 12% more NO<sub>x</sub>, 19% more CO, and 14% less SO<sub>2</sub>. These differences arise from  
254 both actual trends and improved emission accounting. The emission changes would tend to  
255 increase the importance of NO<sub>x</sub> and CO vs. SO<sub>2</sub> in driving intercontinental influence on PM.

256  
257 The intercontinental influence of NO<sub>x</sub> and CO emissions on PM through changes in background  
258 oxidant fields depends in a complex way on the chemical environment in the receptor region  
259 including (1) the relative importance of OH, ozone, and H<sub>2</sub>O<sub>2</sub> as oxidants for SO<sub>2</sub> and NO<sub>x</sub>; (2)  
260 cloudiness; (3) H<sub>2</sub>O<sub>2</sub> limitation of in-cloud SO<sub>2</sub> oxidation by H<sub>2</sub>O<sub>2</sub>; and (4) availability of  
261 ammonia for nitrate PM formation. All of these factors have substantial uncertainty in models,  
262 and the corresponding errors cannot easily be quantified using a single model. The  
263 intercontinental influence on oxidant levels is itself uncertain. The HTAP multi-model  
264 intercomparison of Fiore et al. (2009) shows good agreement between models for  
265 intercontinental influences on ozone, but no such evaluations have been conducted for OH and  
266 H<sub>2</sub>O<sub>2</sub>. In view of the potential for significant impacts presented in this paper, it would be of great

267 interest to carry out a multi-model intercomparison of intercontinental influences of NO<sub>x</sub> and CO  
268 emissions on PM.

269

270 **Acknowledgements**

271 This work was supported by the Electric Power Research Institute (EPRI) and an Environmental  
272 Protection Agency – Science to Achieve Results (EPA-STAR) Program Graduate Fellowship to  
273 Eric Leibensperger. EPRI and EPA have not officially reviewed or endorsed this publication and  
274 the views expressed herein may not reflect those of EPRI and EPA. This work has benefitted  
275 from discussions with Jenny Fisher, Christopher Holmes, Eloïse Marais, and Lin Zhang and  
276 useful comments from Naresh Kumar and Eladio Knipping.

277

278 **References**

- 279 Alexander, B., Park, R.J., Jacob, D.J., Li, Q., Yantosca, R.M., Savarino, J., Lee, C.C.W.,  
280 Thiemens, M.H., 2005. Sulfate formation in sea-salt aerosols: Constraints from oxygen isotopes.  
281 *J. Geophys. Res.* 110, D10307.  
282
- 283 Barrett, S.R.H., Britter, R.E., Waitz, I.A., 2010. Global Mortality Attributable to Aircraft Cruise  
284 Emissions. *Environ. Sci. Technol.* 44, 7736-7742.  
285
- 286 Binkowski, F., Roselle, S., 2003. Models-3 community multiscale air quality (CMAQ) model  
287 aerosol component - 1. Model description. *J. Geophys. Res.* 108, 4183.  
288
- 289 Bouwman, A., Lee, D., Asman, W., Dentener, F., Van Der Hoek, K., Olivier, J., 1997. A global  
290 high-resolution emission inventory for ammonia. *Global Biogeochem. Cy.* 11, 561-587.  
291
- 292 Chin, M., Diehl, T., Ginoux, P., Malm, W., 2007. Intercontinental transport of pollution and dust  
293 aerosols: implications for regional air quality. *Atmos. Chem. Phys.* 7, 5501-5517.  
294
- 295 Choi, Y.-S., Park, R., Ho, C.-H., 2009. Estimates of ground-level aerosol mass concentrations  
296 using a chemical transport model with Moderate Resolution Imaging Spectroradiometer  
297 (MODIS) aerosol observations over East Asia. *J. Geophys. Res.* 114, D04204.  
298
- 299 Chung, S., Seinfeld, J., 2002. Global distribution and climate forcing of carbonaceous aerosols. *J.*  
300 *Geophys. Res.* 107, 4407.  
301
- 302 Dentener, F.J., Crutzen, P.J., 1993. Reaction of N<sub>2</sub>O<sub>5</sub> on tropospheric aerosols: Impact on the  
303 global distributions of NO<sub>x</sub>, O<sub>3</sub>, and OH. *J. Geophys. Res.* 98, 7149-7163.  
304
- 305 Evans, M., Jacob, D., 2005. Impact of new laboratory studies of N<sub>2</sub>O<sub>5</sub> hydrolysis on global  
306 model budgets of tropospheric nitrogen oxides, ozone, and OH. *Geophys. Res. Lett.* 32, L09813.  
307
- 308 Fairlie, T.D., Jacob, D.J., Park, R.J., 2007. The impact of transpacific transport of mineral dust in  
309 the United States. *Atmos. Environ.* 41, 1251-1266.  
310
- 311 Fiore, A.M., Dentener, F.J., Wild, O., Cuvelier, C., Schultz, M.G., Hess, P., Textor, C., Schulz,  
312 M., Doherty, R.M., Horowitz, L.W., Mackenzie, I.A., Sanderson, M.G., Shindell, D.T.,  
313 Stevenson, D.S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I.,  
314 Carmichael, G., Collins, W.J., Duncan, B.N., Faluvegi, G., Folberth, G., Gauss, M., Gong, S.,  
315 Hauglustaine, D., Holloway, T., Isaksen, I.S.A., Jacob, D.J., Jonson, J.E., Kaminski, J.W.,  
316 Keating, T.J., Lupu, A., Marmer, E., Montanaro, V., Park, R.J., Pitari, G., Pringle, K.J., Pyle,  
317 J.A., Schroeder, S., Vivanco, M.G., Wind, P., Wojcik, G., Wu, S., Zuber, A., 2009. Multimodel  
318 estimates of intercontinental source-receptor relationships for ozone pollution. *J. Geophys. Res.*  
319 114, D04301.  
320
- 321 Forster, C., Wandinger, U., Wotawa, G., James, P., Mattis, I., Althausen, D., Simmonds, P.,  
322 O'Doherty, S., Jennings, S., Kleefeld, C., Schneider, J., Trickl, T., Kreipl, S., Jager, H., Stohl, A.,

323 2001. Transport of boreal forest fire emissions from Canada to Europe. *J. Geophys. Res.* 106,  
324 22887-22906.  
325  
326 Heald, C.L., Jacob, D.J., Park, R.J., Alexander, B., Fairlie, T.D., Yantosca, R.M., Chu, D.A.,  
327 2006. Transpacific transport of Asian anthropogenic aerosols and its impact on surface air  
328 quality in the United States. *J. Geophys. Res.* 111, D14310.  
329  
330 Hudman, R.C., Jacob, D.J., Turquety, S., Leibensperger, E.M., Murray, L.T., Wu, S., Gilliland,  
331 A.B., Avery, M., Bertram, T.H., Brune, W., Cohen, R.C., Dibb, J.E., Flocke, F.M., Fried, A.,  
332 Holloway, J., Neuman, J.A., Orville, R., Perring, A., Ren, X., Sachse, G.W., Singh, H.B.,  
333 Swanson, A., Wooldridge, P.J., 2007. Surface and lightning sources of nitrogen oxides over the  
334 United States: Magnitudes, chemical evolution, and outflow. *J. Geophys. Res.* 112, D12S05.  
335  
336 Husar, R., Tratt, D., Schichtel, B., Falke, S., Li, F., Jaffe, D., Gasso, S., Gill, T., Laulainen, N.,  
337 Lu, F., Reheis, M., Chun, Y., Westphal, D., Holben, B., Gueymard, C., McKendry, I., Kuring,  
338 N., Feldman, G., McClain, C., Frouin, R., Merrill, J., DuBois, D., Vignola, F., Murayama, T.,  
339 Nickovic, S., Wilson, W., Sassen, K., Sugimoto, N., Malm, W., 2001. Asian dust events of April  
340 1998. *J. Geophys. Res.* 106, 18317-18330.  
341  
342 Jacob, D., 2000. Heterogeneous chemistry and tropospheric ozone. *Atmos. Environ.* 34, 2131-  
343 2159.  
344  
345 Jaffe, D., Anderson, T., Covert, D., Kotchenruther, R., Trost, B., Danielson, J., Simpson, W.,  
346 Berntsen, T., Karlsdottir, S., Blake, D., Harris, J., Carmichael, G., Uno, I., 1999. Transport of  
347 Asian air pollution to North America. *Geophys. Res. Lett.* 26, 711-714.  
348  
349 Jaffe, D., McKendry, I., Anderson, T., Price, H., 2003. Six 'new' episodes of trans-Pacific  
350 transport of air pollutants. *Atmos. Environ.* 37, 391-404.  
351  
352 Kloster, S., Dentener, F., Feichter, J., Raes, F., van Aardenne, J., Roeckner, E., Lohmann, U.,  
353 Stier, P., Swart, R., 2008. Influence of future air pollution mitigation strategies on total aerosol  
354 radiative forcing. *Atmos. Chem. Phys.* 8, 6405-6437.  
355  
356 Kopacz, M., Jacob, D.J., Fisher, J.A., Logan, J.A., Zhang, L., Megretskaia, I.A., Yantosca, R.M.,  
357 Singh, K., Henze, D.K., Burrows, J.P., Buchwitz, M., Khlystova, I., McMillan, W.W., Gille,  
358 J.C., Edwards, D.P., Eldering, A., Thouret, V., Nedelec, P., 2010. Global estimates of CO  
359 sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS,  
360 SCIAMACHY, TES). *Atmos. Chem. Phys.* 10, 855-876.  
361  
362 Liao, H., Henze, D.K., Seinfeld, J.H., Wu, S., Mickley, L.J., 2007. Biogenic secondary organic  
363 aerosol over the United States: Comparison of climatological simulations with observations. *J.*  
364 *Geophys. Res.* 112, D06201.  
365  
366 Liu, J., Mauzerall, D.L., Horowitz, L.W., 2009a. Evaluating inter-continental transport of fine  
367 aerosols:(2) Global health impact. *Atmos. Environ.* 43, 4339-4347.  
368



369 Liu, J., Mauzerall, D.L., Horowitz, L.W., Ginoux, P., Fiore, A.M., 2009b. Evaluating inter-  
370 continental transport of fine aerosols: (1) Methodology, global aerosol distribution and optical  
371 depth. *Atmos. Environ.* 43, 4327-4338.  
372

373 Mao, J., Jacob, D.J., Evans, M.J., Olson, J.R., Ren, X., Brune, W.H., St. Clair, J.M., Crouse,  
374 J.D., Spencer, K.M., Beaver, M.R., Wennberg, P.O., Cubison, M.J., Jimenez, J.L., Fried, A.,  
375 Weibring, P., Walega, J.G., Hall, S.R., Weinheimer, A.J., Cohen, R.C., Chen, G., Crawford, J.H.,  
376 McNaughton, C., Clarke, A.D., Jaegle, L., Fisher, J.A., Yantosca, R.M., Le Sager, P., Carouge,  
377 C., 2010. Chemistry of hydrogen oxide radicals (HOx) in the Arctic troposphere in spring.  
378 *Atmos. Chem. Phys.* 10, 5823-5838.  
379

380 Martin, R., Jacob, D., Yantosca, R., Chin, M., Ginoux, P., 2003. Global and regional decreases in  
381 tropospheric oxidants from photochemical effects of aerosols. *J. Geophys. Res.* 108, 4097.  
382

383 Olivier, J.G.J., Berdowski, J.J.M., 2001. Global emissions sources and sinks, in: J. Berdowski et  
384 al. (Ed.), *The Climate System*. A. A. Balkema Publishers/Swets and Zeitliner Publishers, Lisse,  
385 Netherlands, pp. 33-78.  
386

387 Park, R., Jacob, D., Chin, M., Martin, R., 2003. Sources of carbonaceous aerosols over the  
388 United States and implications for natural visibility. *J. Geophys. Res.* 108, 4355.  
389

390 Park, R., Jacob, D., Field, B., Yantosca, R., Chin, M., 2004. Natural and transboundary pollution  
391 influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy. *J.*  
392 *Geophys. Res.* 109, D15204.  
393

394 Park, R., Jacob, D., Kumar, N., Yantosca, R., 2006. Regional visibility statistics in the United  
395 States: Natural and transboundary pollution influences, and implications for the Regional Haze  
396 Rule. *Atmos. Environ.* 40, 5405-5423.  
397

398 Peltier, R.E., Hecobian, A.H., Weber, R.J., Stohl, A., Atlas, E.L., Riemer, D.D., Blake, D.R.,  
399 Apel, E., Campos, T.L., Karl, T., 2008. Investigating the sources and atmospheric processing of  
400 fine particles from Asia and the Northwestern United States measured during INTEX B. *Atmos.*  
401 *Chem. Phys.* 8, 1835-1853.  
402

403 Prospero, J., 1999. Long-term measurements of the transport of African mineral dust to the  
404 southeastern United States: Implications for regional air quality. *J. Geophys. Res.* 104, 15917-  
405 15927.  
406

407 Rae, J.G.L., Johnson, C.E., Bellouin, N., Boucher, O., Haywood, J.M., Jones, A., 2007.  
408 Sensitivity of global sulphate aerosol production to changes in oxidant concentrations and  
409 climate. *J. Geophys. Res.* 112, D10312.  
410

411 Shindell, D., Lamarque, J.-F., Unger, N., Koch, D., Faluvegi, G., Bauer, S., Ammann, M.,  
412 Cofala, J., Teich, H., 2008. Climate forcing and air quality change due to regional emissions  
413 reductions by economic sector. *Atmos. Chem. Phys.* 8, 7101-7113.  
414

415 Shindell, D.T., Faluvegi, G., Koch, D.M., Schmidt, G.A., Unger, N., Bauer, S.E., 2009.  
416 Improved Attribution of Climate Forcing to Emissions. *Science* 326, 716-718.  
417  
418 Streets, D., Bond, T., Carmichael, G., Fernandes, S., Fu, Q., He, D., Klimont, Z., Nelson, S.,  
419 Tsai, N., Wang, M., Woo, J., Yarber, K., 2003. An inventory of gaseous and primary aerosol  
420 emissions in Asia in the year 2000. *J. Geophys. Res.* 108, 8809.  
421  
422 Streets, D.G., Zhang, Q., Wang, L., He, K., Hao, J., Wu, Y., Tang, Y., Carmichael, G.R., 2006.  
423 Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific  
424 (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations. *J.*  
425 *Geophys. Res.* 111, D14306.  
426  
427 Task Force on Hemispheric Transport of Air Pollutants, 2007. Hemispheric transport of air  
428 pollution 2007 interim report, in: Keating, T.J., Zuber, A. (Eds.), *Air Pollut. Stud.* 16. U.N. Econ.  
429 Comm. for Europe, New York.  
430  
431 Unger, N., Shindell, D., Koch, D., Streets, D., 2006. Cross influences of ozone and sulfate  
432 precursor emissions changes on air quality and climate. *P. Natl. Acad. Sci. USA* 103, 4377-4380.  
433  
434 van Donkelaar, A., Martin, R.V., Brauer, M., Kahn, R., Levy, R., Verduzco, C., Villeneuve, P.J.,  
435 2010. Global estimates of ambient fine particulate matter concentrations from satellite-based  
436 aerosol optical depth: Development and application. *Environ. Health Perspect.* 118, 847-855.  
437  
438 van Donkelaar, A., Martin, R.V., Leaitch, W.R., Macdonald, A.M., Walker, T.W., Streets, D.G.,  
439 Zhang, Q., Dunlea, E.J., Jimenez, J.L., Dibb, J.E., Huey, L.G., Weber, R., Andreae, M.O., 2008.  
440 Analysis of aircraft and satellite measurements from the Intercontinental Chemical Transport  
441 Experiment (INTEX-B) to quantify long-range transport of East Asian sulfur to Canada. *Atmos.*  
442 *Chem. Phys.* 8, 2999-3014.  
443  
444 Wang, Y., Hao, J., McElroy, M., Munger, J., Ma, H., Chen, D., Nielsen, C., 2009. Ozone air  
445 quality during the 2008 Beijing Olympics: effectiveness of emission restrictions. *Atmos Chem*  
446 *Phys* 9, 5237-5251.  
447  
448 West, J.J., Ansari, A.S., Pandis, S.N., 1999. Marginal PM<sub>2.5</sub>: Nonlinear aerosol mass response  
449 to sulfate reductions in the Eastern United States. *J. Air Waste Manage.* 49, 1415-1424.  
450  
451 Yu, H., Remer, L.A., Chin, M., Bian, H., Kleidman, R.G., Diehl, T., 2008. A satellite-based  
452 assessment of transpacific transport of pollution aerosol. *J. Geophys. Res.* 113, D14S12.  
453  
454 Zhang, L., Jacob, D., Liu, X., Logan, J., Chance, K., Eldering, A., Bojkov, B., 2010.  
455 Intercomparison methods for satellite measurements of atmospheric composition: application to  
456 tropospheric ozone from TES and OMI. *Atmos. Chem. Phys.* 10, 4725-4739.  
457  
458 Zhang, L., Jacob, D.J., Kopacz, M., Henze, D.K., Singh, K., Jaffe, D.A., 2009a. Intercontinental  
459 source attribution of ozone pollution at western U.S. sites using an adjoint method. *Geophys.*  
460 *Res. Lett.* 36, 1-5.

461  
462 Zhang, Q., Streets, D.G., Carmichael, G.R., He, K.B., Huo, H., Kannari, A., Klimont, Z., Park,  
463 I.S., Reddy, S., Fu, J.S., Chen, D., Duan, L., Lei, Y., Wang, L.T., Yao, Z.L., 2009b. Asian  
464 emissions in 2006 for the NASA INTEX-B mission. *Atmos. Chem. Phys.* 9, 5131-5153.

465  
466  
467

468

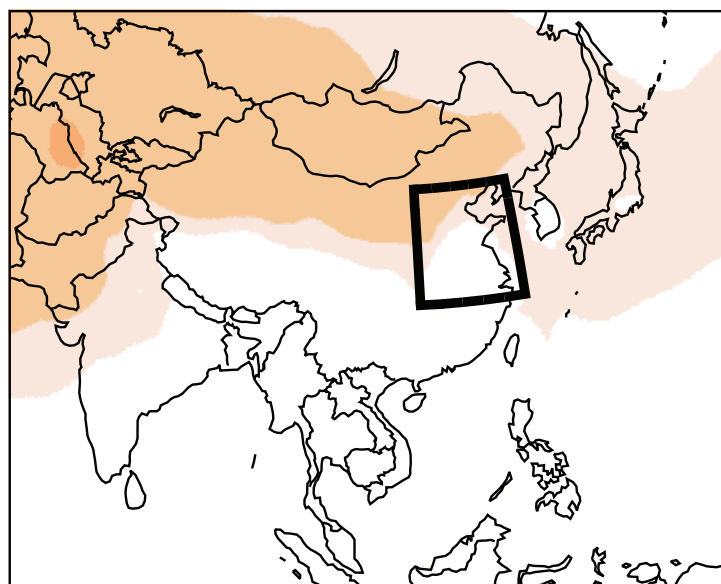
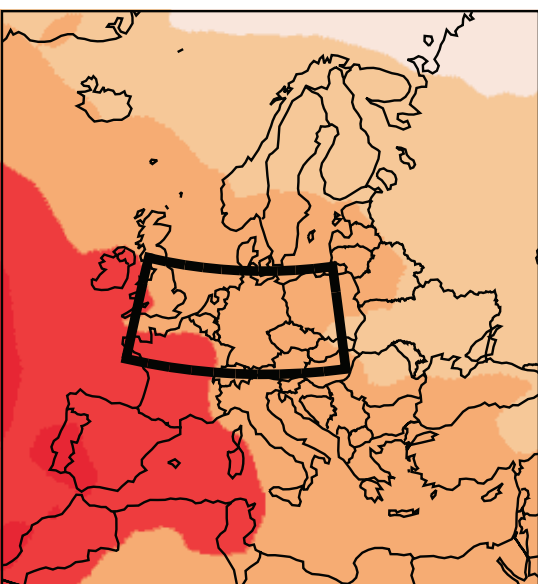
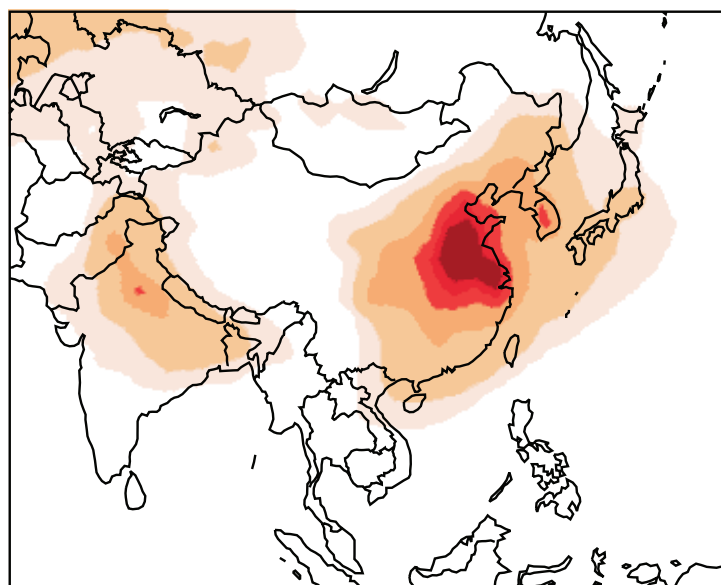
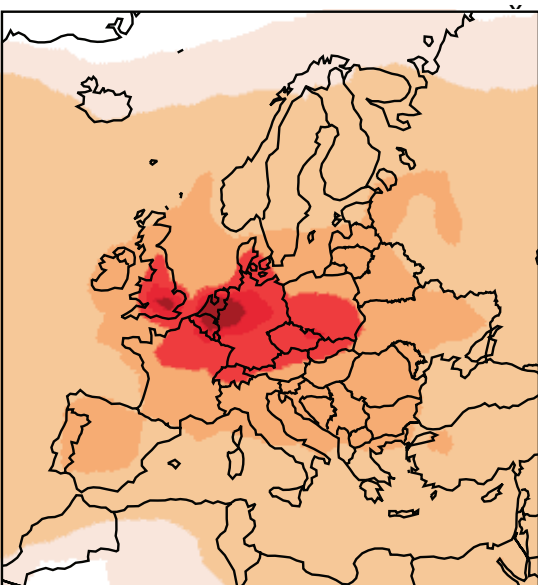
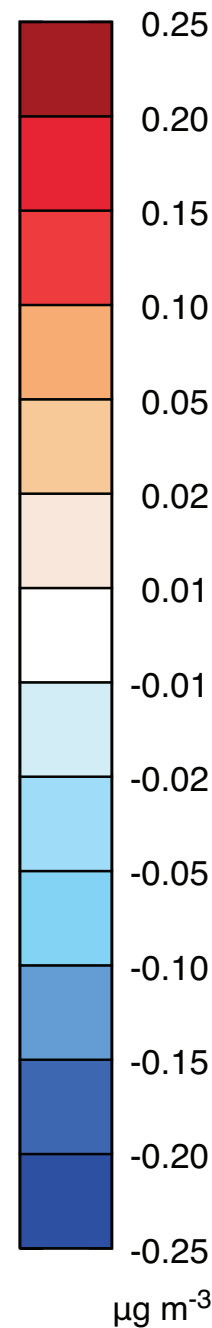
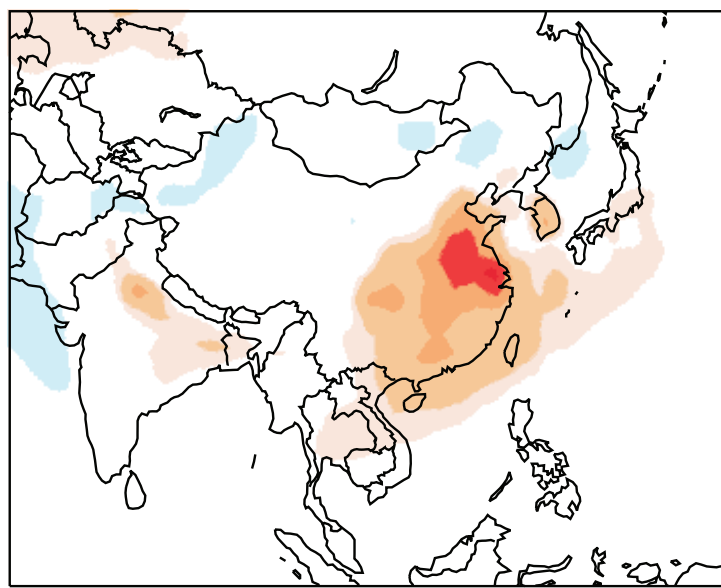
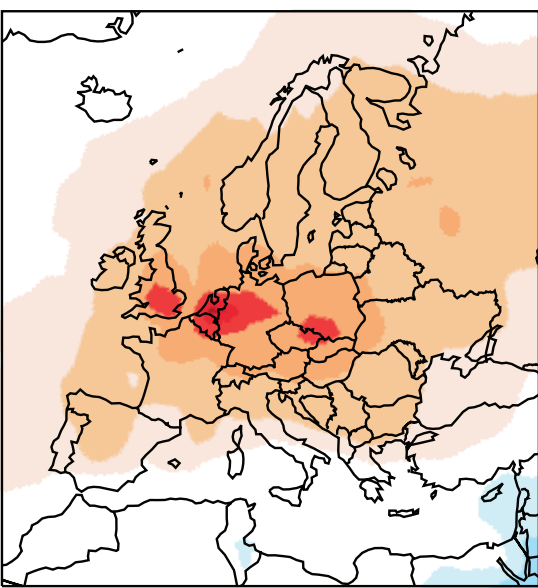
469 **Figure Captions**

470 **Figure 1** - Annual mean enhancements of surface PM concentrations in Europe and Asia from  
471 US anthropogenic emissions of SO<sub>2</sub> (top), NO<sub>x</sub> (middle), and CO (bottom). Values are  
472 differences between the control GEOS-Chem simulation and a sensitivity simulation with US  
473 anthropogenic emissions shut off. Boxes in the top panels outline receptor regions in northern  
474 Europe and eastern China for which speciation and seasonality are shown in Figure 2.

475 **Figure 2** – Speciation of sulfate-nitrate-ammonium (SNA) PM concentrations in surface air for  
476 the receptor regions of Figure 1 (left panels) and corresponding intercontinental enhancements  
477 from US anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, and CO (right panels). Values are seasonal  
478 means from the GEOS-Chem model. Seasonal concentrations have been divided by 100  
479 (Northern Europe) and 200 (Eastern China) to fit on scale.

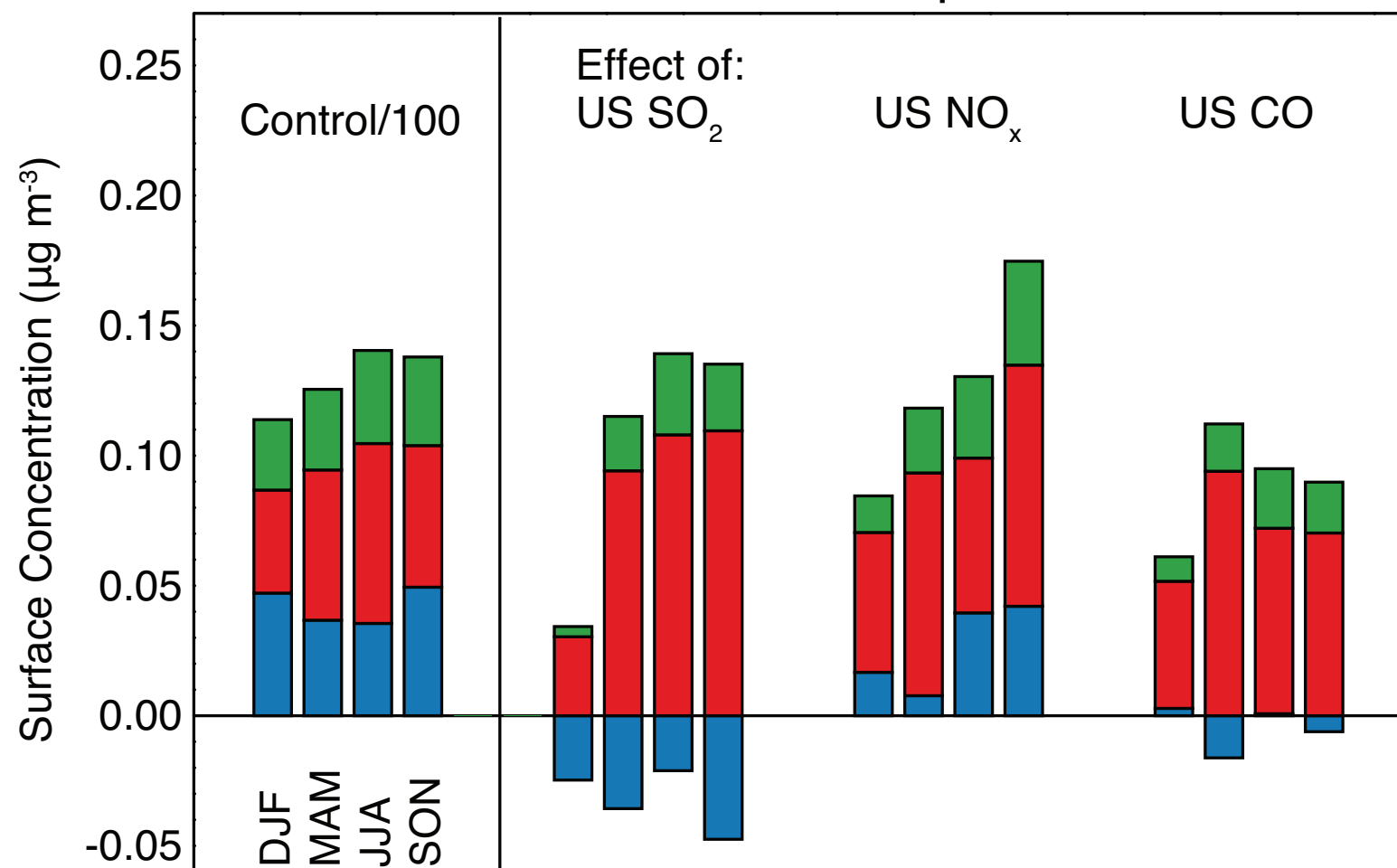
480 **Figure 3** - Annual mean enhancements of surface PM concentrations in Europe and the United  
481 States from Asian anthropogenic emissions of SO<sub>2</sub> (top), NO<sub>x</sub> (middle), and CO (bottom).  
482 Values are differences between the control GEOS-Chem simulation and a sensitivity simulation  
483 with Asian anthropogenic sources shut off. Boxes in the top panels outline the receptor region in  
484 the United States for which speciation and seasonality are shown in Figure 4.

485 **Figure 4** – Speciation of sulfate-nitrate-ammonium (SNA) PM concentrations in surface air for  
486 the receptor regions of Figures 1 and 2 (left panels) and corresponding intercontinental  
487 enhancements from US anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, and CO (right panels). Values are  
488 seasonal means from the GEOS-Chem model. Seasonal concentrations from the control  
489 simulation have been divided by 100 to fit on scale.

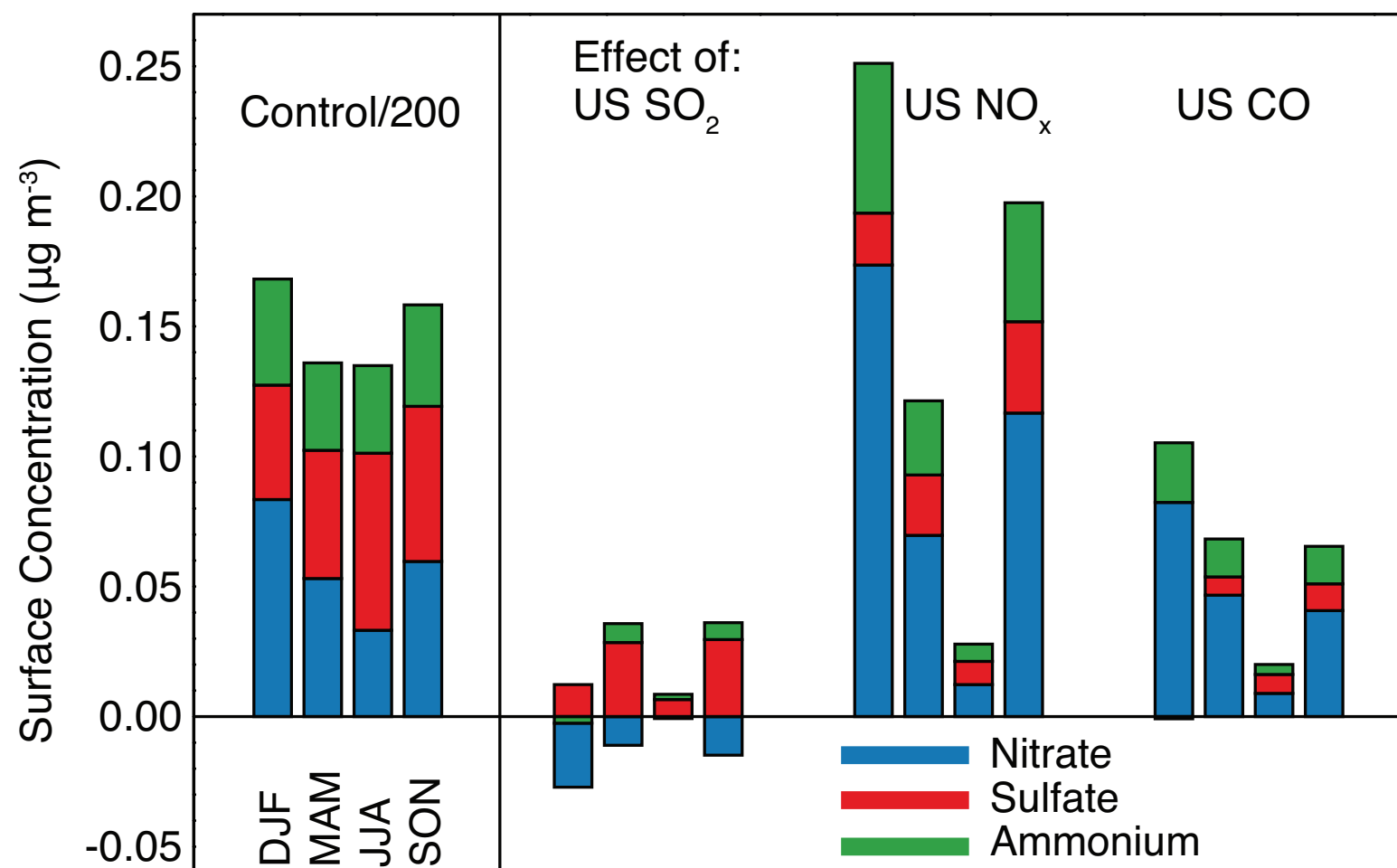
PM<sub>2.5</sub> Enhancement from US SO<sub>2</sub>PM<sub>2.5</sub> Enhancement from US NO<sub>x</sub>PM<sub>2.5</sub> Enhancement from US CO $\mu\text{g m}^{-3}$

Figure

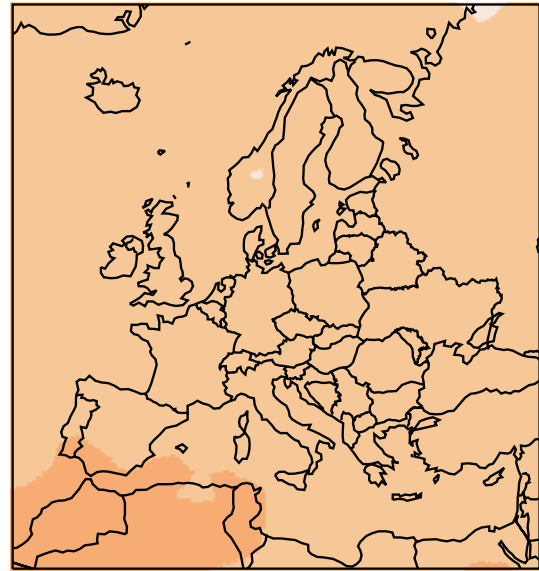
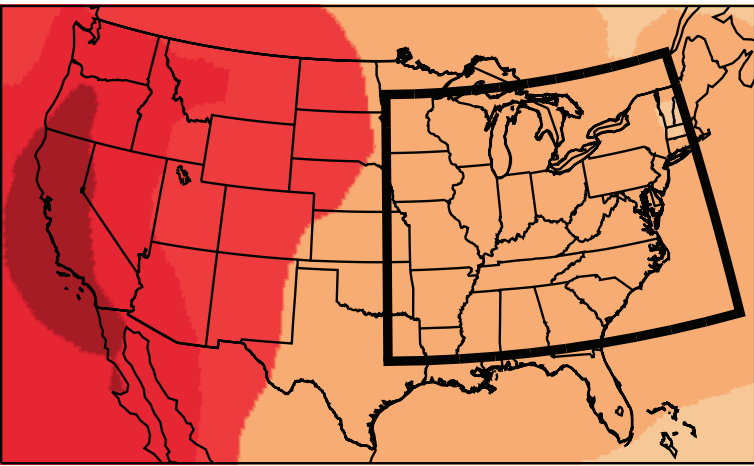
## Northern Europe



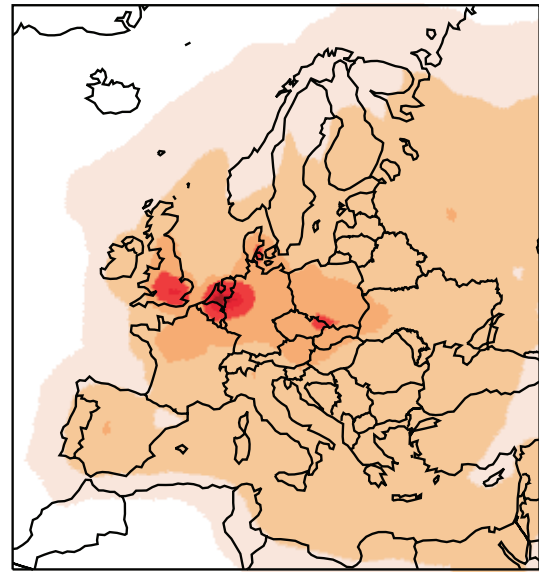
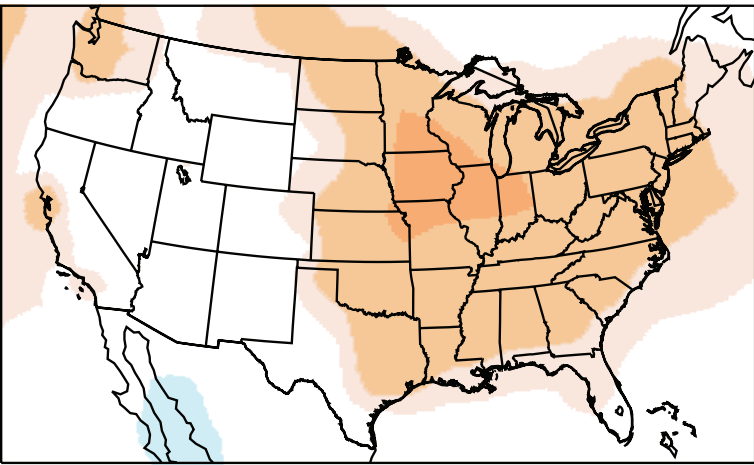
## Eastern China



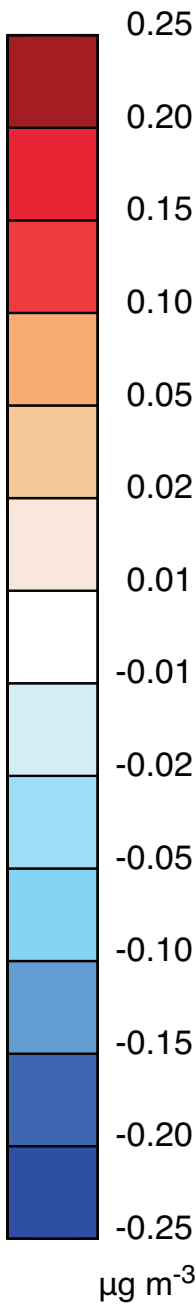
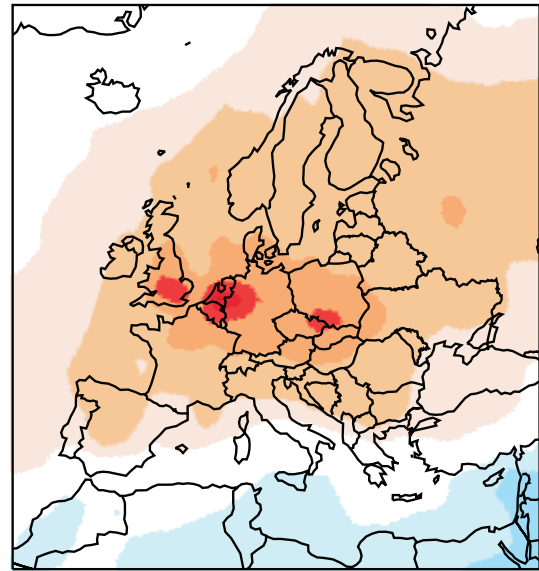
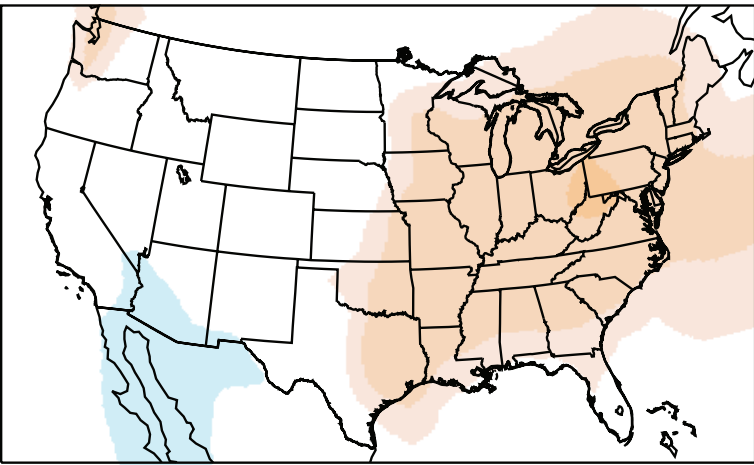
PM<sub>2.5</sub> Enhancement from Asian SO<sub>2</sub>



PM<sub>2.5</sub> Enhancement from Asian NO<sub>x</sub>

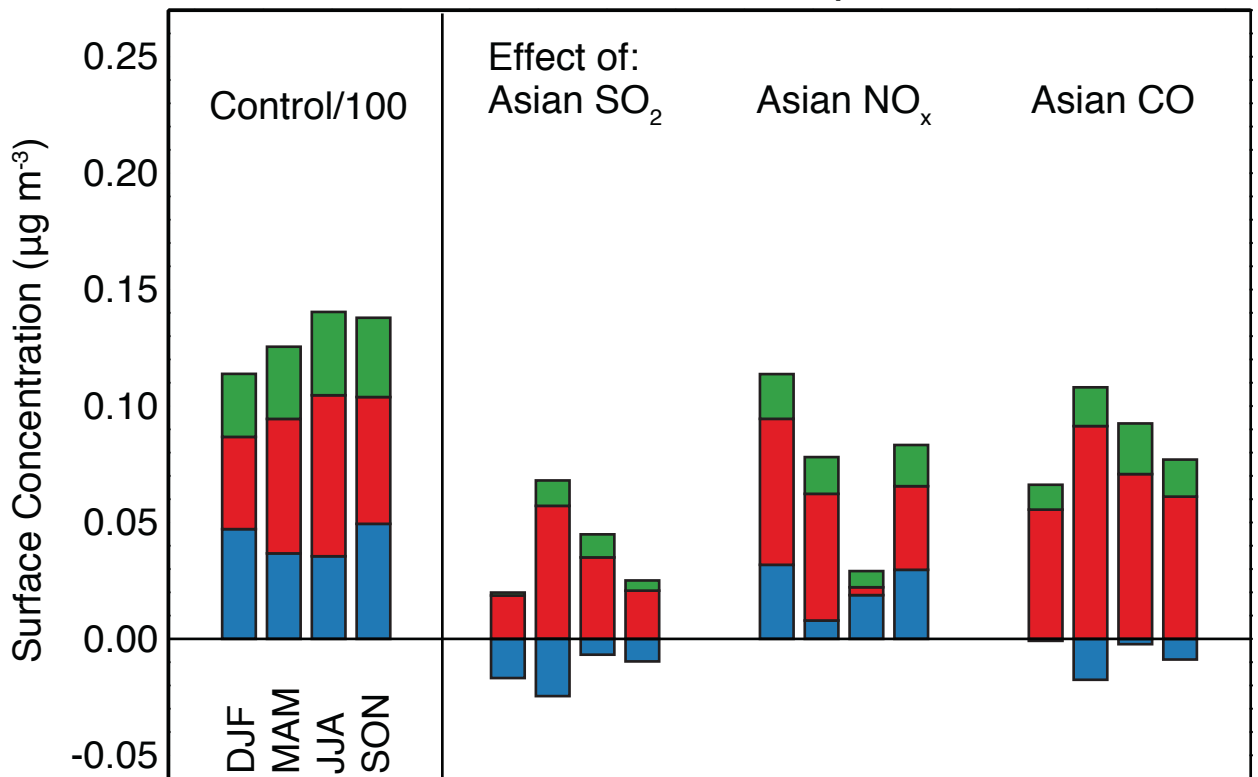


PM<sub>2.5</sub> Enhancement from Asian CO



Figure

## Northern Europe



## United States

