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Abstract

We used a general circulation model of Earth's climate to conduct geoengineering experiments involving stratospheric injection of sulfur dioxide and analyzed the resulting deposition of sulfate. When sulfur dioxide is injected into the tropical or Arctic stratosphere, the main additional surface deposition of sulfate occurs in midlatitude bands, because of strong cross-tropopause flux in the jet stream regions. We used critical load studies to determine the effects of this increase in sulfate deposition on terrestrial ecosystems by assuming the upper limit of hydration of all sulfate aerosols into sulfuric acid. For annual injection of 5 Tg of SO₂ into the tropical stratosphere or 3 Tg of SO₂ into the Arctic stratosphere, the maximum point value of sulfate deposition of approximately 1.5 mEq m⁻² a⁻¹, and the largest additional deposition that would result from geoengineering of approximately 0.05 mEq m⁻² a⁻¹, are not enough to negatively impact most ecosystems.

61 **1. Introduction**

62 Faced with the problem of climate change due to increasing global temperatures, some
63 scientists and policy makers have suggested the deliberate modification of Earth's climate, an
64 activity that has been termed geoengineering. There have been many different suggestions for
65 geoengineering, both recently [e.g., *Angel*, 2006; *Bower et al.*, 2006] as well as historically
66 [*Fleming*, 2007]. However, one method that has received a great deal of recent attention is the
67 suggestion of *Budyko* [1974, 1977], *Dickinson* [1996], and *Crutzen* [2006] to inject gaseous
68 aerosol precursors into the stratosphere. The creation of highly reflective sulfate aerosols in the
69 lower stratosphere would result in some warming of the stratosphere, but the aerosol cloud
70 would also tend to increase the planetary albedo, resulting in cooling of the troposphere and the
71 surface [*Rasch et al.*, 2008a]. *Rasch et al.* [2008b] and *Robock et al.* [2008] calculated climate
72 responses to this aerosol cloud using general circulation models.

73 Geoengineering will, however, invariably have certain undesirable consequences. *Tilmes*
74 *et al.* [2008] and *Robock* [2008a] discussed the negative impact these sulfate aerosols will have
75 on polar stratospheric ozone. *Robock* [2008b] listed 20 potential side effects that could result
76 from this method. Our purpose here is to evaluate one of *Robock's* concerns, that of quantifying
77 the amount of sulfur deposition that would result from two potential scenarios of geoengineering
78 with sulfate aerosols. This is of concern because the sulfate aerosol can hydrate to form sulfuric
79 acid, meaning geoengineering with sulfate aerosols can potentially result in an increase in acid
80 deposition.

81 Acid rain has been studied extensively in terms of its effects on ecosystems. Sulfur is a
82 necessary nutrient for some plants, and the need to add sulfur to crops has long been recognized
83 by farmers [*Hart and Peterson*, 1911]. However, an increase in sulfur deposition will not

84 universally benefit ecosystems, especially ones that are poorly buffered against an increase in
85 acidity. For example, excess acid can decrease or even eliminate freshwater fish populations
86 [Leivestad and Muniz, 1976], cause foliar leaching [Wood and Bormann, 1975], affect plant-
87 parasite interaction [Shriner, 1977], significantly reduce lake bacteria populations [Rao and
88 Dutka, 1983], and, through forest dieback and reduced food supply, can affect forest bird
89 communities [Graveland, 1998]. These, among other potential problems, could present
90 significant ecological concerns, and serve as our motivation for the study of sulfate deposition
91 due to geoengineering.

92 Whether sulfate deposition (both dry and wet) is harmful depends on both the amount of
93 sulfur introduced into the system, the amount of sulfate that is hydrated to form sulfuric acid, and
94 the sensitivity of the ecosystem. We will base our calculations on an upper limit, i.e. that all the
95 sulfur deposition is sulfuric acid. Here we calculate how much additional sulfate would reach
96 the surface from proposed geoengineering and compare this to critical load thresholds for
97 different regions.

98 As of now, most of the discussion of geoengineering with sulfate aerosols has focused on
99 using SO₂ as the preferred sulfate aerosol precursor. Volcanic eruptions can inject a large pulse
100 of SO₂ 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.

103 **2. Experiment**

104 We studied geoengineering with stratospheric sulfate aerosols using ModelE, a general
105 circulation model developed by the National Aeronautics and Space Administration Goddard
106 Institute for Space Studies [Schmidt *et al.*, 2006]. We used the stratospheric version with 4°

107 latitude by 5° longitude horizontal resolution and 23 vertical levels up to 80 km. It is fully
108 coupled to a 4° latitude by 5° longitude dynamic ocean with 13 vertical levels [Russell *et al.*,
109 1995].

110 The aerosol module [Koch *et al.*, 2006] accounts for SO₂ conversion to sulfate aerosols,
111 as well as transport and removal of the aerosols. The chemical model calculates the sulfur cycle
112 in the stratosphere, where the conversion rate of SO₂ to sulfate is based on the respective
113 concentrations of SO₂ and the hydroxyl radical, the latter of which is prescribed [Oman *et al.*,
114 2006]. The dry aerosol effective radius is specified to be 0.25 μm, and the model hydrates the
115 aerosols based on ambient humidity values, resulting in a distribution of hydrated aerosols with
116 an effective radius of approximately 0.30-0.35 μm. Radiative forcing from the aerosols is fully
117 interactive with the atmospheric circulation.

118 Koch *et al.* [2006] thoroughly analyzed the performance of ModelE concerning sulfur
119 deposition from tropospheric sources. The model has some biases in that it produces 50-67% of
120 the observed sulfur deposition in Europe and the East coast of the United States. In the Western
121 United States, the model overpredicts the actual amount by 50-100%, but that region has little
122 sulfur deposition anyway. There are also some other local differences between model output and
123 observed values, but none of these biases is in a location that will affect our conclusions.

124 We proceeded with further analysis of climate simulations performed by Robock *et al.*
125 [2008]. We began with a three-member control ensemble of 20-year runs over which time global
126 greenhouse gas concentrations increased according to the Intergovernmental Panel on Climate
127 Change's A1B scenario [IPCC, 2007]. The greenhouse gas concentrations at the beginning of
128 the simulation were prescribed to be 2007 levels, and they increased to the A1B scenario's
129 estimation of 2026 levels by the end of the simulation.

130 In addition, we used two ensembles, each with three members of 20-year climate
131 simulations, covering the same time period. One involved daily injections of SO₂ into the
132 tropical lower stratosphere (longitude 120°E, latitude 0°, 16-23 km altitude) for a total of 5 Tg
133 per year in addition to the forcing prescribed by the A1B scenario, and one involved daily
134 injections of SO₂ into the Arctic lower stratosphere (longitude 120°E, latitude 68°N, 10-15 km
135 altitude) for a total of 3 Tg per year in addition to the forcing prescribed by the A1B scenario.
136 The value of 5 Tg per year was chosen to correspond to a Mount Pinatubo-sized eruption every 4
137 years, which was a value determined by *Robock et al.* as being sufficient to cause substantial
138 enough cooling to offset the climatic effects of an increase in greenhouse gas concentrations.
139 The smaller value of 3 Tg per year was also chosen by *Robock et al.*, since the goal of the
140 original experiment was to limit the climate response only in the Arctic. The longitude value
141 chosen is arbitrary and irrelevant, since the prevailing general circulation will transport the
142 gas/aerosol cloud around the globe within a matter of weeks.

143 The results of *Robock et al.* [2008] show a globally averaged warming of approximately
144 0.5°C by 2026 over the current climate under the A1B scenario. Under the 3 Tg a⁻¹ Arctic
145 injection case, the globally averaged temperature immediately reduced to 2000 levels and only
146 warmed 0.3°C over the current climate by 2026. Under the 5 Tg a⁻¹ tropical injection case, the
147 globally averaged temperature reduced to 1980 levels and held relatively constant at that level
148 through 2026, resulting in cooling by 0.3°C.

149 **3. Results**

150 Figure 1 shows the annual percent increase in total sulfate deposition, averaged over the
151 second decade of geoengineering. In the tropical injection case, there is an increase in sulfate
152 deposition over much of the globe, with the exception of the tropics (due to poleward

153 stratospheric transport before mixing into the troposphere). As expected, in the Arctic injection,
154 the increase in deposition is mostly confined to the Northern Hemisphere. The majority of the
155 increase is in the form of wet deposition (not shown). In the polluted midlatitudes of the
156 Northern Hemisphere, the increases of sulfate deposition are not noticeable, but in pristine areas,
157 such as Antarctica, they are readily apparent. Although all shaded values in the top panel are
158 statistically significant at a 95% confidence level, for the Arctic injection case, many of the
159 shaded values in the Southern Hemisphere are most likely due to weather noise.

160 Since pristine areas, such as Antarctica, Greenland, and the Southern Pacific Ocean,
161 received very little sulfate deposition in the baseline (A1B) case, additional deposition of tens of
162 percent may not be consequential, so we must evaluate the actual amount of deposition. Figure 2
163 shows that the increases in actual deposition are strongest in midlatitude bands, some as high as
164 $10^{-3} \text{ kg m}^{-2} \text{ a}^{-1}$, due to strong cross-tropopause flux in the jet stream region. Downwind of large
165 urban and industrial areas, we find the largest areas of absolute deposition, since these urban
166 areas are a significant source of sulfate, but they are also the areas of the largest increase in
167 deposition due to geoengineering because they are the jet exit regions, meaning the flux from
168 stratosphere to troposphere is comparatively large in these areas.

169 For the purpose of establishing a reference value for comparison, the baseline surface
170 sulfur emission levels are 135.8 Tg a^{-1} globally [Koch *et al.*, 2006]. Since the additional
171 stratospheric injections are 1-2 orders of magnitude smaller, we might not expect them to be
172 important in any case on a global basis. Dividing the surface emissions by the surface area of the
173 earth, we get an average of $5.41 \times 10^{-5} \text{ kg m}^{-2} \text{ a}^{-1}$. Also according to Koch *et al.*, this sulfate has
174 an average atmospheric lifetime of 6.2 days, meaning levels would be expected to be much

175 higher than this reference value downwind of large urban and industrial areas and much lower
176 (or practically negligible) in unpopulated areas.

177 The notable absence of deposition over some of the continental areas (for example, the
178 Sahara and Western Australia) is because most of the additional sulfate deposition is in the form
179 of wet deposition, and these areas receive little rain. Other seeming gaps in deposition over
180 continents are merely due to the values being small enough that they are obscured by the choice
181 in contouring levels. Model bias may also play a certain role in either enhancing or obscuring
182 these gaps, but we do not have sufficient information to make a detailed analysis effects due to
183 this.

184 These figures only show annually averaged results. There are small regions of larger
185 deposition for certain seasons, but the annual average is sufficient for this analysis.

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

187 The significance of the sulfate deposition increases depends on their potential effects on
188 the ecosystems over which the deposition occurs. The subsequent section is devoted to the
189 potential effects on the ocean, so in this section, we concentrate on terrestrial ecosystems.
190 Although the graphs only show sulfate deposition, for the purposes of establishing an upper limit
191 to potential negative effects, we will assume that all sulfate due to geoengineering reacts to form
192 sulfuric acid.

193 *Kuylenstierna et al.* [2001] used a modeling approach to perform a critical load study on
194 a global scale in which they rank areas by sensitivity to increased acid deposition, a value they
195 determine by evaluating the buffering capacity of each region's soil. Our units of sulfate
196 deposition, $\text{kg m}^{-2} \text{a}^{-1}$, must be converted to the units found in *Kuylenstierna et al.* of
197 $\text{mEq m}^{-2} \text{a}^{-1}$. We use the definitions

198
$$\text{mEq} = \frac{\text{mass (grams)}}{\text{mEq mass (grams)}} \quad \text{and} \quad \text{mEq mass (grams)} = \frac{\text{atomic weight (g/mol)}}{\text{valence} \times 1000}$$

199 The SO_4^{2-} ion has atomic weight 96 g/mole and a valence of 2, giving us mEq mass (grams) of
 200 0.048. So

201
$$\frac{1 \text{ kg}}{\text{m}^2 \cdot \text{a}} \cdot \frac{1000 \text{ g}}{1 \text{ kg}} \cdot \frac{1 \text{ mEq} \times 0.048}{1 \text{ g}} = 48 \frac{\text{mEq}}{\text{m}^2 \cdot \text{a}}$$

202 Figure 3 refers to the 5 Tg a⁻¹ injection scenario. It shows total annual sulfate deposition
 203 (taken as an ensemble average over the second decade of geoengineering) and the annual sulfate
 204 deposition just due to geoengineering (injection minus baseline), both in terms of these new
 205 units. The 5 Tg a⁻¹ injection scenario was chosen because it has larger sulfate deposition than the
 206 Arctic 3 Tg a⁻¹ scenario. The maximum point value for total deposition is approximately 1.5
 207 mEq m⁻² a⁻¹ and the largest point value which is solely the result of geoengineering (injection
 208 minus baseline) is approximately 0.05 mEq m⁻² a⁻¹. According to the critical loading studies of
 209 *Kuylenstierna et al.*, the most sensitive areas of the globe can receive 25-50 mEq m⁻² a⁻¹ of
 210 sulfate deposition before potentially being negatively impacted.

211 In another study, *Skeffington* [2006] takes a very conservative approach to critical
 212 loading. He uses models for many of his results, but he also uses experimental and field
 213 evidence when available. In addition, his purpose is to estimate uncertainty in measurements of
 214 critical loading, so the low ends of his ranges for which loads are considered critical can be seen
 215 as conservative estimates.

216 *Skeffington's* results are given in terms of kEq ha⁻¹ a⁻¹, so we must again perform a
 217 conversion:

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

219 These results, with our conversion factor taken into account, show that our values for acid
 220 deposition over a year, with the possible exception of poorly buffered terrestrial waterways, are
 221 well below critical loading levels (Table 1). In addition, the area in which the total sulfate
 222 deposition exceeds $1 \text{ mEq m}^{-2} \text{ a}^{-1}$ is, according to our model results, very small. However, due to
 223 our grid size, which is especially large when compared to the size of most terrestrial waterways,
 224 there may be localized areas of enhanced deposition from individual precipitation events that we
 225 cannot assess.

226 5. Ocean Acidification

227 One well-known consequence of an increase in carbon dioxide concentrations in the
 228 atmosphere is an increase in the acidity of the oceans, as carbon dioxide dissolves in the oceans,
 229 forming carbonic acid. We wish to compare this resultant acidification with our results for
 230 sulfate deposition to further evaluate significance of our results.

231 *Raven et al.* [2005] estimate that over 500 gigatons (5×10^{17} g) of carbon dioxide has
 232 dissolved in the oceans over the past 200 years. Then knowing that carbonic acid is a weak acid
 233 and that the atomic weight of carbon dioxide is 44 g/mol, we can put this value in terms of mEq
 234 by using our previous definitions. Thus we conclude that 0.1136×10^{20} mEq of carbon dioxide
 235 has dissolved in the ocean. Since the ocean covers approximately 70% of the earth's surface, we
 236 can divide by the surface area covered by the ocean, as well as dividing by the 200 years over
 237 which this process occurred, to get

$$238 \frac{0.1136 \times 10^{20}}{(0.7)(4\pi R_E^2)(200)} = 159 \text{ mEq m}^{-2} \text{ a}^{-1}$$

239 This deposition is two orders of magnitude larger than our highest potential value of sulfuric acid
 240 deposition, again assuming all sulfate due to geoengineering is reacted to form sulfuric acid,

241 leading us to conclude that the increase in acid deposition resulting from geoengineering with
242 stratospheric sulfate aerosols is not significant enough to negatively impact the oceans.

243 **6. Conclusions**

244 Analysis of our results and comparison to the results of *Kuylenstierna et al.* [2001] and
245 *Skeffington* [2006] led to the conclusion that the additional sulfate deposition that would result
246 from geoengineering will not be sufficient to negatively impact most ecosystems. With the
247 exception of terrestrial waterways, every region has a critical loading value a full order of
248 magnitude above the largest potential total amount of acid deposition that would occur under the
249 geoengineering scenarios presented in this paper. Furthermore, our results show that additional
250 sulfate deposition tends to preferentially occur over oceans, meaning the chance of such a
251 sensitive ecosystem receiving enough additional sulfate deposition to suffer negative
252 consequences is very small.

253

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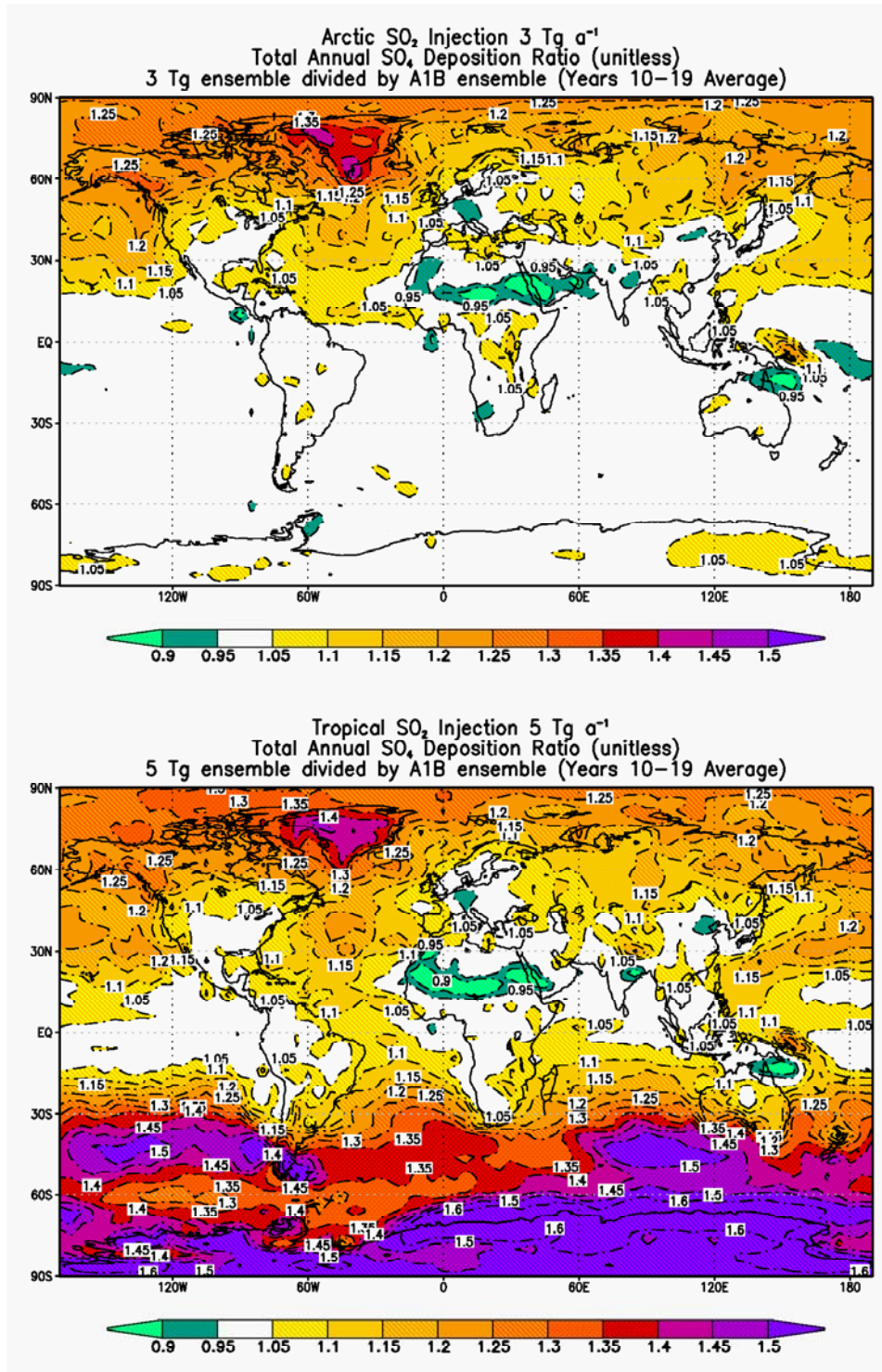
321 **Table 1.** Ranges of critical loading of pollutant deposition (including sulfur) for various sites in
 322 Europe as reported by *Skeffington* [2006].

323

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

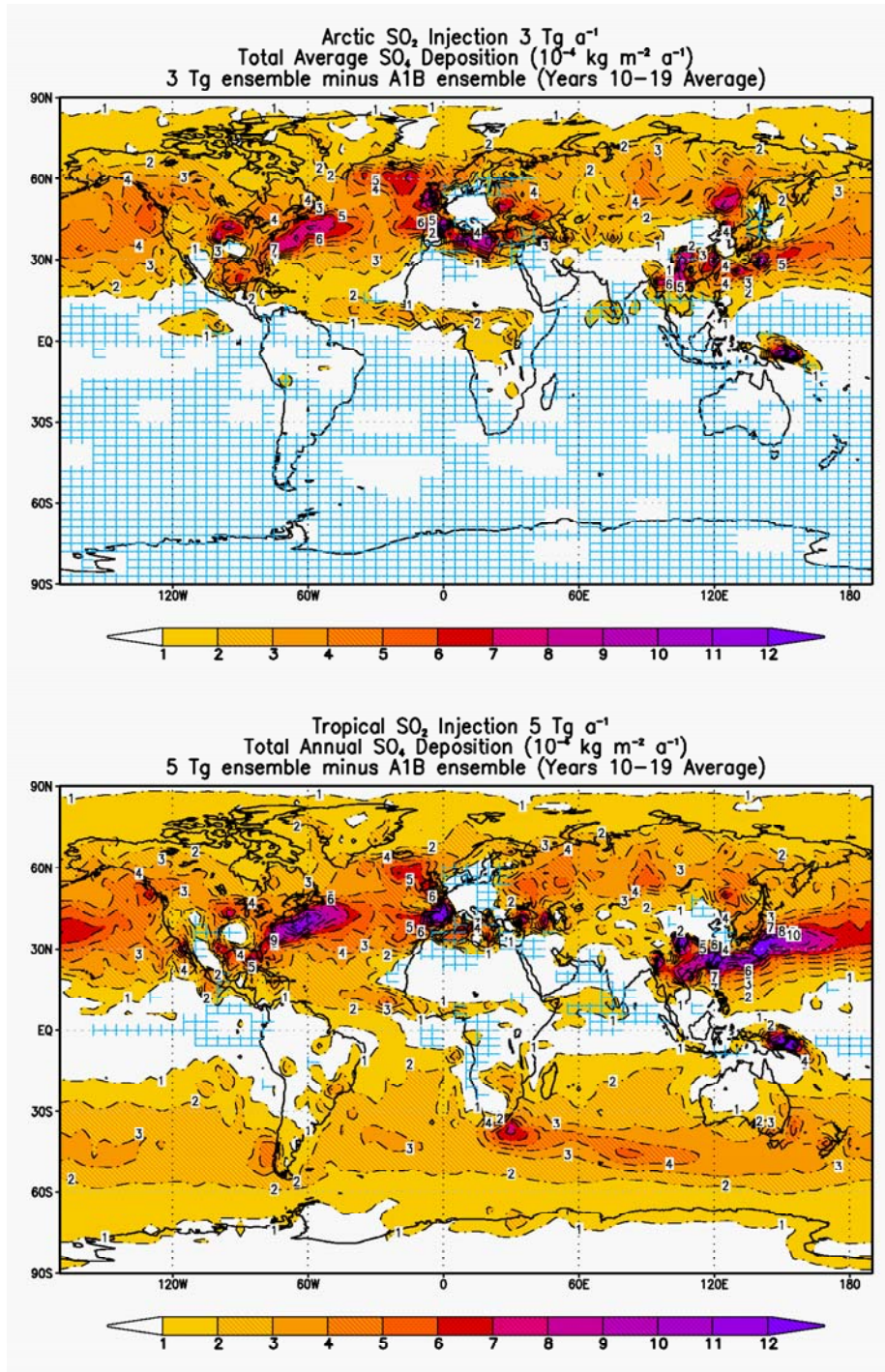
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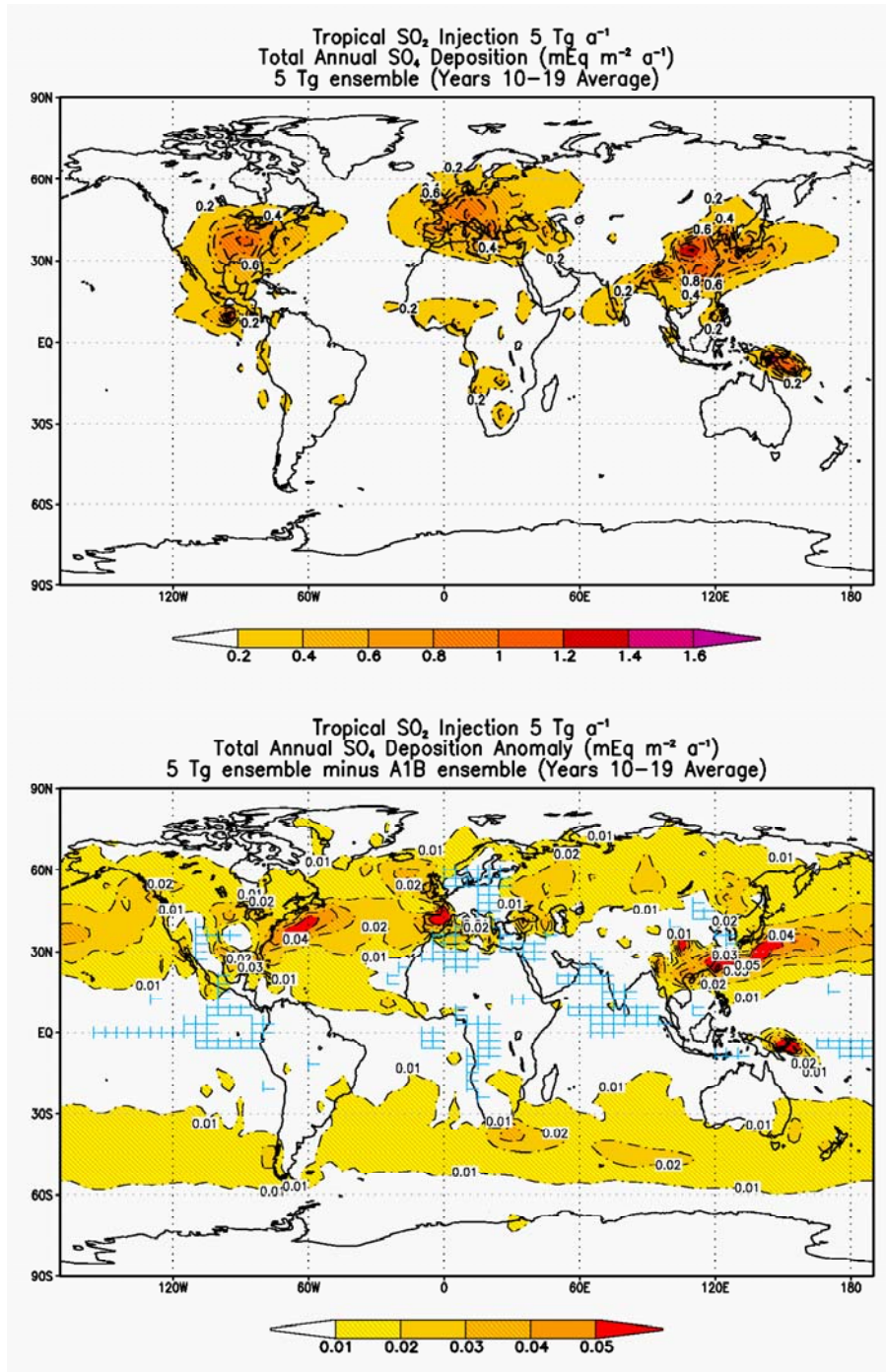
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327 **Figure 1.** Ratios of the geoengineering ensembles (Arctic 3 Tg SO₂ a⁻¹ injection and tropical 5
 328 Tg SO₂ a⁻¹ injection) to the baseline (A1B) ensemble. Both figures show annually averaged total
 329 sulfate deposition averaged over years 10-19 for each experiment. These plots are made from
 330 the model output of the climate simulations performed by *Robock et al.* [2008]. All shaded
 331 values on this figure are statistically significant at a 95% confidence level.
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334 **Figure 2.** Annually averaged total sulfate deposition anomalies (injection minus baseline,
 335 revealing only the additional deposition from geoengineering) for the geoengineering scenarios
 336 of Arctic 3 Tg SO₂ a⁻¹ and tropical 5 Tg SO₂ a⁻¹ injection into the lower stratosphere. The results
 337 are averaged over three ensemble members and for years 10-19 of each experiment. These plots
 338 are made from the model output of the climate simulations performed by *Robock et al.* [2008].
 339 Values not statistically significant at a 95% confidence level are denoted by blue hatching.
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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.