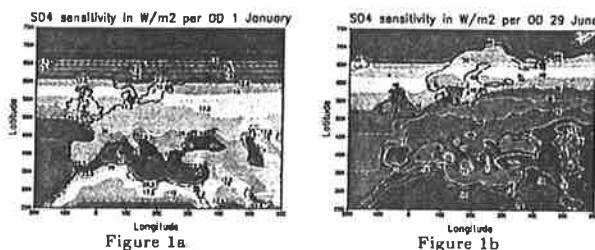


Introduction

Climate effects due to the increasing burden of manmade aerosols are expected to counteract the warming effects due to increasing levels of greenhouse gases by a significant part. Here, we consider the direct effects of sulfate aerosols. The atmospheric lifetime of these aerosols is in the order of a week, depending on meteorological conditions. Hence, their concentrations show a large variability in space and time. Sulfur emissions lead to the formation of sulfate aerosols. These particles are capable of reflecting solar radiation. In addition to this direct effect, aerosols may alter the size distribution of cloud droplets and hence the reflectivity of clouds. The total aerosol effect is not well-known. Uncertainties are largely associated with estimates of the emissions of the precursors and with the parameterizations of chemical processes as well as the modelled transports. In addition, uncertainties in the microphysical properties, such as size distribution and the interactions of aerosols with water vapour and droplets, hamper accurate estimates of the radiative effects.

Here, we focus on the direct radiative forcing sensitivity of sulfate aerosols to insolation parameters and surface conditions. Furthermore, the fields of radiative forcing, using the modelled sulfate aerosol burden over Europe for three selected cases as calculated by the transport/chemistry model TM3 [Jeurken et al., Poster], are presented. This work is performed within the framework of the NRP project MEMORA: Aerosol; cycle and influence on the radiation balance.



Radiative Forcing

The radiative forcing due to sulfate aerosols changes depends on

- surface albedo (R_s)
- average cosine of solar zenith angle (μ_0) during daylight time
- day fraction (f_d = daylight time / daylength)
- cloud cover (C_{cld})

Forcing computations under clear sky conditions are performed using an analytical expression [Van Dorland et al., *J. Geophys. Res.*, 102, 28,079-28,100, 1997]:

$$\Delta F_{clr} = -S_0 f_d k \{4[\mu_0(1-R_s)]^{0.5} [(1-R_s)^{0.5} - [\mu_0]^{0.5}] - r\} \times \Delta SO_4 \quad (1)$$

where ΔF_{clr} is the diurnal average radiative forcing in Wm^{-2} , S_0 is the solar constant ($=1370 W m^{-2}$), $k = 6.8 \times 10^{-2}$ per unity optical depth (OD), $r = 0.05$, and ΔSO_4 is the sulfate aerosol change in terms of optical depth. As a first order approximation, cloud effects can be included by using the factor $(1 - C_{cld})$, implying no direct forcing for total cloudiness.

The clear sky expression is based on calculations using a sophisticated radiative transfer scheme, which is also part of the ECHAM4 climate model, and includes the radiative effects of clouds, water vapour, ozone, 11 aerosol components and the well-mixed greenhouse gases CO_2 , CH_4 , N_2O as well as 16 CFC's, HCFC's, HFC's.

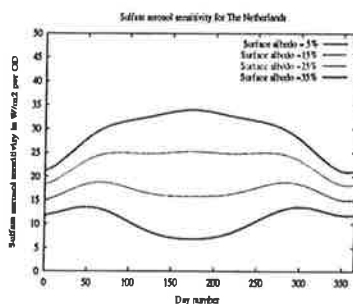
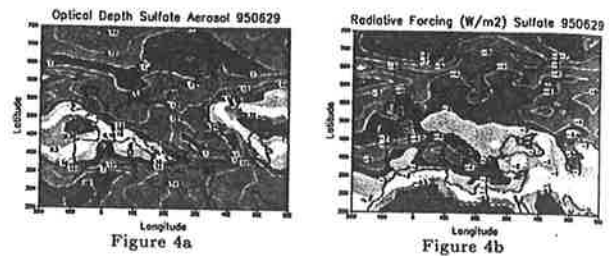


Figure 2



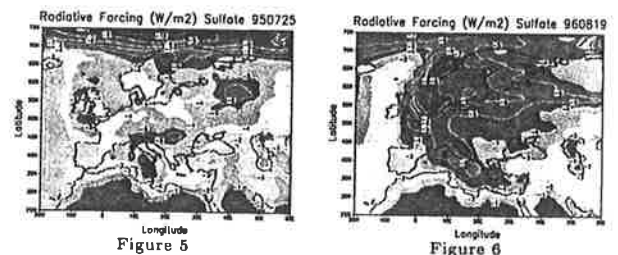
Figure 3



Sensitivity

We examine the clear sky radiative forcing sensitivity, i.e. the forcing for a fixed change of sulfate aerosols, over Europe. In winter (Figure 1a) high latitudes are insensitive to aerosol changes caused by the polar night ($f_d = 0$). We find the largest sensitivities over the subtropical Atlantic Ocean and the Mediterranean Sea of about $25 Wm^{-2}$ per unity optical depth. Oceans with low surface albedos show a larger sensitivity as compared to land surfaces with high surface albedos at the same latitude. In summer, we find sensitivities up to $60 Wm^{-2}$ per unity optical depth (OD) at high latitudes, where $f_d = 1$ (Figure 1b). The seasonal cycle of the sensitivity over the Netherlands is plotted for several values of the surface albedo (Figure 2). Over dark surfaces the sensitivity is largest in June ($35 Wm^{-2}$ per OD) and smallest in December ($21 Wm^{-2}$ per OD). For high surface albedos the situation is reversed.

In a cloudless atmosphere, the backscattered fraction of solar radiation by sulfate aerosols increases for more oblique incident radiation, while the total amount of shortwave radiation at the top of atmosphere decreases. These opposing effects maximize the radiative forcing for a certain zenith angle, which is also dependent on surface albedo. In addition, the sensitivity is strongly influenced by the day fraction.



Case Studies

Within the NRP project several cases have been selected for a detailed investigation, i.e. a comparison between measurements and model results. In this study, we evaluate the modelled fields of sulfate aerosol burden using TM3 in terms of the direct radiative forcing. Hereto, a translation has to be made from aerosol mass into optical depth. This conversion factor depends strongly on the aerosol size distribution and therefore on relative humidity. Here, we use a mass scattering coefficient of $8 m^2 g^{-1}$ in the visual range, corresponding with a typical sulfate (water soluble) aerosol size distribution at a fixed relative humidity of 75%.

In our first case at the 29th of June 1995 (noon), large parts of the European continent are free of clouds (Figure 3). Two spots of high optical depths up to 0.24 are situated in front of the west coast of France and in the vicinity of the Caucasus (Figure 4a). Relative to the other days, quite clean air is found over the Netherlands with optical depths of 0.11. In the cloudy regions of the Atlantic Ocean and Europe, the computed optical depths are considerably lower due to wet deposition. The subsequent radiative forcing under clear sky conditions is shown in Figure 4b. As can be expected from our sensitivity experiments, the forcing is largest over the ocean (values up to $-7 Wm^{-2}$) and lower over the continental areas. In our second case at the 25th of July 1995, peak values of 0.28 (OD) are found over Central Europe, resulting in a radiative forcing of $-6 Wm^{-2}$ (Figure 5). At the 19th of August 1996 (third case), the area covering the North Sea up to Italy is highly polluted with sulfate aerosols with peak values of 0.26 (OD). For the Dutch grid we computed a radiative forcing of $-7 Wm^{-2}$ (Figure 6).

Conclusion

- The analytical expression is a useful tool for estimating the clear sky radiative forcing due to sulfate aerosol changes.
- Locally, the negative radiative forcing due to sulfate aerosols can exceed largely the positive forcing due to greenhouse gases.
- Adding increases of other manmade aerosols, such as nitrate aerosols and organic compounds (measured at ECN, Petten), will result in even larger negative values of the radiative forcing.

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