



Sulfuric acid deposition from stratospheric geoengineering with sulfate aerosols

Ben Kravitz,¹ Alan Robock,¹ Luke Oman,² Georgiy Stenchikov,^{1,3} and Allison B. Marquardt¹

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[1] 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, neither the maximum point value of sulfate deposition of approximately 1.5 mEq m⁻² a⁻¹ nor the largest additional deposition that would result from geoengineering of approximately 0.05 mEq m⁻² a⁻¹ is enough to negatively impact most ecosystems.

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1. Introduction

[2] 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 gaseous aerosol precursors into the stratosphere. The creation of highly reflective sulfate aerosols in the lower stratosphere would result in some warming of the stratosphere, but the aerosol cloud would also tend to increase the planetary albedo, resulting in cooling of the troposphere and the surface [Rasch *et al.*, 2008a]. Rasch *et al.* [2008b] and Robock *et al.* [2008] calculated climate responses to this aerosol cloud using general circulation models.

[3] Geoengineering will, however, invariably have certain undesirable consequences. Tilmes *et al.* [2008] and Robock [2008a] discussed the negative impact these sulfate aerosols will have on polar stratospheric ozone. Robock [2008b] listed 20 potential side effects that could result from this

method. Our purpose here is to evaluate one of Robock's concerns, quantifying the amount of sulfur deposition that would result from two potential scenarios of geoengineering with sulfate aerosols. This is of concern because the sulfate aerosol can hydrate to form sulfuric acid, meaning geoengineering with sulfate aerosols can potentially result in an increase in acid deposition.

[4] Acid rain has been studied extensively in terms of its effects on ecosystems. Sulfur is a necessary nutrient for some plants, and the need to add sulfur to crops has long been recognized by farmers [Hart and Peterson, 1911]. However, an increase in sulfur deposition will not universally benefit ecosystems, especially ones that are poorly buffered against an increase in acidity. For example, 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 our motivation for the study of sulfate deposition due to geoengineering.

[5] Whether sulfate deposition (both dry and wet) is harmful depends on the amount of sulfur introduced into the system, the amount of sulfate that is hydrated to form sulfuric acid, and the sensitivity of the ecosystem. We will base our calculations on an upper limit, i.e., that all the sulfur deposition is sulfuric acid. This is likely an overestimation, since wet deposition in the model accounts for approximately 65% of total sulfate deposition, and dry deposition accounts for the remainder. Moreover, not all

¹Department of Environmental Sciences, Rutgers University, New Brunswick, New Jersey, USA.

²Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, Maryland, USA.

³King Abdullah University of Science and Technology, Thuwal, Saudi Arabia.

sulfate deposition will result in an increase in acid rain. Here we calculate how much additional sulfate would reach the surface from proposed geoengineering and compare this to critical load thresholds for different regions.

[6] As of now, most of the discussion of geoengineering with sulfate aerosols has focused on using SO_2 as the preferred sulfate aerosol precursor. Volcanic eruptions can inject a large pulse of SO_2 into the lower stratosphere, and previous geoengineering studies have considered volcanic eruptions as an analog of geoengineering. However, other precursors, such as hydrogen sulfide, carbonyl sulfide, and ammonium sulfate, could also potentially be used. Regardless, the important factor in determining sulfate deposition is the amount of sulfur injected into the stratosphere. As such, the results presented in this paper need only be scaled appropriately according to the aerosol precursor's molecular weight.

2. Experiment

[7] 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 [Schmidt *et al.*, 2006]. We used the stratospheric version with 4° latitude by 5° longitude horizontal resolution and 23 vertical levels up to 80 km. It is fully coupled to a 4° latitude by 5° longitude dynamic ocean with 13 vertical levels [Russell *et al.*, 1995].

[8] The aerosol module [Koch *et al.*, 2006] accounts for SO_2 conversion to sulfate aerosols, as well as transport and removal of the aerosols. The chemical model calculates the sulfur cycle in the stratosphere, where the conversion rate of SO_2 to sulfate is based on the respective concentrations of SO_2 and the hydroxyl radical, the latter of which is prescribed [Oman *et al.*, 2006]. The dry aerosol effective radius is specified to be $0.25 \mu\text{m}$, and the model hydrates the aerosols on the basis of ambient humidity values, resulting in a distribution of hydrated aerosols with an effective radius of approximately $0.30\text{--}0.35 \mu\text{m}$. Radiative forcing from the aerosols is fully interactive with the atmospheric circulation.

[9] Koch *et al.* [2006] thoroughly analyzed the performance of ModelE concerning sulfur deposition from tropospheric sources. The model has some biases in that it produces 50–67% of the observed sulfur 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 sulfur 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.

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

[11] In addition, we used two ensembles, each with three members of 20-year climate simulations, covering the same time period. One involved daily injections of SO_2 into the tropical lower stratosphere (longitude 120°E , latitude 0° , 16–23 km altitude) for a total of 5 Tg per year in addition to the forcing prescribed by the A1B scenario, and one involved daily injections of SO_2 into the Arctic lower stratosphere (longitude 120°E , latitude 68°N , 10–15 km altitude) for a total of 3 Tg per year in addition to the forcing prescribed by the A1B scenario. The value of 5 Tg per year was chosen to correspond to a Mount Pinatubo–sized eruption every 4 years, which was a value determined by Robock *et al.* [2008] as being sufficient to cause substantial enough cooling to offset the climatic effects of an increase in greenhouse gas concentrations. The smaller value of 3 Tg per year was also chosen by Robock *et al.*, since the goal of the original experiment was to limit the climate response only in the Arctic. The longitude value chosen is arbitrary and irrelevant, since the prevailing general circulation will transport the gas/aerosol cloud around the globe within a matter of weeks.

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

3. Results

[13] Figure 1 shows the annual percent increase in total sulfate deposition, averaged over the second decade of geoengineering. In the tropical injection case, there is an increase in sulfate deposition over much of the globe, with the exception of the tropics (owing to poleward stratospheric transport before mixing into the troposphere). As expected, in the Arctic injection, the increase in deposition is mostly confined to the Northern Hemisphere. The majority of the increase is in the form of wet deposition (not shown). In the polluted midlatitudes of the Northern Hemisphere, the increases of sulfate deposition are not noticeable, but in pristine areas, such as Antarctica, they are readily apparent. Although all shaded values in Figure 1 are statistically significant at a 95% confidence level, for the Arctic injection case, many of the shaded values in the Southern Hemisphere are most likely due to weather noise.

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

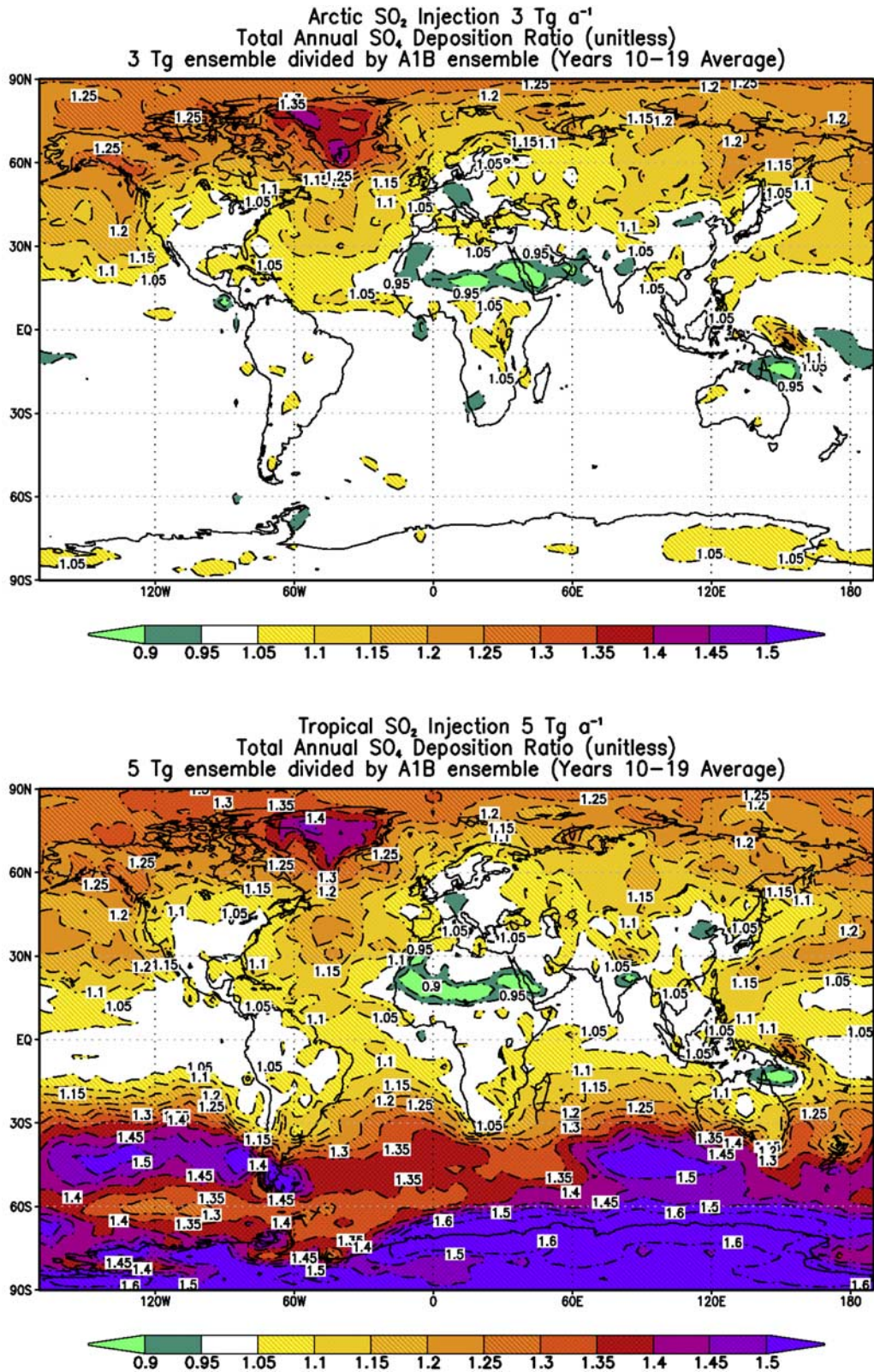


Figure 1. Ratios of the geoengineering ensembles of (top) Arctic 3 Tg SO₂ a⁻¹ injection and (bottom) tropical 5 Tg SO₂ a⁻¹ injection to the baseline (A1B) ensemble. Shown are annually averaged total sulfate deposition averaged over years 10–19 for each experiment. These plots are made from the model output of the climate simulations performed by *Robock et al.* [2008]. All shaded values are statistically significant at a 95% confidence level.

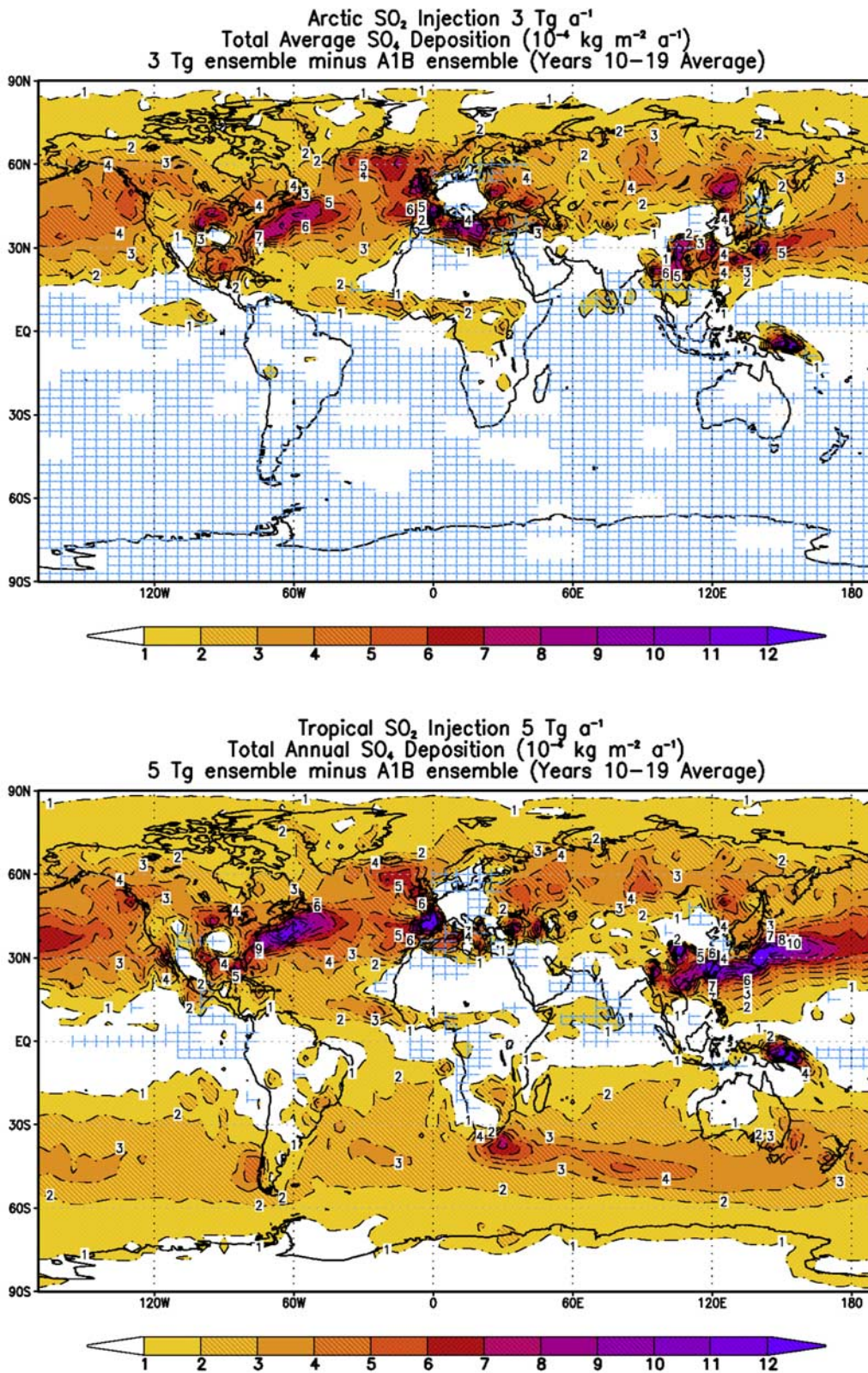


Figure 2. Annually averaged total sulfate deposition anomalies (injection minus baseline, revealing only the additional deposition from geoengineering) for the geoengineering scenarios of (top) Arctic 3 Tg SO₂ a⁻¹ and (bottom) tropical 5 Tg SO₂ a⁻¹ injection into the lower stratosphere. The results are 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.

stratosphere to troposphere is comparatively large in these areas.

[15] For the purpose of establishing a reference value for comparison, the baseline surface sulfur emission levels are 135.8 Tg a^{-1} globally [Koch *et al.*, 2006]. Since the additional stratospheric injections are 1–2 orders of magnitude smaller, we might not expect them to be important in any case on a global basis. Dividing the surface emissions by the surface area of 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 an average atmospheric lifetime of 6.2 days, meaning levels would be expected to be much higher than this reference value downwind of large urban and industrial areas and much lower (or practically negligible) in unpopulated areas.

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

[17] Figure 1 only shows annually averaged results. There are small regions of larger deposition for certain seasons, but the annual average is sufficient for this analysis. However, as greenhouse gas concentrations increase in the future, the strength of the Brewer-Dobson circulation will also increase, resulting in a shorter lifetime for stratospheric aerosols and the need for more sulfur to produce the same climate response [Rasch *et al.*, 2008b], which would cause an increase in sulfate deposition. We have not evaluated the effects that an increase in the strength of stratospheric circulation would have with regard to our study.

4. Impacts of Additional Acid Deposition

[18] The significance of the sulfate deposition increases depends on their potential effects on the ecosystems over which the deposition occurs. Section 5 is devoted to the potential effects on the ocean, so in this section, we concentrate on terrestrial ecosystems. Although the graphs only show sulfate deposition, for the purposes of establishing an upper limit to potential negative effects, we will assume that all sulfate due to geoengineering reacts to form sulfuric acid.

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

$$\text{mEq} = \frac{\text{mass(grams)}}{\text{mEq mass(grams)}} \text{ and}$$

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

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

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

[21] Figure 3 refers to the 5 Tg a^{-1} injection scenario. It shows total annual sulfate deposition (taken as an ensemble average over the second decade of geoengineering) and the annual sulfate deposition just due to geoengineering (injection minus baseline), both in terms of these new units. The 5 Tg a^{-1} injection scenario was chosen because it has larger sulfate deposition than the Arctic 3 Tg a^{-1} scenario, although the results presented in Figure 3 are similar for the Arctic 3 Tg a^{-1} injection case. The maximum point value for total deposition is approximately $1.5 \text{ mEq m}^{-2} \text{ a}^{-1}$, and the largest point value which is solely the result of geoengineering (injection minus baseline) is approximately $0.05 \text{ mEq m}^{-2} \text{ a}^{-1}$. According to the critical loading studies of *Kuylenstierna et al.* [2001], the most sensitive areas of the globe can receive $25\text{--}50 \text{ mEq m}^{-2} \text{ a}^{-1}$ of sulfate deposition before potentially being negatively impacted.

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

[23] *Skeffington's* [2006] results are given in terms of $\text{kEq ha}^{-1} \text{ a}^{-1}$, so we must again perform a conversion:

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

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

5. Ocean Acidification

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

[25] *Raven et al.* [2005] estimated that over 500 Gt ($5 \times 10^{17} \text{ g}$) of carbon dioxide has dissolved in the oceans over the past 200 years. Knowing that carbonic acid is a weak acid and that the atomic weight of carbon dioxide is 44 g/mol, we can put this value in terms of mEq by using our

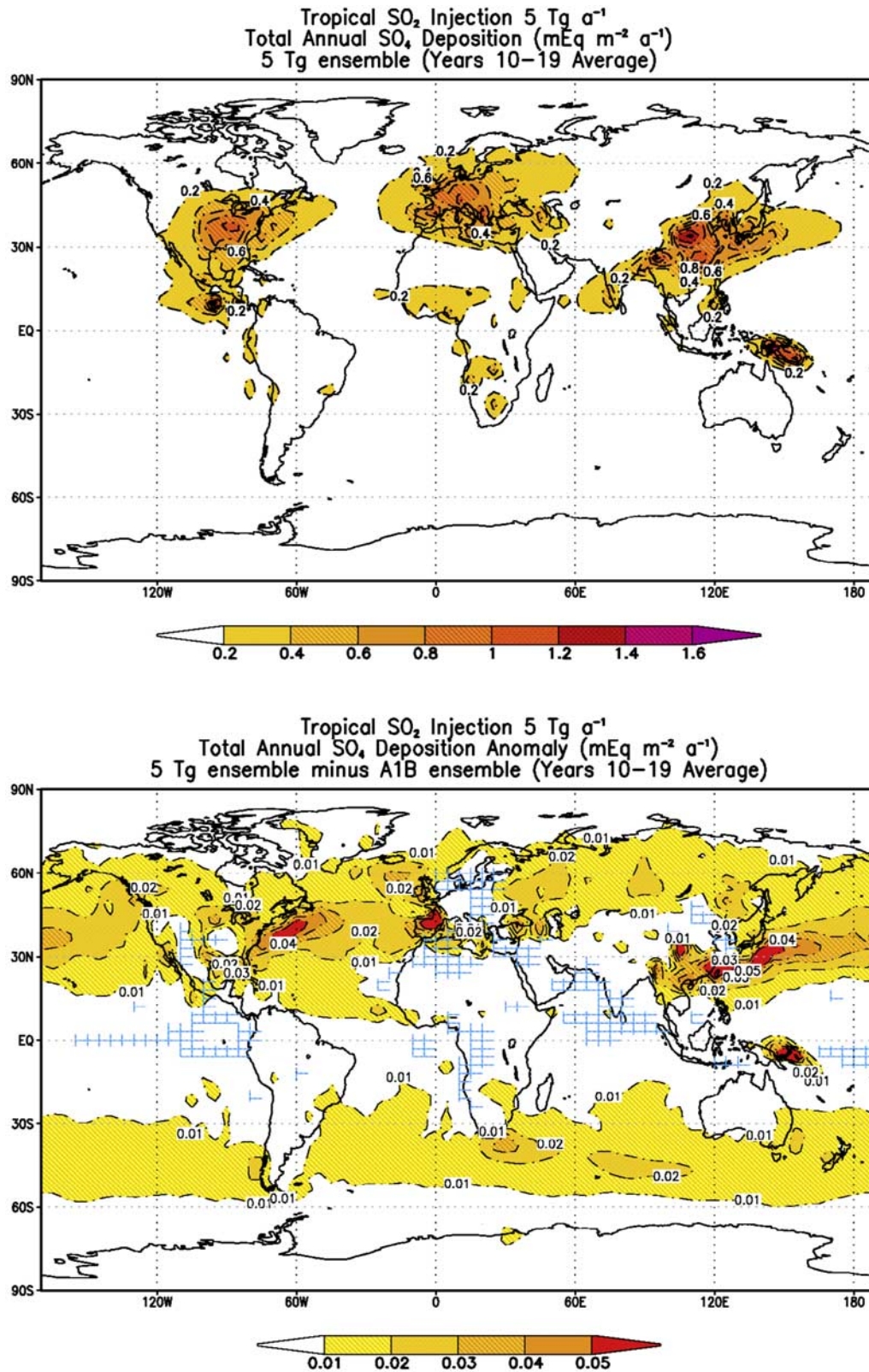


Figure 3. Results for a tropical 5 Tg a^{-1} injection. (top) Total sulfate deposition (geoengineering plus baseline). (bottom) Sulfate deposition anomaly (injection minus baseline, revealing only the additional deposition from geoengineering). The largest total sulfate deposition point value is approximately $1.5 \text{ mEq m}^{-2} \text{ a}^{-1}$, and the largest anomaly point value is approximately $0.05 \text{ mEq m}^{-2} \text{ a}^{-1}$. 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.

Table 1. Ranges of Critical Loading of Pollutant Deposition, Including Sulfur, for Various Sites in Europe as Reported by Skeffington [2006]

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 UK	24–182
Aber in north Wales	32–134
Uhlirska in Czech Republic	260–358
Fåråhall in Sweden	29–134
Several varied sites in China (sulfur only)	63–880
Waterways in Sweden	1–44

previous definitions. Thus we conclude that 1×10^{19} mEq of carbon dioxide has dissolved in the ocean. Since the ocean covers approximately 70% of the Earth's surface, we can divide by the surface area covered by the ocean, as well as dividing by the 200 years over which this process occurred, to get

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

where R_E is the radius of Earth. This deposition is 2 orders of magnitude larger than our highest potential value of sulfuric acid deposition, again assuming all sulfate due to geoengineering is reacted to form sulfuric acid, leading us to conclude that the increase in acid deposition resulting from geoengineering with stratospheric sulfate aerosols is not enough to negatively impact the oceans.

6. Conclusions

[26] Analysis of our results and comparison to the results of Kuylensstierna et al. [2001] and Skeffington [2006] lead to the conclusion that the additional sulfate deposition that would result from geoengineering will not be sufficient to negatively impact most ecosystems, even under the assumption that all deposited sulfate will be in the form of sulfuric acid. However, although these model results are feasible, should geoengineering with sulfate aerosols actually be conducted, local results due to weather variability may differ from the results presented here. With the exception of terrestrial waterways, every region has a critical loading value a full order of magnitude above the largest potential total amount of acid deposition that would occur under the geoengineering scenarios presented in this paper. Furthermore, our results show that additional sulfate deposition tends to preferentially occur over oceans, meaning the chance of such a sensitive ecosystem receiving enough additional sulfate deposition to suffer negative consequences is very small.

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B. Kravitz, A. B. Marquardt, A. Robock, and G. Stenchikov, Department of Environmental Sciences, Rutgers University, 14 College Farm Road, New Brunswick, NJ 08901, USA. (benkravitz@envsci.rutgers.edu; abmarq18@eden.rutgers.edu; robock@envsci.rutgers.edu; gera@envsci.rutgers.edu)
L. Oman, Department of Earth and Planetary Sciences, Johns Hopkins University, 3400 North Charles Street, Baltimore, MD 21218, USA. (oman@jhu.edu)