History of sulfate aerosol radiative forcings

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[1] The history of the global sulfur cycle has been simulated using an emission inventory of SO_2 for 1990 and previously published historical trends in emission on a per country basis. The global- annual-mean radiative forcings due to sulfate aerosols increase (in absolute values) from near-zero and -0.17 Wm^{-2} up to -0.4 and -1 Wm^{-2} between 1850 and 1990, for the direct and indirect effects, respectively. The forcing efficiency (defined as the ratio of the radiative forcing to the anthropogenic sulfate burden) is fairly constant for the direct effect at -150 W(g sulfate)$^{-1}$ but decreases significantly for the indirect effect with increasing sulfate burden. The model results are compared with long-term observations for the period 1980 to 1998 in the U.S. and Europe. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0325 Atmospheric Composition and Structure: Evolution of the atmosphere; 1610 Global Change: Atmosphere (0315, 0325); 3359 Meteorology and Atmospheric Dynamics: Radiative processes

1. Introduction

[2] It is important to determine the history of the various radiative forcings (RFs) in the context of the detection of anthropogenic climate change [*IPCC*, 2001]. *Myhre et al.* [2001] compiled the historical evolution of most of the natural and anthropogenic RFs identified so far. They stressed that the uncertainties are large for the RFs by anthropogenic aerosols because of the uncertainties in emissions and calculations of the RF itself. *Tegen et al.* [2000] computed the trends in aerosol concentrations and direct RF for the period 1950 to 1990. Here we focus on sulfate aerosols because more information on their source are available than for other aerosol species. We present some results on sulfate concentrations, and their direct and indirect effects for the period 1850 to 1990 (1998 over Europe and U.S.).

2. Methodology

2.1. Sulfur Cycle Model

[3] We use the sulfur cycle model implemented in the Laboratoire de Météorologie Dynamique (LMD) general circulation model (GCM). The model is described and evaluated against measurements in *Boucher et al.* [2002]. The model represents many features of the observed concentrations of DMS, SO_2, and sulfate aerosols, although the sulfate concentrations are underestimated over Europe in winter and overestimated over North America in summer and autumn. Model simulations are performed at a horizontal resolution of 96×72 and 19 vertical layers. The RFs are computed as the differences in top-of-atmosphere shortwave radiative flux between two simulations, with and without anthropogenic sulfur emissions. In these simulations sulfate aerosols do not feedback on the radiation and cloud fields (i.e., sulfate radiative effects are purely diagnostic) so that all simulations have the same meteorology. We consider that neither the oxidant fields (except H_2O_2 which is calculated interactively in our model) nor the natural sources of sulfur species (including volcanoes) have changed during the period under study (1850–1998). Therefore the trends simulated in our model only account for changes in the anthropogenic sulfur emissions, which constitutes the focus of this study. All the results presented in this study are annual averages after a 6 month spin up time.

[4] We follow *Boucher and Anderson* [1995] for the parametrization of the sulfate aerosol direct effect, assuming a chemical composition of ammonium sulfate, a log-normal size distribution with a mean geometric volume diameter $D_{gv} = 0.3$ μm and a geometric standard deviation $\sigma_g = 2.0$. The aerosol size distribution and optical properties depend on the relative humidity in the clear-sky fraction of a GCM grid-box, following the laboratory measurements of *Tang and Munkelwitz* [1994].

[5] One usually distinguishes the first aerosol indirect effect (an increase in cloud droplet number concentration at fixed liquid water content) from the second aerosol indirect effect (induced by a reduction in the precipitation efficiency). Here we only consider the first indirect effect and follow the parametrization of *Boucher and Lohmann* [1995]. We use their relationship D to compute the cloud droplet number concentration (CDNC, in droplets per cm^{-3}) from the sulfate mass (m_{SO_4}, in μg SO_4 m^{-3}):

$$\mathrm{CDNC} = 10^{2.21+0.41\ \log(m_{SO_4})} \quad (1)$$

Although empirical, this parametrization predicts RFs of comparable magnitude to those from more elaborate "mechanistic" parametrizations of the aerosol indirect effect [*IPCC*, 2001]. It is appropriate for the purpose of this study since we are interested primarily in the time evolution of the RF.

2.2. Emissions

[6] There are some uncertainties in the emission strengths of all sulfur compounds, including industrial SO_2. Commonly used emission inventories include GEIA for base year 1985 [*Benkovitz et al.*, 1996] -as in *Boucher et al.* [2002]- and EDGAR for base year 1990 [*Olivier et al.*, 1996]. Regional inventories have also been developed in recent years. Because different methodologies and input data have been used, the differences between the 1985 GEIA and 1990 EDGAR inventories do not represent a "real" evolution in the emissions. Instead one should use a same methodology to derive the sulfur emissions for different years. Historical emission inventories have been built by *Örn et al.* [1996] at a $5° \times 5°$ resolution for 1860 to 1980 and by *Lefohn et al.* [1999] on a per country basis for 1850 to 1990.

[7] In our CONTROL simulation we use the EDGAR V2.0 database [*Olivier et al.*, 1996] for the year 1990 and scale the emissions country by country using the data by *Lefohn et al.* [1999], in order to build $1° \times 1°$ emission inventories for some other years

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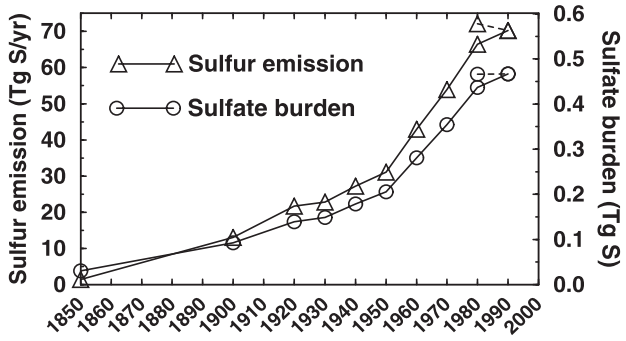

Figure 1. Global- annual-mean emission of anthropogenic SO_2 (Tg S yr^{-1}) and anthropogenic sulfate burden simulated by the model (Tg S) for the period 1850 to 1990. The solid and dashed lines are for the CONTROL and EPA-EMEP simulations, respectively.

(i.e., 1850, 1900, and 1920 to 1990 with a step of 10 years). The emission strength of SO_2 at a year y is obtained from

$$F(\lambda,\phi,y) = \frac{\sum_c G_c(y)\ \alpha_c(\lambda,\phi)}{\sum_c G_c(1990)\ \alpha_c(\lambda,\phi)} F_{\mathrm{EDGAR}}(\lambda,\phi) \qquad (2)$$

where F_{EDGAR} is the EDGAR emission strength, $G_c(y)$ the emission strength for country c and year y according to *Lefohn et al.* [1999], and $\alpha_c(\lambda, \phi)$ the area fraction of country c in the grid-box determined by its latitude λ and longitude ϕ (with a resolution of 1°). There are few countries with missing data. In such cases, the emission strength of a neighbouring country with similar economy was used. When data for the year 1990 were missing, the trend obtained from earlier years was extrapolated to 1990. As there is no information on the time evolution of sulfur emissions from shipping, this source was left out of this study. We consider that a small fraction (5%) of the SO_2 is directly emitted as sulfate and add a small industrial source of H_2S set to 4% of the SO_2 source [*Watts*, 2000; *Boucher et al.*, 2002].

[8] Recent data by the U.S. Environmental Protection Agency [*EPA*, 2000; *Husain et al.*, 1998] and the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) [*Mylona*, 1996; *Vestreng and Støren*, 2000] suggest a more rapid decrease in sulfur emissions over U.S. and Europe than in the Lefohn et al. database. In a second set of simulations for years 1980 and 1998 (referred to as EPA-EMEP), the $G_c(y)$ terms of Equation (2) are prescribed from *EPA* [2000] for the U.S., from *Vestreng and Støren* [2000] for European countries (i.e., Western Europe and former USSR republics west to Russia), and from (or extrapolated in time from) the Lefohn et al. data for the rest of the countries.

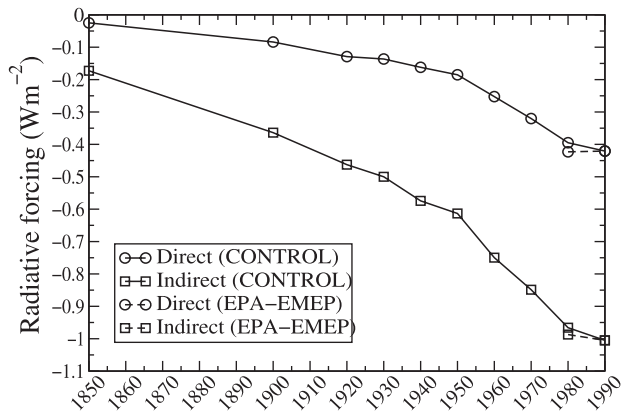

Figure 2. Time evolution of the global- and annual-mean direct and indirect RFs by sulfate aerosols (Wm^{-2}). The solid and dashed lines are for the CONTROL and EPA-EMEP simulations, respectively.

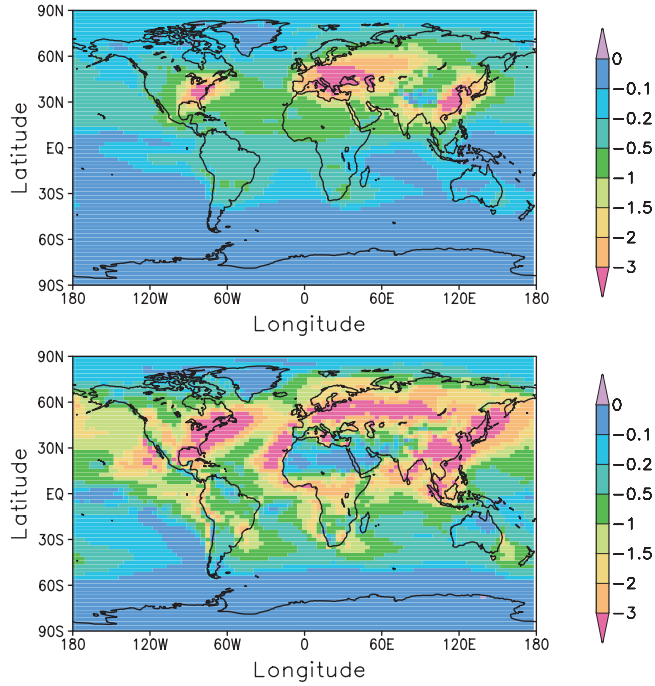

Figure 3. Direct (upper panel) and indirect (lower panel) RFs by sulfate aerosols (Wm^{-2}) for the year 1990.

3. Discussion

[9] The natural burden of sulfate aerosols is estimated to be 0.19 Tg S. The anthropogenic burden of sulfate aerosols is represented on Figure 1 along with the emission strength. It increases from 0.03 Tg S in 1850 to 0.47 Tg S in 1990. The NH to SH ratio in burden decreases from 7.2 to 5.8 between 1920 and 1990 (when, in the same time, the NH to SH ratio in emission decreases from 50 to 13). The anthropogenic sulfate burden increases less rapidly than the emission strength, because only a fraction of the sulfur dioxide is oxidized into sulfate (roughly 70% in our model). Note however that the ratio of anthropogenic sulfate burden to the SO_2 emission strength does not change significantly from 1900 to 1990, indicating that the efficiency of SO_2 to sulfate oxidation has not decreased, despite the increasing SO_2 concentrations.

[10] The time evolution of the RFs by sulfate aerosols is represented on Figure 2. The global- and annual-mean RFs reach -0.42 and -1.0 Wm^{-2} in 1990, for the direct and indirect effects, respectively. In contrast to the direct RF, the indirect RF is not negligible in 1850, reducing the RF over the period 1850 to 1990 compared to the period from pre-industrial times to 1990. The NH to SH ratios in RFs decrease from maximum values of 9.0 and 6.0 in 1920 to 6.8 and 3.7 in 1990, for the direct and indirect effects, respectively. The spatial distributions of the two effects are quite different as illustrated in Figure 3 for the year 1990. While the direct effect is concentrated in the midlatitudes of the Northern Hemisphere and above the continents, the indirect effect shows a more widespread distribution with a significant contribution over the oceans.

[11] In contrast to our CONTROL simulation, the EPA-EMEP simulation shows a flattening of both the direct and indirect RFs

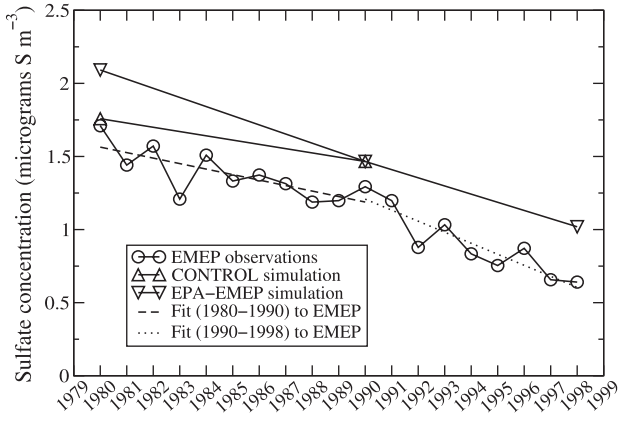

Figure 4. Sulfate concentrations (μg S m^{-3}) observed and simulated under the two emission scenarii. The values are annual means averaged over the 18 stations of the EMEP network having continuous measurements from 1980 to 1998.

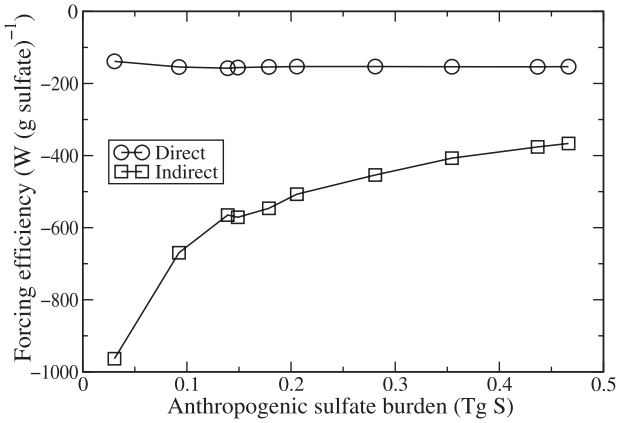

Figure 6. Forcing efficiencies (W (g sulfate)$^{-1}$) for the direct and indirect effects of sulfate aerosols as a function of the anthropogenic sulfate burden (Tg S).

between 1980 and 1990 (Figure 2). The direct RF decreases slightly following the decrease in sulfate burden, while the indirect RF increases slightly because of a larger forcing efficiency of the sulfate in the regions of increasing compared to the regions of decreasing concentrations. We follow *Tegen et al.* [2000] and compare the simulated sulfate concentrations over Europe to the EMEP measurements [*Hjellbrekke*, 2001] and the wet deposition flux over the U.S. to the NADP measurements (National Atmospheric Deposition Program, see http://nadp.sws.uiuc.edu). In these comparisons, only sites with continuous measurements over the period 1980 to 1998 have been considered. The model overestimates the mean sulfate concentrations at EMEP sites by 10–25% between 1980 to 1990 (Figure 4). The observed (negative) slope of the trend is intermediate between those simulated in the CONTROL and EPA-EMEP runs. For the period 1980 to 1998, the simulated slope in the EPA-EMEP run is close to the observed one. There is a good agreement (<10%) for the wet deposition flux of sulfur simulated and observed over the NADP network (Figure 5). The trend is not as strong in both simulations as in the observations. This discrepancy may be due to a stronger decrease in the emissions than implemented in the model or to changes in the geographical patterns of the emissions or precipitation not accounted for in the model. It is difficult from this limited set of comparisons to select the better of our two scenarios. However, it seems that the observed decrease in sulfate concentrations over Europe and deposition fluxes over U.S. is as large or larger than implied by our EPA-EMEP scenario.

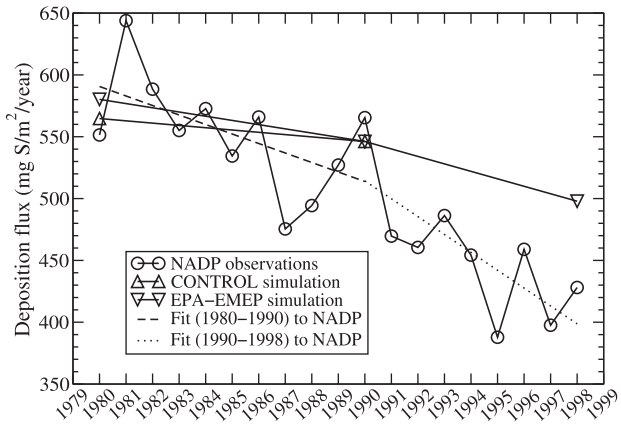

Figure 5. Wet deposition flux (mg S m^{-2} yr^{-1}) observed and simulated under the two emission scenarii. The values are annual means averaged over the 57 stations of the NADP network having continuous measurements from 1980 to 1998.

[12] The forcing efficiency is defined as the ratio of the global- and annual-mean RF (in Wm^{-2}) to the anthropogenic sulfate burden (in g m^{-2}), as introduced by *Boucher and Anderson* [1995]. As shown in Figure 6, the forcing efficiency for the sulfate aerosol direct effect is fairly constant at -150 W(g sulfate)$^{-1}$. It is close to the forcing efficiency of -125 W(g sulfate)$^{-1}$ calculated in *Boucher and Anderson* [1995] for an earlier version of our GCM (with off-line sulfur cycle) and on the low side of the range reported by *Haywood and Boucher* [2000]. Such differences in the forcing efficiency among global models have not been eluded yet, but seem to be due to differences in the treatment of relative humidity and cloud effects, rather than in the radiative transfer itself [*Boucher et al.*, 1998]. In contrast to the direct effect, the forcing efficiency of the sulfate indirect effect shows a large reduction from 1850 to 1990, from -963 to -366 W(g sulfate)$^{-1}$. This illustrates the strong non-linearities of the aerosol indirect effect induced by the aerosol-to-CDNC and CDNC-to-cloud albedo relationships.

4. Conclusions

[13] We have computed the RFs due to sulfate aerosols over the period 1850 to 1990. The direct RF increased (in absolute value) from near-zero to -0.42 Wm^{-2}, while the indirect RF increased from -0.17 to -1.0 Wm^{-2} but with a decreasing efficiency. The decrease in SO_2 emissions over U.S. and Europe results in a global-mean RF due to sulfate aerosols which stays fairly constant between 1980 and 1990. There is a shift in the RF patterns from U.S., Europe, Russia, Northern Atlantic Ocean and parts of Africa to South-East Asia and the Indian and Pacific Oceans. This work needs to be extended to other aerosol species, in particular carbonaceous aerosols for which the history of emissions is becoming available.

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