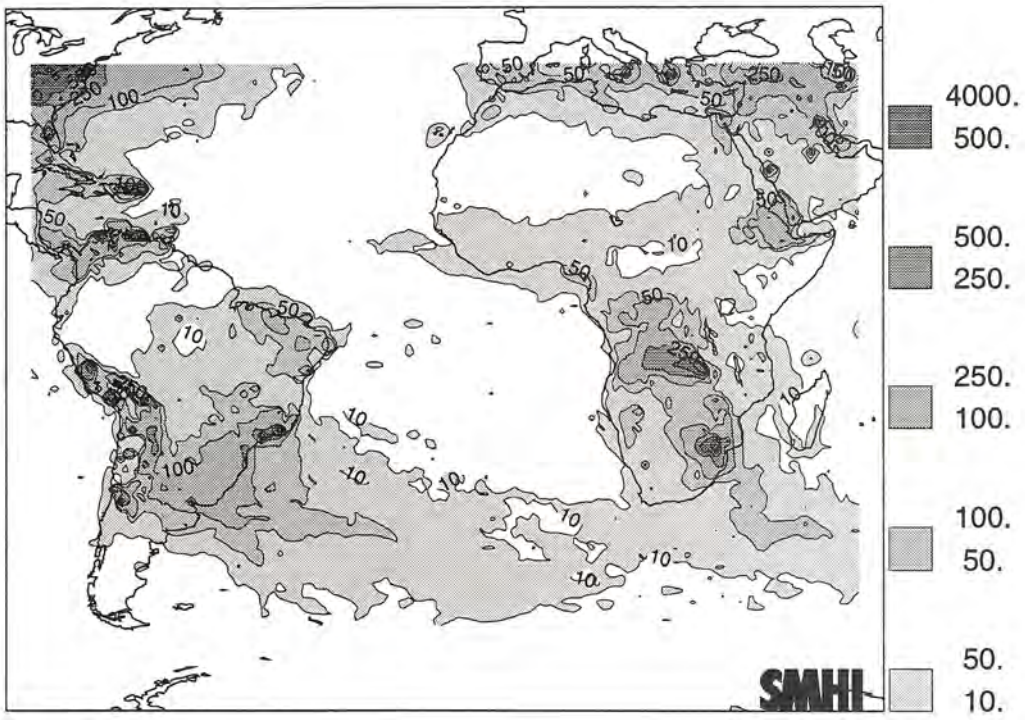
SMHI RMK
No.73, December 1996



Modelling of anthropogenic sulfur deposition for the African and South American continents

Lennart Robertson

SMHI REPORTS METEOROLOGY and CLIMATOLOGY

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CA-Tryck AB Norrköping 1996

Report Summary / Rapportsammanfattning

Issuing Agency/Utgivare

Swedish Meteorological and Hydrological Institute
S-601 76 NORRKÖPING
Sweden

Report number/Publikation

RMK No. 73

Report date/Utgivningsdatum

December 1996

Author (s)/Författare

Lennart Robertson

Title (and Subtitle/Titel

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Abstract/Sammandrag

Acidification problems in developing countries are expected to become more prevalent in the coming decades. Assessments of means of abatement strategies are likely to become of vital interest. This paper presents some preliminary results of modelling of acidic deposition due to anthropogenic emissions of sulfur in Africa and South America, using emission data for 1987. The calculations indicate that the anthropogenic perturbation of the sulfur deposition exceeds 100 mg S $m^{-2}yr^{-1}$ for southern Africa, areas around the Red Sea, Caribbean Islands, northernmost South America, central Andes, and the eastern parts of Brazil and Argentina. Substantial areas around the major source regions receive more than 250 mg S $m^{-2}yr^{-1}$. Even higher values are calculated for areas surrounding large cities as Carracas, Lima, Rio de Janeiro and Sao Paulo, Pretoria and around the borderline between Zaire and Zambia.

Key words/sök-, nyckelord

Sulfur deposition, Africa, South America, Atmospheric modelling

Supplementary notes/Tillägg

Number of pages/Antal sidor

10

Language/Språk

English

ISSN and title/ISSN och titel

0347-2116 SMHI Reports Meteorology Climatology

Report available from/Rapporten kan köpas från:

SMHI
S-601 76 NORRKÖPING
Sweden

Modelling of anthropogenic sulfur deposition to the African and South American continents

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Abstract. Acidification problems in developing countries are expected to become more prevalent in the coming decades. Assessments of means of abatement strategies are likely to become of vital interest. This paper presents some preliminary results of modelling of acidic deposition due to anthropogenic emissions of sulfur in Africa and South America, using emission data for 1987. The calculations indicate that the anthropogenic perturbation of the sulfur deposition exceeds 100 mg S $m^{-2}yr^{-1}$ for southern Africa, areas around the Red Sea, Caribbean Islands, northenmost South America, central Andes, and the eastern parts of Brazil and Argentina. Substantial areas around the major source regions receive more than 250 mg S $m^{-2}yr^{-1}$. Even higher values are calculated for areas surrounding large cities as Carracas, Lima, Rio de Janeiro and São Paulo, Pretoria and around the borderline between Zaire and Zambia.

1. Introduction

Anthropogenic sulfur emissions in the developing parts of the world are likely to increase substantially in the future and projections for the coming decades indicate that severe environmental problems are likely to occur (Rodhe *et al.*, 1992). This is especially pronounced for the East Asian area where some of the most rapid growing economies are found today, but may also be true for some regions over the African and South American continents. Previous efforts to model the distribution and deposition of sulfur compounds over this area have used coarser resolution models (e.g Langner and Rodhe 1991, Jacobs *et al.* 1995). The present study was initiated 1994 within the project "Acidic Deposition in Developing Countries" by the Stockholm Environment Institute (SEI). The objective is to provide estimates of sulfur deposition with high horizontal resolution appropriate for evaluating critical loads in developing countries, which is the main task of the project conducted by SEI. The role of SMHI is to provide the deposition estimates by applying the atmospheric transport model MATCH at high resolution. We acknowledge the Swedish International Development Cooperation Agency (Sida) for the support of this project.

2. The atmospheric transport model

The MATCH (Mesoscale Atmospheric Transport and CHemistry model) model has been developed as a tool for air pollution assessment studies on different scales with support from the Swedish Environmental Protection Agency (Swedish EPA) (Langner *et al.*, 1996). It has primarily been used as a basis for decision making concerning environmental protection, such as assessment studies over Sweden (on a resolution of 20×20 km) and for high resolution applications within subregions within Sweden (5×5 km) on commission by local environmental authorities. Studies for larger areas in Europe have recently been initiated (e.g Persson and Ullerstig, 1996).

MATCH is an Eulerian three-dimensional "off-line" model, which means that the meteorological data needed, to describe transport, transformation and deposition processes, are taken from some external source and fed into the model at regular time intervals (see section 4). The model used here includes horizontal and vertical transport, vertical diffusion, dry deposition, wet scavenging and chemical transformations. The model design enables a flexible choice of horizontal and vertical grid, principally defined by the input meteorological data. For the application presented here the model has 10 layers in the vertical extending up to 8.5 km on a latitude-longitude horizontal grid. Approximate heights to the different model levels are given in Figure 1. The advective transport is treated by a modified fourth order Bott scheme in the horizontal (Bott, 1989), and a zero order upstream scheme in the vertical. Vertical diffusion is based on K-theory where exchange coefficients follow suggestions by (Holtslag *et al.*, 1991) for neutral and weakly unstable conditions.

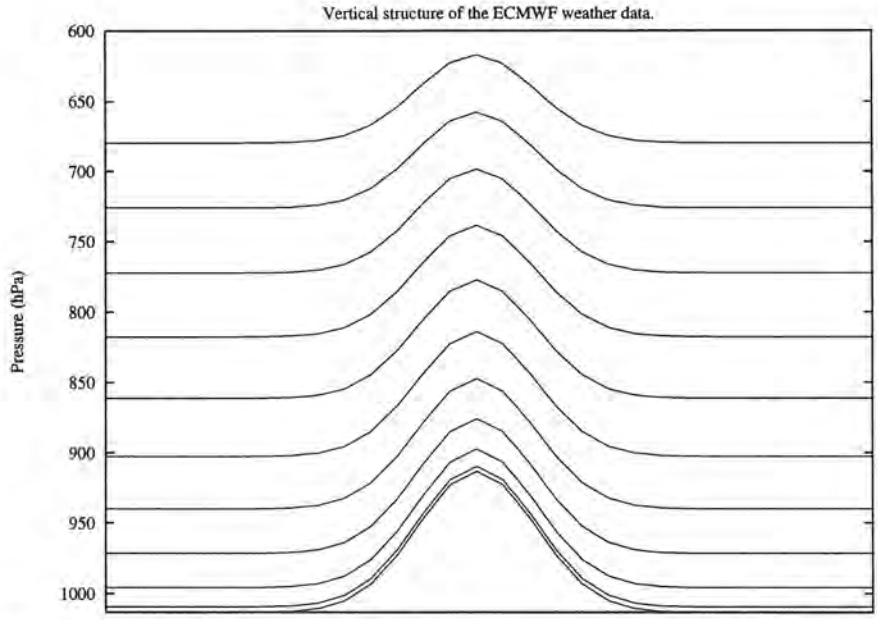

Fig. 1. Approximate heights to various model levels as defined by the ECMWF input weather data. Heights are given as pressure levels (hPa). The lowest contour assumes the ground surface.

TABLE I

Sink parameters for SO_2 and SO_4^{2-},respectively. The scavenging time-scale is related to a precipitation intensity of 1 mm/hour.

SO_2		
Scavenging time-scale	Dry deposition velocities (cm/s)	
(hours)	Land	Sea
2.8	0.5 - 0.8	0.8

SO_4^{2-}		
Scavenging time-scale	Dry deposition velocities (cm/s)	
(hours)	Land	Sea
1.0	0.1	0.05

For convective conditions the vertical diffusion is assumed to be dependent on a convective turnover time-scale defined by the boundary layer height and the convective velocity-scale. Vertical mixing by deep convection is not treated so far.

The chemistry scheme employed is a simplified linear transformation of SO_2 to SO_4^{2-} (Eliassen and Saltbones, 1983). The gase-phase transformation is modeled using a seasonal varying bulk transformation rate with a mean value of $3 \cdot 10^{-6}$ s^{-1} and an amplitude of $2 \cdot 10^{-6}$ s^{-1}. The wet-phase transformation of SO_2 is parameterized by introducing a scavenging coefficient for SO_2, enhancing the deposition and implicitly treating the wet-phase chemistry at a bulk rate. Table I shows the deposition parameters applied. The dry deposition velocity for SO_2 over land areas has a diurnal variation within the range presented. The rates of this simplified scheme are similar to those used for European conditions.

3. Emission data

Emissions of SO_x are taken from the Global Emission Inventory Activity (GEIA) (Benkowitz *et al.*, 1993) (see Figure 2). The inventory takes fuel combustion and industrial activities into account, valid for year 1987. Biomass burning is not included in this study. The total emissions of SO_x within the model area is 13.3 Tg S yr^{-1}, divided into 3.0 Tg S yr^{-1} for Africa, 4.1 Tg S yr^{-1} for South America and 6.2 Tg S yr^{-1} for surrounding areas within the model domain (S-E USA, the Mediterranean area, Middle East and Saudi Arabia). Figure 3 shows these numbers together with the emissions for Asia, Europe and North America for comparison. All numbers are derived from the GEIA inventory of anthropogenic emissions (biomass burning excluded). The emissions are assumed to be partitioned into 95% gaseous sulfur dioxide (SO_2) and 5% particulate sulfate (SO_4^{2-}) before used in the calculations.

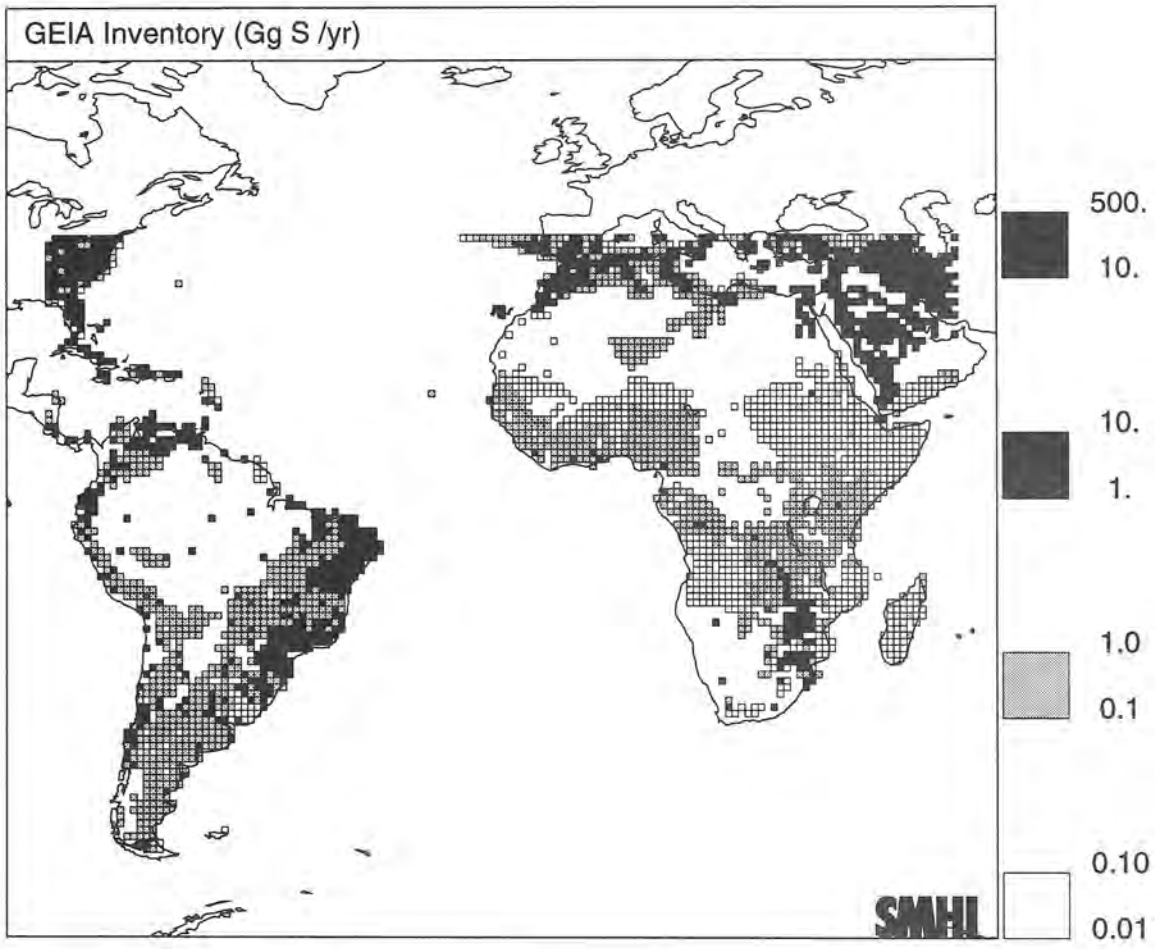

Fig. 2. Anthropogenic emissions used on $1° \times 1°$ resolution (Gg S yr^{-1}). Based on the GEIA inventory (Benkovitz, 1993).

4. Meteorological data

The forcing weather information is taken from European Centre for Medium Range Forecasts (ECMWF) global analyses (available at 6 hour intervals). No precipitation analysis is available in the ECMWF data, so 6 hour ECMWF precipitation forecasts were utilized. For computational reasons we have limited the calculations to a few shorter periods which roughly comprise a representative year. The selected periods are in January, April, July and September (1994) with a total number of 122 days. These periods include winter and summer monsoons, and the intermediate

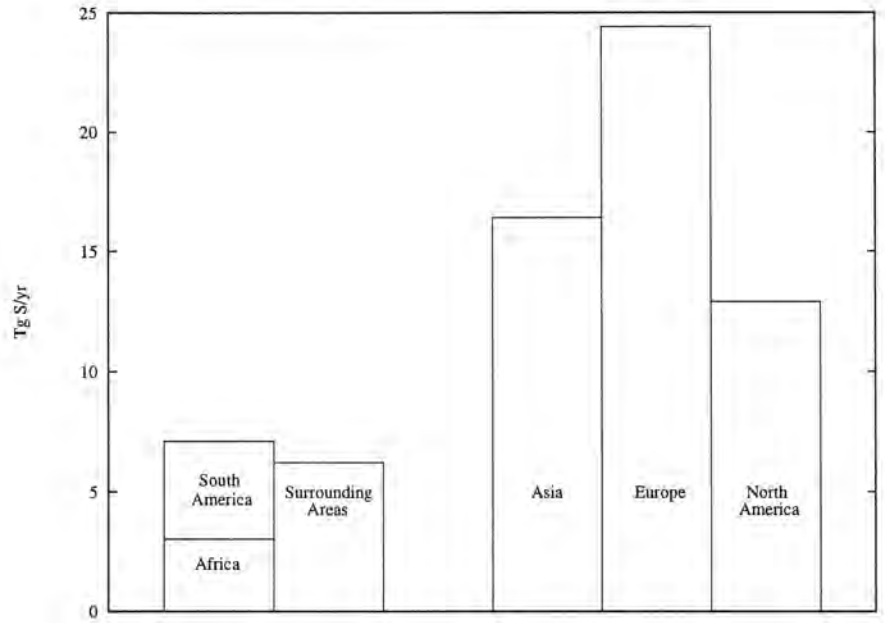

Fig. 3. Emission budgets for Africa and South America and surrounding areas (S-E USA, Mediterranean area, Middle East and Saudi Arabia) used in the simulations. For comparison emissions for Asia shown (50 °E - 160 °E, 30 °S - 50 °N), Europe and North America is shown. Based on the GEIA inventory (Benkovitz, 1993).

dry periods. Provided that the selected periods make up a representative subset of a complete year, calculated depositions should arithmetically underestimate the annual deposition by a factor $365/122 = 3$. Of specific interest are the precipitation intensities used to parameterize the wet scavenging. Figure 4 shows the accumulated precipitation used in this study, scaled to represent annual precipitation amounts, and thus comparable with analyzed annual precipitation data (Rudolf, 1996) as shown in Figure 5. The precipitation pattern derived from the ECMWF data shows greate similarities with analyzed precipitation pattern. The amounts tend however to be underestimated primarily over the Sahara region, southern and northern Atlantic. In general we could though be fairly confident in calculated wet depositions, as an approximation of a full year simulation.

5. Results and Discussion

5.1. Concentration and deposition of sulfur compounds

The calculated spatial distribution of concentrations and depositions are shown in Figures 8 to 11 as annual SO_4^{2-} and SO_2 mean concentrations, annual wet deposition of SO_4^{2-} and annual total deposition of sulfur, respectively.

From Figure 11 we can draw the following conclusions of the contributions from anthropogenic emissions to the total depositions of sulfur. The deposition load exceeds 10 mg S $m^{-2}yr^{-1}$ for most parts of the two continents, with exception for the Sahara desert, central Africa, Somalia, upper part of Amazona Basin, Surinam, French Guiana, and the southernmost part of South America. Exceedances of 100 mg S $m^{-2}yr^{-1}$ are calculated for the Caribbean Islands, northernmost South America, Central Andes, northern Argentina and western Brazil, together with the very northern parts of Africa, areas adjacent to southern Red Sea, and central part of southern Africa. Largest depositions are calculated for a minor number of spots as western Caribbean Islands (Puerto Rico),

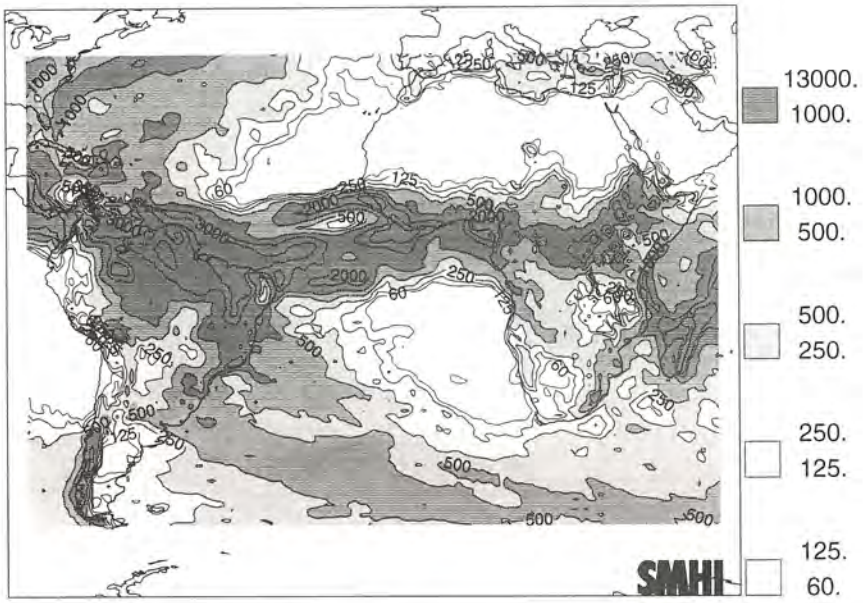

Fig. 4. Acummulated precipitation from used ECMWF weather data, and scaled to represent annual precipitation (mm yr^{-1}).

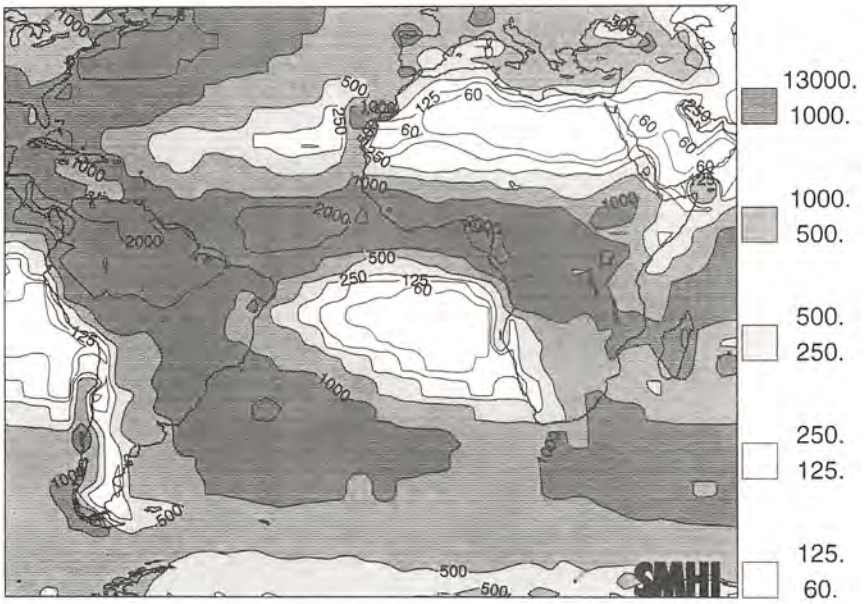

Fig. 5. Annual precipitation estimates for 1988 (Rudolf, 1993)

the area around Caracas in Venezuela, the areas around Lima and Arequipa in Peru, some spots along the eastern border of Chile, the areas around Rio de Janeiro and São Paulo in Brazil, an area that is crossed by the borderline of Zaire and Zambia, and for Transvaal in South Africa.

In Table II, and the Figures 6 and 7 the results are presented as spatial averages for a sample of countries within the model domain. In Table II are corresponding estimates for East Asia (Robertson *et al.*, 1996) and Europe (Iversen *et al.*, 1991) given for comparison.

This study was restricted to anthropogenic emissions (biomass burning excluded). Estimates of emissions of sulfur from biomass burning are of the order of 2-3 Tg Syr^{-1} (Langner and Rodhe, 1991) globally. Assuming that three quarters of this originate in either Africa or South America implies that biomass burning is a significant source compared to the fossil fuel source. Natural

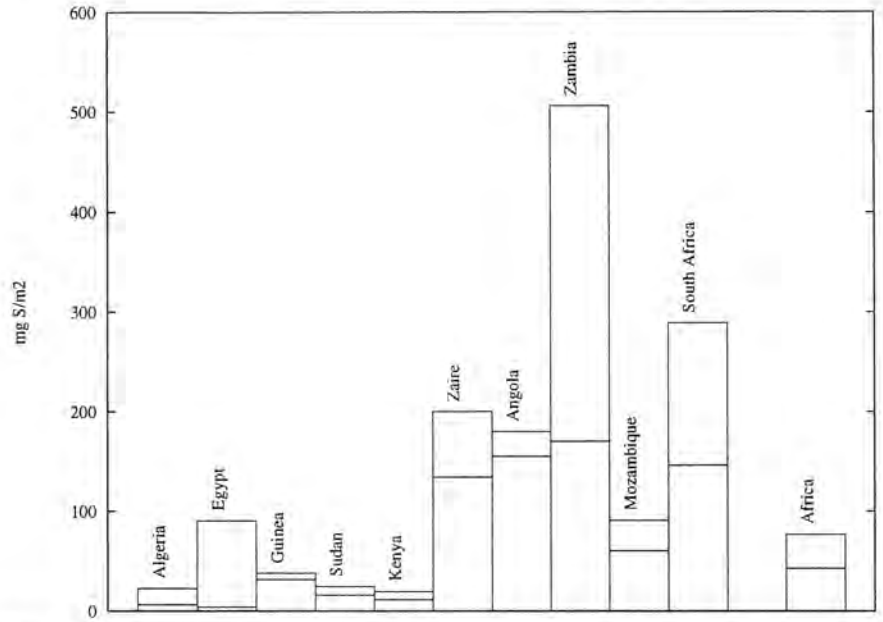

Fig. 6. Box diagram of the areal mean deposition for a sample of countries in Africa, together with the areal mean deposition for the whole continent (the rightmost box) in mg S m^{-2}. Each box i divided into two parts denoting wet deposition (bottom) and dry depostion (top).

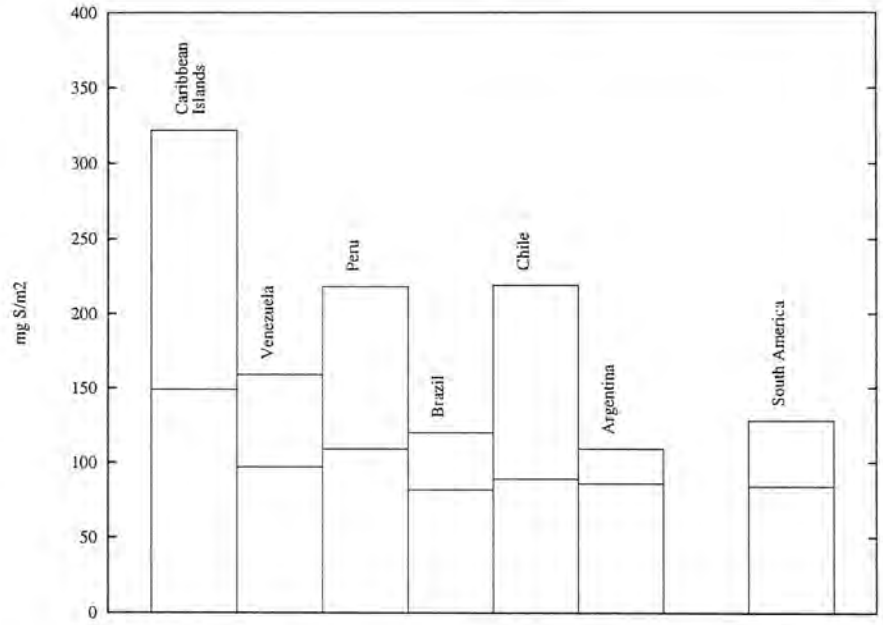

Fig. 7. Box diagram of the mean deposition for a sample of countries in South America, together with the areal mean deposition for the whole continent (the rightmost box) in mg S m^{-2}. Each box i divided into two parts denoting wet deposition (bottom) and dry depostion (top). Note the different scale from previous figure.

sources are expected to contribute of the order of $\sim$ 100 mg S $m^{-2}yr^{-1}$ over sea areas (Galloway and Gaudry, 1984), where the calculated contributions from anthropogenic emissions in this study are less then 10 mg S $m^{-2}yr^{-1}$ for most parts of the Atlantic, but increasing up to the same order as the natural contribution for the southern Atlantic.

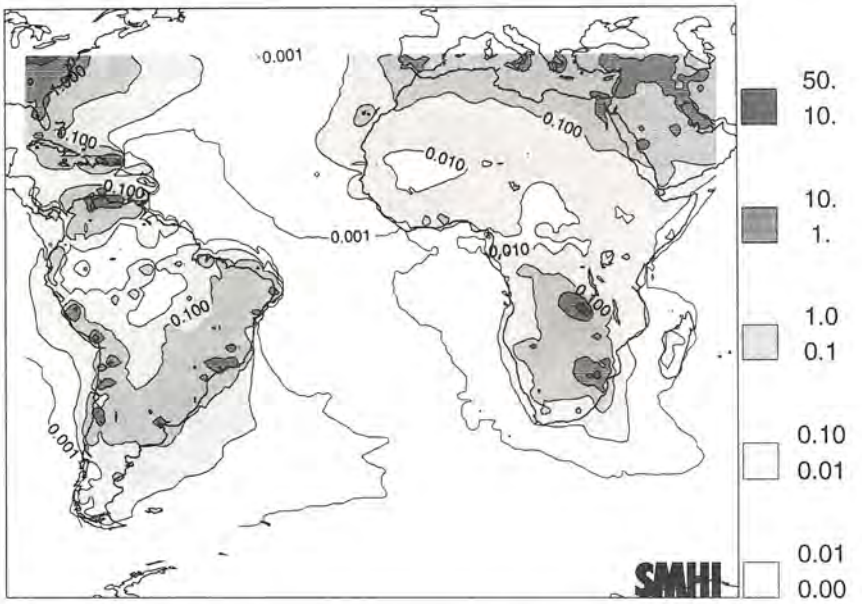

Fig. 8. Annual mean concentration of SO_2 in μg S m^{-3}.

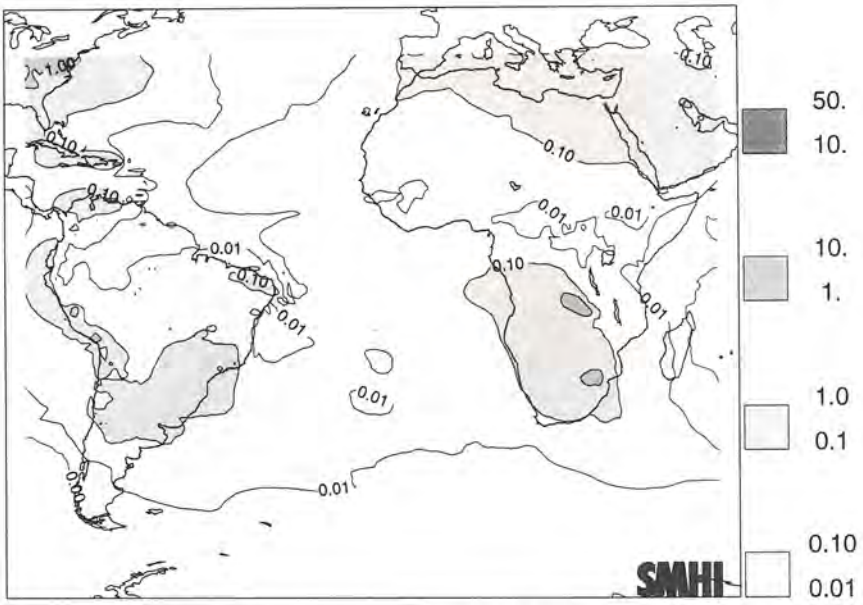

Fig. 9. Annual mean concentration of SO_4^{2-} in μg S m^{-3}.

5.2. Validation

Table III shows some comparisons with measured concentrations in precipitation. A complete validation of the model calculations is not possible to perform on these data, due to different periods for which calculations and measurements are valid and by limitations in the calculations to anthropogenic contributions. Rodhe *et al.* (1981) conclude that natural sources significantly contributes to their measurements for East Africa, which is not contradicted by the calculations. The importance of natural or biomass burning sources become clear as the calculated contribution to the sulphur concentration in precipitation is just ~30 % of the measured one. The importance of biomass burning sources are also most likely true for the measurements from the Ivory Coast (Lacaux *et al.*, 1987). The measured and calculated values for Congo are almost identical which indicate an

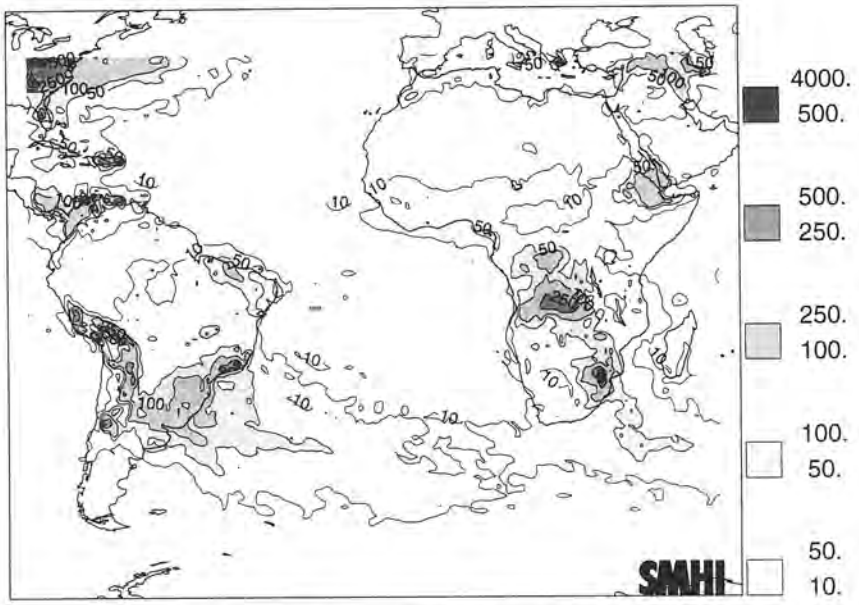

Fig. 10. Annual wet deposition of sulfur in mg S m^{-2}

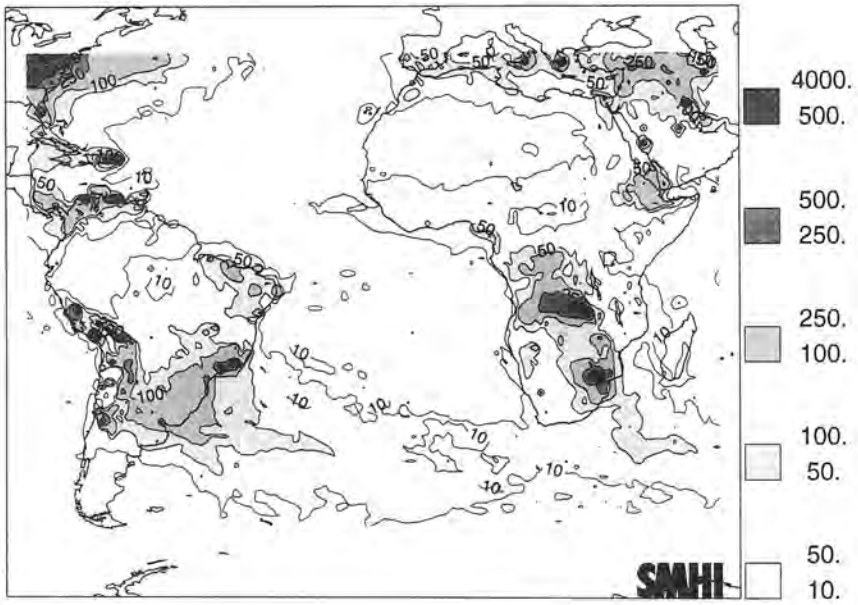

Fig. 11. Annual total deposition of sulfur in mg S m^{-2}

overestimation by the model. The calculations versus measurements for northern South America are more ambiguous but it could be concluded that the model calculations and measurements are within the same order of magnitude, which also is the case for the Amazona Basin. The quite large discrepancy for Bermuda are likely due to influence from sources over the North American continent that are just to a very limited extent included in this study.

It is concluded that the calculations in general is within the same order of magnitude as measurements, while any conclusions of the details could not be made.

TABLE II

Average deposition of sulfur for a sample of countries and areas. Units: mg S $m^{-2}yr^{-1}$.

Country	Dry deposition SO_2	Dry deposition SO_4^{2-}	Wet deposition	Total deposition
Africa	30	3.7	47	77
Algeria	15	3.0	6.5	24
Angola	17	7.3	156	181
Egypt	124	9.1	3.9	90
Guinea area[1]	5.8	0.5	32	38
Kenya	7.3	0.5	12	19
Mozambique	28	2.5	61	91
South Africa	130	13	146	189
Sudan	6.1	2.5	16	25
Zaire	55	7.9	137	200
Zambia	319	16	170	506
South America	42	2.9	84	128
Argentina	19	3.0	86	109
Brazil	19	2.6	82	120
Caribbean Islands	170	3.6	149	322
Chile	124	6.4	89	219
Peru	101	7.8	109	218
Venezuela	59	3.0	97	159
Europe[2]				$\sim$ 1300
Poland				$\sim$ 4000
Scandinavia				$\sim$ 500
Spain				$\sim$ 500
The Black Triangle				$\sim$ 14000
Western Germany				$\sim$ 2500
Asia[3]				$\sim$ 300
Cambodia				34
India				280
Myanmar				170
Southern China				950
Thailand				460

(1) Guinea, Sierra Leone, Liberia, Ivory Coast, Ghana, Togo, Dahomey, Nigeria, Cameroon

(2) Iversen *et al.*, 1991

(3) Robertson *et al.*, 1996

6. Conclusions

This study provides quantitative estimates of concentration and deposition of sulfur compounds due to anthropogenic emissions for Africa and South America. The study does not present a full picture of the deposition load since biomass burning and natural sources are not included. (From a modelling point of view it should be stressed that transformation rates are not accounting for the various conditions that are expected throughout the area). Deep convection is neither treated which is expected to be an important transport path in the tropics. However, comparisons with available precipitation chemistry data qualitatively indicate that the calculations are reasonable.

TABLE III

Model estimates versus measurements. Units μeq/l.

Area	Model	Measurement	Reference
Ivory Coast	1.4	10.0	Lacaux *et al.*, 1987
East Africa	4.4	15.6	Rodhe *et al.*, 1981
Congo	10	10.5	Lacaux *et al.*, 1992
Bermuda	6.6	26-34	Galloway *et al.*, 1988
Venezuela			Sanhuza *et al.*, 1992
Camburito	9.0	9.0	
La Paragua	1.5	4.9	
J. de Tigre	16	2.1	
Chaguarama	15	4.6	
Guri	8.0	2.9	
Venezuela	8-16	3.0	Galloway *et al.*, 1982
Amazona Basin	2.5	3.5	Andrea *et al.*, 1988

Acknowledgements

Financial support was obtained from Sida through SEI, under the project "Acidification studies in Africa and South America". Special thanks to Joakim Langner and Åsa Gerger for careful review of the paper and to Laura Gallardo, Department of Meteorology at Stockholm University, for pointing out the available measurements for model comparisons.

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ISSN 0347-2116