MODELLING OF AEROSOL CHEMISTRY AND TRANSPORT OVER EUROPE AND COMPARISON WITH MEASUREMENTS

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Introduction

Because of the short lifetime of aerosols (days-weeks),the radiative forcing associated with aerosols varies strongly spatially and temporally (see *van Dorland et al.*, poster). Current measurement networks of sulfate and other aerosol species are unable to provide a representative picture of the aerosol distribution in time and space. Therefore, models are needed that describe the formation and transport of aerosols. Measurements, in return, are needed to validate these models. Satellite remote sensing potentially can provide measurements of column integrated aerosol optical properties on local, regional and global scales. In this work the joint KNMI and IMAU chemical tracer transport model (CTM) TM3 that describes the global cycle of sulfate aerosols is compared to GOME retrievals of aerosol optical thickness. The possible role of nitrate is briefly discussed. This work is part of the NRP project: *Aerosol; cycle and influence on the radiation balance*

Modelling the Sulfur Cycle

The CTM TM3, describes the transport of chemical tracers by observed winds, convective fluxes and boundary layer diffusion taken from the numerical weather prediction model of the ECMWF. Sulfate aerosol is formed by gas phase and cloud phase oxidation of SO_2. SO_2 is either emitted from anthropogenic sources or formed by oxidation of DMS. The main loss mechanism of sulfate aerosol is scavenging by precipitation.

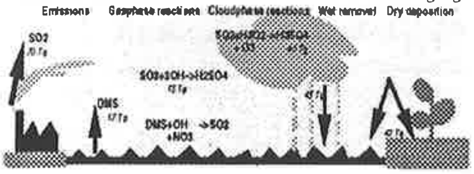

Figure 1 The Sulfur cycle in TM3 and annual global budgets.

Figure 2. shows that with its 6 hour temporal resolution the model is unable to capture the measured peak concentration of sulfate but the average tendency is well simulated.

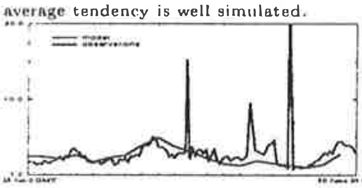

Figure 2 Modelled Sulfate concentrations (ug/m^3) compared to measurements at ECN, Petten for 25-30 June 1995 (see Hensen et al., poster).

Aerosol Retrieval from GOME

The Global Ozone Monitoring Experiment (GOME) is a spectrometer on board the ERS-2 satellite which measures radiances emanating from the atmosphere at wavelengths between 240 and 790 nm, with a spectral resolution of 0.2 to 0.4 nm. The pixel size of GOME is 320x40 km^2. From the GOME spectra five wavelength bands were selected, with effective wavelengths of 342, 355, 368, 388, and 400 nm. In the aerosol retrieval algorithm the satellite measured radiance is fitted to radiative transfer calculations. To separate atmospheric and surface contributions to radiances, assumptions on the surface albedo are used. Aerosol is assumed to be a mixture between an anthropogenic and a seasalt type. The retrieval produces the relative contribution of the two types as well as the the optical depth of the aerosols. The retrieval works well for cloud free cells.

Results

The aerosol retrieval algorithm was applied to GOME data over Europe, for 11 to 22 August 1997. To identify cloud-free regions the Polarization Measuring Devices (PMDs) of GOME were used and data from AVHRR.

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The aerosol retrieval algorithm described above was used to determine the aerosol optical depth.

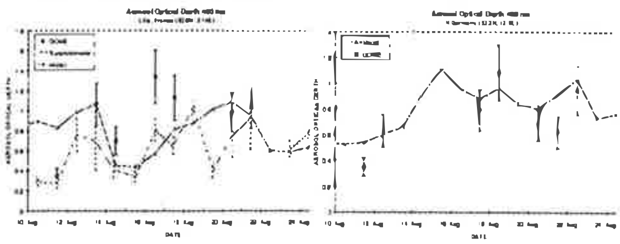

Figure 3a and 3b

To compare the satellite retrieval to the model, the sulfate column as derived by the model was translated into the aerosol optical depth at 400 nm :

$AOD_{400} = [SO_4]\alpha_{SO_4} f(RH) p_{SO_4}^{-1}$

where AOD is the total aerosol optical depth at 400 nm, $[SO_4]$ is the column burden of sulfate mass, α_{SO_4} is the scattering mass efficiency of sulfate aerosol at 400 nm, at low relative humidity, $f(RH)$ is the enhancement factor due to hygroscopic growth and p_{SO_4} is the fraction of the light scattering caused by sulfate. To compute the following values are used: $\alpha_{SO_4}(400nm) = 7.5m^2g^{-}1$, $f(RH) = 1.5$ and $p_{SO_4} = 0.5$. Figure 3a shows that the GOME retrieval and the sunphotometer data correlate very well. However the satellite retrieval is approximately 20% higher than the sunphotometer. The model data follows the trend of the sunphotometer and the satellite retrieval, except for 10, 11, and 19 August. The correlation in Figure 3b between the satellite retrieval and the model is 0.82.

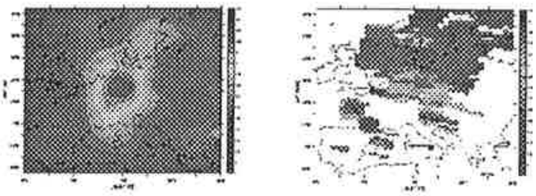

Figure 4a modelled AOD and 4b GOME retrieval of AOD for 20-22 August.

Focusing on Northern Europe, it can be seen that the spatial distribution of the aerosol optical depth is very well predicted by the model. The maximum in both retrieval and model image are over Germany. However, over most of the image, the model derived aerosol optical depths are somewhat higher than the satellite retrieved values.

Nitrate Aerosol

Nitrate aerosols may gain future importance. Due to air pollution abatement in the USA and Western Europe, the emissions of sulfur gases from fossil fuel use have been reduced by a factor of 2 to 4 since the 1970s. Emissions of nitrogen oxides have remained relatively constant during this period. The relative importance of nitrate versus sulfate aerosols is therefore increasing in most industrialized regions of Europe and North America.

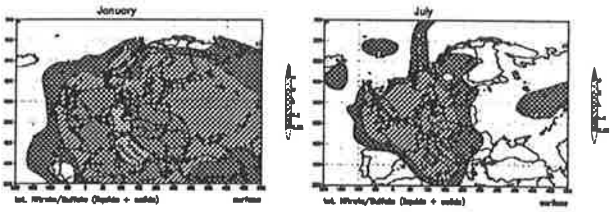

Figure 5 Ratio Nitrate/Sulphate in aerosol for January and July as calculated with the equilibrium model.

Volatile aerosol species are not yet treated in global models due to the lack of reliable measurements and because of difficulties in treating the complex chemistry. We have developed a parameterization to simulate in global models the chemical composition of atmospheric aerosol particles, the gas-partitioning between the gas phase and the liquid-solid phases, and the aerosol associated water. The parameterization, based on a relationship between activity coefficients and relative humidity, has been derived by non-linear curve fitting using the results from a recent version of the thermodynamic gas/aerosol equilibrium model SCAPE. First results indicate that over large area in Europe nitrate aerosol concentrations are of the same order of magnitude as sulfate concentrations especially in winter (see also Figure 5).