1 # Sulfuric Acid Deposition from Stratospheric Geoengineering
2 # with Sulfate Aerosols

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47   **Abstract**

48   We used a general circulation model of Earth's climate to conduct geoengineering

49   experiments involving stratospheric injection of sulfur dioxide and analyzed the resulting

50   deposition of sulfate. When sulfur dioxide is injected into the tropical or Arctic stratosphere, the

51   main additional surface deposition of sulfate occurs in midlatitude bands, because of strong

52   cross-tropopause flux in the jet stream regions. We used critical load studies to determine the

53   effects of this increase in sulfate deposition on terrestrial ecosystems by assuming the upper limit

54   of hydration of all sulfate aerosols into sulfuric acid. For annual injection of 5 Tg of SO_2 into the

55   tropical stratosphere or 3 Tg of SO_2 into the Arctic stratosphere, the maximum point value of

56   sulfate deposition of approximately 1.5 mEq m^{-2} a^{-1}, and the largest additional deposition that

57   would result from geoengineering of approximately 0.05 mEq m^{-2} a^{-1}, are not enough to

58   negatively impact most ecosystems.

1. Introduction

59

60   Faced with the problem of climate change due to increasing global temperatures, some

61   scientists and policy makers have suggested the deliberate modification of Earth's climate, an

62   activity that has been termed geoengineering. There have been many different suggestions for

63   geoengineering, both recently [e.g., *Angel*, 2006; *Bower et al.*, 2006] as well as historically

64   [*Fleming*, 2007]. However, one method that has received a great deal of recent attention is the

65   suggestion of *Budyko* [1974, 1977], *Dickinson* [1996], and *Crutzen* [2006] to inject gaseous

66   aerosol precursors into the stratosphere. The creation of highly reflective sulfate aerosols in the

67   lower stratosphere would result in some warming of the stratosphere, but the aerosol cloud

68   would also tend to increase the planetary albedo, resulting in cooling of the troposphere and the

69   surface [*Rasch et al.*, 2008a]. *Rasch et al.* [2008b] and *Robock et al.* [2008] calculated climate

70   responses to this aerosol cloud using general circulation models.

71   Geoengineering will, however, invariably have certain undesirable consequences. *Tilmes*

72   *et al.* [2008] and *Robock* [2008a] discussed the negative impact these sulfate aerosols will have

73   on polar stratospheric ozone. *Robock* [2008b] listed 20 potential side effects that could result

74   from this method. Our purpose here is to evaluate one of *Robock*'s concerns, that of quantifying

75   the amount of sulfur deposition that would result from two potential scenarios of geoengineering

76   with sulfate aerosols. This is of concern because the sulfate aerosol can hydrate to form sulfuric

77   acid, meaning geoengineering with sulfate aerosols can potentially result in an increase in acid

78   deposition.

79   Acid rain has been studied extensively in terms of its effects on ecosystems. Sulfur is a

80   necessary nutrient for some plants, and the need to add sulfur to crops has long been recognized

81   by farmers [*Hart and Peterson*, 1911]. However, an increase in sulfur deposition will not

82   universally benefit ecosystems, especially ones that are poorly buffered against an increase in

83   acidity. For example, excess acid can decrease or even eliminate freshwater fish populations

84   [*Leivestad and Muniz*, 1976], cause foliar leaching [*Wood and Bormann*, 1975], affect plant-

85   parasite interaction [*Shriner*, 1977], significantly reduce lake bacteria populations [*Rao and*

86   *Dutka*, 1983], and, through forest dieback and reduced food supply, can affect forest bird

87   communities [*Graveland*, 1998]. These, among other potential problems, could present

88   significant ecological concerns, and serve as our motivation for the study of sulfate deposition

89   due to geoengineering.

90   Whether sulfate deposition (both dry and wet) is harmful depends on both the amount of

91   sulfur introduced into the system, the amount of sulfate that is hydrated to form sulfuric acid, and

92   the sensitivity of the ecosystem. We will base our calculations on an upper limit, i.e., that all the

93   sulfur deposition is sulfuric acid. This is likely an overestimation, since wet deposition in the

94   model accounts for approximately 65% of total sulfate deposition, and dry deposition accounts

95   for the remainder. Moreover, not all sulfate deposition will result in an increase in acid rain.

96   Here we calculate how much additional sulfate would reach the surface from proposed

97   geoengineering and compare this to critical load thresholds for different regions.

98   As of now, most of the discussion of geoengineering with sulfate aerosols has focused on

99   using SO_2 as the preferred sulfate aerosol precursor. Volcanic eruptions can inject a large pulse

100   of SO_2 into the lower stratosphere, and previous geoengineering studies have considered

101   volcanic eruptions as an analog of geoengineering. However, other precursors, such as hydrogen

102   sulfide, carbonyl sulfide, and ammonium sulfate, could also potentially be used. Regardless, the

103   important factor in determining sulfate deposition is the amount of sulfur injected into the

104   stratosphere. As such, the results presented in this paper need only be scaled appropriately

105   according to the aerosol precursor's molecular weight.

106   **2. Experiment**

107   We studied geoengineering with stratospheric sulfate aerosols using ModelE, a general

108   circulation model developed by the National Aeronautics and Space Administration Goddard

109   Institute for Space Studies [*Schmidt et al.,* 2006]. We used the stratospheric version with 4°

110   latitude by 5° longitude horizontal resolution and 23 vertical levels up to 80 km. It is fully

111   coupled to a 4° latitude by 5° longitude dynamic ocean with 13 vertical levels [*Russell et al.*,

112   1995].

113   The aerosol module [*Koch et al.*, 2006] accounts for SO_2 conversion to sulfate aerosols,

114   as well as transport and removal of the aerosols. The chemical model calculates the sulfur cycle

115   in the stratosphere, where the conversion rate of SO_2 to sulfate is based on the respective

116   concentrations of SO_2 and the hydroxyl radical, the latter of which is prescribed [*Oman et al.*,

117   2006]. The dry aerosol effective radius is specified to be 0.25 µm, and the model hydrates the

118   aerosols based on ambient humidity values, resulting in a distribution of hydrated aerosols with

119   an effective radius of approximately 0.30-0.35 µm. Radiative forcing from the aerosols is fully

120   interactive with the atmospheric circulation.

121   *Koch et al.* [2006] thoroughly analyzed the performance of ModelE concerning sulfur

122   deposition from tropospheric sources. The model has some biases in that it produces 50-67% of

123   the observed sulfur deposition in Europe and the East coast of the United States. In the Western

124   United States, the model overpredicts the actual amount by 50-100%, but that region has little

125   sulfur deposition anyway. There are also some other local differences between model output and

126   observed values, but none of these biases is in a location that will affect our conclusions.

127   We proceeded with further analysis of climate simulations performed by *Robock et al.*

128   [2008]. We began with a three-member control ensemble of 20-year runs over which time global

129   greenhouse gas concentrations increased according to the Intergovernmental Panel on Climate

130   Change's A1B scenario [*IPCC*, 2007]. The greenhouse gas concentrations at the beginning of

131   the simulation were prescribed to be 2007 levels, and they increased to the A1B scenario's

132   estimation of 2026 levels by the end of the simulation.

133   In addition, we used two ensembles, each with three members of 20-year climate

134   simulations, covering the same time period. One involved daily injections of SO_2 into the

135   tropical lower stratosphere (longitude 120°E, latitude 0°, 16-23 km altitude) for a total of 5 Tg

136   per year in addition to the forcing prescribed by the A1B scenario, and one involved daily

137   injections of SO_2 into the Arctic lower stratosphere (longitude 120°E, latitude 68°N, 10-15 km

138   altitude) for a total of 3 Tg per year in addition to the forcing prescribed by the A1B scenario.

139   The value of 5 Tg per year was chosen to correspond to a Mount Pinatubo-sized eruption every 4

140   years, which was a value determined by *Robock et al.* as being sufficient to cause substantial

141   enough cooling to offset the climatic effects of an increase in greenhouse gas concentrations.

142   The smaller value of 3 Tg per year was also chosen by *Robock et al.*, since the goal of the

143   original experiment was to limit the climate response only in the Arctic. The longitude value

144   chosen is arbitrary and irrelevant, since the prevailing general circulation will transport the

145   gas/aerosol cloud around the globe within a matter of weeks.

146   The results of *Robock et al.* [2008] showed a globally averaged warming of

147   approximately 0.5°C by 2026 over the current climate under the A1B scenario. Under the

148   3 Tg a^{-1} Arctic injection case, the globally averaged temperature immediately reduced to 2000

149   levels and only warmed 0.3°C over the current climate by 2026. Under the 5 Tg a^{-1} tropical

150 injection case, the globally averaged temperature reduced to 1980 levels and held relatively

151 constant at that level through 2026, resulting in cooling by 0.3°C.

152 **3. Results**

153   Figure 1 shows the annual percent increase in total sulfate deposition, averaged over the

154 second decade of geoengineering. In the tropical injection case, there is an increase in sulfate

155 deposition over much of the globe, with the exception of the tropics (due to poleward

156 stratospheric transport before mixing into the troposphere). As expected, in the Arctic injection,

157 the increase in deposition is mostly confined to the Northern Hemisphere. The majority of the

158 increase is in the form of wet deposition (not shown). In the polluted midlatitudes of the

159 Northern Hemisphere, the increases of sulfate deposition are not noticeable, but in pristine areas,

160 such as Antarctica, they are readily apparent. Although all shaded values in the top panel are

161 statistically significant at a 95% confidence level, for the Arctic injection case, many of the

162 shaded values in the Southern Hemisphere are most likely due to weather noise.

163   Since pristine areas, such as Antarctica, Greenland, and the Southern Pacific Ocean,

164 received very little sulfate deposition in the baseline (A1B) case, additional deposition of tens of

165 percent may not be consequential, so we must evaluate the actual amount of deposition. Figure 2

166 shows that the increases in actual deposition are strongest in midlatitude bands, some as high as

167 10^{-3} kg m^{-2} a^{-1}, due to strong cross-tropopause flux in the jet stream region. Downwind of large

168 urban and industrial areas, we find the largest areas of absolute deposition, since these urban

169 areas are a significant source of sulfate, but they are also the areas of the largest increase in

170 deposition due to geoengineering because they are the jet exit regions, meaning the flux from

171 stratosphere to troposphere is comparatively large in these areas.

172   For the purpose of establishing a reference value for comparison, the baseline surface

173   sulfur emission levels are 135.8 Tg a^{-1} globally [*Koch et al.*, 2006]. Since the additional

174   stratospheric injections are 1-2 orders of magnitude smaller, we might not expect them to be

175   important in any case on a global basis. Dividing the surface emissions by the surface area of

176   Earth, we get an average of 5.41 x 10^{-5} kg m^{-2} a^{-1}. Also according to *Koch et al*., this sulfate has

177   an average atmospheric lifetime of 6.2 days, meaning levels would be expected to be much

178   higher than this reference value downwind of large urban and industrial areas and much lower

179   (or practically negligible) in unpopulated areas.

180   The notable absence of deposition over some of the continental areas (for example, the

181   Sahara and Western Australia) is because most of the additional sulfate deposition is in the form

182   of wet deposition, and these areas receive little rain. Other seeming gaps in deposition over

183   continents are merely due to the values being small enough that they are obscured by the choice

184   in contouring levels. Model bias may also play a certain role in either enhancing or obscuring

185   these gaps, but we do not have sufficient information to make a detailed analysis effects due to

186   this.

187   These figures only show annually averaged results. There are small regions of larger

188   deposition for certain seasons, but the annual average is sufficient for this analysis. However, as

189   greenhouse gas concentrations increase in the future, the strength of the Brewer-Dobson

190   circulation will also increase, resulting in a shorter lifetime for stratospheric aerosols and the

191   need for more sulfur to produce the same climate response [*Rasch et al.*, 2008b], which would

192   cause an increase in sulfate deposition. We have not evaluated the effects that an increase in the

193   strength of stratospheric circulation would have with regard to our study.

194   **4. Impacts of Additional Acid Deposition**

195   The significance of the sulfate deposition increases depends on their potential effects on

196   the ecosystems over which the deposition occurs. The subsequent section is devoted to the

197   potential effects on the ocean, so in this section, we concentrate on terrestrial ecosystems.

198   Although the graphs only show sulfate deposition, for the purposes of establishing an upper limit

199   to potential negative effects, we will assume that all sulfate due to geoengineering reacts to form

200   sulfuric acid.

201   *Kuylenstierna et al.* [2001] used a modeling approach to perform a critical load study on

202   a global scale in which they rank areas by sensitivity to increased acid deposition, a value they

203   determine by evaluating the buffering capacity of each region's soil. Our units of sulfate

204   deposition, kg m^{-2} a^{-1}, must be converted to the units found in *Kuylenstierna et al.* of

205   mEq m^{-2} a^{-1}. We use the definitions

206
$$\text{mEq} = \frac{\text{mass (grams)}}{\text{mEq mass (grams)}} \quad \text{and} \quad \text{mEq mass (grams)} = \frac{\text{atomic weight (g/mol)}}{\text{valence x 1000}}$$

207   The SO_4^{2-} ion has atomic weight 96 g/mole and a valence of 2, giving us mEq mass (grams) of

208   0.048. So

209
$$\frac{1\,\text{kg}}{\text{m}^2 \cdot \text{a}} \cdot \frac{1000\,\text{g}}{1\,\text{kg}} \cdot \frac{1\,\text{mEq x 0.048}}{1\,\text{g}} = 48 \frac{\text{mEq}}{\text{m}^2 \cdot \text{a}}.$$

210   Figure 3 refers to the 5 Tg a^{-1} injection scenario. It shows total annual sulfate deposition

211   (taken as an ensemble average over the second decade of geoengineering) and the annual sulfate

212   deposition just due to geoengineering (injection minus baseline), both in terms of these new

213   units. The 5 Tg a^{-1} injection scenario was chosen because it has larger sulfate deposition than the

214   Arctic 3 Tg a^{-1} scenario, although the results presented in Figure 3 are similar for the Arctic

215   3 Tg a^{-1} injection case.   The maximum point value for total deposition is approximately

216   1.5 mEq m^{-2} a^{-1} and the largest point value which is solely the result of geoengineering (injection

217   minus baseline) is approximately 0.05 mEq m^{-2} a^{-1}. According to the critical loading studies of

218   *Kuylenstierna et al.*, the most sensitive areas of the globe can receive 25-50 mEq m^{-2} a^{-1} of

219   sulfate deposition before potentially being negatively impacted.

220   In another study, *Skeffington* [2006] takes a very conservative approach to critical

221   loading. He uses models for many of his results, but he also uses experimental and field

222   evidence when available. In addition, his purpose is to estimate uncertainty in measurements of

223   critical loading, so the low ends of his ranges for which loads are considered critical can be seen

224   as conservative estimates.

225   *Skeffington*'s results are given in terms of kEq ha^{-1} a^{-1}, so we must again perform a

226   conversion:

227   $$\frac{1 \text{ kEq}}{\text{ha} \cdot \text{a}} \cdot \frac{10^6 \text{ mEq}}{1 \text{ kEq}} \cdot \frac{1 \text{ ha}}{10^4 \text{ m}^2} = 100 \frac{\text{mEq}}{\text{m}^2 \cdot \text{a}} \text{ .}$$

228   These results, with our conversion factor taken into account, show that our values for acid

229   deposition over a year, with the possible exception of poorly buffered terrestrial waterways, are

230   well below critical loading levels (Table 1). In addition, the area in which the total sulfate

231   deposition exceeds 1 mEq m^{-2} a^{-1} is, according to our model results, very small. However, due to

232   our grid size, which is especially large when compared to the size of most terrestrial waterways,

233   there may be localized areas of enhanced deposition from individual precipitation events that we

234   cannot assess.

235   **5. Ocean Acidification**

236   One well-known consequence of an increase in carbon dioxide concentrations in the

237   atmosphere is an increase in the acidity of the oceans, as carbon dioxide dissolves in the oceans,

238 forming carbonic acid. We wish to compare this resultant acidification with our results for

239 sulfate deposition to further evaluate significance of our results.

240 *Raven et al.* [2005] estimated that over 500 Gt (5×10^{17} g) of carbon dioxide has

241 dissolved in the oceans over the past 200 years. Knowing that carbonic acid is a weak acid and

242 that the atomic weight of carbon dioxide is 44 g/mol, we can put this value in terms of mEq by

243 using our previous definitions. Thus we conclude that 1×10^{19} mEq of carbon dioxide has

244 dissolved in the ocean. Since the ocean covers approximately 70% of the earth's surface, we can

245 divide by the surface area covered by the ocean, as well as dividing by the 200 years over which

246 this process occurred, to get

247
$$\frac{1 \times 10^{19}}{(0.7)(4\pi R_E^{\ 2})(200)} = 140 \ \text{mEq m}^{-2} \ \text{a}^{-1}$$

248 This deposition is two orders of magnitude larger than our highest potential value of sulfuric acid

249 deposition, again assuming all sulfate due to geoengineering is reacted to form sulfuric acid,

250 leading us to conclude that the increase in acid deposition resulting from geoengineering with

251 stratospheric sulfate aerosols is not enough to negatively impact the oceans.

6. Conclusions

253 Analysis of our results and comparison to the results of *Kuylenstierna et al.* [2001] and

254 *Skeffington* [2006] lead to the conclusion that the additional sulfate deposition that would result

255 from geoengineering will not be sufficient to negatively impact most ecosystems, even under the

256 assumption that all deposited sulfate will be in the form of sulfuric acid. However, although

257 these model results are feasible, should geoengineering with sulfate aerosols actually be

258 conducted, local results due to weather variability may differ from the results presented here.

259 With the exception of terrestrial waterways, every region has a critical loading value a full order

260 of magnitude above the largest potential total amount of acid deposition that would occur under

261   the geoengineering scenarios presented in this paper. Furthermore, our results show that

262   additional sulfate deposition tends to preferentially occur over oceans, meaning the chance of

263   such a sensitive ecosystem receiving enough additional sulfate deposition to suffer negative

264   consequences is very small.

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References

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271 Angel, R. (2006), Feasibility of cooling the Earth with a cloud of small spacecraft near the inner

272   Lagrange point (L1), *Proc. Nat. Acad. Sci., 103*, 17,184-17,189.

273 Bower, K., T. Choularton, J. Latham, J. Sahraei, and S. Salter (2006), Computational assessment

274   of a proposed technique for global warming mitigation via albedo-enhancement of marine

275   stratocumulus clouds, *Atm. Res.*, *82*, 328-336.

276 Budyko, M. I. (1974), *Climate and Life* (Academic Press, New York, NY), 508 pp.

277 Budyko, M. I. (1977), *Climatic Changes* (American Geophysical Union, Washington, DC), 261

278   pp.

279 Crutzen, P. (2006), Albedo enhancement by stratospheric sulfur injections: A contribution to

280   resolve a policy dilemma? *Climatic Change*, *77*, 211-219.

281 Dickinson, R. E. (1996), Climate engineering: A review of aerosol approaches to changing the

282   global energy balance, *Climatic Change*, *33*, 279-290.

283 Fleming, J. R. (2007), The climate engineers, *Wilson Quarterly*, *Spring 2007*, 46-60.

284 Graveland, J. (1998), Effects of acid rain on bird populations, *Environ. Rev.*, *6*(1), 41-54,

285   doi:10.1139/er-6-1-41.

286 Hart, E. B. and W. H. Peterson (1911), The sulfur requirements of farm crops in relation to the

287   soil and air supply, *J. Am. Chem. Soc.*, *33*(4), 549-564, doi:10.1021/ja02217a015.

288 IPCC (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working*

289   *Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate*

290   *Change*, S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor

291   and H. L. Miller, Eds., (Cambridge University Press, Cambridge, United Kingdom and New

292   York, NY, USA), 996 pp.

293 Koch, D., G. A. Schmidt, and C. V. Field (2006), Sulfur, sea salt, and radionuclide aerosols in

294   GISS ModelE, *J. Geophys. Res.*, *111*, D06206, doi:10.1029/2004JD005550.

295 Kuylenstierna, J. C. I., H. Rodhe, S. Cinderby, and K. Hicks (2001), Acidification in developing

296   countries: ecosystem sensitivity and the critical load approach on a global scale, *Ambio*,

297   *30*(1), 20-28.

298 Leivestad, H., and I. P. Muniz (1976), Fish kill at low pH in a Norwegian river, *Nature*, *259*,

299   391-392, doi:10.1038/259391a0.

300 Oman, L., A. Robock, G. L. Stenchikov, T. Thordarson, D. Koch, D. T. Shindell, and C. Gao

301   (2006), Modeling the distribution of the volcanic aerosol cloud from the 1783-1784 Laki

302   eruption, *J. Geophys. Res.*, *111*, D12209, doi:10.1029/2005JD006899.

303 Rao, S. S., and B. J. Dutka (1983), Influence of acid precipitation on bacterial populations in

304   lakes, *Hydrobiologia*, *98*(2), 153-157, doi:10.1007/BF02185633.

305 Rasch, P. J., et al. (2008a), An overview of geoengineering of climate using stratospheric

306   sulphate aerosols, *Phil. Trans. Royal Soc. A.*, *366*, 4007-4037, doi:10.1098/rsta.2008.0131.

307 Rasch, P. J., P. J. Crutzen, and D. B. Coleman (2008b), Exploring the geoengineering of climate

308   using stratospheric sulfate aerosols: The role of particle size, *Geophys. Res. Lett.*, *35*,

309   L02809, doi:10.1029/2007GL032179.

310 Raven, J. A., et al. (2005), Ocean acidification due to increasing atmospheric carbon dioxide,

311   *Royal Society*, London, UK.

312 Robock, A. (2008a), Whither geoengineering? *Science*, *320*, 1166-1167.

313 Robock, A. (2008b), 20 reasons why geoengineering may be a bad idea, *Bull. Atomic Scientists*,

314   *64*(2), 14-18, 59, doi:10.2968/064002006.

315   Robock, A., L. Oman, and G. Stenchikov (2008), Regional climate responses to geoengineering

316   with tropical and Arctic SO$_2$ injections, *J. Geophys. Res.*, *113*, D16101, doi:10.1029/

317   2008JD010050.

318   Russell, G. L., J. R. Miller, and D. Rind (1995), A coupled atmosphere-ocean model for transient

319   climate change, *Atmos.-Ocean*, *33*, 683-730.

320   Schmidt, G. A., et al. (2006), Present day atmospheric simulations using GISS ModelE:

321   Comparison to in situ, satellite and reanalysis data, *J. Climate*, *19*, 153-192.

322   Shriner, D. S. (1977), Effects of simulated rain acidified with sulfuric acid on host-parasite

323   interactions, *Water, Air, & Soil Pollution*, *8*(1), 9-14, doi:10.1007/BF00156719.

324   Skeffington, R. A. (2006), Quantifying uncertainty in critical loads: (A) literature review, *Water,*

325   *Air, and Soil Pollution*, *169*, 3-24.

326   Tilmes, S., R. Müller, and R. Salawitch (2008), The sensitivity of polar ozone depletion to

327   proposed   geoengineering   schemes,   *Science*,   *320*(5880),   1201-1204,   doi:10.1126/

328   science.1153966.

329   Wood, T., and F. H. Bormann (1975), Increases in foliar leaching caused by acidification of an

330   artificial mist, *Ambio*, *4*, 169-171.

331 **Table 1.** Ranges of critical loading of pollutant deposition (including sulfur) for various sites in

332 Europe as reported by *Skeffington* [2006].

333

| Region	Critical Load [mEq m^{-2} a^{-1}]
Coniferous forests in Southern Sweden	13-61
Deciduous forests in Southern Sweden	15-72
Varied sites in the UK	24-182
Aber in North Wales	32-134
Uhlirska in the Czech Republic	260-358
Fårahall in Sweden	29-134
Several varied sites in China (sulfur only)	63-880
Waterways in Sweden	1-44

334
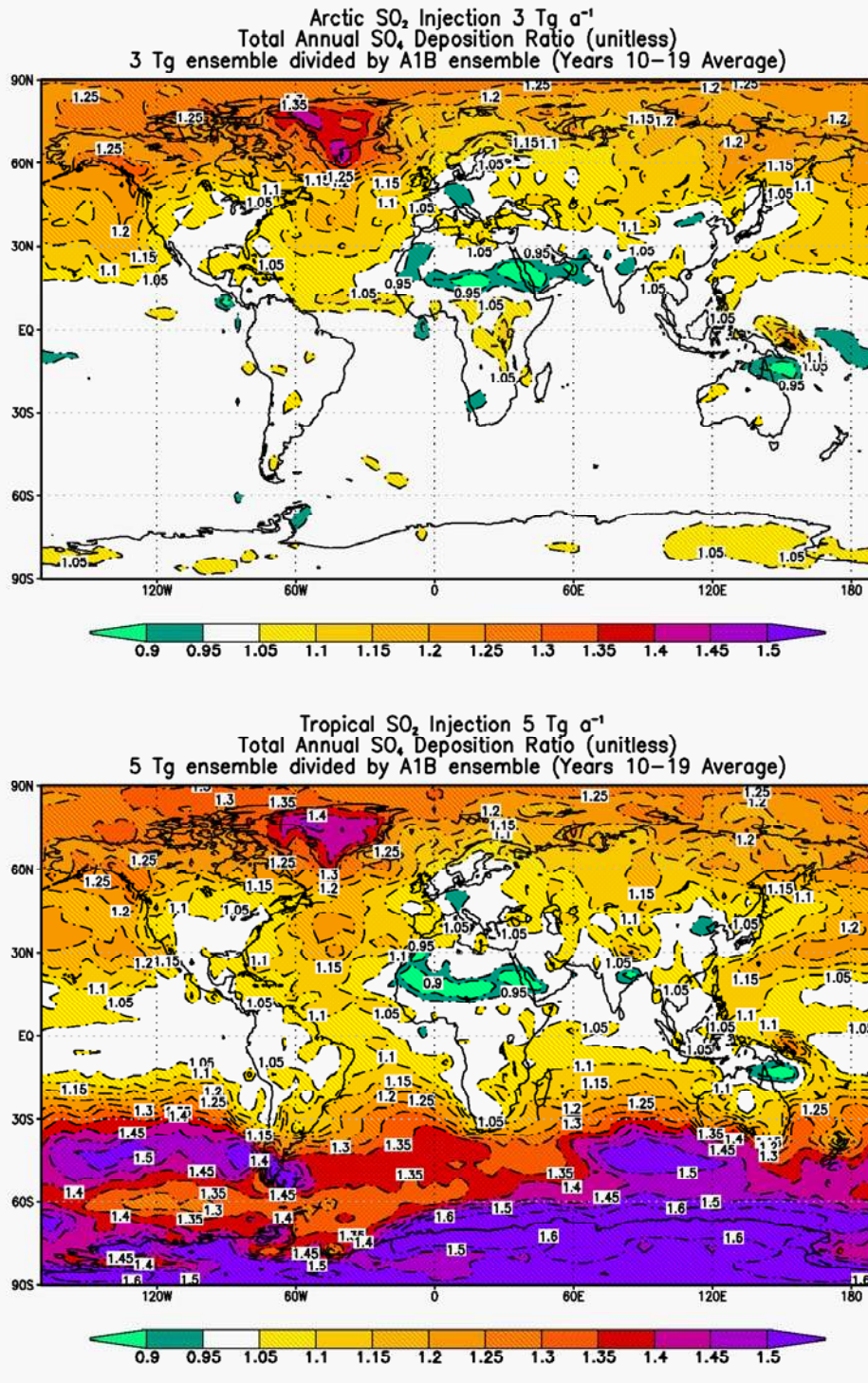
Figure 1. Ratios of the geoengineering ensembles (Arctic 3 Tg SO_2 a^{-1} injection and tropical 5 Tg SO_2 a^{-1} injection) to the baseline (A1B) ensemble. Both figures show annually averaged total sulfate deposition averaged over years 10-19 for each experiment. These plots are made from the model output of the climate simulations performed by *Robock et al.* [2008]. All shaded values on this figure are statistically significant at a 95% confidence level.

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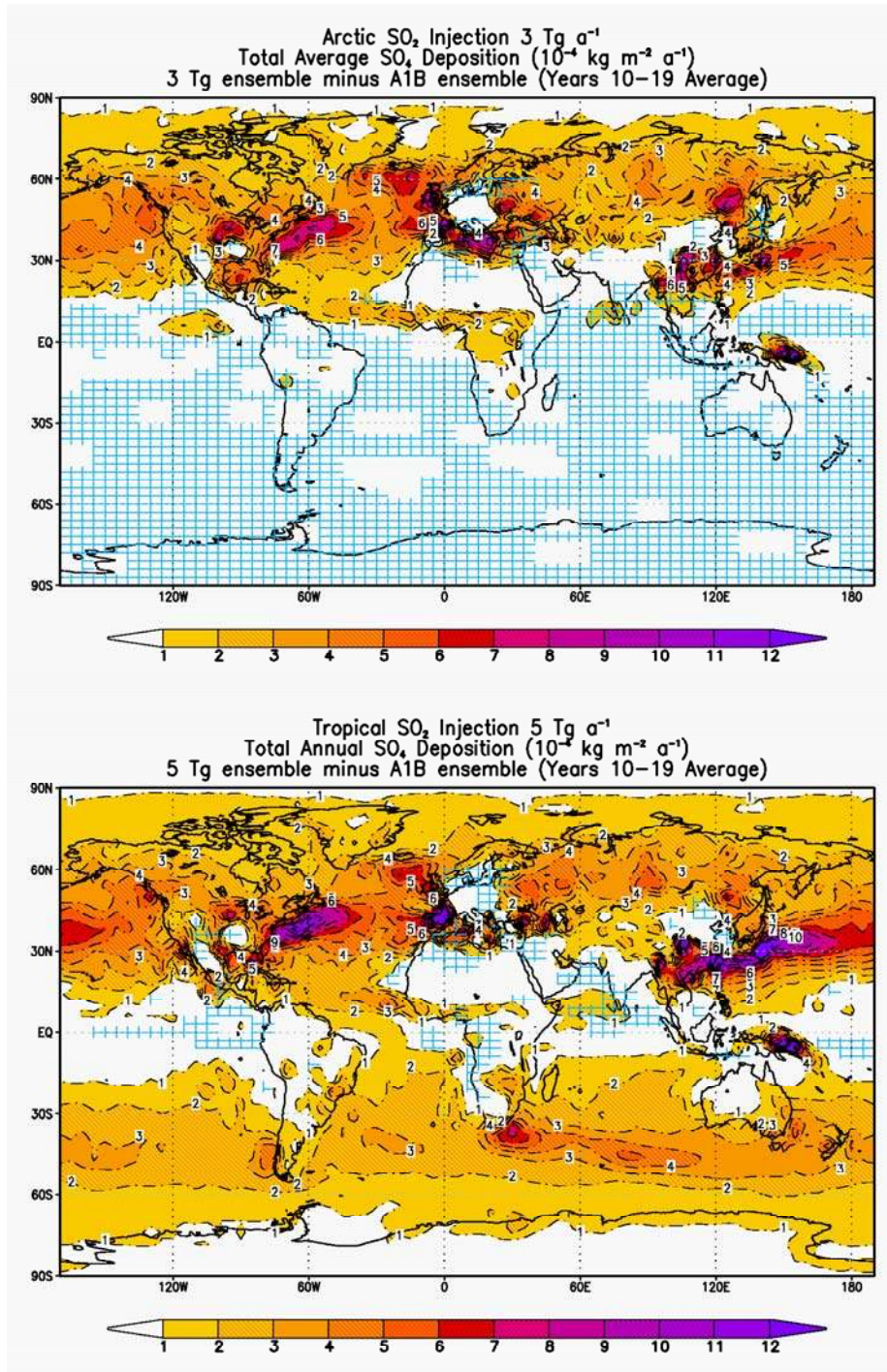
Figure 2. Annually averaged total sulfate deposition anomalies (injection minus baseline, revealing only the additional deposition from geoengineering) for the geoengineering scenarios of Arctic 3 Tg SO_2 a^{-1} and tropical 5 Tg SO_2 a^{-1} injection into the lower stratosphere. The results are averaged over three ensemble members and for years 10-19 of each experiment. These plots are made from the model output of the climate simulations performed by *Robock et al.* [2008]. Values not statistically significant at a 95% confidence level are denoted by blue hatching.
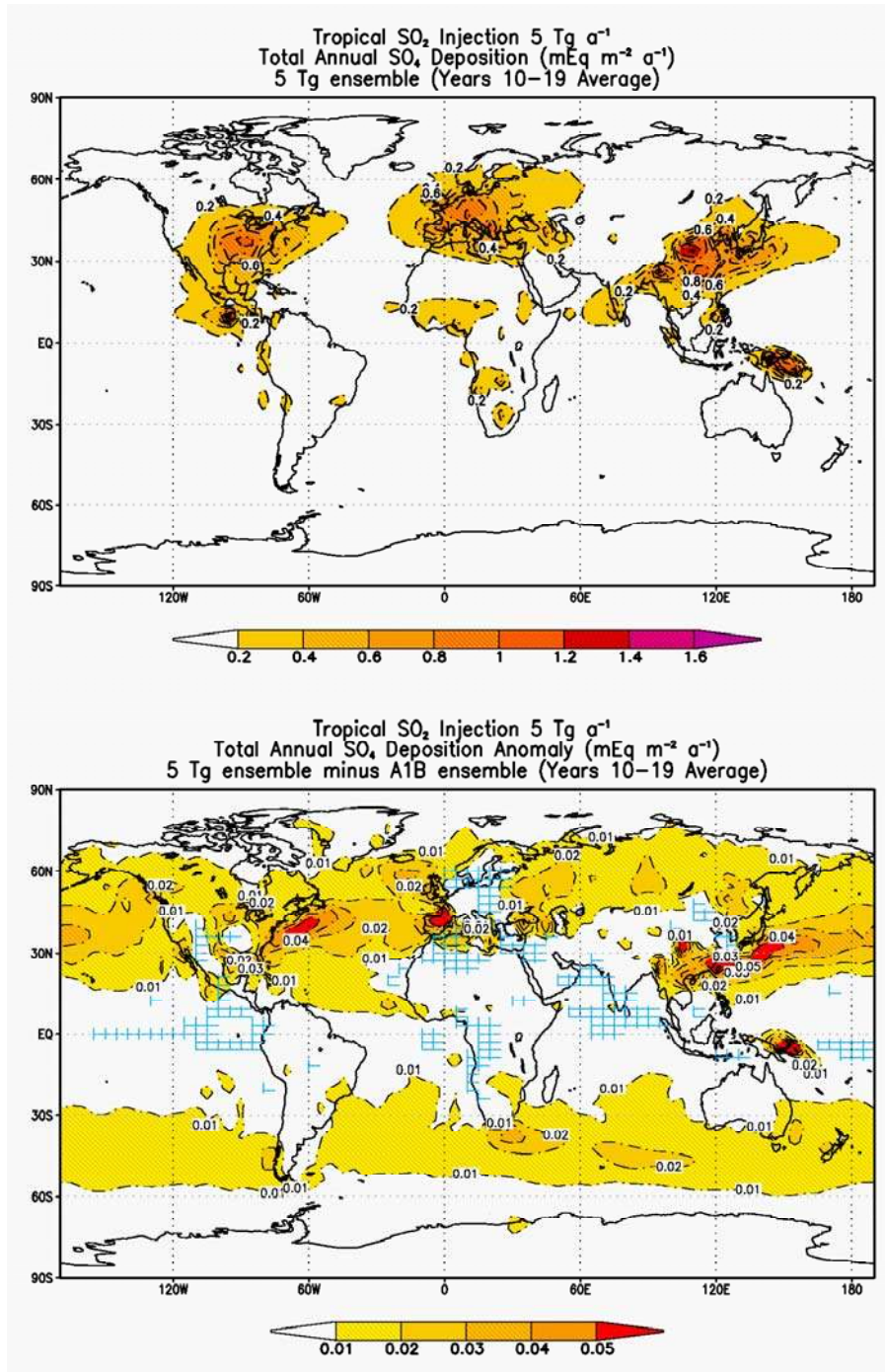
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349
Figure 3. Both figures show results for a tropical 5 Tg a^{-1} injection. The top figure shows total sulfate deposition (geoengineering plus baseline). The bottom figure shows sulfate deposition anomaly (injection minus baseline, revealing only the additional deposition from geoengineering). The largest total sulfate deposition point value is approximately 1.5 mEq m^{-2} a^{-1}, and the largest anomaly point value is approximately 0.05 mEq m^{-2} a^{-1}. These plots are made from the model output of the climate simulations performed by *Robock et al.* [2008], averaged over three ensemble members and years 10-19 for each experiment. Values not statistically significant at a 95% confidence level are denoted by blue hatching.