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Anthropogenic sources of NO_x, SO_x, CO and CH_4. Geographical distribution

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ANTHROPOGENIC SOURCES OF NO_x, SO_x, CO and CH_4.

GEOGRAPHICAL DISTRIBUTION

by

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Abstract

Available energy and industry statistics from different international organizations were used, together with various results from recent publications to produce world maps of the anthropogenic emissions of NO_x, SO_x, CO and CH_4. These emissions are distributed onto a 5° x 5° resolution latitude-longitude grid, suitable for use in a three-dimensional chemical/transport model of the atmosphere.

Résumé

Des cartes mondiales des émissions anthropogéniques de NO_x, SO_x, CO et CH_4 ont été élaborées à l'aide de statistiques industrielles et énergétiques publiées par différentes organisations internationales, ainsi que de divers résultats de travaux récents. Ces émissions sont distribuées sur une grille latitude-longitude de résolution 5° x 5°, et peuvent être utilisées dans un modèle tri-dimensionnel de l'atmosphère.

Samenvatting

Beschikbare energie- en industriestatiestieken van verschillende internationale organisaties werden gebruikt samen met verscheidene resultaten van recente publicaties om wereldkaarten van de antropogene emissies van NO_x, SO_x, CO en CH_4 samen te stellen. Deze emissies zijn verdeeld over een breedte-lengte resolutierooster van 5°x 5° en kunnen gebruikt worden in een driedimensionaal model van de atmosfeer.

Zusammenfassung

Verfügbare Energie- und Industriestatistiken von verschiedenen internationalen Organisationen wurden benützt zusammen mit verschiedenen Resultaten von rezenten Publikationen um Weltkarten der anthropogenen Emissionen von NO_x, SO_x, CO und CH_4 zusammen zu stellen. Diese Emissionen sind verteilt über einen Breite-Länge Resolutionrost von 5°x 5° und können gebraucht werden in einem driedimensionalen Modell der Atmosphäre.

1. INTRODUCTION

Human activities, mainly fossil fuel burning and, to a lesser extent, industrial processes and waste disposal, emit substantial amounts of pollutants to the atmosphere, changing its composition significantly. On the global scale, the magnitude of these emissions is generally comparable to the magnitude of the emissions associated with biological and other natural processes. On a regional scale, anthropogenic emissions often exceed natural emissions. Very high pollutants levels correspond to the world's most densely populated and industrialized areas. It is therefore important to assess, as accurately as possible the global strength of these emissions and their geographical distribution.

Four important species are considered here : nitrogen oxides (NO_x), sulfur oxides (SO_x), carbon monoxide (CO) and methane (CH_4). NO_x, CO and CH_4 take part in photochemical production of ozone in the troposphere. They also control the atmospheric concentration of the hydroxyl radical (OH). OH represents the most efficient way of removal from the atmosphere for many species. High levels of tropospheric ozone and precipitation acidity are harmful to the biosphere, by damaging trees and crops, and threatening man's health. Finally, methane and ozone are greenhouse gases and therefore contribute to the warming of the planet.

Source maps for these pollutants are presented in this paper. The estimated fluxes are distributed on a 5° x 5° resolution latitude-longitude grid, and can be used as part of surface boundary conditions in a three-dimensional chemical/transport model of the atmosphere. These data are available on computer diskettes and can be requested from the author.

An outline of the methods applied is first presented in section 2. Each species is then separately discussed in sections 3 to 6.

2. GENERAL METHOD

The world has been divided into three large regions : North America, which includes the United States and Canada; the other members of the Organization of Economic Co-operation and Development (OECD), including Western Europe, Japan, Australia and New Zealand; and the non-OECD countries. These three regions differ from each other by the prevailing concern in air pollution problems. The most detailed emission estimates are made in North America [National Acid Precipitation Assessment Program (NAPAP), 1987; U.S. Environmental Protection Agency (U.S.EPA), 1986; United States-Canada Memorandum of Intent, 1982].

Most emission rates have been reduced in the U.S. since the early seventies as a consequence of the implementations of the Clean Air Act Amendements. This is reflected by the evolution of the U.S. emissions since 1970 (Table 1). In contrast, hardly any pollutant controls exist in the third region. The second region stands somewhere in between, emission controls having been introduced in only a limited number of countries. Emission data for the main air pollutants have been collated and published by the OECD for most countries of this organization [OECD, 1989]. Those data which have been used in this work are listed in Table 2. It must be emphasized, however that their reliability is questionable. Estimation and measurements methods may differ from country to country. The definition itself of "total sources" may vary. Attempt was made to take this into account by correcting the OECD emission estimates, when thought to be necessary (and possible).

When pollutant-emission data was not available (for countries of the third region), the contributions from the various known sources were computed and then summed to give the country's emissions. Assessing the individual contributions was done by using statistics for energy consumption and industrial production (summarized in tables 3 and 4), and emission factors from recent publication.

Table 1.-Emission estimates of CO, NO_x and SO_x in the U.S. from 1970 to 1984, in millions of tons (Source : U.S. EPA 1986).

	total	transportation	fuel combustion	industry	disposal waste	Miscell. uncontrolable
		controlable emissions				
1970 : CO	98.8	71.8	4.4	9.0	6.4	7.2
SO_x	28.2	0.6	21.3	6.2	0.0	0.1
NO_x	18.1	7.6	9.1	0.7	0.4	0.3
1980 : CO	76.2	52.7	7.4	6.3	2.2	7.6
SO_x	23.2	0.9	18.8	3.5	0.0	0.0
NO_x	20.4	9.2	10.2	0.7	0.1	0.2
1984 : CO	69.9	48.5	8.3	4.9	1.9	6.3
SO_x	21.4	0.9	17.4	3.1	0.0	0.0
NO_x	19.7	8.7	10.1	0.6	0.1	0.2

Table 2.-OECD emission estimates of NO_x, SO_x and CO in 1980, in millions of tons (Derived from OECD, 1989).

	SO_x	NO_x	CO
Canada	4650	1900	9928
U.S.	23200	20300	76100
Japan	3400	3600	13600
Australia	1600	940	3950
New Zealand	100	90	600
Austria	350	210	1126
Belgium	856	317	839
Denmark	452	245	832
Finland	584	284	660
France	3550	2561	6620
West Germany	3200	3000	11708
Greece	800	155	740
Iceland	6	13	30
Ireland	217	70	497
Italy	3500	1600	5850
Luxembourg	24	23	60
Netherlands	400	553	1450
Norway	150	203	608
Portugal	266	166	800
Spain	3250	850	4200
Sweden	502	318	1550
Switzerland	126	196	711
U.K.	4800	2264	4999

Table 3.- Fossil fuels and electricity consumption by type (in millions of terajoules/year) and regional breakdown (in percents of the world consumption). (Source : UN, 1986).

	World (millions TJ)	North America (%)	Other OECD (%)	Non-OECD (%)
Solid fuels	90.4	20.5	17.0	62.5
Hard Coal	78.7	22.4	17.0	60.6
Lignite	11.7	7.7	17.0	75.3
Liquid fuels	113.8	29.9	28.3	41.7
Gasoline	30.2	47.2	22.2	30.6
Gas-diesel oils	30.7	22.4	35.7	41.8
Residual oils	28.7	12.2	26.5	61.3
Others	24.2	38.9	28.8	32.3
Natural gas	61.3	30.6	18.0	51.4
Fossil fuels	265.5	26.9	22.1	51.1
Electricity	36.1	30.5	28.6	41.0

Table 4.- Industrial productions relevant for SO_x, CO and CH_4 source estimation (in millions of tons/year) and regional breakdown (in percents of the world production). (Sources : UN, 1986; UN, 1988; American Iron and Steel Institute, 1987).

	World (millions of tons)	North America (%)	Other OECD (%)	Non-OECD (%)
Copper (smelted)	8.5	16.3	22.2	61.5
Copper (refined)	9.4	20.9	27.8	51.3
Sulphuric acid	132	27.9	24.8	47.4
Steel	715	12.6	34.2	53.2
Pig iron (total)	497	10.1	34.7	55.2
Pig iron (foundry)	8.5	13.9	38.2	47.8
Carbon black	4.1	32.3	42.5	25.2
Ammonia	86	19.5	10.6	69.9
Hard coal	3194	24.1	12.0	63.9
Lignite	1225	7.6	18.0	74.4

Once a pollutant relase has been assigned to a country (or to a part of it), apportioning among the model gridpoints the country occupies was made according to population distribution, except when parts of the total source had a significantly different distribution. Population and other distributions were deduced from various atlas and encyclopedias economic maps.

3. NO_x

Fossil fuel burning is the greatest contributor to anthropogenic NO_x. Transportation alone represents about 40 p.c. of the total emission. The other sources, primarily petroleum refining and manufacture of nitric acid and cement, account for only about 5 p.c. of the total world source [Logan, 1983].

The U.S. and Canadian yearly sources of NO_x amount to about 19 and 2 x 10^6 tons of NO_2, respectively [Logan, 1983; U.S. EPA, 1986; NAPAP, 1987; OECD, 1989], while other OECD countries account for about 18 x 10^6 tons of NO_2 [OECD, 1989]. Emission control implementations have been installed on new automobiles in the U.S., but their effect on total emissions are relatively small (see Table 1). NAPAP's emission data for individual states of the U.S. were used in this study.

The NO_x release in the rest of the world was estimated from statistics for consumption of fossil fuels [United Nations (UN), 1986] and from emission factors reported by Logan [1983]. These are listed in Table 6. The resulting emission is 33 x 10^6 tons of NO_2/year for non-OECD countries.

These results are summarized in Table 5. The estimated global NO_x source, 72 x 10^6 tons of NO_2/year, agrees with previous results [e.g. Logan, 1983; Hameed and Dignon, 1988]. Its geographical distribution is showed on figure 1.

Table 5.-Emissions estimates by pollutant and by regions, in millions of tons/year. The transportation source is indicated into brackets (See text for details).

	World	North America	Other OECD	Non-OECD
NO_x	72(27)	21(9)	18(9)	33(9)
SO_x	184(3)	26(1)	28(1)	130(1)
CO	373(193)	80(56)	60(32)	233(105)
CH_4	120(0)	33(0)	24(0)	63(0)

Table 6.- NO_x emissions factors for combustion processes in kg (NO_2)/ton of product, except if indicated (Condensed from Logan, 1983).

Coal	
Hard coal	5.9
Lignite	2.9
Oil	
Gasoline vehicles	27
Diesel vehicles and engines	30-56
Combustion (stationary)	2.3-10
Natural gas	6.4 $(kg/10^3\ m^3)$
Industrial processes (world source)	5×10^6 tons/year

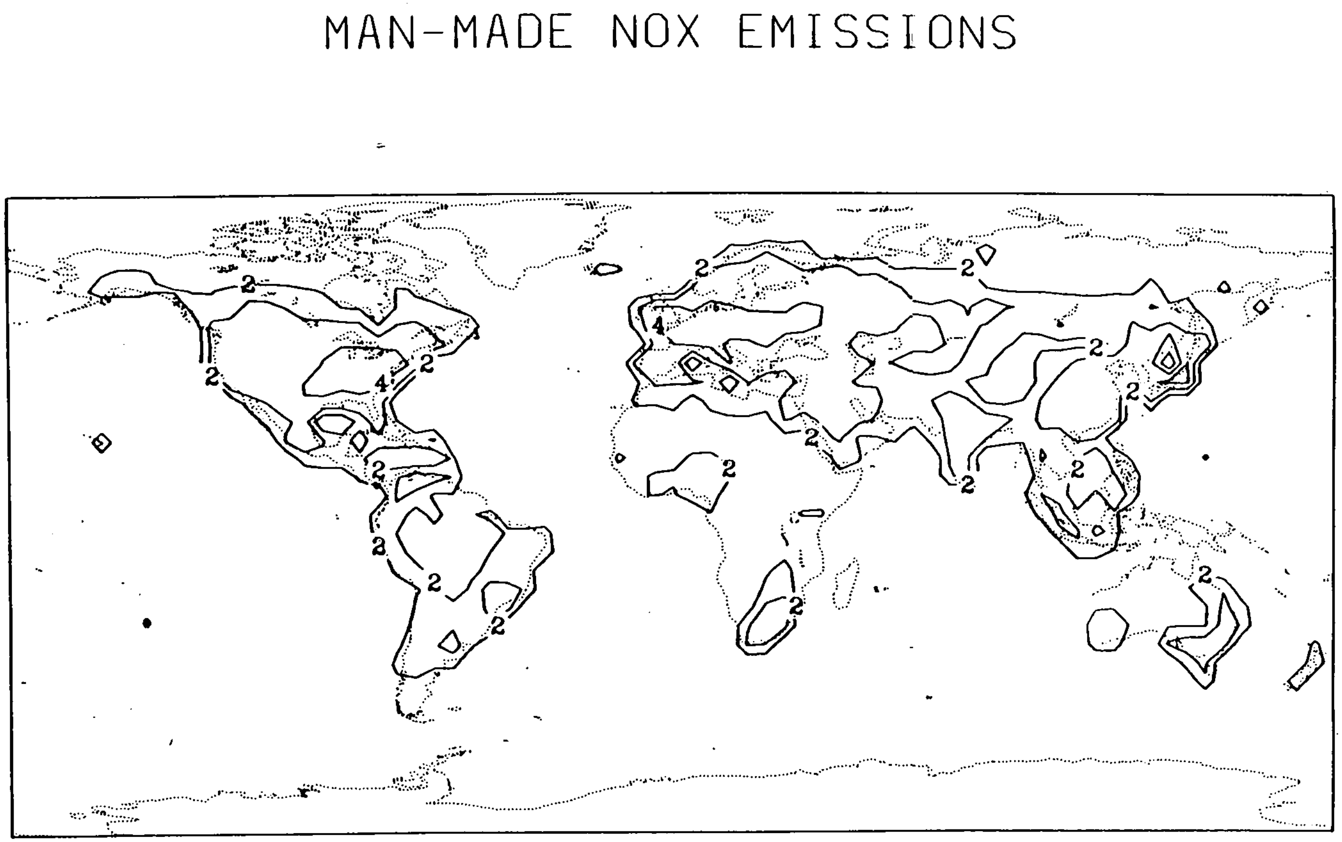


Fig. 1. Anthropogenic NO_x emissions (log).

4. SO_x

Coal burning accounts for at least 60 to 70 p.c. of the global man-made source of SO_x. The rest originates mainly from oil refining and burning, ore smelting and refining, and sulphuric acid manufacturing [Cullis and Hirschler, 1980].

As for NO_x, NAPAP and OECD data were used. Implemented controls appear to be more efficient for SO_x than for NO_x. Emissions in the U.S. have decreased from 28.2 x 10^6 tons of SO_2 in 1970 to 21.4 x 10^6 tons of SO_2 in 1984 (Table 1). Reductions of fuel sulphur content are also responsible for a significant lowering in SO_x emissions in other OECD countries. They amounted to 28 x 10^6 tons of SO_2 in 1980 [OECD, 1989].

Emission factors from Cullis and Hirschler [1980] and statistics for consumption of fossil fuel and for production of copper and sulphuric acid [UN, 1986 and 1988] were employed to estimate SO_x release from non-OECD countries. The calculated emission, 130 x 10^6 tons of SO_2, seems large when compared with OECD countries emissions (Table 5). However, activities with the highest SO_x emission factors, namely hard coal, lignite and residual fuel oil burning, and copper smelting (Table 7) are concentrated in non-OECD countries and comprise a large proportion of the world consumption or production, 61, 75, 61 and 62 percent, respectively (Tables 3 and 4). Furthermore, as for the other pollutants, emission factors are believed to be generally higher in non-OECD than in OECD countries, because of the lack of environmental controls and policy, and of inadequate maintenance of vehicles and machinery.

Figure 2 shows the geographical distribution of the global SO_x source, estimated to be 184 x 10^6 tons of SO_2 per year.

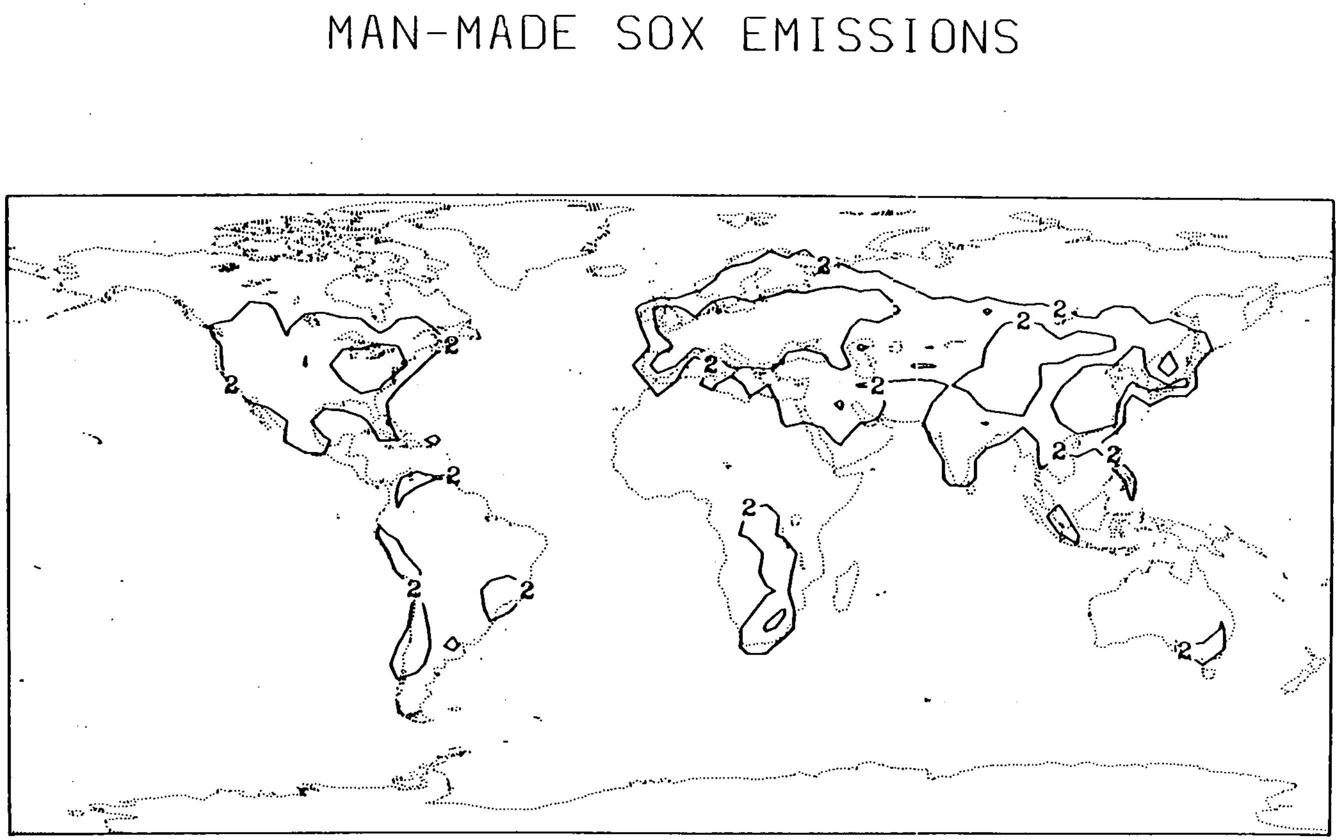


Fig. 2. Anthropogenic SO_x emissions (log).

Table 7.- Emissions factor for SO_x, in kg (SO_2)/ton of product, except if indicated (Source : Cullis and Hirschler, 1980).

Coal	
Hard coal	48.2
Lignite	35.6
Coke and briquettes	5.4
Oil	
Gasoline	0.72
Gas-diesel fuel oils	4.47
Residual fuel oils	36.0
Petroleum coke	13.5
Refining	2.0
Ore smelting	
Copper (smelted)	2000
Copper (refined)	300
Lead	400
Zinc	200
Others	
Sulphuric acid	24
Sulphur	2
Paper/pulp	2
Refuse (world source)	8×10^5 tons/year

Table 8.- CO emission factors in kg (CO)/ton of product, except if indicated (Condensed from Logan et al., 1981).

Coal (kg/ton)	
Hard coal	27
Lignite	3.7
Oil (kg/m^3)	
Gasoline vehicles	360
Diesel vehicles	26-35
Combustion (stationary)	4
Natural gas (kg/10^3 m^3)	750
Industrial processes (kg/ton)	
Pig iron production	80
Pig iron, foundry	72.5
Steel production	69.5 kg/m^3
Petroleum refining	39.2
Ammonia production	100
Miscellaneous (world source)	15 x 10^6 tons/year
Waste disposal (world source)	12 x 10^6 tons/year

5. CO

Exhausts from gasoline vehicles constitute more than half the world's anthropogenic source of CO. The remainder comes from the other combustion processes, plus various industrial activities such as metallurgy and oil refining, and waste disposal [Logan et al., 1981].

According to the U.S. EPA [1986], CO is, among the gaseous constituents considered here, the one for which regulation policy seems to be the most efficient, since U.S. emissions have decreased from 100 to 70 x 10^6 tons of CO/year between 1970 and 1984 (Table 1). Similar and even greater reductions have occured during the same period in other OECD countries, particularly Western Germany and the Netherlands. The CO released from OECD countries, except North America, amount to about 62 x 10^6 tons [OECD, 1989].

Statistics from the United Nations and from the American Iron and Steel Institute [1987], and the CO emission factors from Logan et al. [1981] were used to derive the CO source in other countries. The total non-OECD source is estimated to be 233 x 10^6 tons of CO/year. The emission factors are listed in table 8. Note that these factors are subject to considerable uncertainties. For example, Cullis and Hirschler [1989] reported a CO emission factor for gasoline vehicles which is about two times higher than the emission factor from Logan et al. [1981]. Recent studies suggest that only a small fraction of cars in use account for most of the emissions [Stedman and Bishop, 1989]. It must also be remembered that, as for the other pollutants, the only available measurements were made in North America and Western Europe.

The estimated world yearly source of CO is then 373 x 10^6 tons (Table 5). Its geographical distribution is shown in figure 3.

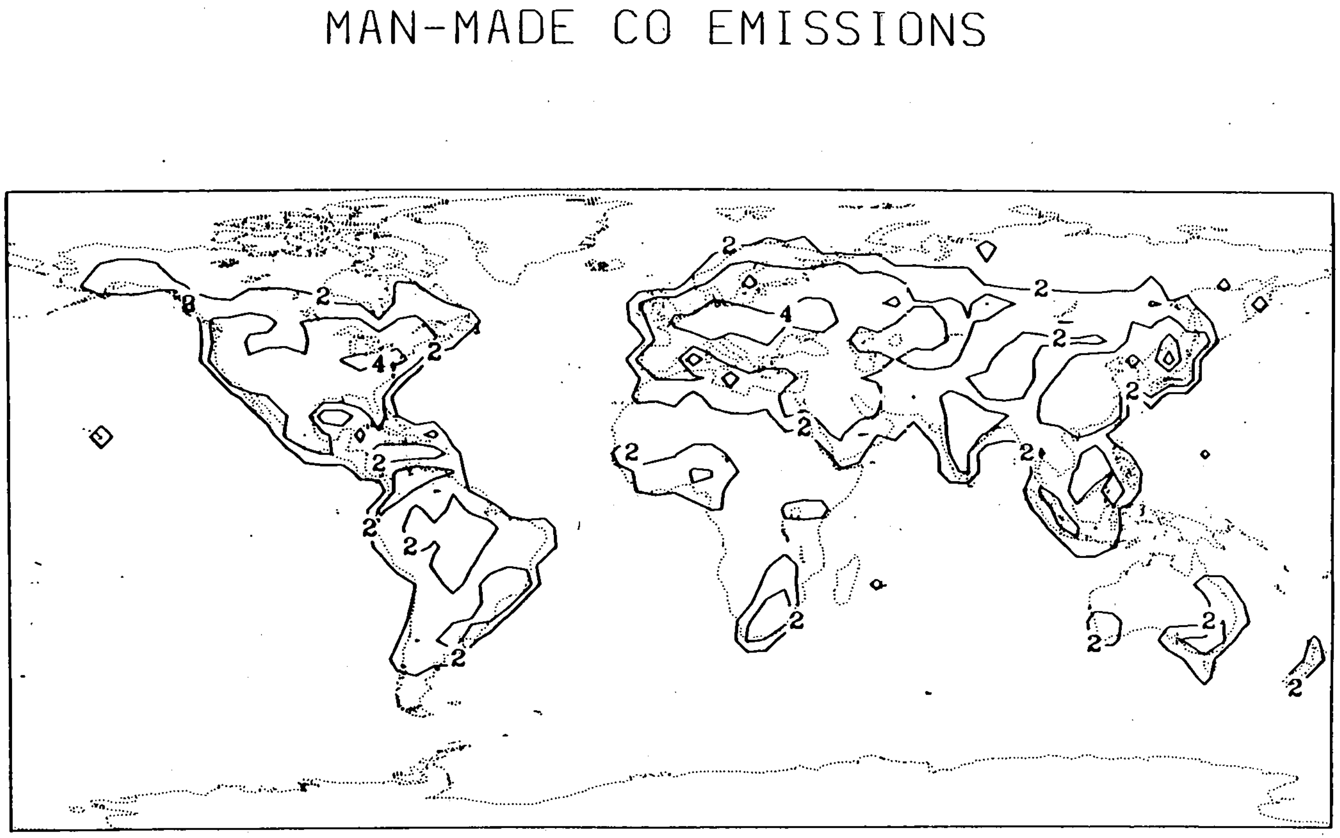
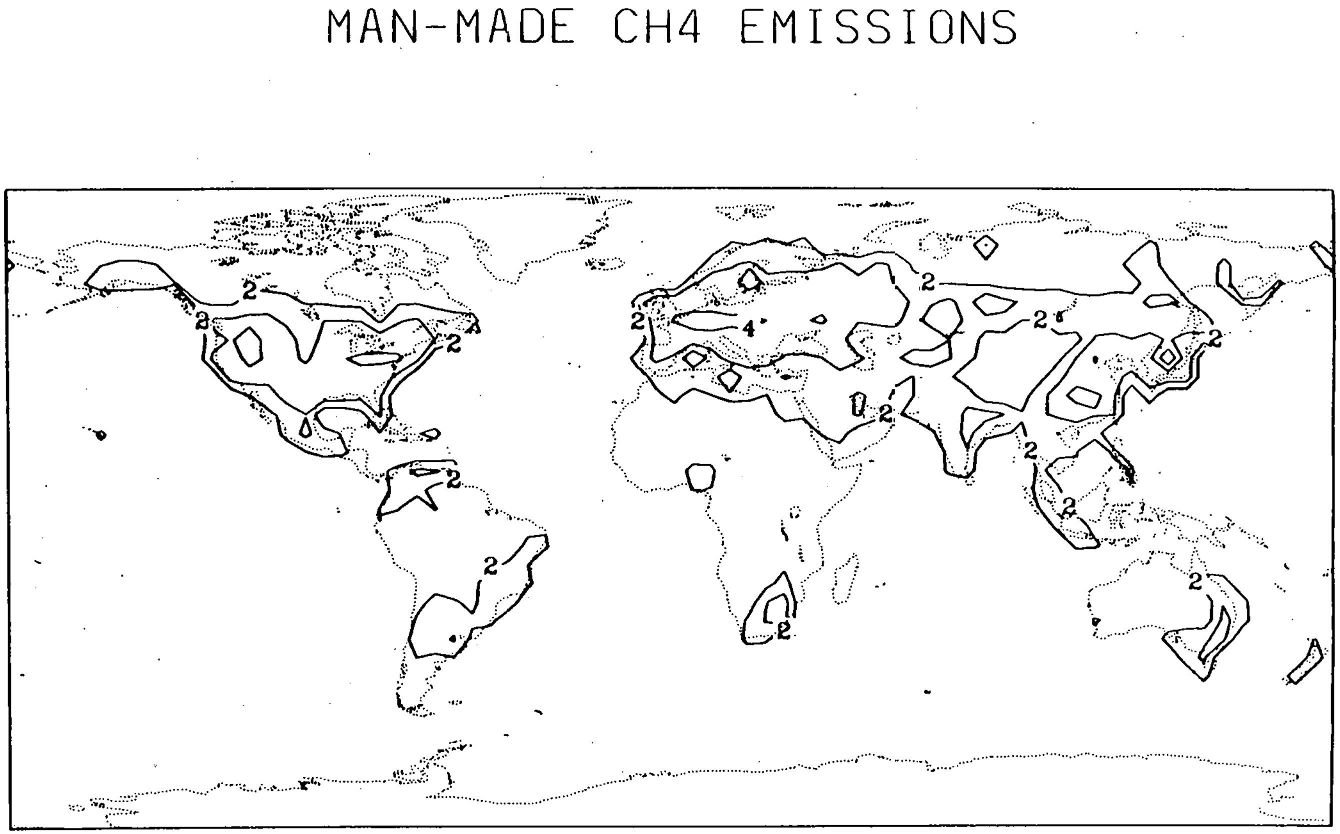


Fig. 3. Anthropogenic CO emissions (log).

6. CH_4

The purely anthropogenic sources of methane are coal mining, natural gas exploitation and waste disposal. They account for about 120 x 10^6 tons of CH_4/year, or 20 p.c. of the global budget [Cicerone and Oremland, 1988]. Biogenic but man-related sources, such as rice paddy fields, enteric fermentation in animals and biomass burning were not considered in this work.

The 35 x 10^6 tons of CH_4 released each year by coal mining according to Cicerone and Oremland [1988] were distributed as coal (= hard coal + lignite) production [UN, 1988], while the 45 x 10^6 tons of CH_4 from gas exploitation (drilling, venting and transmission) were distributed as natural gas production [UN, 1986]. Because of the lack of available data concerning waste, of 40 x 10^6 tons of CH_4 arising from landfills was distributed as the electricity consumption [UN, 1986], to account for the fact that landfills are associated with populated areas of industrial countries.



MAN-MADE CH4 EMISSIONS

Fig. 4. Anthropogenic CH_4 emissions (log).

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