



Regional and global trends in sulfate aerosol since the 1980s

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[1] In the last two decades anthropogenic SO₂ emissions have decreased across Europe and North America but have increased across Asia. Long-term surface observations suggest that atmospheric sulfate concentrations have followed trends in sulfur emissions more closely across Asia, than across the USA and Europe. We use a global model of chemistry and aerosol to understand changes in the regional sulfur budget between 1985 and 2000. For every 1% decrease in SO₂ emissions over Europe and the USA the modelled sulfate column burden decreased by 0.65%, while over Asia a 1% increase in SO₂ resulted in a 0.88% increase in sulfate. The different responses can be explained by the availability of oxidant in cloud. We find that because emissions have moved southward to latitudes where in-cloud oxidation is less oxidant limited, the 12% reduction in global SO₂ emissions between 1985 and 2000 caused only a 3% decrease in global sulfate. **Citation:** Manktelow, P. T., G. W. Mann, K. S. Carslaw, D. V. Spracklen, and M. P. Chipperfield (2007), Regional and global trends in sulfate aerosol since the 1980s, *Geophys. Res. Lett.*, *34*, L14803, doi:10.1029/2006GL028668.

1. Introduction

[2] The atmospheric sulfur cycle is important for both climate and air quality. Sulfate aerosols can affect the radiative balance of the Earth through scattering solar radiation and by increasing cloud albedo and lifetime [Intergovernmental Panel on Climate Change, 2001]. Sulfate is produced from the oxidation of SO₂, which has strong sources across North America, Europe and East Asia. Anthropogenic emissions of SO₂ have decreased across North America and Europe since the 1980s following the introduction of cleansing technology and a switch to cleaner fuels, while emissions have increased across Asia in response to economic growth. These changes have resulted in a southward shift in the global distribution of northern hemisphere sulfur emissions. Although atmospheric concentrations of sulfate and SO₂ have responded to this change, long term measurements suggest that the magnitude of the response has varied regionally. Across the UK, France and Germany between 1980 and 2000 sulfate mass concentrations decreased by 50–70% in response to 90% reductions in emissions and measured concentrations of SO₂ [Lövlad *et al.*, 2004]. Across eastern USA sulfate concentrations decreased by 26% between 1989 and 1995 as SO₂ concentrations decreased by 35% [Holland *et al.*, 1998]. Similarly, in Canada, decreases in sulfate are lower

than those of SO₂, a pattern that is repeated in several North American studies reviewed by Reid *et al.* [2001]. Contrary to this, Prospero *et al.* [2003] observed that anthropogenic sulfate concentrations at Midway Island, Pacific had grown at approximately the same rate as SO₂ emissions from East Asia. This relationship between sulfur source and resulting sulfate burden must be understood if we are to deduce climate forcing from future emissions and scenarios, as well as meet air quality targets.

[3] A number of models have been used to understand the atmospheric sulfur cycle (reviewed in Barrie *et al.* [2001], and more recently Berglen *et al.* [2004] and Pham *et al.* [2005]). Berglen *et al.* [2004] used the OsloCTM2 model to understand how the global sulfur cycle had responded to a change in the spatial distribution of emissions between 1985 and 1996. Global anthropogenic SO₂ emissions were practically unchanged over this period, but the peak northern hemisphere source shifted from 30°N–60°N to 15°N–50°N. However, current estimates suggest that SO₂ emissions have now decreased below 1985 values. Stern [2006] estimated that global anthropogenic emissions decreased by 24% between 1987 and 2000, while Streets *et al.* [2006] estimated a 13% decrease from 1988 to 2000. It is hence timely to re-evaluate the effect on measured and modelled sulfur burdens. In this paper we use a global model to quantify changes in the sulfur budget following the decrease in global anthropogenic emissions in the period 1985–2000. We also examine the modelled changes in SO₂ and sulfate over this period and compare them against long-term observations.¹

2. Model Description

[4] We use the aerosol sub-model being developed for the UK Chemistry and Aerosol (UKCA) model (<http://www.ukca.ac.uk>). The scheme is implemented in the global offline 3-D chemical transport model, TOMCAT, incorporating a 7 sulfur species chemistry scheme (see Spracklen *et al.* [2005] for details). The runs in this paper have 2.8° × 2.8° horizontal resolution and 31 hybrid σ -p levels extending from the surface to 10 hPa. Large scale atmospheric transport and meteorology is specified from European Centre for Medium-Range Weather Forecasts (ECMWF) analyses at 6-hourly intervals.

[5] The UKCA aerosol scheme is multicomponent and multi-modal, transporting particle number and component mass concentrations in several log-normal modes. It follows the framework of the M7 model [Vignati *et al.*, 2004] carrying sulfate, sea salt, black carbon, organic carbon and dust in 7 internally mixed modes. For the runs shown

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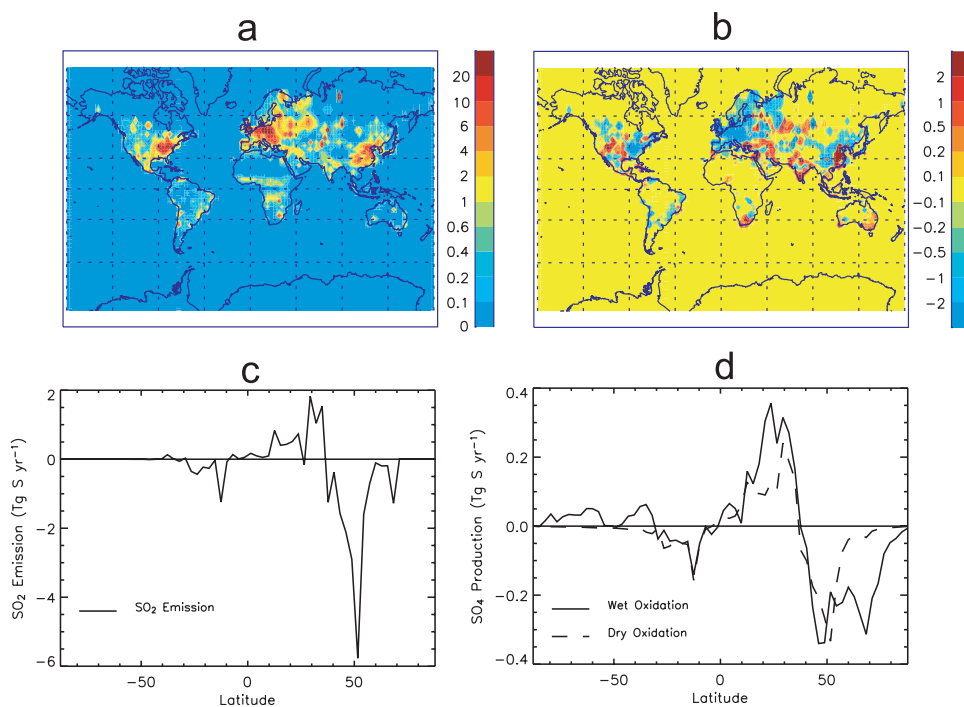


Figure 1. (a) Anthropogenic SO₂ emission inventory for 1985 and (b) absolute change in emission by 2000. Units = mg (S) m⁻² day⁻¹. (c) Change in zonal mean emission of SO₂ and (d) change in zonal mean production of sulfate through in-cloud oxidation (solid line) and dry oxidation (dotted line). Units = Tg (S) yr⁻¹.

here, dust is not included and the geometric standard deviation is fixed for each mode.

[6] The model incorporates full aerosol microphysics including nucleation, condensation, coagulation and cloud processing, together with dry and wet deposition of gas and aerosol. Anthropogenic SO₂, volcanic SO₂ [Halmer *et al.*, 2002], oceanic dimethyl sulfide (DMS) [Kettle *et al.*, 1999; Nightingale *et al.*, 2000], anthropogenic primary sulfate, sea spray [Gong, 2003], primary OC/BC from biofuel and fossil fuel [Bond *et al.*, 2004], as well as biomass burning SO₂ and OC/BC [Van der Werf *et al.*, 2003] emissions are included in the model. Concentrations of the oxidants OH, NO₃, H₂O₂, HO₂ and O₃ are interpolated temporally using 6-hourly monthly mean 3-D concentrations from TOMCAT runs with a comprehensive tropospheric chemistry scheme [Arnold *et al.*, 2005]. SO₂ is oxidised in cloud by H₂O₂ and O₃ assuming a pH of 4 or 5, depending on the concentration of SO₂ in cloud. There is a pseudo-coupling of oxidants and sulfur species: Concentrations of H₂O₂ are prescribed initially, but are allowed to deplete due to reaction with SO₂ and replenish from HO₂. The replenished concentration of H₂O₂ is not allowed to exceed the initial prescribed concentration. In-cloud oxidation is assumed to occur only in low clouds, which are specified from monthly mean International Satellite Cloud Climatology Project data, while convective and dynamic rain remove aerosol through nucleation scavenging.

[7] To simulate the effect of the shift in SO₂ emissions on the sulfur cycle, the GEIA 1b [Benkovitz, 1996] and AEROCOM [Cofala *et al.*, 2005] SO₂ emission inventories for 1985 (S85) and 2000 (S00) were used. International shipping was removed from each inventory for consistency and the emission heights of surface (<100 m) and elevated

(>100 m) sources were consistent for the two scenarios. The seasonality from the GEIA emissions was applied to the annual AEROCOM emissions. The S85 anthropogenic SO₂ emission inventory and the change in SO₂ emission by S00, are shown as an average over June and December in Figures 1a and 1b. Total global anthropogenic SO₂ emissions decrease by 21% between S85 and S00, which is in good agreement with the 24% decrease estimated by Stern [2006] over the period of 1987 to 2000.

[8] The results presented here are a mean over June and December following a nine week model spin up prior to each of these months. Year 2000 meteorology and oxidants were used for both emission scenarios. With the exception of the anthropogenic SO₂ emissions inventory, all other emissions including natural sulfur are identical for both S85 and S00.

3. Results and Discussion

3.1. Modelled and Observed Changes in Regional Sulfate

[9] Modelled sulfur budgets and burdens corresponding approximately to the USA (30°–50°N and 70°–125°W), W. Europe (35°–75°N and 10°W–40°E) and E. Asia (20°–50°N and 100°–130°E) are presented in Table 1. For W. Europe and the USA the mean decrease in anthropogenic emissions between S85 and S00 is 3.4% and 1.9% per year, which matches the estimates of EMEP [Lövlad *et al.*, 2004] for W. Europe, and the EPA for the USA [U.S. EPA, 2001]. For E. Asia, emissions increase by 2.5% per annum, which agrees with the estimate of Streets *et al.* [2000] for the annual growth in emissions from E. Asia between 1985 and 1997.

Table 1. Modelled Regional Sulfur Budgets, Column Burden, and Surface Concentration Calculated for S85 Across Europe, USA, and E. Asia^a

	W. Europe		USA		E.Asia	
	S85	S00, %	S85	S00, %	S85	S00, %
SO₂						
Emission	3.7	-47.4	2.8	-26.3	3.36	31.5
Dry oxidation	0.26	-50.0	0.30	-22.8	0.42	44.1
In-cloud oxidation	0.68	-22.6	0.57	-9.2	0.7	30.3
Deposition	1.96	-56.8	1.42	-30.1	1.87	20.9
Column burden	4.5	-53.9	2.6	-20.4	4.1	13.9
Surface conc	0.94	-57.5	1.03	-25.8	2.01	23.4
H ₂ O ₂ fraction	0.74	0.71	0.79	0.77	0.91	0.93
SO ₂ export fraction	0.22	0.22	0.18	0.17	0.11	0.14
Sulfate						
Emission	0.09	-51.1	0.07	-29.0	0.08	37.4
Production	0.95	-30.5	0.86	-13.8	1.12	35.7
Deposition	0.82	-31.4	0.64	-10.0	0.9	40.0
Column burden	2.2	-31.5	1.6	-16.9	2.6	27.7
Surface conc	0.92	-31.6	0.64	-15.0	1.13	40.7
SO ₄ export fraction	0.21	0.21	0.31	0.27	0.25	0.23

^aModelled regional sulfur budgets, $\text{mg (S) m}^{-2} \text{ day}^{-1}$; column burden, mg (S) m^{-2} ; surface concentration, mg (S) m^{-3} . The percentage change (S00, %) in the budgets/burdens between the S85 and S00 scenario is also shown. Note that the SO₂ emission includes SO₂ production from DMS. The fraction of in-cloud oxidation which occurs through H₂O₂ is presented for each scenario. Finally, the fraction of the source exported and so not oxidised or deposited in each region is shown for SO₂ and sulfate (SO₄) in 1985 and 2000. The atmospheric column extends to 10 hPa.

[10] For W. Europe and the USA, total SO₂ emissions have decreased by 47.4% and 26.3% over a 15 year period, yet the modelled sulfate column burden has declined by only 31.5% and 16.9% respectively. We therefore find that between 1985 and 2000, a 1% decrease in SO₂ emission

over both W. Europe and the USA results in approximately 0.65% decrease in sulfate column burden. The response of sulfate is similar at the surface, where concentrations decreased by 0.65% and 0.57% per 1% reduction in SO₂ emission across W. Europe and the USA respectively. Simulated SO₂ concentrations have, however, decreased more rapidly than sulfate. We find that per 1% reduction in SO₂ emission, surface SO₂ concentrations decreased by 1.2% across W. Europe and by 0.98% across the USA.

[11] Similar disproportionate changes in SO₂ and sulfate are apparent in long-term measurements over W. Europe and the USA. For example, *Lövblad et al.* [2004] present sulfate measurements across EMEP monitoring sites in the UK, France and Germany, which show that sulfate concentrations decreased by 50%, 50% and 70% respectively in response to a 90% reduction in SO₂ emissions between 1980 and 2000 (equivalent to 0.55–0.77% change in sulfate for a 1% change in SO₂ emission). By contrast, SO₂ concentrations across the same regions were observed to have decreased in line with emissions. This is demonstrated in Figure 2, which shows that the modelled and observed contribution of sulfate to total atmospheric sulfur (SO₂ + sulfate) increased across W. Europe between 1985 and 2000. *Holland et al.* [1998] present measurements across 34 rural sites in the eastern USA showing that SO₂ concentrations decreased by 35% (median) between 1989 and 1995 while sulfate concentrations decreased by 26% (a 0.75% change in sulfate per 1% SO₂ change).

[12] The modelled and observed trends are quite different for Asia. Emissions from E. Asia increase by 31.5% between 1985 and 2000 causing the modelled sulfate column burden to increase by 27.7% (a 0.88% change in sulfate for a 1% change in SO₂ emission). Modelled surface

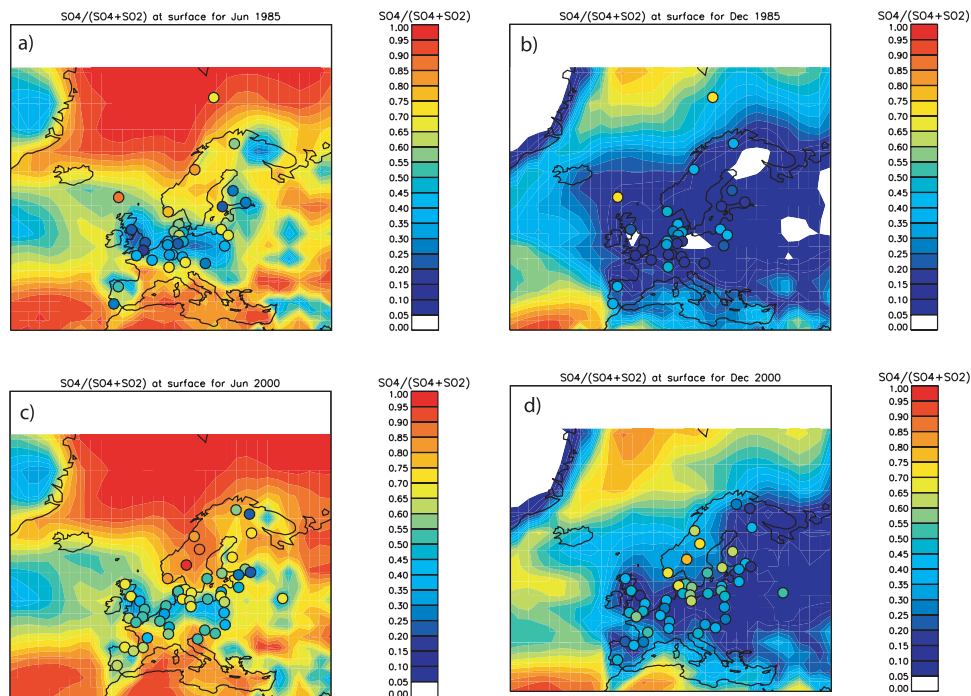


Figure 2. The fraction of atmospheric sulfur (SO₂ + SO₄) which exists in the form of sulfate across W. Europe in (a) June 1985, (b) December 1985, (c) June 2000, and (d) December 2000. The simulated fraction is shown by the colored contours. The filled circles represent EMEP sites and observations for the same time period.

Table 2. Modelled Average Concentration of OH, H₂O₂, and O₃ Below 600 hPa for W. Europe, USA, and E. Asia^a

	W. Europe	USA	E. Asia
OH	1063	1675	1901
H ₂ O ₂	269	310	397
O ₃	41	42	40

^aOH, 10³ molecules per cc; H₂O₂, pptv; O₃, ppbv.

sulfate concentrations increase by 1.3% per 1% increase in SO₂ emission. Measurements from Midway Island in the Pacific also indicate that anthropogenic sulfate has increased approximately at the same rate as SO₂ emissions from East Asia. *Prospero et al.* [2003] used concentrations of methanesulfonate (MSA) at Midway to estimate the contribution of DMS to sulfate. All non DMS derived sulfate was assumed to originate from Asia as the contribution from other continental sources was expected to be minimal in springtime. Concentrations of mean springtime anthropogenic sulfate in 1993–94 were 1.85 times higher than for 1981 and 1983. *Streets et al.* [2000] estimated that emissions from East Asia increased by a factor of 1.61 over this same time period.

[13] Although we have not performed a full statistical analysis of modelled and observed trends, there is general model-observation agreement that in contrast to Asia, decreases in sulfate over W. Europe and the USA are less than decreases in SO₂ emissions and SO₂ concentrations. We now use the model to diagnose the cause of this difference and explore the impact on global sulfate.

3.2. Modelled Regional Budgets

[14] The relationship between SO₂ emission and sulfate burden can be understood by analysing the sulfur species budget. Results are presented in Table 1 (Refer to auxiliary material for global maps of SO₂ oxidation and SO₂ deposition under each emission scenario).

[15] Sulfate is produced from the dry oxidation of SO₂ by OH and from in-cloud oxidation of SO₂ by H₂O₂ and O₃. For W. Europe and the USA, decreases in SO₂ emissions have brought about a similar reduction in dry oxidation. However, the 47.4% and 26.3% reduction in emissions across W. Europe and the USA result in only a 22.6% and 9.2% decrease in the sulfate produced by in-cloud oxidation. These changes imply that over W. Europe and the USA, the conversion efficiency of SO₂ to sulfate in clouds is increasing as SO₂ emissions decrease. In contrast, Table 1 shows that for E. Asia, SO₂ emissions and in-cloud sulfate production increase by 31.5% and 30.3%. The differing response of sulfate to changes in SO₂ over the higher latitude European/US regions and lower latitude E. Asia is largely explained by the availability of H₂O₂ to oxidise SO₂ in cloud. We find that E. Asia is less oxidant-limited than the higher latitude regions. This is also demonstrated by dry oxidation, which although less important for sulfate production than in-cloud oxidation, increases at a greater rate than emissions across E. Asia between 1985 and 2000. Table 2 shows that regional OH and H₂O₂ concentrations are greater across E. Asia than at higher latitudes.

[16] The potential impact of oxidant limitation on long-term changes in sulfate was highlighted by *Dutkiewicz et al.* [1995]. They found that total available H₂O₂ could only

oxidise a small fraction of the SO₂ which existed in cloud throughout NE USA. Our model shows that the westward spread of emissions to cleaner regions of the USA (Figures 1a and 1b) also caused an increase in the oxidation efficiency of SO₂ in cloud. However, between S85 and S00 the absolute change in the modelled column sulfate burden west of 93°W is only 14% of that to the east. The limited availability of oxidant in the east is therefore the more dominant factor in controlling US sulfate changes. There is also a change in the spatial distribution of emissions from E. Asia between both inventories, although the eastward shift of the source leads to an increase in the emission density. This should reduce the exposure of SO₂ to oxidants, yet simulated oxidation still increases at approximately the same rate as emissions from this region.

[17] The relationship between SO₂ emission and sulfate burden could also be explained by a change in the proportion of SO₂ and sulfate exported from each region. We can estimate export from the difference between the regional sources and sinks for atmospheric sulfur. We find that export from our defined regions accounts for 11–22% of the SO₂ emitted, and 21–31% of the sulfate produced. This would suggest that the loss of sulfur from each region is small compared to the regional loss which occurs through deposition and oxidation. Furthermore, between S85 and S00, there is no change in SO₂ and sulfate export from W. Europe, and only small changes in export for the USA and E. Asia.

[18] Long-term changes in the amplitude of the seasonal variation in sulfate provides further evidence for the role of oxidant limitation. CASTNet measurements at sites that have annual data from 1988–1999 show an average decrease of 0.04 μg m⁻³ yr⁻¹ (1.6% yr⁻¹) for winter and 0.21 μg m⁻³ yr⁻¹ (3.0% yr⁻¹) for summer [*Mueller*, 2003]. For the USA, we calculate a decrease of 0.6% yr⁻¹ and 0% yr⁻¹ in surface and column sulfate during winter, and a decrease of 1.1% yr⁻¹ and 0.8% yr⁻¹ in surface and column sulfate during summer. The relative decrease in observed and simulated summertime sulfate is therefore almost twice as large as for winter. This long-term decrease in the seasonal amplitude of sulfate concentration can be explained by the limited supply of H₂O₂ in winter relative to summer across the mid to high latitudes.

[19] Zonal-mean changes in SO₂ emission and sulfate production are shown in Figures 1c and 1d. A large decrease in SO₂ emissions is seen between 35°N to 60°N, with a small increase between 35°N to 10°N. The latitudinal change in emissions leads to a reduction in both dry and wet oxidation of SO₂ across mid to high latitudes, and an increase in production from 35°N to the equator. We find that although the increase in emissions across the tropics and sub tropics is substantially outweighed by the decrease in emissions at higher latitudes, there is almost a global net cancellation in sulfate production. Table 3 shows that the modelled global sulfate burden declined by only 3.2% between 1985 and 2000 in response to a 12.3% reduction in global sulfur emissions.

[20] Changes in the emission of particulate carbon are not included here, but may also have had an effect on sulfate production since BC/OC allow the heterogeneous oxidation of SO₂. The increase in BC/OC across E. Asia, and decrease

Table 3. Modelled Global Sulfur Budget, Burden, and Lifetime Calculated for S85 and S00^a

	S85	S00	change, %
SO ₂			
Emission	105.1	92.2	-12.3
Dry oxidation	11.4	10.6	-7.0
In-cloud oxidation	40.9	40.1	-1.9
Deposition	54.9	43.3	-21.1
Burden	0.44	0.34	-21.3
Lifetime	1.5	1.3	-10.3
H ₂ O ₂ fraction	0.82	0.84	
Sulfate			
Emission	1.5	1.9	-21.0
Production	52.3	50.7	-3.1
Deposition	53.1	51.4	-3.2
Burden	0.44	0.43	-3.2
Lifetime	3.0	3.0	0.0

^aModelled global sulfur budget, T_g (S) yr⁻¹; burden, T_g (S); lifetime, days. The percentage change in the budget, burden and lifetime between each emission scenario is also presented. The fraction of in-cloud oxidation which occurs through H₂O₂ is shown for each scenario. Note that the SO₂ emission includes SO₂ production from DMS.

in particulates across W. Europe and the USA may also have been important in controlling sulfate changes.

[21] One limitation of our study is that we have used year 2000 oxidant fields for both the S85 and S00 simulations. Since the fate of sulfur is controlled by oxidant concentrations, regional changes in oxidants between S85 and S00 will have affected sulfate concentrations. Unger *et al.* [2006] studied the effect of changes in oxidant concentrations between 1995 and 2030 on the global sulfate burden using 1995 SO₂ emissions and the GISS GCM model. In their model, changing oxidants led to an increase in the surface sulfate burden by up to 20% across China and India but by less than 5% across W. Europe. The burden was practically unchanged in North America. Their work suggests that long-term changes in oxidants are likely to amplify the effect described here: that is, barely changing the oxidant limitation at mid/high latitudes but reducing the already rather weak limitation at lower latitudes.

4. Conclusion

[22] We have used a global model of aerosol processes to simulate regional and global changes in the sulfur cycle between 1985 and 2000. Model sulfate concentrations have declined at a slower rate than sulfur emissions across W. Europe and the USA, mainly as a result of the increased availability of H₂O₂ in clouds. Sulfate concentrations across Asia are calculated to have risen more in line with the growth in the sulfur source. These modelled responses of sulfate to changing emissions of SO₂ are supported by observations of long-term changes in annual mean sulfate and the seasonal cycle of sulfate.

[23] Although our global model cannot capture the complex regional changes that have occurred over the last two decades [e.g., Holland *et al.*, 1998; Lövblat *et al.*, 2004], it does suggest that gross shifts in emissions from high to lower latitudes will have significantly affected changes in global sulfate.

[24] The consequence of a latitude-dependent SO₂ oxidation efficiency is that an equatorward shift in SO₂ emissions

to regions with a greater oxidation efficiency has resulted in only a 3.2% decline in the global sulfate burden from a 12.3% reduction in the global sulfur source. Previous studies [Berglen *et al.*, 2004] found that with changes in SO₂ emission, conversion to sulfate was damped in both Europe and E. Asia. Our model suggests that the lesser damping in E. Asia combined with the shift of emissions to that region means that global sulfate may be declining much more slowly than global SO₂ emissions.

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