

Global Inventory of Sulfur Emissions With $1^{\circ} \times 1^{\circ}$ Resolution

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A global inventory of gaseous sulfur emissions with $1^{\circ} \times 1^{\circ}$ resolution is described. Emissions from fuel combustion and industrial activities are estimated for countries where no detailed inventories are available by using economic data for individual sulfur-emitting activities, sulfur emission factors, and information on sulfur recovery. Fuel sulfur contents are specified as a function of fuel type and country of origin and are conserved during international trading. This procedure for estimating emissions reproduces well existing inventories for countries in Europe and North America, suggesting that it can be applied with some confidence to other countries. Emissions from biomass burning, volcanoes, and oceans are derived from existing data bases and are distributed with fine spatial resolution. Emissions from terrestrial vegetation are computed as a function of leaf area index, temperature, and solar radiation. The global emission of sulfur gases in 1980 is estimated to be 102 Tg S yr^{-1} , apportioned among fuel combustion and industrial activities (76%), marine biosphere (12%), volcanoes (9%), biomass burning (2%), and terrestrial biosphere (1%). Detailed breakdowns of anthropogenic and natural sources are given for individual countries and regions. Anthropogenic sources account for 84% of total sulfur emissions in the northern hemisphere and for 50% in the southern hemisphere. Biomass burning dominates emissions in central Africa during the dry season but is of minor importance elsewhere. Smelters dominate anthropogenic emissions in the Arctic and in the southern hemisphere. Volcanoes are significant contributors to the sulfur budget in Central America, the East Indies, and some subarctic regions.

1. INTRODUCTION

This paper describes a $1^{\circ} \times 1^{\circ}$ global inventory of gaseous sulfur emissions for use in atmospheric models. Development of the inventory is motivated by the need to understand the origin of atmospheric sulfate in the context of several regional and global environmental issues, in particular acid deposition [Schwartz, 1989], visibility [Barrie *et al.*, 1981], and climate [Shaw, 1983; Charlson *et al.*, 1987, 1992a]. Sulfate is produced in the atmosphere by oxidation of reduced sulfur gases and is also emitted directly by mechanical processes (sea spray, soil dust). Atmospheric oxidation is the most interesting source of sulfate from an environmental standpoint, because it generates acidity and because it produces submicron aerosol. The main sulfur gases emitted to the atmosphere are SO_2 (combustion, industrial, volcanoes), dimethylsulfide (biota), and H_2S (principally biota). Emissions of COS, CS_2 , methyl mercaptan, and dimethyldisulfide are relatively small [Andreae, 1992] and will not be considered here. Dimethylsulfide and H_2S are oxidized in the atmosphere to SO_2 , and SO_2 is in turn oxidized to sulfate which is removed by deposition. The time scales for these processes range from hours to a few weeks.

Understanding the variability of sulfate concentrations in the atmosphere requires a three-dimensional model of atmospheric transport and chemistry including fine resolution of emissions. The present emission inventory is designed for this purpose. We place particular emphasis on estimating anthropogenic emissions in countries outside Europe and North America; this work is described in section 2. Distributions of other sulfur sources are derived in section 3. Prominent features of the inventory are discussed in section 4. A copy of the inventory with $1^{\circ} \times 1^{\circ}$ spatial resolution and monthly temporal resolution is available from D.J. Jacob.

2. EMISSIONS FROM FUEL COMBUSTION AND INDUSTRIAL ACTIVITIES

Procedure

The standard procedure for computing sulfur emissions from fuel combustion and industrial activities is to multiply the level of activity (e.g., consumption or production rate of a commodity) by a sulfur emission factor which depends on the type of activity, the sulfur content of the commodity, and the extent of sulfur recovery [Environmental Protection Agency, 1985]. Detailed emission inventories for Europe, the Soviet Union, North America, and Japan were compiled by government agencies in the 1980s (Table 1). We focus on estimating emissions for the rest of the world, where few data are available. Global emission inventories have been reported by Cullis and Hirschler [1980] and Varhelyi [1985], among others, but include only limited spatial resolution. Our approach is to develop a general procedure for estimating sulfur emissions from individual countries in 1980 and to evaluate the procedure by comparison with existing inventories for countries where such data are available.

The principal sulfur-emitting activities are coal and petroleum combustion, petroleum refining, and smelting of copper, lead, and zinc ores [Cullis and Hirschler, 1980]. Annual 1980 consumption totals of hard coal, lignite, crude petroleum, 15 refined petroleum products, and wood fuel in individual countries are obtained on computer tape from the United Nations [1989]. The tape includes additional data for many countries on fuel imports (including country of origin) and fuel use (energy or nonenergy, industrial or residential). Annual 1980 totals of metal production in individual countries are obtained from the Statistical Yearbook [United Nations, 1985].

Sulfur emission factors for individual activities, assuming no sulfur recovery, are adopted from recommendations by the Environmental Protection Agency [1985] and are listed in Table 2. The emission factors for fuel consumption depend on the fuel sulfur content, which varies widely with the type and origin of the fuel. We compiled from recent surveys the average sulfur contents of hard coal, lignite, and petroleum produced in individual

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TABLE 1. Major Data Bases Used in the Construction of the Inventory

Data Base	Domain	Resolution	Year	Reference
Anthropogenic sulfur emission ^a	western Europe ^b	50x50 km ²	1980	1 (OECD)
	eastern Europe ^c	country	1980	2 (IIASA)
	Soviet Union	300x300 km ²	1980	3
	Soviet Union	27 regions + 611 point sources	1989	4
Population	North America	1/6°x1/4°	1980	5 (NAPAP)
	North America	1/6°x1/4°	1985	6 (NAPAP)
	Japan	80x80 km ²	1983	7
	global	1°x1°	1980	8
Fuel production and consumption ^d	global	country	1980	9
Metal production ^e	global	country	1980	10
Sulfur recovery ^f	global	country	1980	11 (USBM)
Biomass burning ^g	global	1°x1°, monthly	1980	8
Volcanic eruptions	global	locations, dates	1980	12
Plume sizes of noneruptive volcanoes ^h	global	locations	1981-1982	13
Surface wind speed ⁱ	global	2°x2°, monthly	climatological	14 (COADS)
Sea surface temperature	global	4°x5°, monthly	climatological	15
DMS seawater concentration	global	zonal, season	climatological	16
Vegetation type, cultivation intensity	global	1°x1°	1969-1980	17
Normalized differential vegetation index	global	1°x1°, monthly	1983-1984	18
Soil type	global	1°x1°		19

References are: 1, *Lubkert and De Tilly* [1989]; 2, *Alcamo et al.* [1987]; 3, *Semb* [1985]; 4, *Berlyand* [1990]; 5, *Environmental Protection Agency* [1986]; 6, *Saeger et al.* [1989]; 7, *Fujita et al.* [1991]; 8, *J.A. Logan and J.E. Dignon* (manuscript in preparation, 1992); 9, *United Nations* [1989]; 10, *United Nations* [1985]; 11, *Morse* [1984]; 12, *Sirkin et al.* [1981]; 13, R. E. Stoiber (personal communication, 1990); 14, *Woodruff et al.* [1987]; 15, *Robinson and Bauer* [1981]; 16, *Bates et al.* [1987]; 17, *Matthews* [1983]; 18, *Fung et al.* [1987]; 19, *Zobler* [1986]. Acronyms are: OECD, Organization for Economic and Cooperative Development; IIASA, International Institute for Applied Systems Analysis; NAPAP, National Acid Precipitation Assessment Program; USBM, United States Bureau of Mines; COADS, Comprehensive Ocean-Atmosphere Data Set.

^a Fossil fuel combustion and industrial activities.

^b Austria, Belgium, Denmark, Finland, France, W. Germany, Greece, Ireland, Italy, Luxembourg, Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, and United Kingdom.

^c Albania, Bulgaria, Czechoslovakia, E. Germany, Hungary, Poland, Romania, Turkey, and Yugoslavia.

^d Data for hard coal, lignite, crude petroleum, 15 refined petroleum products, and wood fuel. The following activities and transactions are included: gross production, production of secondary products from refineries, total imports, exports, changes in stocks, losses, consumption for non-energy use, residential consumption, and consumption by coke ovens. The *Energy Statistics Yearbook [United Nations, 1984]* contains a large subset of these data. Annual consumption totals of each fuel in each country are computed as the balances between inflows (domestic production, imports, decreases in stocks) and outflows (exports, increases in stocks), following the procedure recommended by the *United Nations* [1984]. Data on trade between countries are given by the *United Nations* [1989] for certain years later than 1980. We use 1987 data, which we scale to total imports for each country in 1980.

^e Primary copper production in 38 countries, secondary copper production in 36 countries, primary lead production in 41 countries, secondary lead production in 14 countries, and primary zinc production in 33 countries.

^f Amounts of sulfur recovered from petroleum refining and individual smelting activities.

^g Data for total carbon burned.

^h Data for 116 volcanoes.

ⁱ At 10 m altitude.

countries (Table 3). Data on trade between individual countries [*United Nations, 1989*] are used to establish the origins and hence the sulfur contents of the hard coal and crude petroleum consumed within a particular country. We can trace in this manner the country of origin for 94% of the hard coal and 96% of the crude petroleum traded internationally (which account for 10% and 50% of the world production totals of these two fuels, respectively). Trade data are less complete for lignite, but only 0.4% of the world production is exported from the producing countries.

Petroleum sulfur may be emitted to the atmosphere during either refining or combustion. It is best not to separate these two processes in the inventory because emission factors for petroleum

refining are unreliable [*Environmental Protection Agency, 1985*]. We calculate the total sulfur emission from petroleum uses in a given country by a four-step process. First, we obtain a preliminary value by considering that all the sulfur present in the crude petroleum consumed by the country is emitted. Second, we subtract the sulfur recovered during refining, using 1980 data for individual countries published by the United States Bureau of Mines [*Morse, 1984*]. Third, we subtract the sulfur that is not emitted due to exports, losses, increases in stocks, and nonenergy uses of domestically refined petroleum products (Table 4). Fourth, we add the sulfur present in refined petroleum products originating from sources other than domestic refining (imports, decreases in

TABLE 2. Sulfur Emission Factors From Fuel Combustion and Industrial Activities

Activity	Emission Factor
Hard coal combustion, industrial ^a	97.5% S
Hard coal combustion, residential ^a	77.5% S
Hard coal combustion, coking ^a	33% S
Lignite combustion	75% S
Petroleum uses	
nonenergy uses	0% S
combustion (kerosene)	15% S
combustion (other petroleum products)	100% S
Wood fuel combustion	72.5 g m ⁻³
Primary copper smelting	1060 kg ton ⁻¹
Secondary copper smelting ^b	225 kg ton ⁻¹
Primary lead smelting	149 kg ton ⁻¹
Secondary lead smelting	42.6 kg ton ⁻¹
Primary zinc smelting ^c	490 kg ton ⁻¹

Emission factors are for uncontrolled emissions with no sulfur recovery and are taken from the *Environmental Protection Agency* [1985] unless otherwise indicated. Units are: % S = percent weight of sulfur in the fuel emitted during consumption; g m⁻³ = g S emitted per m³ of wood burned; kg ton⁻¹ = kg S emitted per metric ton of metal produced.

^a Hard coal includes bituminous coal, subbituminous coal, and anthracite; bituminous coal is the predominant form. Sulfur emission factors are for bituminous coal.

^b Data are from *Cullis and Hirschler* [1980].

^c Zinc ore is mainly ZnS [*Environmental Protection Agency*, 1985]. We assume that all the SO₂ produced in the roaster reaction (2ZnS + 3 O₂ → 2ZnO + 2SO₂) is emitted.

stocks) and used for energy. The sulfur contents of the refined products are scaled from the global average values in Table 4 to reflect the sulfur content of the domestically refined crude petroleum. Data for each country on imports, exports, changes in stocks, losses, and uses of the individual refined petroleum products are taken from the *United Nations* [1989] tape.

Emission factors for smelting of ores should depend on the sulfur content of the ore, but sulfur content data are generally lacking for countries outside Europe and North America. We therefore adopt fixed emission factors based on average sulfur contents for United States ores (Table 2). The resulting emission estimates for each country, and for each activity, are reduced by the amount of sulfur recovered [Morse, 1984]. We find that 36% of total sulfur in metal ores is recovered globally.

The above procedure yields a global source of 88.4 Tg S yr⁻¹ from fuel use and industrial activities in 1980, apportioned among individual activities as given in Table 5. *Cullis and Hirschler* [1980] reported a global source of 103.6 Tg S yr⁻¹ in 1976; our estimate is lower, partly because we account for sulfur recovery, and partly because we use a lower emission factor for lignite combustion. *Varhelyi* [1985] reported a global source of 79.2 Tg S yr⁻¹ in 1979; our estimate is higher because we use a higher emission factor for industrial combustion of hard coal. *Dignon and Hameed* [1989] reported a global source of 62.8 Tg S yr⁻¹ in 1980 from fossil fuel consumption only; our corresponding estimate is higher, 81.6 Tg S yr⁻¹, because we use higher emission factors for both coal and petroleum.

We can improve our estimate of the global source in 1980 by using existing 1980 inventories for Europe (Organization for Economic and Cooperative Development (OECD); International Institute for Applied Systems Analysis (IIASA)), the Soviet Union [Semb, 1985], and North America (National Acid Precipitation Assessment Program (NAPAP)), plus a 1983 inventory for Japan [Fujita et al., 1991]. These inventories (referenced in Table 1) were compiled by government agencies, which presumably had

good access to local data on sulfur contents and emission controls. We thus obtain an improved global source estimate of 77.6 Tg S yr⁻¹ in 1980, including 11.0 Tg from western Europe, 9.4 Tg from eastern Europe, 12.5 Tg from the Soviet Union, 14.9 Tg from North America, 0.6 Tg from Japan, and 29.2 Tg from the rest of the world (computed using our procedure).

In the 1°x1° inventory we choose to replace the 1980 emission inventories for the Soviet Union and North America with more recent inventories: 1989 for the Soviet Union [Berlyand, 1990] and 1985 for North America (NAPAP). These more recent inventories were constructed with improved methodology, particularly in the case of the Soviet Union [Berlyand, 1990]. They provide probably a best estimate of the distribution of emissions for purposes of atmospheric sulfur modeling. The secular trend of emissions must be kept in mind, however. Emissions in North America decreased by 10-15% between 1980 and 1985 according to NAPAP, and emissions in the Soviet Union decreased perhaps by 33% between 1980 and 1989 (Semb [1985] gives 12.5 Tg S yr⁻¹ in 1980, and Berlyand [1990] gives 8.4 Tg S yr⁻¹ in 1989). The decrease of emissions in the Soviet Union could be explained by improved sulfur recovery over the past decade [Mytona, 1989; Edwards, 1990]. The global source in our 1°x1° inventory is 71.0 Tg S yr⁻¹, 6.6 Tg S yr⁻¹ less than our best estimate for 1980, due to the use of the more recent inventories for the Soviet Union and North America.

The existing inventories for western Europe, North America, and Japan include spatial resolution finer than 1°x1° (Table 1). For the rest of the world, we distribute emissions over the 1°x1° grid as follows. The Soviet Union inventory of Berlyand [1990] divides the country into 27 economic regions and specifies in addition emissions from 611 point sources (industrial or urban centers); we retain information on the locations of the 71 highest individual point sources (those with emissions > 2x10⁴ tons S yr⁻¹) and distribute evenly the remaining emissions in each economic region. Emissions from fuel use in countries outside western Europe, the Soviet Union, North America, and Japan are distributed on a per capita basis using a 1°x1° population map (J. A. Logan and J. E. Dignon, manuscript in preparation, 1992). Emissions from metal production in those countries are assigned to the locations of copper, lead, and zinc smelters [Jones et al., 1972; Laclavere, 1978 (Zaire); Instituto Geografico Militar, 1981 (Chile); Division of National Mapping, 1987 (Australia)].

Evaluation

Our procedure for estimating national emission inventories can be evaluated by comparing emission estimates for individual countries in Europe, the Soviet Union, and North America with existing 1980 inventories. Results are shown in Figure 1. The existing inventories are reproduced to within a factor of 2 for all countries except the Netherlands and Canada. Excellent agreement is found for countries in eastern Europe. There is slightly more scatter for countries in western Europe but the agreement is still reasonably good. No significant differences in the quality of the estimate are found between coal-burning and oil-burning economies.

The high emission estimate for the Netherlands appears to be caused by inadequate accounting of emission controls. Petroleum accounts for 90% of sulfur emissions in that country according to our estimate. Based on the sulfur recovery data of Morse [1984], it would appear that only 10% of petroleum sulfur in the Netherlands is recovered during refining; a larger fraction could however be retained by scrubbing of combustion gases, which is not included in the Morse [1984] data. Inadequate accounting of scrubbers

TABLE 3. Sulfur Contents of Fuels

Country	World Production, %	Sulfur Content, % weight	Reference
Hard Coal			
Australia	2.7	1.1	1
China	21.8	1.6	1
W. Germany	3.5	1.1	1
India	4.0	2.3	2
Poland	7.1	0.8	1
South Africa	4.3	0.6	3
Soviet Union	18.1	1.8	1
United Kingdom	4.8	1.4	1
United States	26.0	2.5	1
Others	6.9	2.4	1,2,4,5
World	100	1.8	
Lignite			
Australia	3.2	2.2	1
Bulgaria ^a	2.9	0.7	
Canada ^a	1.5	0.7	
China ^a	2.2	0.7	
Czechoslovakia	8.8	1.4	3
East Germany	29.7	0.9	6
West Germany	12.6	1.4	1
Greece	2.1	1.0	3
Hungary	2.2	3.1	3
Poland	3.4	1.1	3
Romania ^a	2.5	0.7	
Soviet Union ^a	15.4	0.7	
Spain	1.4	7.5	1
Turkey	1.3	3.3	3
United States	3.9	0.7	7
Yugoslavia	4.3	2.4	3
Others	2.6	1.8	1,3,8
World	100	1.3	
Crude Petroleum			
Algeria	1.6	0.2	9
Canada	2.4	0.5	9
China	3.6	0.5	10
Indonesia	2.6	0.1	9
Iran	2.4	1.6	9
Iraq	4.4	1.9	9
Kuwait	2.8	2.5	10
Libya	3.0	0.3	9
Mexico	3.4	2.6	9
Nigeria	3.5	0.2	9
Saudi Arabia	16.6	2.0	9
Soviet Union	20.2	1.4	10
United Arab Emirates	2.8	0.6	9
United Kingdom	2.7	0.5	10
United States	14.2	0.7	9
Venezuela	3.9	2.1	9
Others	10.2	0.5	9,10,11
World	100	1.2	

1980 production data are from *United Nations* [1989]. Countries lumped as "Others" account individually for less than 1% of world production. Sulfur contents are averages for each country and are given as percent weight of sulfur in the fuel. A complete country-by-country listing of sulfur contents is available from the authors. References are: 1, J. Vernon, International Energy Agency (IEA) Coal Research (personal communication, 1989); 2, *Varhelyi* [1985]; 3, *Reidick* [1981]; 4, *Attar and Corcoran* [1977]; 5, *Ridley* [1982]; 6, *Moller* [1984]; 7, *Environmental Protection Agency* [1985]; 8, *World Coal* [1983]; 9, *Carrales and Martin* [1975]; 10, *Oil and Gas Journal Data Book* [1987]; 11, *Cass* [1977].

^a No sulfur content data are available; a value of 0.7% S is assumed [*Environmental Protection Agency*, 1985].

is also a likely source of error for the United States, where we overestimate emissions from hard coal combustion by a factor 1.8 (hard coal accounts for 72% of total United States emissions in the NAPAP 1980 inventory). Countries outside of Europe, North America, and Japan made little use of scrubber technology in 1980, therefore our estimates for these countries should be minimally affected.

The underprediction of Canadian emissions is due in part to an unusually high contribution in the NAPAP 1980 inventory from minor sources not considered in our estimate: natural gas consumption, nickel and iron production, paper manufacturing, and some other industrial processes. In addition, our estimate of emissions from copper smelters in Canada ($0.13 \text{ Tg S yr}^{-1}$) is much lower than the NAPAP value (0.8 Tg S yr^{-1}); in order to match the

TABLE 4. Sulfur Contents of Refined Petroleum Products

Product	Sulfur Content, % weight	Reference
Aviation gasoline*	0.022	1
Motor gasoline*	0.036	2
Bitumen/asphalt	2.3	3
Gas-diesel oils*	0.022	2
Feedstocks ^a	0.033	
Jet fuel*	0.048	2
Kerosene*	0.320	2
Lubricants	2.3	3
Naphthas	0.033	3
Petroleum coke	2.25	2
Petroleum waxes ^b	2.3	
Residual fuel oils*	1.8	2
White/industrial spirit ^c	0.022	
Liquefied petroleum gas*	0	4
Others ^d	1.2	

Percent weight of sulfur in the product. Values are global averages. The products used mainly for energy are identified with asterisks. References are: 1, *Considine* [1977]; 2, *Cullis and Hirschler* [1980]; 3, *Oil and Gas Journal Data Book* [1987]; 4, *Environmental Protection Agency* [1985]

- ^a Assumed same as for naphthas.
- ^b Assumed same as for lubricants.
- ^c Assumed same as for aviation fuel.
- ^d Assumed same as for crude petroleum.

TABLE 5. Global Sulfur Source From Fuel Combustion and Industrial Activities

Activity	Global Emission	
	This work, Tg S yr ⁻¹	<i>Cullis and Hirschler</i> [1980], Tg S yr ⁻¹
Hard coal combustion	44.6	44.1
Coking of coal	0.9	1.3
Lignite combustion	10.2	16.5
Petroleum uses	25.9	29.2
Wood fuel combustion	0.1	
Copper smelting	5.4	9.4
Lead smelting	0.3	0.8
Zinc smelting	1.1	0.5
Other		1.8
Total	88.4	103.6

The global source of 88.4 Tg S yr⁻¹ given here differs from our best estimate of 77.6 Tg S yr⁻¹ for 1980 and from the value of 71.0 Tg S yr⁻¹ used in the 1°x1° inventory. We obtained 88.4 Tg S yr⁻¹ by applying globally our procedure for estimating national emissions from 1980 economic data. The improved, lower global estimates were obtained by superseding national estimates for Europe, the Soviet Union, North America, and Japan with existing inventories. The *Cullis and Hirschler* [1980] data are for 1976.

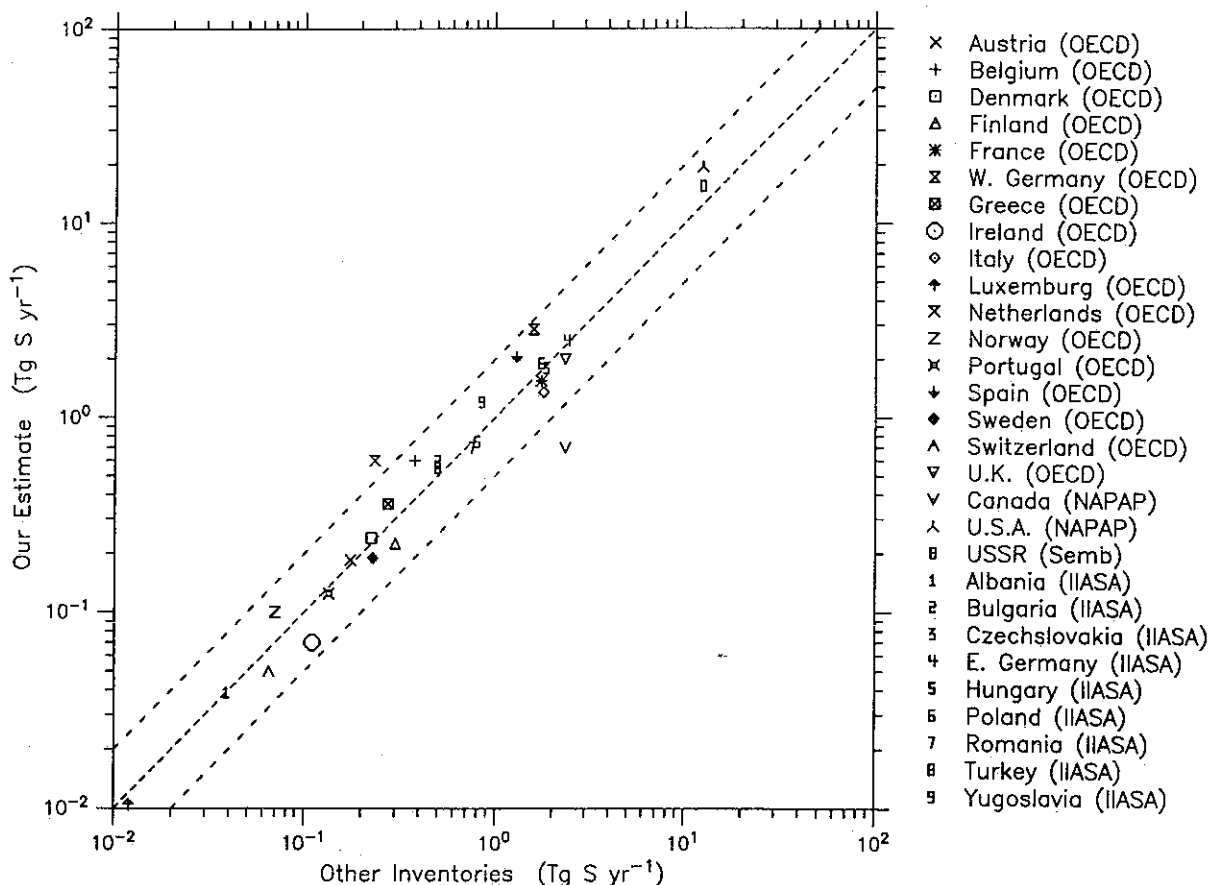


Fig. 1. National sulfur emissions from fuel combustion and industrial activities. The values estimated with the procedure of section 2 are compared to existing 1980 inventories for western Europe (OECD) [Lubkert and DeTilly, 1990], eastern Europe (IIASA) [Alcamo et al., 1987], North America (NAPAP) [Environmental Protection Agency, 1986], and the Soviet Union [Semb, 1985]. The 1:1, 1:2, and 2:1 lines are shown.

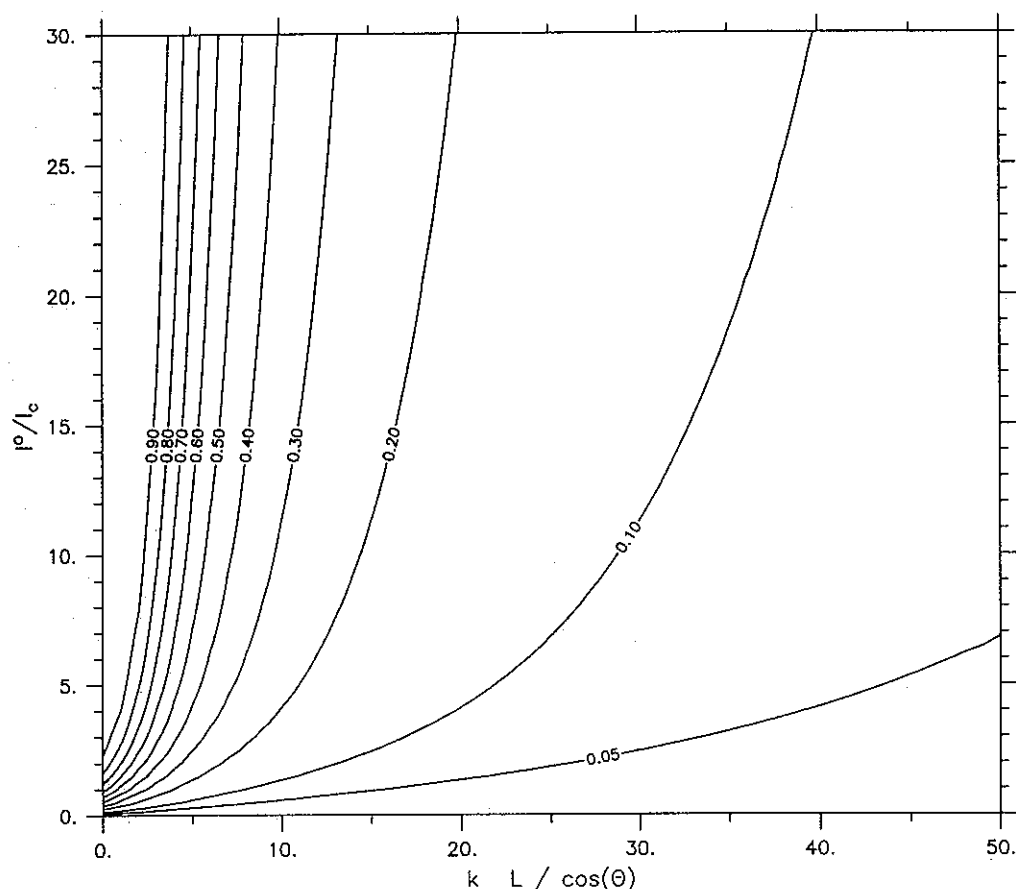


Fig. 2. Light correction factor ω for sulfur emission from a vegetation canopy (equation (5)) as a function of the dimensionless variables $kL/\cos\theta$ and P°/I_c . Here k is the light extinction coefficient normalized to the leaf

area index, L is the leaf area index of the canopy, θ is the solar zenith angle, P° is the solar radiation flux at canopy top, and I_c is the light dependence coefficient of sulfur emission in equation (3).

NAPAP value, taking into account the 0.9 Tg S yr^{-1} recovered from Canadian copper smelters in 1980 according to Morse [1984], we would need to increase the emission factor for copper smelting by 70% from the value in Table 2. Uncertainties on the sulfur contents of copper ores could possibly be an important source of error in countries where smelters account for a large fraction of sulfur emissions (e.g., Chile, Peru, Zaire, Zambia).

3. OTHER EMISSIONS

Terrestrial Biosphere

Vegetation and soils emit a number of sulfur compounds, principally H_2S and dimethylsulfide (DMS) [Andreae, 1992]. Measurements of sulfur emission fluxes have been reported recently for various ecosystems. Andreae *et al.* [1990] measured 24-hour average fluxes for the Amazon forest in the wet season of 45 and $1.3 \text{ ng S m}^{-2} \text{ min}^{-1}$ from canopy and soil, respectively. Dry season measurements at the same site [Andreae and Andreae, 1988] indicated similar canopy emissions but soil emissions were much higher, about $16 \text{ ng S m}^{-2} \text{ min}^{-1}$. Bingemer *et al.* [1992] reported total (canopy + soil) emission fluxes in the range $32\text{--}54 \text{ ng S m}^{-2} \text{ min}^{-1}$ for the Congo rainforest during the dry season. Lamb *et al.* [1987] measured canopy emissions in the range $10\text{--}60 \text{ ng S m}^{-2} \text{ min}^{-1}$ for forests and crops in the United States in summer, and mean soil emissions of 15 and $217 \text{ ng S m}^{-2} \text{ min}^{-1}$ for mollisol (organic-poor) and histisol (organic-rich) soils, respectively. Hines and Morrison [1992] reported a mean emission of $1.6 \text{ ng S m}^{-2} \text{ min}^{-1}$ for Alaskan tundra in summer.

We model the emission of sulfur from a vegetation canopy as a function of the leaf area index L , the surface air temperature T , and the solar radiation flux P° at canopy top. The total emission flux ϕ from the canopy, per unit area of land surface, is given by:

$$\phi = \int_0^L \psi f(T) g(I) d\lambda \quad (1)$$

$$f(T) = \exp[\alpha(T-298)] \quad (2)$$

$$g(I) = 1 - \exp[-I/I_c] \quad (3)$$

$$I = P^\circ \exp[-k\lambda/\cos\theta] \quad (4)$$

Here ψ is the emission flux per unit area of leaf at 298 K and under light-saturated conditions; and I is the local solar radiation flux inside the canopy which is dependent on the leaf area index overhead λ , the solar zenith angle θ , and a canopy light extinction coefficient k normalized to the leaf area index. The functions $f(T)$ and $g(I)$, with coefficients $\alpha = 0.1 \text{ K}^{-1}$ and $I_c = 100 \mu\text{E m}^{-2} \text{ s}^{-1}$, are inferred from laboratory data [Fall *et al.*, 1988]. We assume that ψ , T , and k are vertically uniform within the canopy, and combine equations (1)–(4) to write a compact expression for the flux:

$$\phi = \psi L \omega \exp[\alpha(T-298)] \quad (5)$$

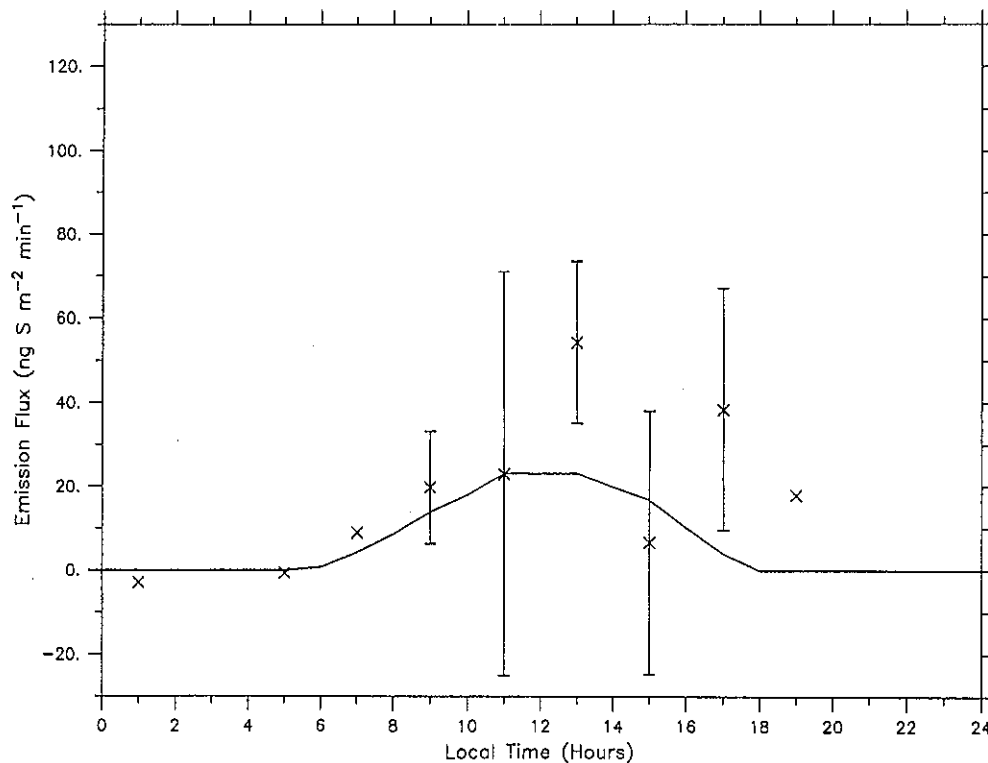


Fig. 3. Diurnal cycle of DMS emission from the Amazon forest canopy in the wet season. The solid line shows values computed from equation (5) with data for T and I^0 from Fitzjarrald *et al.* [1990]. The crosses with er-

ror bars show values derived by Andreae *et al.* [1990] from observed concentration gradients at canopy top and a flux-gradient relationship.

TABLE 6. Soil Emission Fluxes

Soil Type	H ₂ S Emission		DMS Emission	
	ϕ_0 , ng S m ⁻² min ⁻¹	α , K ⁻¹	ϕ_0 , ng S m ⁻² min ⁻¹	α , K ⁻¹
Mollisol	0.86	0.04	0.48	0.10
Histisol	19	0.06	0.68	0.18

Data are from Lamb *et al.* [1987]. The soil emission flux ϕ (ng S m⁻² min⁻¹) is calculated as $\phi = \phi_0 \exp[\alpha(T-298)]$, where T is the surface air temperature (K).

where ω is a light correction factor ($0 \leq \omega < 1$) dependent on the dimensionless variables $kL/\cos\theta$ and I^0/I_c . Values of ω obtained by integration of equation (1) are shown in Figure 2. We assume a uniform angular distribution of leaves so that $k = 0.5$ [Verstraete, 1987].

Values of ψ are fitted to match the average midday canopy fluxes ϕ reported by Andreae *et al.* [1990] for the Amazon forest in the wet season: $\phi = 130$ ng S m⁻² min⁻¹ for H₂S and $\phi = 22$ ng

S m⁻² min⁻¹ for DMS, with local conditions $L = 8$, $T = 302$ K, $\theta = 13^\circ$, and $I^0 = 2500$ $\mu\text{E m}^{-2} \text{s}^{-1}$. We obtain $\psi = 13$ ng S m⁻² leaf min⁻¹ for H₂S and $\psi = 2.2$ ng S m⁻² leaf min⁻¹ for DMS. Figure 3 compares the diurnal cycle of DMS emission computed from equation (5), for the Amazon forest in the wet season, to values derived by Andreae *et al.* [1990] from measured DMS concentration gradients at canopy top and a flux-gradient relationship. The two estimates are reasonably consistent.

Vegetative emissions are allowed only during the growing season, which is defined using satellite measurements of the normalized difference vegetation index (NDVI) [Fung *et al.*, 1987]. The distribution of vegetation types is taken from the 1°x1° map of Matthews [1983], and leaf area indices for different vegetation types are taken from Leith [1975]. Temperature and solar radiation are specified with 4°x5° spatial resolution and fine temporal resolution using output from a general circulation model [Hansen *et al.*, 1983]. The resulting mean emission fluxes of total sulfur (H₂S + DMS) are in the range 20-60 ng S m⁻² min⁻¹ for forests and crops during the growing season, in general agreement with

TABLE 7. Global Sulfur Source From Volcanoes (Tg S yr⁻¹)

	Sulfur Emission		
	This work 1980	This work 1964-1972 (9-year average)	Stoiber <i>et al.</i> [1987]
Eruptive (plume height ≤ 9 km)	5.9	4.5	5.1
Eruptive (plume height > 9 km)	1.1	0.6	0.8
Noneruptive	2.7	2.7	3.4
Total	9.6	7.8	9.3

The inventory of Stoiber *et al.* [1987] is based on a 20-year record for eruptions with VEI ≤ 4 and a 400-year record for eruptions with VEI > 4 . Eruptions with VEI ≤ 3 have a plume height ≤ 9 km [Simkin *et al.*, 1981].

TABLE 8. Global Emission of Sulfur Gases in 1980 (Tg S yr⁻¹)

Source	Global	North Hemisphere	South Hemisphere
Fuel combustion and industrial activities	77.6	69.8	7.8
Biomass burning	2.3	1.3	1.0
Volcanoes	9.6	7.6	2.0
Marine biosphere	11.9	5.3	6.6
Terrestrial biosphere	0.9	0.5	0.3
Total	102.2	84.5	17.7

the observations cited above. Fluxes computed for Alaskan tundra in summer are low (4 ng S m⁻² min⁻¹), consistent with the observations of *Hines and Morrison* [1992].

Soil emission fluxes are computed as a function of surface air temperature using exponential regressions constructed by *Lamb et al.* [1987] to fit their observations for mollisol and histisol soils

(Table 6). A 1°x1° global inventory of soil types is used [*Zobler*, 1986]. We conservatively extend the *Lamb et al.* [1987] regression for mollisol soils to all nonhistisol soils, which are for the most part organic poor.

The resulting global emission of sulfur from the terrestrial biosphere is 0.91 Tg S yr⁻¹, including 0.86 Tg S yr⁻¹ from vegetation and 0.05 Tg S yr⁻¹ from soils. This value is considerably lower than older estimates, which were based on early measurements of high soil emission fluxes that have since shown to be in error [*Andreae*, 1992]. A recent inventory by *Bates et al.* [1992] gives a global emission flux of 0.35 Tg S yr⁻¹ from the terrestrial biosphere (0.24 Tg S yr⁻¹ from vegetation, 0.11 Tg S yr⁻¹ from soils); the vegetative source is conservative as the authors extrapolated the low end of vegetation flux measurements.

Biomass Burning

We estimate biomass burning emissions by extrapolating data from aircraft measurements in biomass burning plumes over the

TABLE 9. National and Regional Sulfur Emission Totals in 1980

Country or Region	Total Emission, Tg S yr ⁻¹	Source and Percent Contribution to Total				
Angola	0.060	BRN 75	VEG 20			
Botswana	0.054	BRN 76	COA 17	VEG 7		
Madagascar	0.049	BRN 78	VEG 10	PET 9		
Mozambique	0.068	BRN 57	PET 16	VEG 16	COA 9	
Namibia	0.087	CU 49	BRN 39	PB 7		
South Africa	0.834	COA 61	PET 16	CU 15	BRN 6	
Tanzania	0.053	BRN 66	VEG 15	PET 10	VOL 5	
Zaire	0.672	CU 68	VOL 12	BRN 9	VEG 5	
Zambia	0.809	CU 91				
Zimbabwe	0.123	COA 53	CU 25	BRN 17		
Others	0.024	BRN 67	COA 23	VEG 6		
Regional Total, Southern Africa	2.832	CU 49	COA 22	BRN 14	PET 6	
Benin	0.012	BRN 87	VEG 8			
Burkina Faso	0.018	BRN 86	VEG 9			
Cameroon	0.027	BRN 68	VEG 16	PET 12		
Central African Republic	0.049	BRN 86	VEG 13			
Chad	0.067	BRN 85	VEG 10			
Congo	0.017	BRN 60	VEG 37			
Ethiopia	0.071	BRN 58	VOL 23	VEG 9	PET 6	
Gabon	0.010	VEG 41	BRN 36	PET 21		
Ghana	0.022	BRN 48	PET 34	VEG 10	WDF 8	
Guinea	0.016	BRN 65	PET 20	VEG 12		
Ivory Coast	0.038	BRN 45	PET 43	VEG 10		
Kenya	0.075	BRN 43	PET 34	VOL 15		
Liberia	0.014	PET 54	BRN 30	VEG 14		
Mali	0.047	BRN 87	VEG 11			
Mauritania	0.011	BRN 70	VEG 20	PET 7		
Niger	0.044	BRN 88	VEG 10			
Nigeria	0.092	BRN 71	PET 10	VEG 7	WDF 6	
Senegal	0.032	BRN 49	PET 43	VEG 6		
Sierra Leone	0.009	BRN 45	PET 40	VEG 9	WDF 6	
Somalia	0.031	BRN 76	VEG 13	PET 8		
Sudan	0.136	BRN 82	PET 9	VEG 7		
Uganda	0.020	BRN 85	VEG 8			
West Sahara	0.012	BRN 82	VEG 15			
Others	0.029	BRN 50	PET 40	VEG 6		
Regional Total, Central Africa	0.899	BRN 69	PET 14	VEG 10		
Algeria	0.053	PET 43	ZN 26	COA 19	BRN 7	VEG 5
Bahrain	0.066	PET 100				
Egypt	0.219	PET 93				
Iran	0.460	PET 89	COA 5			

TABLE 9. (continued)

Country or Region	Total Emission, Tg S yr ⁻¹	Source and Percent Contribution to Total					
Iraq	0.151	PET 94					
Israel	0.100	PET 100					
Kuwait	0.149	PET 100					
Lebanon	0.030	PET 80	BRN 19				
Morocco	0.076	PET 66	COA 20	PB 8			
Saudi Arabia	0.350	PET 99					
Syria	0.151	PET 97					
Tunisia	0.030	PET 72	BRN 16		PB 9		
South Yemen	0.028	PET 98					
Others	0.075	PET 92	VEG 6				
Regional Total, Middle East and North Africa	1.939	PET 92					
China	9.893	COA 90					
India	2.767	COA 86	PET 7				
Indonesia	0.309	VOL 67	PET 15	VEG 9		BRN 6	
North Korea	1.027	COA 86	LIG 5				
South Korea	0.796	COA 53	PET 35		CU 7		
Philippines	0.223	PET 60		VOL 34			
Singapore	0.246	PET 100					
Taiwan	0.360	PET 65	COA 33				
Others	0.634	PET 48	COA 27	BRN 10		VEG 7	
Regional Total, Eastern Asia ^a	16.257	COA 79	PET 11				
Australia	1.467	LIG 38	COA 21	PET 10	CU 9	BRN 9	ZN 6
New Zealand	0.095	COA 41	PET 31	VOL 20			
Papua N. Guinea	0.158	VOL 90		VEG 7			
Solomon Islands	0.060	VOL 100					
Vanuatu	0.042	VOL 99					
Others	0.011	PET 73	COA 24				
Regional Total, Oceania	1.834	LIG 31	COA 19	VOL 14	PET 10	CU 7	BRN 7
Bahamas	0.032	PET 100					
Costa Rica	0.183	VOL 96					
Cuba	0.111	PET 98					
Dominican Republic	0.027	PET 96					
Guatemala	0.038	VOL 44	PET 42	VEG 7	BRN 6		
Jamaica	0.043	PET 98					
Mexico	1.433	PET 81					
Nicaragua	0.071	VOL 81	PET 10	BRN 5			
Panama	0.029	PET 92					
Others	0.054	PET 44	VOL 41	BRN 7	VEG 7		
Regional Total, Central America	2.020	PET 72	VOL 17				
Argentina	0.326	PET 67	BRN 16	COA 6			
Bolivia	0.057	BRN 54	VEG 22	PET 17			
Brazil	2.348	COA 50	PET 31	BRN 10	VEG 5		
Chile	1.375	CU 85	VOL 6				
Colombia	0.127	PET 46	BRN 22	VEG 13	COA 12		
Peru	0.516	CU 80	BRN 6	ZN 6			
Venezuela	0.187	PET 81	BRN 11	VEG 7			
Others	0.131	PET 46	BRN 28	VEG 15	VOL 10		
Regional Total, South America	5.066	CU 32	PET 25	COA 25	BRN 9		

Only individual sources contributing more than 5% of the national or regional total are listed. Countries lumped as "Others" account individually for less than 1% of the regional total. Eruptive emissions from volcanoes are not included. COA: Hard coal, LIG: lignite, PET: petroleum, WDF: wood fuel, CU: copper, ZN: zinc, PB: lead, BRN: biomass burning, VOL: non-eruptive volcanic emissions, VEG: terrestrial vegetation.

^a Not including Japan; *Fujita et al.* [1991] give a total anthropogenic emission from Japan of 0.6 Tg S yr⁻¹ in 1983.

Global Distribution of Gaseous Sulfur Emissions (1980)

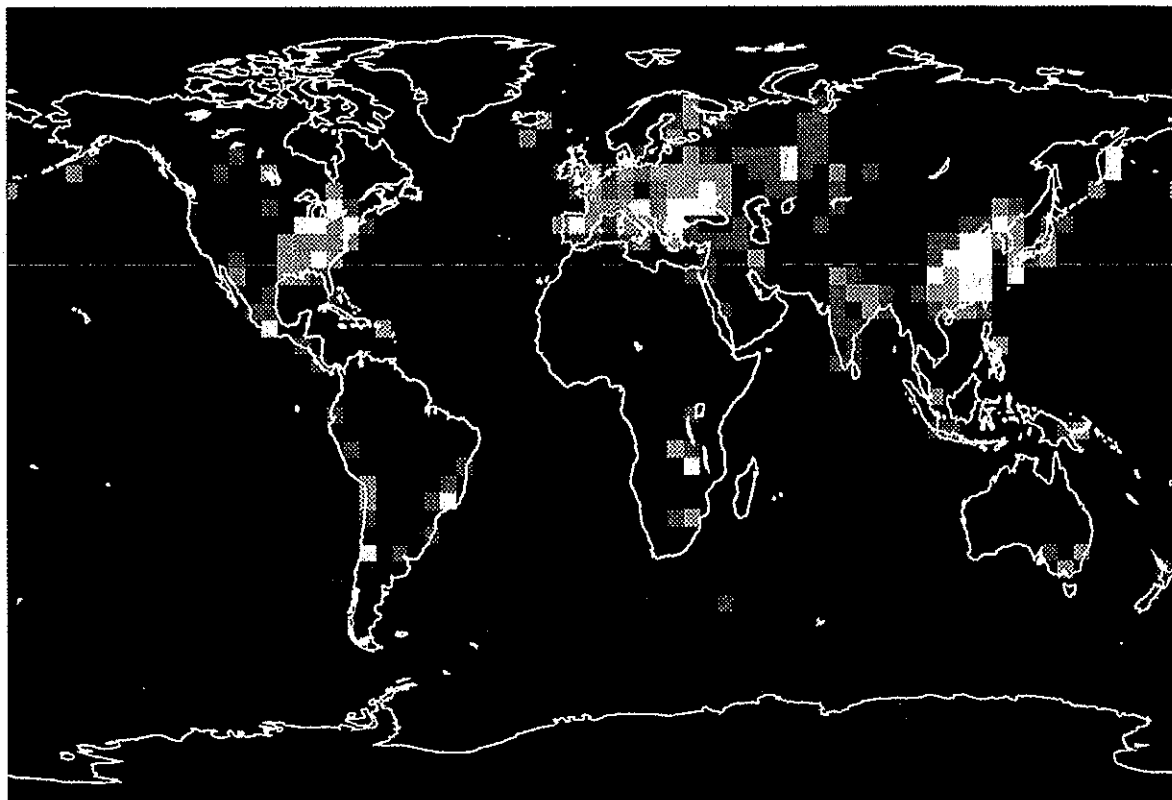
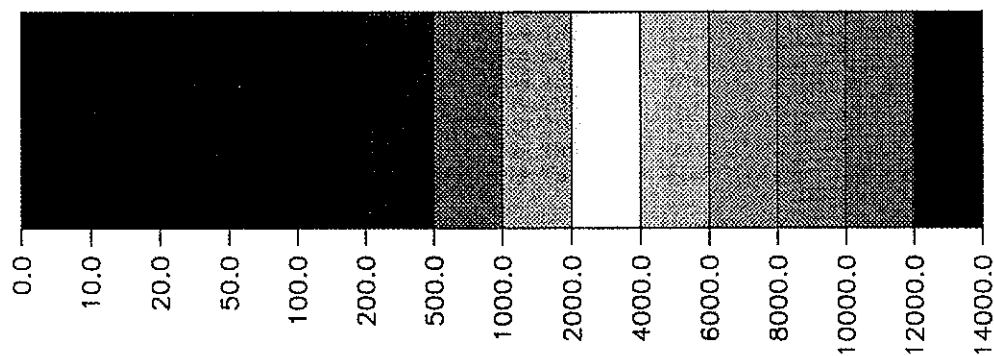
Emission Flux ($\text{Kg S km}^{-2} \text{ yr}^{-1}$)

Plate 1. Global distribution of sulfur emissions with $4^\circ \times 5^\circ$ resolution. Data are yearly means for 1980 except for anthropogenic emissions from the Soviet Union (1989), North America (1985), and Japan (1983). The

maxima reflect the eruption of Mount St. Helens (Washington), smelting activities in Norilsk (Siberia), and coal combustion in East Germany and Czechoslovakia.

Amazon Basin [Andreae *et al.*, 1988]. The mean sulfur/ CO_2 and CO/CO_2 enhancement ratios observed in the plumes were $0.85 \text{ g S kg}^{-1} \text{ CO}_2$ and $0.085 \text{ kg C kg}^{-1} \text{ C}$, respectively; assuming that CO and CO_2 account for all of the carbon burned, the sulfur emission factor is $0.78 \text{ g S kg}^{-1} \text{ C}$ burned. We specify the distribution of carbon burned using the $1^\circ \times 1^\circ$ global inventory of J.A. Logan and J.E. Dignon (manuscript in preparation, 1992), which assimilates

information on deforestation, savanna burning, slash burn agriculture, and wildfires in individual countries. Biomass burning in the tropics is concentrated in the last 3 months of the dry season, which is defined as the driest continuous 5-month period in the year and is specified using climatological precipitation data with $4^\circ \times 5^\circ$ resolution [Shea, 1986]. Biomass burning at latitudes 23° - 40° extends from September to April and biomass burning at lati-

tudes above 40° extends from June to August. J.A. Logan and J.E. Dignon give a global estimate for total carbon burned of 2860 Tg C yr⁻¹, yielding a global sulfur emission from biomass burning of 2.3 Tg S yr⁻¹.

Volcanoes

Volcanic emissions are of two types, eruptive and noneruptive. Emissions from erupting volcanoes are specified following Stoiber et al. [1987] as a function of the type of eruption (episodic or continuous), the location (at a convergent plate margin or not), and the volcanic explosivity index (VEI). The VEI is a logarithmic measure of the magnitude of the eruption including volume of ejecta and plume height [Newhall and Self, 1982]. Global data on the locations, dates, durations, types, and VEIs of volcanic eruptions are taken from Simkin et al. [1981]. The sulfur emitted during volcanic eruptions is assumed to disperse into the atmosphere at the top of the plume; strong eruptions (VEI ≥ 4) inject sulfur directly into the stratosphere. A specific emission estimate of 1.1 Tg S is used for the major eruption of Mount St. Helens in 1980 [Millan et al., 1985].

Noneruptive emissions are treated as a continuous release of sulfur at the mouths of active volcanoes. The locations and altitudes of active volcanoes are taken from Simkin et al. [1981]. Sulfur emission data for a few volcanoes are available from Malin-

conico [1987] and R. E. Stoiber (personal communication, 1990). For other volcanoes we estimate emissions as a function of plume size following the procedure of Stoiber et al. [1987]. Plume size data for 116 nonerupting volcanoes in 1981-1982 were provided by R. E. Stoiber.

Table 7 compares volcanic emission estimates for 1980 and for 1964-1972 to the inventory of Stoiber et al. [1987], which averages eruption statistics over many years. The comparison indicates that 1980 was not an atypical year. We find slightly lower noneruptive emissions than Stoiber et al. [1987], due in part to our use of actual emission data for some volcanoes and in part to differences in the cataloging of plume sizes. Our global sulfur source from volcanoes in 1980 is 9.6 Tg S; of that amount 2.7 Tg is from nonerupting volcanoes, 5.9 Tg is emitted into the troposphere by eruptions, and 1.1 Tg is injected into the stratosphere (Mount St. Helens).

Marine Biosphere

The main sulfur gas emitted from the oceans is DMS [Andreae, 1985, 1986; Saltzman and Cooper, 1988]. Surface waters are greatly supersaturated in DMS, hence the emission flux can be computed as the product of the surface water DMS concentration and a sea-to-air transfer velocity. The sea-to-air transfer velocity is usually parameterized as a function of the surface wind speed

Zonal Distribution of Sulfur Emissions

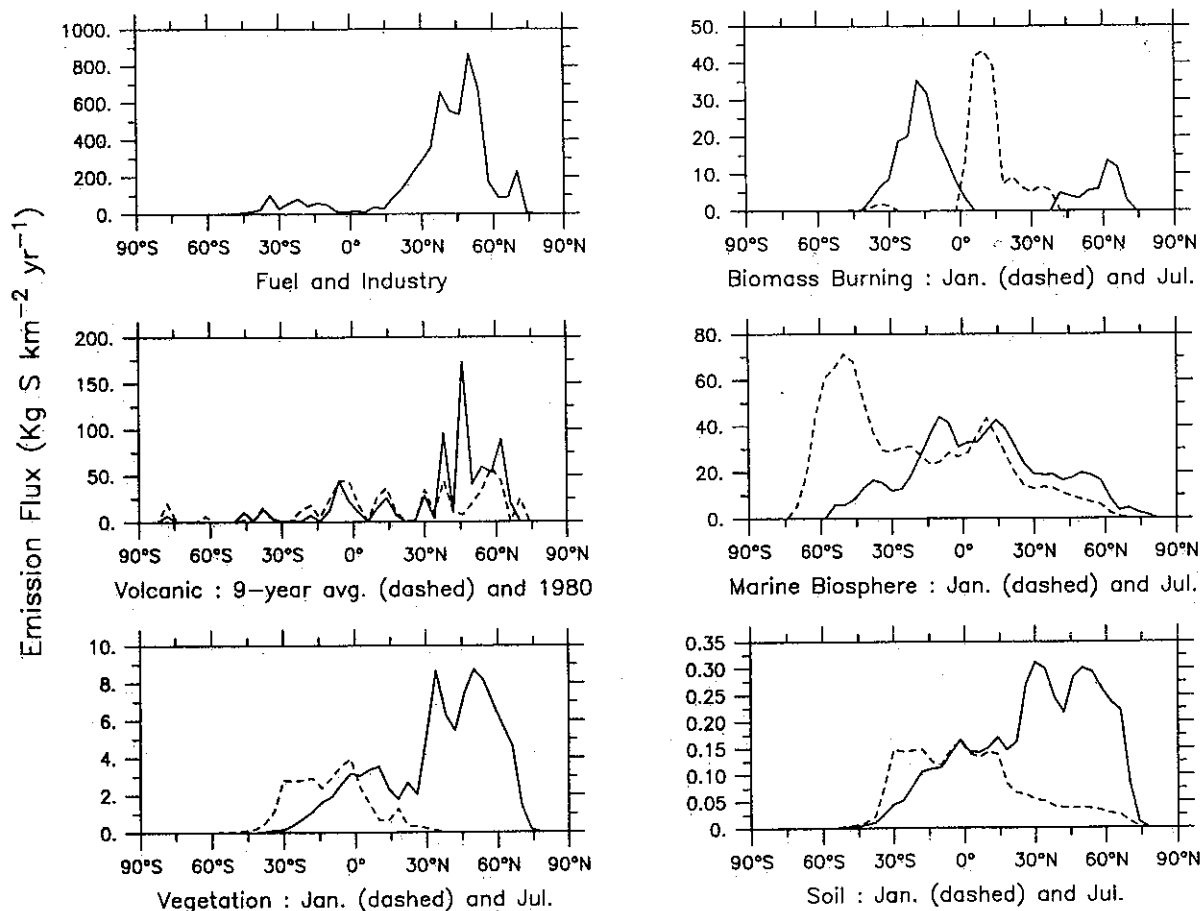


Fig. 4. Zonally averaged sulfur emissions from individual sources as a function of latitude. Emissions from fuel combustion and industrial activities are yearly means for 1980 except for the Soviet Union (1989), North America (1985), and Japan (1983). Emissions from biomass burning, the

marine biosphere, and the terrestrial biosphere are monthly means for January and July. Volcanic emissions are means for 1980 and for 1964-1972 (9 years).

(10 m altitude) and the Schmidt number $Sc = \nu/D$, where ν is the kinematic viscosity of seawater and D is the molecular diffusivity of DMS [Liss and Merlivat, 1986]. Global inventories of DMS emission have been compiled previously by Bates *et al.* [1987] and Andreae [1990], using data for DMS seawater concentrations and climatological wind speeds. These authors report values of 16 and 19–54 Tg S yr⁻¹, respectively. The discrepancy reflects largely differences in the assumed seawater DMS concentrations.

We construct a two-dimensional time-dependent map of DMS emissions by using climatological surface wind speeds from COADS (Comprehensive Ocean-Atmosphere Data Set) available on a global grid with 2°x2° spatial resolution and monthly temporal resolution [Woodruff *et al.*, 1987]. The COADS data set is a compilation of ship measurements at 10 m altitude. We further

adopt the seawater DMS concentrations of Bates *et al.* [1987] as a function of latitude and season and compute sea-to-air transfer velocities with the formulation of Liss and Merlivat [1986]. Values of ν and D are specified as a function of sea surface temperature using data from Knauss [1978] for ν and the Nernst-Einstein equation for D . Climatological data for sea surface temperatures are taken from Robinson and Bauer [1981] with 4°x5° spatial resolution and monthly temporal resolution. The resulting global DMS emission is 11.9 Tg S yr⁻¹, 25% lower than the estimate of Bates *et al.* [1987], largely because of differences in the computation of sea-to-air transfer velocities. Considering that models for computing the sea-to-air transfer velocity are uncertain by at least a factor of 2 [Liss and Merlivat, 1986; Erickson, 1989; Upstill-Goddard *et al.*, 1990], it appears that the emission flux of

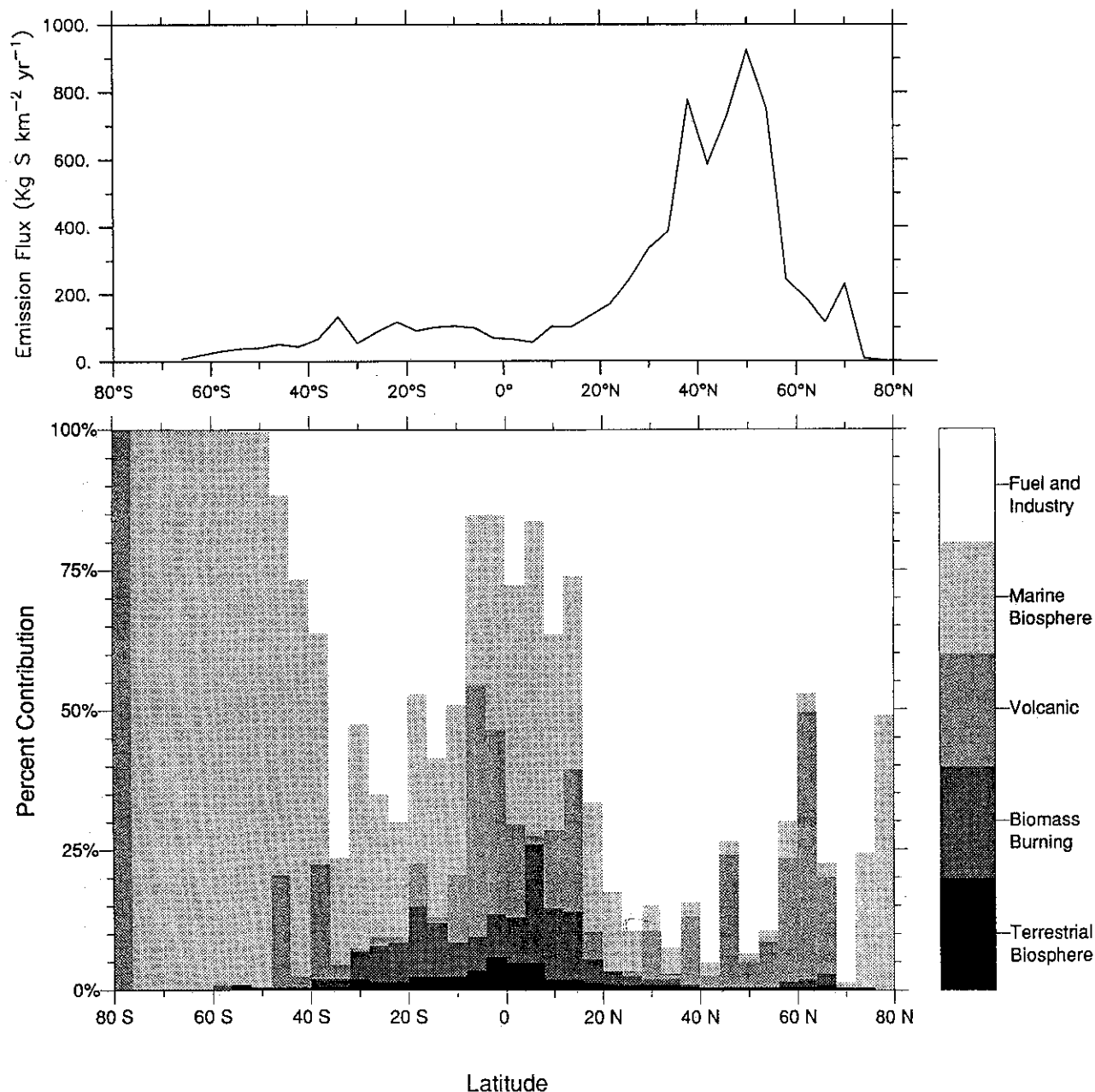


Fig. 5. Yearly mean sulfur emission as a function of latitude. The top panel shows total emission; the bottom panel shows the relative contributions from individual sources. Data are for 1980 except for anthropogenic

emissions from the Soviet Union (1989), North America (1985), and Japan (1983).

DMS cannot in any case be constrained strongly using DMS surface water concentrations. An independent constraint would be to use data on atmospheric sulfate concentrations and deposition fluxes over the oceans; however, Bates *et al.* [1987] point out that interpretation of such data is made difficult by influences from long-range transport of continental sulfur. Global model simulations of atmospheric sulfur may help to address this difficulty.

4. GEOGRAPHICAL DISTRIBUTION OF EMISSIONS

We estimate a global sulfur emission flux of 102.2 Tg S yr⁻¹ in 1980 (Table 8), apportioned among fuel combustion and industrial activities (76.1%), marine biosphere (11.7%), volcanoes (9.2%), biomass burning (2.2%), and terrestrial biosphere (0.9%). Table 9 gives a detailed breakdown of major anthropogenic and natural sources for individual countries and continental regions outside Europe, the Soviet Union, North America, and Japan. Only noneruptive volcanic emissions are included in Table 9 in order to emphasize the effects of regional source distributions on the sulfate concentrations in surface air. Prominent features of the geographical distribution of sources are illustrated further in Plate 1 and Figures 4-5. Plate 1 shows the global distribution of yearly mean emissions with 4°x5° resolution, Figure 4 shows the zonally averaged distributions of individual sources, and Figure 5 gives the relative contributions from each source to the total sulfur emission in each latitudinal band.

We see that sulfur emissions in the Arctic are dominated by smelting activities in Norilsk, Siberia. Emissions at northern mid-latitudes are mainly anthropogenic, but volcanoes are important in some subarctic regions (Kamchatka, Iceland, Aleutians). There was unusually high eruptive activity at northern mid-latitudes in 1980; the three major peaks in Figure 4 are due to the eruptions of Etna (Italy), St. Helens (United States), and Hekla (Iceland).

Emissions in the tropics are diverse. Continental and oceanic emissions are of comparable magnitude on a zonal average basis (Figure 5). The continental emissions are principally anthropogenic: petroleum (Central America, North Africa, Middle East), coal (Brazil, India), biomass burning (Central Africa), and copper smelters (South America, southern Africa). Biomass burning accounts for 69% of yearly mean emissions in central Africa. Biomass burning emissions are also high in southern Africa but are subordinate to copper smelters on the regional scale. Volcanic emissions are significant in some tropical regions (in particular Central America and the East Indies). Emissions from the terrestrial biosphere peak at the equator but are still relatively small, accounting there for only 6% of zonal average emissions.

Anthropogenic sources account for 50% of total sulfur emission in the southern hemisphere, reflecting major contributions from copper smelters (Peru, Chile, Zambia, Zaire) and coal combustion (southeastern Australia, South Africa). This result suggests that sulfate concentrations in the southern hemisphere may be widely influenced by human activity, with possible implications for climate [Schwartz, 1988; Charlson *et al.*, 1992a,b].

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