

The radiative effect of the anthropogenic influence on the stratospheric sulfate aerosol layer

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ABSTRACT

Stratospheric sulfate aerosols have a cooling effect on the Earth's surface. Sulfur aerosols from large volcanic eruptions are often the dominant source, while non-volcanic background stratospheric sulfate aerosols are supposed to mainly originate from carbonyl sulfide (OCS). Several recent studies indicate, however, that this latter source is too small to account for the observed background stratospheric aerosol concentration. Based on model calculations we suggest that most of the lower stratospheric sulfate aerosol concentration is of anthropogenic origin. We estimate a global mean radiative forcing due to the anthropogenic influence on the stratospheric aerosol layer of -0.05 W m^{-2} . This represents a new climate forcing mechanism and emphasizes anthropogenic sulfur emission as an important cooling mechanism.

1. Introduction

Atmospheric aerosols influence the radiative balance directly by scattering and absorption of radiation and indirectly through the cloud microphysics, affecting the radiative effect of clouds (IPCC 2001). Increase in the abundance of tropospheric sulfate aerosols due to human activity has led to an uncertain but significant cooling effect (IPCC 2001). In volcanically active periods eruptions are the major source of stratospheric aerosols (Robock, 2000). The last major volcanic eruption, at Mount Pinatubo in 1991, had a significant influence on the radiative balance and led to significant cooling on the Earth's surface in the subsequent 1–3 yr and a maximum cooling of about 0.5 K after about 18 months (Minnis et al. 1993; Hansen et al. 1997; Soden et al. 2002). Based on analysis in a volcanically quiescent period Junge et al. (1961) found that there is a background stratospheric aerosol layer, and Crutzen (1976) proposed that carbonyl sulfide (OCS) could be an important source of stratospheric sulfate aerosols. Extensive research has revealed that this layer has its maximum between 15 to 25 km, depending on latitude (Hofmann, 1990; Thomason et al. 1997). Observations in volcanically quiescent periods indicate an increase in the background stratospheric sulfate aerosol layer at some locations (Hofmann and Rosen, 1980; Sedlacek et al. 1983; Hofmann, 1990); satellite observations have shown the global picture (Thomason et al. 1997). Up until now there has been considerable uncertainty as to whether this is caused by

anthropogenic activity or if the trend is influenced by volcanic eruptions. Various explanations have been proposed to describe the increase in stratospheric sulfate aerosols, either as due to an increase in the source of OCS or to aircraft emissions of sulfur (Turco et al. 1980; Hofmann, 1991). Particular attention has been given to OCS and the large uncertainties in the sources of OCS. Aydin et al. (2002) recently published ice core measurements which showed that pre-industrial OCS concentrations were 25% lower than the current atmospheric values. This is consistent with the detailed evaluation of anthropogenic and natural sources of OCS given by Watts (2000). Nevertheless, modelling studies indicate that the current atmospheric concentrations of OCS are too small to account for the observed background stratospheric sulfate aerosols (Chin and Davis, 1995; Weisenstein et al. 1997; Kjellström, 1998; Pitari et al. 2002). This finding is strengthened by recent isotopic measurements, suggesting that OCS is not the major source for the background stratospheric sulfate aerosols (Leung et al. 2002). We show that anthropogenic SO_2 emission is the main contributor to the stratospheric sulfate aerosols and we provide an estimate of radiative forcing as a consequence of the human-induced changes in stratospheric sulfate aerosols.

2. Method

We have used an atmospheric chemistry transport model (the Oslo CTM2) to model stratospheric sulfate distribution. The Oslo CTM2 is used with a T21 horizontal resolution ($5.625^\circ \times 5.625^\circ$) and 19 vertical layers extending from the surface up to 2 hPa. The input data driving the model are provided by the

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Integrated Forecast System from ECMWF, updated every third hour. T. F. Berglen (pers. comm.) has a detailed description of the sulfur cycle in Oslo CTM2. The Oslo CTM2 has extensive ozone chemistry and includes six sulfur components (dimethyl sulfoxide (DMS), SO₂, sulfate, OCS, H₂S and methanesulfonic acid (MSA)), which are calculated interactively with the ozone chemistry. A detailed chemical scheme is included (Berntsen and Isaksen, 1997). Soluble tracers are removed by wet deposition in two ways—large-scale rainfall and convection. Large-scale rainfall takes into account the solubility of the actual tracer and how much water is rained out from a cloud to determine the mass of tracer removed by rain (Berge, 1993). Convection in Oslo CTM2 is solved using a so-called ‘elevator’ where mass is redistributed vertically based on the convective mass fluxes given in the meteorological input data. The fraction of liquid water in the ‘elevator’ and the solubility of the tracers are used to determine the convective wash-out. In addition, SO₂ and sulfate undergo sub-cloud scavenging as described by Martin (1984) and Berge (1993) respectively. We estimate that 46% of SO₂ is lost by dry deposition, 43% by aqueous phase oxidation, 9% by gas phase oxidation and 2% by wet scavenging. Sulfate is removed by large-scale precipitation (79%), by convection (6%) and by dry deposition (15%).

Natural and anthropogenic emissions of ozone precursors and sulfur compounds are included in the model estimates. The simulations are performed using meteorological data from 1996. The SO₂ emission is representative for the same year (anthropogenic SO₂ 71.2 Tg(S), DMS 12.0 Tg(S), volcanic SO₂ 8.0 Tg(S), biomass burning 2.3 Tg(S), H₂S 0.8 Tg(S)). The surface level mixing ratio of OCS is held constant at 500 parts per trillion (ppt). In the stratosphere OCS is lost by photolysis and reaction with O(³P). The simulations represent background stratospheric sulfate aerosol conditions with an average volcanic emission into the troposphere; since we are studying the background stratospheric sulfate aerosols we have not included any major volcanic eruptions. A constant size distribution for the stratospheric sulfate aerosols is adopted, which results in a sedimentation velocity of 2.5 cm h⁻¹ for the aerosols. With this small sedimentation velocity we see that for the background stratospheric sulfate aerosols this effect is of minor importance. However, for stratospheric aerosols of volcanic origin conditions are quite different since the size is larger and the sedimentation velocity increases substantially with aerosol size (Seinfeld and Pandis 1998). Calculations show that the model represents tropospheric–stratospheric exchange processes reasonably well (Danilin et al. 1998; Bregman et al. 2001; Gauss et al. 2003).

The optical properties of the stratospheric aerosols are modelled with Mie theory based on the size distribution and refractive indices. The refractive indices used in the solar region depend on the aerosol acid weight percentage (Palmer and Williams, 1975) and in the terrestrial region on the aerosol acid weight percentage and temperature (Myhre et al. 2003). In the calculations we adopt the global and seasonal distribution of aerosol

acid weight percentage from SAGE II (Yue et al. 1994). The densities used in the calculation of the optical properties depend on aerosol acid weight percentage as well as temperature (Myhre et al. 2003). The sulfate aerosols are modelled with a mono log-normal size distribution with a geometric mean radius of 0.081 μm and geometric standard deviation of 2.0, within the range of measurements (Hofmann, 1990; Strevermayer et al. 2000).

3. Results

3.1. Anthropogenic influence on stratospheric sulfate aerosols

Four sets of simulations (see Table 1) were performed to estimate the anthropogenic contribution to the stratospheric sulfate aerosols layer. Figure 1 shows the modelled vertical profile of stratospheric sulfate aerosols at 0°E, 44°N compared with observed vertical profiles of stratospheric sulfate aerosols at the same location. The results are shown for simulations (i), (ii) and (iii). The vertical profile in Fig. 1 depicts a good agreement with the observations in the case where all sulfur emissions are included (simulation (i)). However, for simulation (ii) the amount of modelled stratospheric sulfate aerosol is a factor of ten smaller than that observed. With OCS as the only sulfur species (simulation (iii)) the model is not able to reproduce the maximum observed amount of stratospheric sulfate aerosols in the lower stratosphere at about 15 km. The global mean vertical distribution of OCS is shown in Fig. 2 and shows a strong decrease in the stratosphere, which is in accordance with the observations of OCS (Chin and Davis, 1995; Kettle et al. 2002). The model and the observations show a negligible decrease below 10 km, but already at 25 km the mixing ratio of OCS is below 200 ppt, shown in Fig. 2 as well as in the observations (Chin and Davis, 1995; Kettle et al. 2002).

Figure 3 illustrates the modelled zonal mean vertical profile of sulfate. It demonstrates that the model simulation (i) is able to reproduce the stratospheric sulfate aerosols in a layer between 15 and 25 km including the latitudinal

Table 1. Description of calculations

Case	Description
(i)	All natural and anthropogenic sulfur emissions are included
(ii)	Only natural sulfur emissions are included, with a tropospheric OCS abundance of 500 ppt
(iii)	Only OCS are included with a tropospheric abundance of 500 ppt
(iv)	Only natural sulfur emissions are included and OCS with a tropospheric abundance of 373 ppt according to pre-industrial concentrations (Aydin et al. 2002)

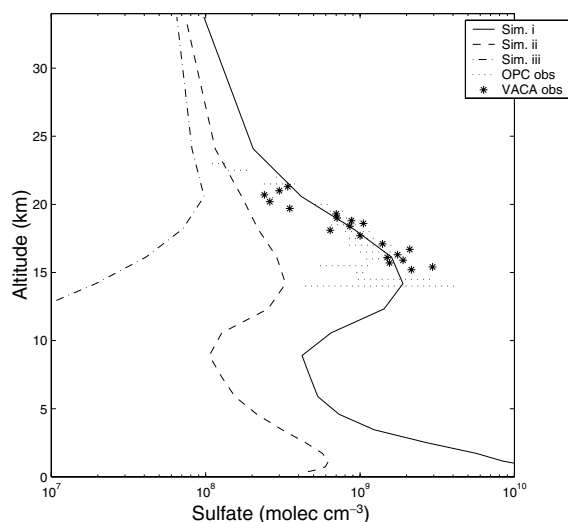


Fig 1. Comparison of vertical profile of sulfate from the model (for simulations (i), (ii), and (iii)) and observations (Arnold et al., 1998) at 0°E, 44°N. Two different instruments [volatile aerosol composition analyzer (VACA) and optical particle counter (OPC)] are used. For OPC the measured interval is indicated and for VACA the individual measurements are shown.

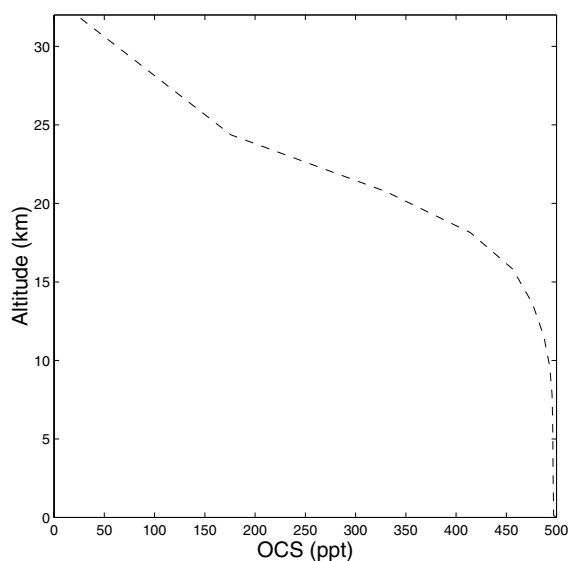


Fig 2. Global mean modelled vertical profile of OCS.

variation, with a higher abundance of stratospheric sulfate aerosols in the Northern Hemisphere than in the Southern Hemisphere. The pattern is in agreement with earlier measurements (Thomason et al. 1997), although the hemispheric difference in stratospheric sulfate aerosols is larger than in our model results. This illustrates that in the case where all sulfur emissions are included; the model reproduces the main features of the distribution of stratospheric sulfate aerosols.

Figure 4 shows the global mean vertical profile of sulfate for the four different calculations. The results clearly indicate

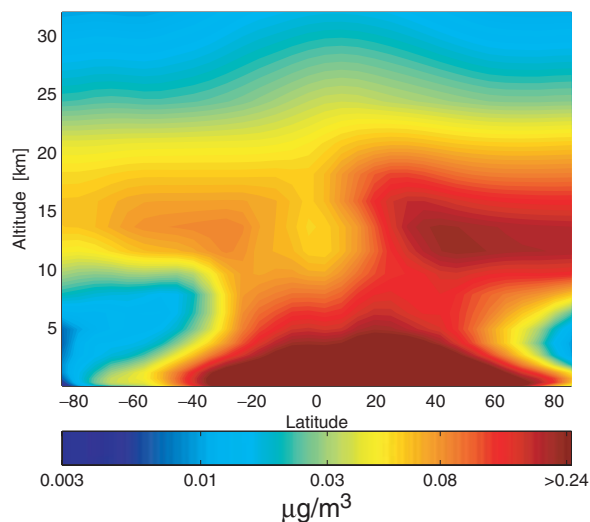


Fig 3. Modelled annual and zonal mean vertical distribution of sulfate ($\mu\text{g m}^{-3}$).

that the main source of stratospheric sulfate aerosols is anthropogenic. This is justified by the difference between simulations (i) and (ii). In calculations with all sulfur emissions included (simulation (i)) the stratospheric sulfate aerosols are up to three times higher than in the case with only natural sources (simulation (ii)). In the simulation with only OCS as the sulfur component (simulation (iii)) the contribution to stratospheric sulfate aerosols is important at 30 km, but not significant at lower altitudes. Consequently the difference in stratospheric sulfate aerosols between simulations (ii) and (iv), which only differ in the tropospheric OCS abundance, is small. Thus the stratospheric sulfate distribution is not sensitive to the concentration of OCS. Based on this consideration our interpretation of Fig. 1

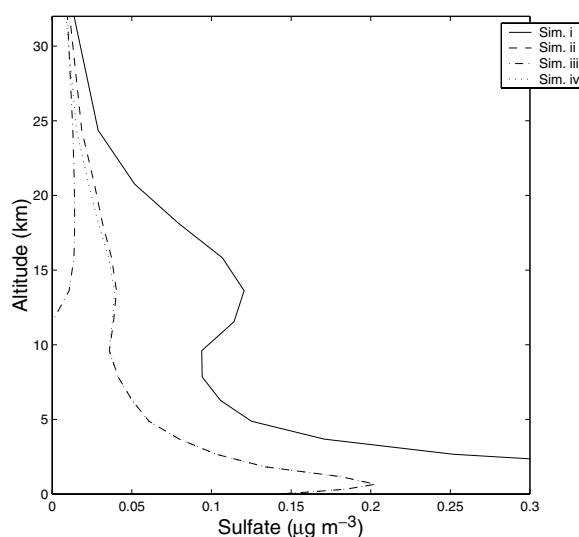


Fig 4. Global mean vertical profile of four model simulations (i)–(iv) as defined in Table 1.

is that transport of sulfate aerosols from the troposphere to the lower stratosphere is required to explain the current background distribution of stratospheric sulfate aerosols. Such transport of aerosols is also supported by observations (Brock et al. 1995; Lelieveld et al. 2002). This is consistent with the conclusions in previous model studies (Chin and Davis, 1995; Kjellström 1998) that OCS is not the main contributor to stratospheric sulfate aerosols, and with observations (Leung et al. 2002). Importantly, our conclusion is consistent with recent isotopic measurements (Leung et al. 2002), which show that the contribution of OCS to the stratospheric sulfate aerosol sulfur budget is small. In fact, the isotopic measurements indicate an even smaller OCS contribution to the stratospheric sulfate aerosol sulfur budget, only 6% versus about 10% in the global and annual mean model results. In the model the contribution from OCS is stronger at higher altitudes.

3.2. Radiative forcing

Climate change over the last 150 yr has been caused by several radiative forcing mechanisms, with the increase in the greenhouse gases being the dominating process (IPCC 2001). The anthropogenic influence on the stratospheric sulfate aerosol layer is an additional radiative forcing mechanism not previously reported. We have used radiative transfer schemes for solar and thermal infrared radiation to calculate the radiative forcing due to stratospheric sulfate aerosols of anthropogenic origin (Myhre and Stordal, 2001), and followed the definition given in IPCC (2001). The geographical distribution of the radiative forcing of anthropogenically produced stratospheric sulfate aerosols is shown in Fig. 5. The radiative forcing calculations are based on the difference between simulations (i) and (ii), but note that only the stratospheric part of the difference is taken into account in the calculations. The pattern of radiative forcing due to anthropogenic stratospheric sulfate aerosols is more homogeneous than for the anthropogenic influence on the tropospheric sulfate. Nevertheless, the strongest forcing is over middle and high latitudes. The global and annual mean net radiative forcing due to anthropogenically produced stratospheric sulfate aerosols is -0.05 W m^{-2} , and slightly weaker than -0.07 W m^{-2} in the Northern Hemisphere. The short-wave forcing is -0.07 W m^{-2} , which dominates the net radiative forcing. For stratospheric aerosols of volcanic origin the short-wave forcing dominates in a similar way (Lacis et al. 1992). The long-wave forcing is 0.02 W m^{-2} , with a small contribution from an increase in the stratospheric temperature, which is a part of the radiative forcing (IPCC 2001).

The assumption of a constant size distribution of the stratospheric sulfate aerosols influences the radiative forcing results. However, the dependence on size distribution is only modest and the size distribution must be significantly different from what we have used in this study before the forcing differs by more

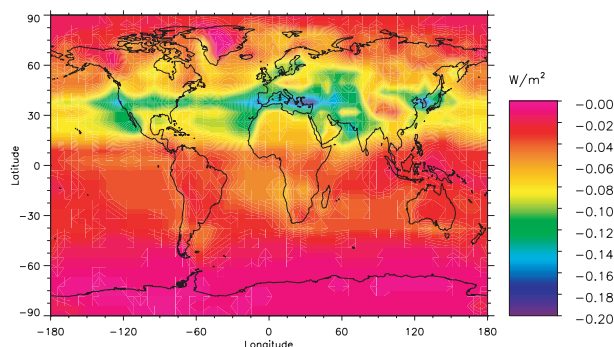


Fig. 5. Geographical distribution of the annual mean radiative forcing due to stratospheric sulfate aerosols of anthropogenic origin.

than 15–20%. Adopting the size distribution used in Kjellström (1998) with a geometric mean radius of $0.0725 \mu\text{m}$ and geometric standard deviation of 1.86 results in almost identical radiative forcing (less than 1% difference).

4. Summary and conclusion

We find SO_2 to be the major source of stratospheric sulfate aerosols, with OCS contributing only as a minor fraction. Furthermore, since anthropogenic emissions of SO_2 are much larger than natural sources, human emission of SO_2 must have influenced stratospheric sulfate aerosols, and we present an estimate of the radiative effect. The increase in the anthropogenic SO_2 emissions was particularly large during the period from 1950 to mid 1980. Significantly more than 50% of the contribution from anthropogenic emissions occurred during this period (Lefohn et al. 1999; Schlesinger et al. 1992). The SO_2 emissions over the USA and Europe have been reduced significantly in the last two decades (Vestreng and Støren, 2000). However, this reduction has been compensated by a strong increase in SO_2 emissions in Asia over this period (Streets et al. 2000a, 2003). The strong increase in Asian emissions lasted up to 1995, with some indication of a reduction the last few years (Carmichael et al. 2002). Emissions from ships are steadily increasing (Streets et al. 2000b; Endresen et al. 2003). In summary the global mean trend in anthropogenic SO_2 emissions over the last 15–20 yr has probably been small and may even be close to zero. Therefore small changes over this period in background stratospheric sulfate aerosols is expected. This is in agreement with measurements, where a small or no change was observed during the last 15–20 yr in the background stratospheric sulfate aerosols (Bingen et al. 2004).

The present study demonstrates that OCS, which has been assumed to be the main source for background stratospheric sulfate aerosols, is likely to be a minor contributor to the stratospheric sulfate aerosol layer compared with the contribution from anthropogenic emissions. We propose that the oxidation of SO_2 to sulfate compounds and transport from the troposphere into the stratosphere are the main sources of stratospheric background

aerosols. Changes in stratospheric sulfate aerosols with anthropogenic activity as the main source have several important consequences. In addition to the discussed influence on climate, there is also an effect on stratospheric ozone chemistry. Stratospheric ozone depletion due to chlorofluorocarbon emissions occurs via heterogeneous reactions on stratospheric sulfate aerosols (Solomon, 1999). With the results presented here the human contribution to stratospheric ozone depletion is likely to be stronger than previously assumed.

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