# Acid Deposition from Stratospheric Geoengineering with Sulfate Aerosols

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Submitted to *Geophysical Research Letters*

October, 2008

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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 is injected into the tropical or Arctic stratosphere, the main additional surface deposition 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 acid deposition on terrestrial ecosystems. For annual injection of 3 Tg of $SO_2$ into the Arctic stratosphere or 5 Tg of $SO_2$ into the tropical stratosphere, the maximum total local surface acid deposition of approximately 1.5 mEq $m^{-2}$ $a^{-1}$, and the maximum anomaly of approximately 0.05 mEq $m^{-2}$ $a^{-1}$, are not enough to negatively impact most ecosystems.

## 1. Introduction

Faced with the problem of climate change due to increasing global temperatures, some scientists and policy makers have suggested the deliberate modification of Earth's climate, an activity that has been termed geoengineering. There have been many different suggestions for geoengineering, both recently [e.g., *Angel*, 2006; *Bower et al.*, 2006] as well as historically [*Fleming*, 2007]. However, one method that has received a great deal of recent attention is the suggestion of *Budyko* [1974, 1977], *Dickinson* [1996], and *Crutzen* [2006] to inject aerosols into the stratosphere. Injecting precursors of highly reflective sulfate aerosols into the lower stratosphere would increase the planetary albedo, resulting in cooling [*Rasch et al.*, 2008a]. *Rasch et al.* [2008b] and *Robock et al.* [2008] calculated climate responses using general circulation models.

,Geoengineering, however, will invariably have certain undesirable consequences. *Tilmes et al.* [2008] and *Robock* [2008a] discuss the negative impact these sulfate aerosols will have on polar stratospheric ozone. *Robock* [2008b] lists 20 potential side effects that could result from this method. Our purpose here is to evaluate one of *Robock*'s suggestions, by calculating the amount of acid deposition that would result from geoengineering.

Acid rain has been studied extensively in terms of damage to ecosystems. Excess acid can decrease or even eliminate freshwater fish populations [*Leivestad and Muniz*, 1976], cause foliar leaching [*Wood and Bormann*, 1975], affect plant-parasite interaction [*Shriner*, 1977], significantly reduce lake bacteria populations [*Rao and Dutka*, 1983], and, through forest dieback and reduced food supply, can affect forest bird communities [*Graveland*, 1998]. These, among other potential problems, could present significant ecological concerns, and serve as motivation for the study of acid deposition due to geoengineering.

Whether acid deposition (both dry and wet) is harmful depends on both the amount of acid and the sensitivity of the ecosystem. Here we calculate how much additional sulfuric acid would reach the surface from proposed geoengineering, and compare this to critical load thresholds for different regions.

## 2. Experiment

We studied geoengineering with stratospheric sulfate aerosols using ModelE, a general circulation model developed by the National Aeronautics and Space Administration Goddard Institute for Space Studies. We used the stratospheric version with 4° latitude by 5° longitude horizontal resolution and 23 vertical levels up to 80 km [*Schmidt et al.,* 2006]. It is fully coupled to a 4° latitude by 5° longitude dynamic ocean with 13 vertical levels [*Russell et al.*, 1995]. The aerosol module [*Koch et al.*, 2006] accounts for $SO_2$ conversion to sulfate aerosols, as well as transport and removal of the aerosols. Radiative forcing from the aerosols is fully interactive with the atmospheric circulation. We defined the dry aerosol effective radius as 0.25 μm, which creates hydrated sulfate aerosols with an effective radius of approximately 0.30-0.35 μm.

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

*Robock et al.* [2008] performed a three-member ensemble of 20-year climate simulations under the Intergovernmental Panel on Climate Change A1B scenario [*IPCC*, 2007]. In addition, for geoengineering simulations, *Robock et al.* performed two ensembles, each with three

103 members of 20-year climate simulations, which were compared to the A1B scenario ensemble.
104 One involved daily injections of $SO_2$ into the Arctic lower stratosphere (latitude 68°N) for a total
105 of 3 Tg per year in addition to the A1B forcing, and one involved daily injections into the
106 tropical lower stratosphere (latitude 0°N) for a total of 5 Tg per year in addition to the A1B
107 forcing.

108 **3. Results**

109 Figure 1 shows the annual percent increase in total deposition, averaged over the second
110 decade of geoengineering, over much of the globe, with the exception of the tropics (due to
111 poleward stratospheric transport before mixing into the troposphere). The increases are broad in
112 spatial scope, often reaching 135% of the A1B values. As expected, in the Arctic injection, the
113 increase in deposition is mostly confined to the Northern Hemisphere, whereas both hemispheres
114 are affected in a tropical injection. The majority of the increase is in the form of wet deposition
115 (not shown). In the polluted midlatitudes of the Northern Hemisphere, the increases of acid
116 deposition are not noticeable, but in pristine areas, such as Antarctica, they are. However, as
117 there was little acid deposition in pristine areas in the A1B case, additional deposition of tens of
118 percent may not be consequential, so we must evaluate the actual amount of deposition. Figure 2
119 shows that the increases in actual deposition are strongest in midlatitude bands, frequently as
120 high as 5-10 x $10^{-4}$ kg $m^{-2}$ $a^{-1}$, due to strong cross-tropopause flux in the jet stream region. There
121 are small regions of larger deposition for certain seasons, but the annual average is sufficient for
122 this analysis.

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

124 The significance of the acid deposition increases depends on their potential effects on
125 terrestrial ecosystems. *Kuylenstierna et al.* [2001] used a modeling approach to perform a

126 critical load study on a global scale in which they rank areas by sensitivity to increased acid
127 deposition, a value they determine by evaluating the buffering capacity of each region's soil.
128 Our units of sulfate deposition, kg $m^{-2}$ $a^{-1}$, must be converted to the units found in *Kuylenstierna*
129 *et al.* of mEq $m^{-2}$ $a^{-1}$. We use the definitions

130 $$\text{mEq} = \frac{\text{mass (grams)}}{\text{mEq mass (grams)}} \quad \text{and} \quad \text{mEq mass (grams)} = \frac{\text{atomic weight (g/mol)}}{\text{valence x 1000}}$$

131 The $SO_4^{2-}$ ion has atomic weight 96 g/mole and a valence of 2, giving us mEq mass (grams) of
132 0.048. So

133 $$\frac{1\,\text{kg}}{\text{m}^2 \cdot \text{a}} \cdot \frac{1000\,\text{g}}{1\,\text{kg}} \cdot \frac{1\,\text{mEq x } 0.048}{1\,\text{g}} = 48 \frac{\text{mEq}}{\text{m}^2 \cdot \text{a}}.$$

134 Figure 3 shows total annual deposition (taken as an ensemble average over the second 10
135 years) and the anomaly of total deposition when compared to the A1B scenario, both in terms of
136 these new units. This scenario (5 Tg $a^{-1}$ injection) was chosen because it has the largest point
137 values of sulfate deposition of all scenarios. The maximum total deposition in a given year is
138 approximately 1.5 mEq $m^{-2}$ $a^{-1}$ and the maximum anomaly is approximately 0.05 mEq $m^{-2}$ $a^{-1}$.
139 According to the critical loading studies of *Kuylenstierna et al.*, the most sensitive areas of the
140 globe can receive 25-50 mEq $m^{-2}$ $a^{-1}$ of sulfate deposition before potentially being negatively
141 impacted.

142 In another study, *Skeffington* [2006] takes a very conservative approach to critical load
143 studies by accounting for many means by which a region can be impacted by pollutant
144 deposition, where sulfate is defined as only one pollutant that is considered. He uses models for
145 much of his results, but also uses experimental and field evidence when available. In addition,
146 his purpose is to estimate uncertainty in measurements of critical loading, so the low ends of his
147 ranges for which a load is considered critical can be seen as conservative estimates.

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

150 $$\frac{1\,\mathrm{kEq}}{\mathrm{ha}\cdot\mathrm{a}}\cdot\frac{10^6\,\mathrm{mEq}}{1\,\mathrm{kEq}}\cdot\frac{1\,\mathrm{ha}}{10^4\,\mathrm{m}^2}=100\frac{\mathrm{mEq}}{\mathrm{m}^2\cdot\mathrm{a}}.$$

151 *Skeffington*'s results, with our conversion factor taken into account, show that our values for acid
152 deposition over a year, with the possible exception of waterways, are well under critical loading
153 levels (Table 1). In addition, the area in which our results exceed 1 mEq $m^{-2}$ $a^{-1}$ of sulfate
154 deposition is very small. However, due to our grid size, which is especially large when
155 compared to the size of most waterways, there may be localized areas of enhanced deposition
156 from individual precipitation events that we cannot assess.

157 **5. Conclusions**

158 Analysis of our results and comparison to the results of *Kuylenstierna et al.* [2001] and
159 *Skeffington* [2006] leads to the conclusion that the additional acid deposition that would result
160 from geoengineering will not be sufficient to negatively impact most ecosystems. With the
161 exception of waterways, every region has a critical loading value a full order of magnitude above
162 the total amount of acid deposition that would occur under the geoengineering scenario. Even
163 waterways would receive at most 0.05 mEq $m^{-2}$ $a^{-1}$ in additional sulfate deposition, meaning only
164 those waterways which are most sensitive to small amounts of additional deposition would be
165 negatively impacted.

166

167 **Acknowledgments.** We thank Greg Carmichael for pointing us to relevant references on acid
168 deposition. Model development and computer time at Goddard Institute for Space Studies are
169 supported by National Aeronautics and Space Administration climate modeling grants. This
170 work is supported by NSF grant ATM-0730452.

171 **References**

172 Angel, R. (2006), Feasibility of cooling the Earth with a cloud of small spacecraft near the inner
173 Lagrange point (L1), *Proc. Nat. Acad. Sci., 103*, 17,184-17,189.
174 Bower, K., T. Choularton, J. Latham, J. Sahraei, and S. Salter (2006), Computational assessment
175 of a proposed technique for global warming mitigation via albedo-enhancement of marine
176 stratocumulus clouds, *Atm. Res.*, *82*, 328-336.
177 Budyko, M. I. (1974), *Climate and Life* (Academic Press, New York, NY), 508 pp.
178 Budyko, M. I. (1977), *Climatic Changes* (American Geophysical Union, Washington, DC), 261
179 pp.
180 Crutzen, P. (2006), Albedo enhancement by stratospheric sulfur injections: A contribution to
181 resolve a policy dilemma? *Climatic Change*, *77*, 211-219.
182 Dickinson, R. E. (1996), Climate engineering: A review of aerosol approaches to changing the
183 global energy balance, *Climatic Change*, *33*, 279-290.
184 Fleming, J. R. (2007), The climate engineers, *Wilson Quarterly*, *Spring 2007*, 46-60.
185 Graveland, J. (1998), Effects of acid rain on bird populations, *Environ. Rev.*, *6*(1), 41-54,
186 doi:10.1139/er-6-1-41.
187 IPCC (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working*
188 *Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate*
189 *Change*, S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor
190 and H. L. Miller, Eds., (Cambridge University Press, Cambridge, United Kingdom and New
191 York, NY, USA), 996 pp.
192 Koch, D., G. A. Schmidt, and C. V. Field (2006), Sulfur, sea salt, and radionuclide aerosols in
193 GISS ModelE, *J. Geophys. Res.*, *111*, D06206, doi:10.1029/2004JD005550.

194 Kuylenstierna, J. C. I., H. Rodhe, S. Cinderby, and K. Hicks (2001), Acidification in developing
195 countries: ecosystem sensitivity and the critical load approach on a global scale, *Ambio*,
196 *30*(1), 20-28.
197 Leivestad, H., and I. P. Muniz (1976), Fish kill at low pH in a Norwegian river, *Nature*, *259*,
198 391-392, doi:10.1038/259391a0.
199 Rao, S. S., and B. J. Dutka (1983), Influence of acid precipitation on bacterial populations in
200 lakes, *Hydrobiologia*, *98*(2), 153-157, doi:10.1007/BF02185633.
201 Rasch, P. J., et al. (2008a), An overview of geoengineering of climate using stratospheric sulfate
202 aerosols, *Phil. Trans. Royal Soc. A.*, doi:10.1098/rsta.2008.0131, published online, 31 pp.
203 Rasch, P. J., P. J. Crutzen, and D. B. Coleman (2008b), Exploring the geoengineering of climate
204 using stratospheric sulfate aerosols: The role of particle size, *Geophys. Res. Lett.*, *35*,
205 L02809, doi:10.1029/2007GL032179.
206 Robock, A. (2008a), Whither geoengineering? *Science*, *320*, 1166-1167.
207 Robock, A. (2008b), 20 reasons why geoengineering may be a bad idea, *Bull. Atomic Scientists*,
208 *64*(2), 14-18, 59, doi:10.2968/064002006.
209 Robock, A., L. Oman, and G. Stenchikov (2008), Regional climate responses to geoengineering
210 with tropical and Arctic $SO_2$ injections, *J. Geophys. Res.*, *113*, D16101, doi:10.1029/
211 2008JD010050.
212 Russell, G. L., J. R. Miller, and D. Rind (1995), A coupled atmosphere-ocean model for transient
213 climate change, *Atmos.-Ocean*, *33*, 683-730.
214 Schmidt, G. A., et al. (2006), Present day atmospheric simulations using GISS ModelE:
215 Comparison to in situ, satellite and reanalysis data, *J. Climate*, *19*, 153-192.

216 Shriner, D. S. (1977), Effects of simulated rain acidified with sulfuric acid on host-parasite
217 interactions, *Water, Air, & Soil Pollution*, *8*(1), 9-14, doi:10.1007/BF00156719.
218 Skeffington, R. A. (2006), Quantifying uncertainty in critical loads: (A) literature review, *Water,*
219 *Air, and Soil Pollution*, *169*, 3-24.
220 Tilmes, S., R. Müller, and R. Salawitch (2008), The sensitivity of polar ozone depletion to
221 proposed geoengineering schemes, *Science*, *320*(5880), 1201-1204, doi:10.1126/
222 science.1153966.
223 Wood, T., and F. H. Bormann (1975), Increases in foliar leaching caused by acidification of an
224 artificial mist, *Ambio*, *4*, 169-171.
225

225 **Table 1.** Ranges of critical loading of pollutant deposition (including sulfur) for various sites in
226 Europe as reported by *Skeffington* [2006].
227

| **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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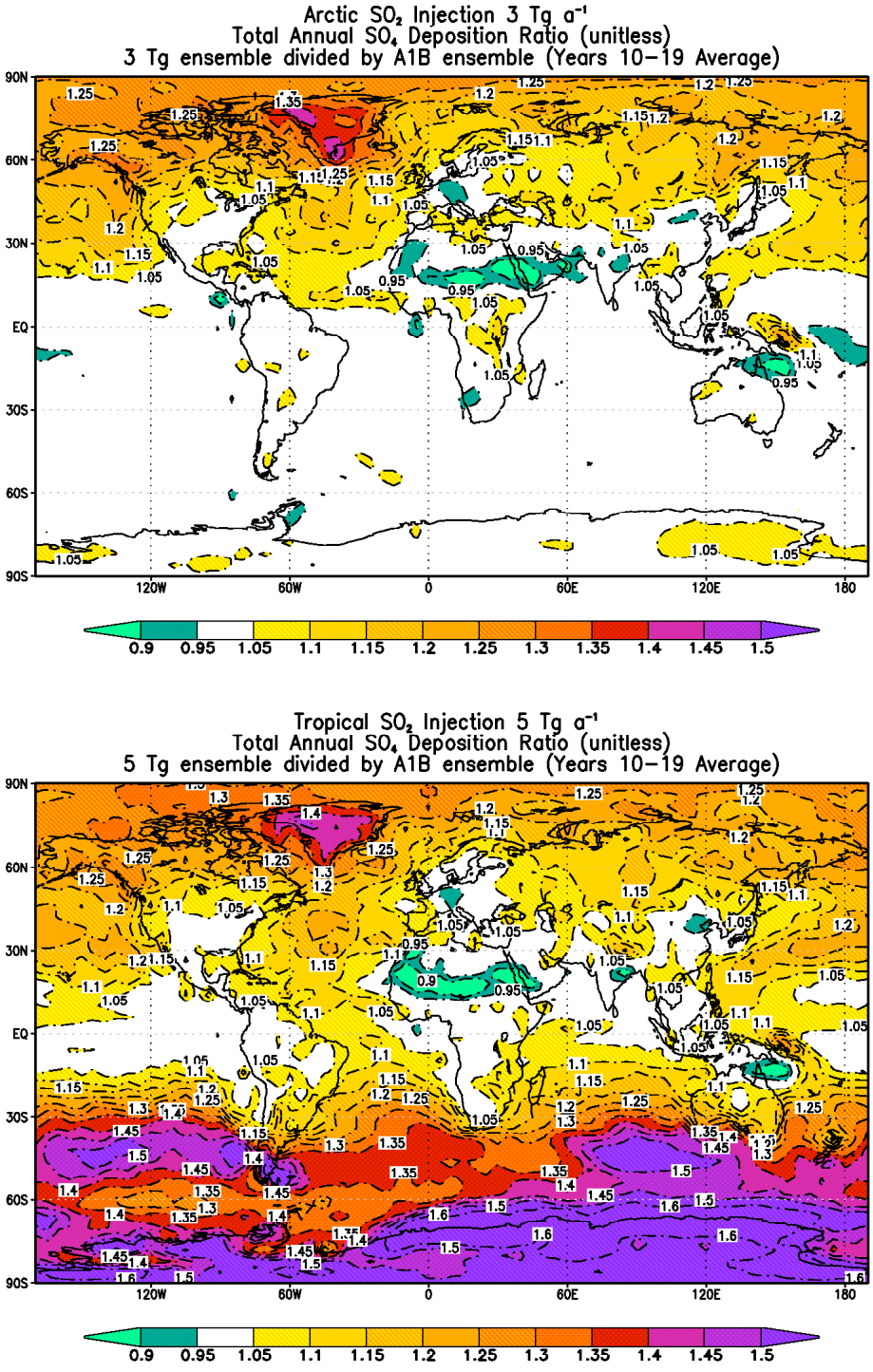


**Figure 1.** Ratios of the geoengineering ensembles (Arctic 3 Tg $SO_2$ $a^{-1}$ injection and tropical 5 Tg $SO_2$ $a^{-1}$ injection) to the A1B ensemble. Both figures show annually averaged total sulfate deposition averaged over years 10-19 for each experiment. These plots are made from the model 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.

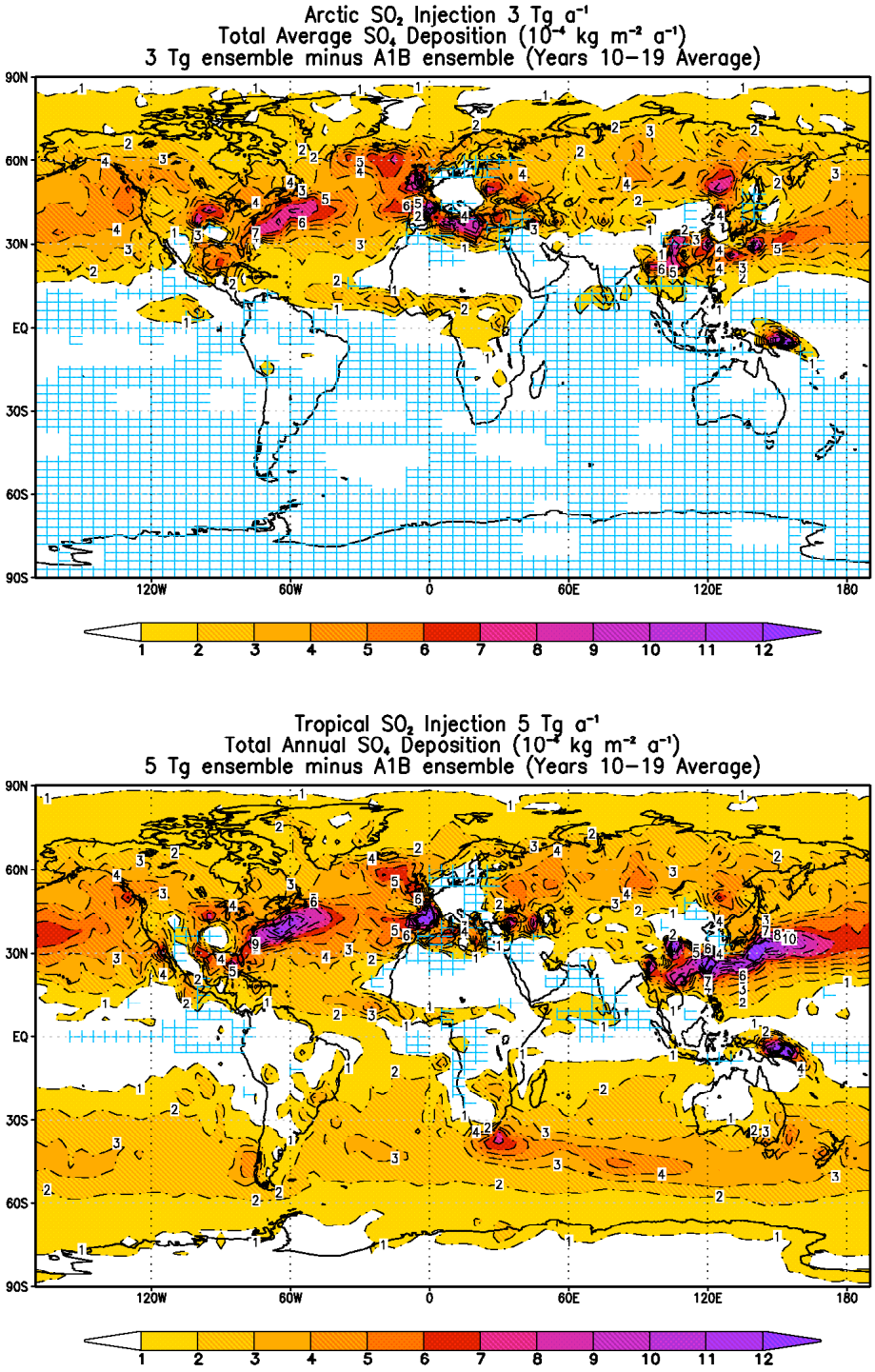


**Figure 2.** Annually averaged total sulfate deposition anomalies for the geoengineering scenarios of Arctic 3 Tg $SO_2$ $a^{-1}$ and tropical 5 Tg $SO_2$ $a^{-1}$ injection into the lower stratosphere. The results are averaged over three ensemble members and for years 10-19 of each experiment. These 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.

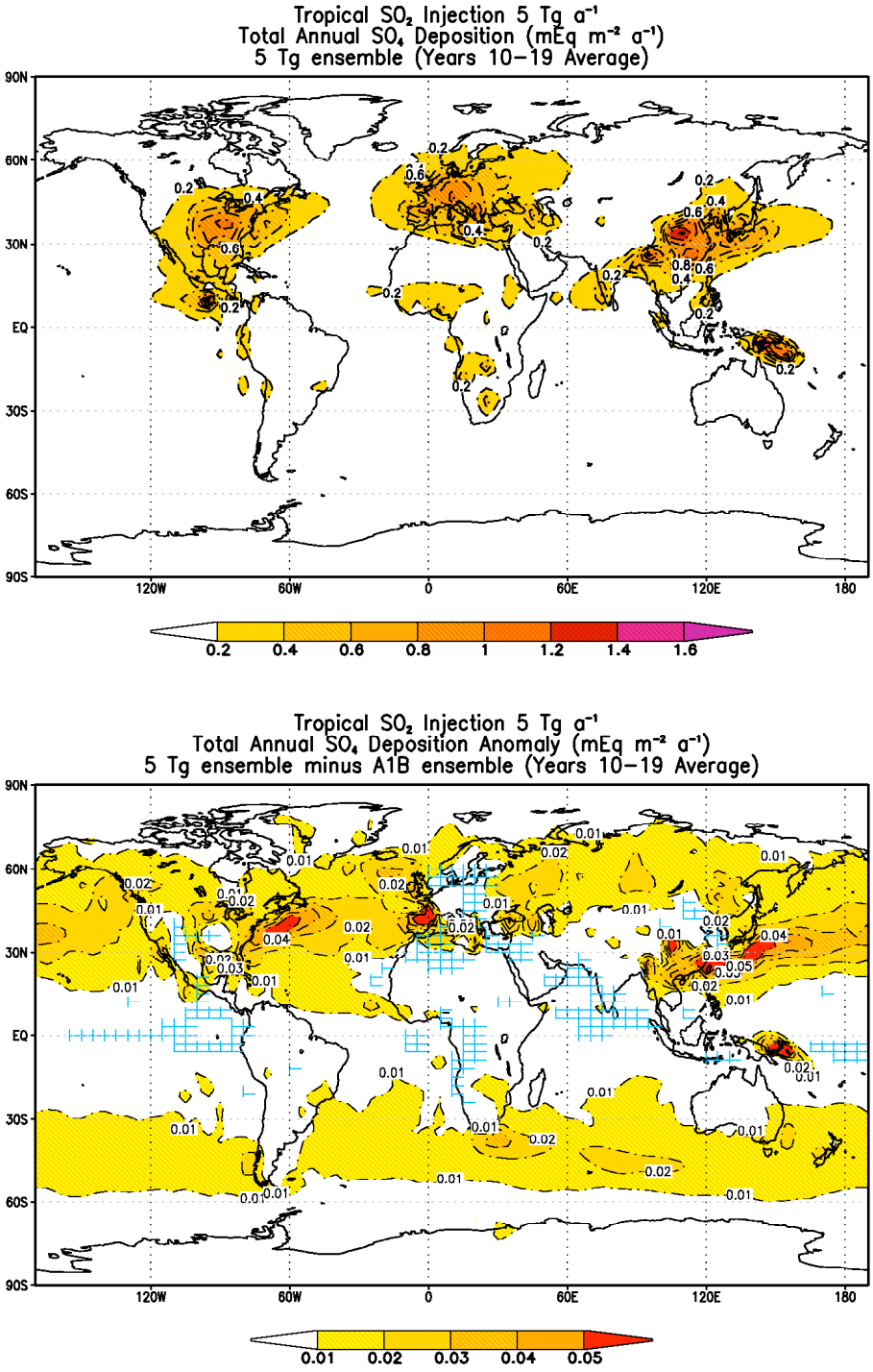


**Figure 3.** Total sulfate deposition and total deposition anomaly for a tropical 5 Tg $a^{-1}$ injection. The largest total sulfate deposition value is approximately 1.5 mEq $m^{-2}$ $a^{-1}$, and the largest anomaly value is approximately 0.05 mEq $m^{-2}$ $a^{-1}$. These plots are made from the model 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.