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A 1° × 1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990

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Abstract. An anthropogenic emissions data set has been constructed for CO_2, CO, CH_4, nonmethane volatile organic compounds, SO_2, NO_x, N_2O, and NH_3 spanning the period 1890–1990. The inventory is based on version 2.0 of the Emission Database for Global Atmospheric Research (EDGAR 2.0). In EDGAR the emissions are calculated per country and economic sector using an emission factor approach. Calculations of the emissions with 10 year intervals are based on historical activity statistics and selected emission factors. Historical activity data were derived from the Hundred Year Database for Integrated Environmental Assessments (1890–1990) supplemented with other data and our own estimates. Emission factors account for changes in economical and technological developments in the past. The calculated emissions on a country basis have been interpolated onto a 1° × 1° grid. This consistent data set can be used in trend studies of tropospheric trace gases and in environmental assessments, for example, the analysis of historical contributions of regions and countries to environmental forcing like the enhanced greenhouse gas effect, acidification, and eutrofication. The database focuses on energy/industrial and agricultural/waste sources; for completeness, historical biomass-burning estimates where added using a simple and transparent approach.

1. Introduction

To quantify current developments and assess possible future scenarios, it is essential to try to understand past anthropogenic changes. No doubt, human activities have always modified the natural environment; however, during the past century the intensity and scale of these modifications have increased dramatically. Emissions of greenhouse gases and their precursors have been identified as significant driving forces of global changes [*Intergovernmental Panel on Climate Change (IPCC)*, 1995] that occur on a wide range of spatial and temporal scales and dimensions and often differ among regions. In spite of their importance, there have been few attempts to estimate long-term historical emission time series (especially before 1970), let alone on a relatively detailed sectoral basis and on a high-resolution grid basis. The Carbon Dioxide Information and Analysis Center (CDIAC) presents a good estimate of historical CO_2 emissions from fossil fuel combustion for the period 1950–1990 on a 1° × 1° grid based on United Nations (UN) energy data [*Andres et al.*, 1997] (In a recent publication, *Andres et al.* [1999] report on an exercise similar to that described in this paper to extend their data sets of energy consumption on country levels to 1751.) *Gschwandtner et al.* [1985] estimated emissions of sulfur and nitrogen oxides by the United States for the period 1900–1980, partly on a subnational (state) level. In 1996 the *Environmental Protection Agency (EPA)* [1996] presented an overview of historical emissions for several greenhouse gases and other pollutants for the United States in the period 1900–1995. *Mylona* [1996] presented sulfur emissions for several European countries (including Russia and Turkey) for the period 1880–1990. A detailed global study of sulfur emissions from 1850 to 1990 with data per country and for some sectors has been presented by *Lefohn et al.* [1996, 1999].

The information presented in this paper is to our knowledge the first attempt to construct a global gridded trace gas emission database on a sectoral basis with a time frame of 100 years using a consistent and transparent methodology for a set of trace gases that are relevant for global atmospheric chemistry. However, one should be aware of the limitations of such a large-scale historical emission inventory. Information on activities and emission factors in the past is limited and uncertain and sometimes nonexistent, leading in some cases to scaling back of current activity rates using indicators and the application of global aggregated emission factors. This study includes emissions of three groups of anthropogenic sources: (1) energy/industry, (2) agriculture/waste, and (3) biomass burning, and builds on knowledge that is currently available on these three source categories. We focused on construction of a detailed sectoral energy/industry data set; in addition, a more aggregated agriculture/waste data set was compiled. For completeness we included a data set on biomass burning, using a simplified, however, transparent method that can be improved when more information becomes available. A further discussion of limitations of this emission inventory can be found in section 4.2. Finally, we note that the distinction between anthropogenic and natural sources is not always clear. We have neglected secondary effects of human activities on natural sources such as methane emissions from wetland changes.

2. Methodology

This study builds on the data and methodology of the Emission Database for Global Atmospheric Research (EDGAR 2.0) [*Olivier*

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Table 1. Overview of Anthropogenic Source Categories Distinguished in This Study Together With an Overview of Their Importance for Emissions of Gases Considered Here[a]

	CO_2	CO	CH_4	NMVOC	SO_2	N_2O	NO_x	NH_3
			Energy					
Fossil fuel combustion	>30	10–30	<5	10–30	>30	5–<10	>30	<5
Fossil fuel production	<5	NI/NS	10–30	10–30	NI/NS	NI/NS	NI/NS	NI/NS
Biofuel combustion	(10–30)	10–30	5–<10	10–30	<5	<5	5–<10	5–<10
			Industry					
Industrial processes	<5	<5	<5	>30	10–30	10–30	5–<10	<5
			Agriculture					
Agricultural land	NI/NS	NI/NS	10–30	NI/NS	NI/NS	>30	10–30	10–30
Animals	NI/NS	NI/NS	10–30	NI/NS	NI/NS	>30	NI/NS	>30
			Biomass Burning					
Savannah burning	NI/NS	10–30	<5	5–<10	NI/NS	<5	5–<10	5–<10
Deforestation	5–<10	10–30	<5	<5	NI/NS	<5	5–<10	5–<10
			Waste					
Agricultural waste burning	NI/NS	10–30	<5	5–<10	<5	<5	5–<10	5–<10
Landfills	NI/NS	NI/NS	10–30	NI/NS	NI/NS	NI/NS	NI/NS	5–<10

[a] Importance determination is based on EDGAR 2.0 1990 emission numbers [*Olivier et al.*, 1999a, 1999b]. Contribution given as percent of total; NI/NS indicates not included, no source.

et al., 1999a]. In EDGAR, emissions are calculated on the basis of information stored in the system: activity data, emission factors, and other explanatory variables. The underlying information is organized by (sub)source category, by country or region, or as gridded maps, for a number of sources by season. The database has been designed in a modular fashion using a so-called process approach [*Laan and Bruinsma*, 1993]. In general, emissions are first calculated on a country basis by multiplying activity levels by compound-specific emission factors. These emission factors define the source strength as emission per unit time and per unit activity of the process. The process approach allows the required level of detail to be included through defining a tree of subprocesses in which emission factors are adopted from the parent process if no factor is explicitly specified at the lower level. This inference of emission factors (either through the process tree or the location tree or, subsequently, from a previous year) and related maps (via the process tree) efficiently and transparently defines emission factors and spatial allocation functions. Using specific definitions of sources and regions as groups of subprocesses and countries, respectively, for each compound, emission tables per region and source type can be generated. In addition, thematic maps on a $1° \times 1°$ grid are used by relating a specific grid map to each subprocess defined as the spatial allocation function to convert per process total country emissions to gridded emissions per process involved.

For EDGAR 2.0, 1990 data on national activities were selected on the basis of generally accepted statistical data assembled by international organizations that have performed consistency checks on the data. Thus activity data have been derived, for example, from the International Energy Agency (IEA, energy data), the UN (industrial production and consumption), and the Food and Agriculture Organization (FAO, agricultural data). For biomass burning, agricultural waste burning, and biogenic land-related sources, gridded data were used as basic activity data. Emission factors are either defined uniformly for all countries, for example, for CO_2, or are evaluated for individual countries or groups of countries (regions). In some cases, such as for road traffic, emission estimates for individual countries were used as well as independently defined activity levels to derive country-specific emission factors. When available, major point sources are included in version 2.0 as distribution parameters by combining these per source categories in so-called thematic maps. A population density map was used as the default when no source-specific map was available. Unless stated otherwise, the population map provided by J. A. Logan (personal communication, 1993) was used as a default when no source-specific map was available or when point source data were only available for a few countries. A more detailed description of the data sources used is given by *Olivier et al.* [1996, 1999a, 1999b].

The emissions of CO_2, CO, CH_4, nonmethane volatile organic compounds (NMVOC), SO_2, NO_x, N_2O, and NH_3 for the period 1890–1990 with 10 year intervals presented in this publication have been computed using an emission factor approach. The activity data were taken from international statistics included in the Hundred Year Database for Integrated Environmental Assessments (1890–1990) (HYDE) [*Klein Goldewijk and Battjes*, 1997] supplemented with other data and our own estimates. Historical emission factors per process are based on the emission factors for uncontrolled sources in EDGAR 2.0 for 1990. The databases describe anthropogenic source categories such as fossil fuel production and combustion, industrial production, agricultural practices, waste handling, and land use-related activities. An overview of these categories and their importance for present emissions can be found in Table 1. Please note that the emission inventories for 1990 in EDGAR 2.0 were compiled using more complete and more detailed source categories (see also section 2.6).

2.1. Energy Use

Within the energy sector, three emission source categories are distinguished (Table 1): production and combustion of fossil fuels and burning of biofuels. In the remainder of this paper we define important sources as those that contribute more than 30% to the total emission and significant sources as those that contribute between 10 and 30% to the total emission. Fossil fuel combustion is an important source of CO_2, SO_2, and NO_x and a significant source of CO and NMVOC. Fossil fuel production contributes significantly to CH_4 and NMVOC emissions, and biofuel combustion is a significant source of CO_2 (gross), CO, and NMVOC emissions (see Table 1). However, not all sectors contribute to the emission of each compound. For instance, the analysis by *Olivier et al.* [1999a] showed that the emission factors for combustion differ not only between regions but also between sector and fuel-type combinations. Therefore a further breakdown was made in the fossil fuel combustion sector (i.e., use of coal,

oil, and gas for power generation, industry, transport, and residential energy use including services) and the biofuel combustion sector (industrial and residential sectors). In the fuel production sector we distinguish between the production of hard and brown coal, oil, and gas.

2.1.1. Fossil fuel combustion. Fossil fuel combustion activity data were included for three periods (1890–1920, 1930–1960, and 1970–1990). For the years 1970, 1980, and 1990, IEA energy statistics were used that include annual energy statistics on a country level followed by the breakdown into many detailed fuel types and sectors [*IEA*, 1994].

For the years 1930, 1940, 1950, and 1960 an energy consumption data set was constructed on the basis of a study by *Darmstadter* [1971]. Darmstadter provided annual consumption statistics for three main fuel types (solids, liquids, and gas) and totals of electricity and hydroelectricity production by region and for some countries without sectoral information for selected years only; (i.e., 1925, 1929, 1930, 1933, 1937, 1938, 1950, 1953, 1955, 1957, 1960, 1961, 1962, 1963, 1964, and 1965). The amount of fossil fuels (coal, oil, and gas) needed in the power-generating sector to meet electricity production was determined by subtracting hydroelectricity production from electricity data taking into account the efficiency in electricity production in the past from *Etemad et al.* [1991]. Darmstadter presented the mixture of coal, oil, and gas (in percentages) in the years 1929 and 1965 that was used to produce electricity. The fuel mix applied in the power generation sector was estimated for each year by linear extrapolation of the fuel mix in 1929 and 1960 as presented by Darmstadter.

The amount of coal, oil, and gas used for other purposes than power generation (transport, industry, and residential) was determined by subtracting the amount and type of fuel used for power generation from the total fuel consumption of each fuel type. For 1930 and 1940, no "year-specific" data were included in the *Darmstadter* [1971] study. Linear extrapolation of the 1929 and 1933 values yielded the data for 1930, and likewise, 1940 is based on linear extrapolation of 1938 and 1950 values. Detailed sectoral data were available in the IEA data used for the period 1970–1990. The sectoral split of 1970 was scaled back in time for the years 1930, 1940, 1950, and 1960 by using indicators per sector that can be associated with the fuel use in the sector industry (value-added industry), transport (number of vehicles), and residential (gross domestic product (GDP) per capita). The procedure used for this was to divide the consumption of fuel type per sector in 1970 by the indicator value of 1970 for each region and multiply the result by the indicator value for the years 1930, 1940, 1950, and 1960. The indicators from HYDE were used as driving factors. Value-added industry is an indicator for the contribution of the industrial sector to the GDP of a country. The sum of the value added of the economic sectors agriculture, industry, and service together form the total GDP of a country. Value-added industry and GDP per capita are based on *World Bank* [1993], which presents time series for most of the world from 1970 until 1990. For the period 1890–1970 the historical estimates from *Maddison* [1994] were used. The numbers of vehicles were derived from *Mitchell* [1992, 1993, 1995].

For the years 1890, 1900, 1910, and 1920 the energy consumption per fuel, per sector, and per region were scaled back in time by using UN coal, oil, and gas production used by *Darmstadter* [1971] as an indicator for the fuel consumption. *Etemad et al.* [1991] also presents fuel production figures, which are comparable with the data included by Darmstadter. Energy consumption per fuel type per sector was scaled by multiplying the average fraction of fuel used in each sector (electricity generation, transport, industry, and residential) in the period 1925–1929 with fossil fuel production in the years 1890–1920.

Emissions in the pre-1970 period were calculated by using per sector per fuel type globally uniform emission factors based on emission factors in 1990 in EDGAR 2.0 [*Olivier et al.*, 1999a] in those regions without emission control. We assumed that these values reflect the uncontrolled emission factors of equipment in the pre-1970 period.

Fuel- and sector-specific emission factors for CH_4, NO_x, CO, and NMVOC from stationary sources in EDGAR 2.0 were adopted from the Long-Term Ozone Simulation (LOTOS) database developed for European countries as described by *Builtjes* [1992] and from *EPA* [1996] data for the United States. For the rest of the world we assumed that emissions were essentially unabated, and we used emission factors from LOTOS valid for eastern Europe. For road transport in 1990 country-specific emission factors for CH_4, NO_x, CO, NMVOC, and SO_2 were based on *IPCC* [1994], which are essentially the same as those used by *Andres et al.* [1996]. The factors for N_2O were also from *IPCC* [1994], except for those for road transport, which were based on *Olivier* [1993] and *De Soete* [1993]. For more details on the data sources for the 1990 emission factors, see Table 2.

Since SO_2 emissions from coal largely depend on the fuel sulfur content (for which regional information is available), regional emission factors were used, which represent uncontrolled emission of SO_2. For the period 1970–1990, per sector per fuel/type the non-CO_2 emission factors were interpolated between the 1970 regional emission factors for uncontrolled equipment and the 1990 regional per country values estimated in EDGAR 2.0 as described by *Olivier et al.* [1999a].

2.1.2. Fossil fuel production. Activity data on the production of hard coal, brown coal, crude oil, and natural gas were included on a country basis using input from *Etemad et al.* [1991]. CH_4 emissions from coal mining were calculated with the emission factors used for the 1990 coal production in EDGAR 2.0, which are based on *Smith and Sloss* [1992] (see Table 2). A distinction between surface and underground mining was made. Since further historical information for the period 1890–1990 is lacking, we assumed a constant ratio between underground and surface mining, with values equal to the ratio per country in the year 1990 in the EDGAR 2.0 data set [*Olivier et al.*, 1999a]. CO_2, CH_4, and NMVOC emissions from oil production were calculated with emission factors included in the EDGAR 2.0 data set for 1990. The CO_2 country-specific emission factors were based on estimated emissions from *Marland et al.* [1994]. The aggregated NMVOC emission factors were calculated from emission estimates by *Little* [1989]. CH_4 emissions from natural gas production and transmission are calculated with emission factors taken from *Ebert et al.* [1993], as applied in EDGAR 2.0.

2.1.3. Biofuel combustion. Reliable activity data of historical biofuel combustion are not included in HYDE and are rarely found in literature. Even for the present data, the information is very uncertain. Therefore we applied a simple approach for this source (assuming that biofuel use in industrialized countries was not substantially higher in the past century than at present). To estimate emissions from biofuel use in the period 1890–1980, the EDGAR 1990 values were extrapolated back in time by dividing the biofuel use in 1990 per country by the rural population per country in 1990 and multiplying this with the rural population per country for the years 1890–1980. For most countries the total biomass use per country in 1990 has been taken from *Hall et al.* [1994], resulting in a global total of 50 EJ for 1990, which is considerably higher than most FAO estimates. A possible explanation for this discrepancy is that FAO statistics are based on market figures, whereas in many countries these may not be representative. Rural population data for the years 1890–1980 were taken from the HYDE database, based on *United Nations (UN)* [1995], *Urquhart and*

Table 2. Emission Factors in Rounded Figures as Used for Energy-Related Emissions 1890–1970[a]

	CO_2, kg C GJ^{-1}	CO, g C GJ^{-1}	CH_4, g C GJ^{-1}	NMVOC, g C GJ^{-1}	SO_2, g S GJ^{-1}	N_2O, g N GJ^{-1}	NO_x, g N GJ^{-1}	NH_3, g N GJ^{-1}
				Fossil Fuel Combustion				
Power Plants								
Coal	26	9	1	2	450	0.6	122	···
Oil	19	9	2	3	600	0.4	67	···
Gas	15	9	1	5	10	0.1	46	···
Domestic								
Coal	26	2100	225	200	Regional[b]	0.9	25	···
Oil	19	13	8	3	200	0.4	15	···
Gas	15	27	···	10	5	0.1	15	···
Industry								
Coal	26	60	8	20	550	0.9	82	2.5
Oil	19	9	2	2	400	0.4	18	···
Gas	15	13	4	5	10	0.1	34	···
Transport								
Coal	26	64	8	20	450	0.9	82	···
Oil	19	4300	15	1300	100	0.4	183	0.1
Gas	15	···	···	10	10	0.1	···	···
				Fossil Fuel Production				
Brown coal	···	···	24	···	···	···	···	···
Hard coal — surface	···	···	840	···	···	···	···	···
Hard coal — underground	···	···	77	···	···	···	···	···
Oil	Country[c]	···	···	1800	···	···	···	···
Gas	···	···	660	···	···	···	···	···
				Biofuel Combustion				
Industry	30	510	8	65	5	0.9	31	45
Domestic	30	2000	300	800	5	1.3	24	45

[a] Resulting aggregated regional emission factors for 1990 presented by *Olivier et al.* [1999a, 1999b, 2000]. Primary data sources for the 1990 emission factors: CO_2, *IPCC* [1994] and *Andres et al.* [1996], also see *Marland et al.* [1999]; N_2O, *Bouwman et al.* [1995, and references therein] and *Olivier* [1993]; petrol-fueled cars equipped with catalytic converters, *De Soete* [1993]; NH_3, *Bouwman et al.* [1997]; SO_2: J. Berdowski (personal communication,1995) (except Japan: *Kato and Akimoto* [1992]). Primary data sources for other factors: all sectors road and aircraft, *Builtjes* [1992, and references therein] (except United States: *EPA* [1996]); NO_x in Japan, *Kato and Akimoto* [1992]; and road transport: *Samaras and Veldt* [1993] (except United States: *EPA* [1996]).

[b] Domestic coal (g SO_2-S GJ^{-1}): Canada, 550; United States, 350; Latin America, 500; Africa, 300; OECD Europe, 450; eastern Europe, 450; FSU, 400; Middle East, 700; India region, 300; China region, 500; east Asia, 350; Oceania, 400; and Japan, 250.

[c] Country-specific emission factors [*Olivier et al.*, 1999b].

Buckley [1965], *U.S. Bureau of Census* [1990], *Grigg* [1997], and our own estimates. The result of scaling biofuel with rural population data was consistent with results obtained in Africa by *Marufu et al.* [1999]. We distinguish between industrial and domestic emissions. The calculation of emissions from biofuel combustion is based on globally uniform emission factors (Table 2). Emission factors are taken from EDGAR 2.0: CO, CH_4, and NMVOC from *Veldt and Berdowski* [1995], N_2O from fuel wood from *Smith et al.* [1993], and NO_x and NH_3 from LOTOS [*Builtjes*, 1992]. For CO_2, gross emission factors are from *IPCC* [1994], and the SO_2 emission factors are provided by J. Berdowski (personal communication, 1995).

2.2. Industrial Processes

The following industrial processes are included in this study: the production of iron, steel, copper, nitric acid, adipic acid, and cement and solvent use. For 1890–1980, production data for iron, steel, copper, and cement by country are taken from the HYDE database. Additional estimates were derived from activity data pertaining to solvents, adipic acid production, and nitric acid production. As shown in Table 1, industrial processes are an important source of NMVOC emissions (solvents) and a significant source of SO_2 and N_2O emissions (copper and adipic/nitric acid production, respectively).

Data in HYDE on iron production are taken from *Mitchell* [1992, 1993, 1995]. *Mitchell* [1992, 1993, 1995] and *Organization for Economic Co-operation and Development (OECD)* [1972, 1975a, 1975b, 1992] provided data for steel production figures as used in HYDE. Emissions from iron production were calculated using default world emission factors for CO, CH_4 [*Builtjes*, 1992; *Olivier et al.*, 1999b], and NMVOC [*Olivier et al.*, 1999a]. Table 3 presents emission factors used for the calculation of industrial emissions. CO and NO_x emissions from steel production are calculated with regional emission factors based on the LOTOS database [*Builtjes*, 1992], taking into account steel production in different furnace types. NMVOC emissions from steel production were calculated with a global default emission factor from *Olivier et al.* [1999a]. Copper production data in the period 1890–1955 are based on *Schmitz* [1979], and those for the period 1960–1990 on *Metallgesellschaft* [1991]. SO_2 emission factors for copper production were taken from *Olivier et al.* [1996, 1999a]. Finally, historical data on cement production are based on *Woytinski and Woytinski* [1953], *Marland et al.* [1994], and *Solomon* [1994]; the emission factor for CO_2 emissions from cement production is from *Marland and Rotty* [1984].

2.2.1. Adipic acid and nitric acid. The production of adipic acid became significant after World War II. Scaling the 1990 country data from EDGAR 2.0 to the year 1950 (for which no production was assumed), using population numbers as an indicator yields a first estimate of adipic acid production. Nitric acid is mainly used in fertilizer production. The production of nitric acid has become significant since 1930 [*Mitchell*, 1992, 1993, 1995]. During the construction of EDGAR 2.0, *Olivier et al.* [1999a] found that these statistics and data provided from industry are inconsistent. Therefore statistics of N fertilizer production *FAO* [1991] were adopted as a proportionality factor for nitric acid production. FAO presents data for the period 1961–

Table 3. Emission Factors in Rounded Figures as Used for Industrial Emissions

	CO_2, kton C kton^{-1}	CO, kg C kton^{-1}	CH_4, kg C kton^{-1}	NMVOC, kg C kton^{-1}	SO_2, ton S kton^{-1}	N_2O, kg N kton^{-1}	NO_x, kg N kton^{-1}	NH_3, kg N kton^{-1}
Iron	...	4	675	100	...	...	...	...
Steel			...		...	...		...
OHF	...	21		100			...	
BOF	...	6		...			30	
EAF	...	4		200			61	
Copper	...	...	...	...				
Canada					79			
United States					9			
Latin America					1,060			
Africa					910			
OECD Europe					641			
Eastern Europe					1,100			
FSU					1,100			
Middle East					1,470			
India region					470			
China region					960			
East Asia					1,060			
Oceania					560			
Japan					11			
Nitric acid	...	...	...	...	...	16	...	...
Adipic acid								
Global	...	...	...	...	...	147	...	...
United States						110		
Canada						185		
Cement	136	...	...	...	...	...	...	...
Solvents	...	...	...	1,000,000	...	...	...	...

1990; the 1970 value was extrapolated to zero in 1930. The emission factors for N_2O emissions from adipic and nitric acid production were based on *Reimer et al.* [1992].

2.2.2. Solvent use. Historical data on solvent use are not available. The 1990 data included in EDGAR 2.0 were adopted to estimate the solvent use in the period 1890–1970. The 1990 solvent data from EDGAR 2.0 suggest a relationship between GDP and solvent use. Therefore historical solvent use was scaled to GDP in time. Historical GDP data are presented in HYDE on a regional scale, being used to estimate the regional historic solvent use production by each category. The solvent use by country is based on the 1990 country share to the total regional consumption of solvents.

2.3. Agriculture (Nonburning)

We distinguish between agricultural emissions from rice cultivation, fertilizer use, and domestic animals. According to Table 1 these sectors are important sources of N_2O and NH_3 (fertilizer use and animals) and significant sources of CH_4 (rice). The area under rice cultivation was adopted from the HYDE database, as based on *Mitchell* [1992, 1993, 1995]. Data gaps between years were filled by linear extrapolation. For the period 1890–1970 a world default emission factor was applied to calculate CH_4 emissions from rice fields, and for the period 1970–1990, regional emission factors were used. Both global and regional emission factors were adopted from *Kreileman and Bouwman* [1994].

Fertilizer consumption was taken from FAO statistics [*FAO*, 1991] for the period 1961–1990. Since chemical fertilizers were not widely used before 1950, the 1965–1961 FAO trend is extrapolated to the year 1950. Emissions of N_2O and NH_3 result from fertilizer use. The global emission factors for N_2O and NH_3 emissions were adopted from *Bouwman et al.* [1995, 1997]. NO_x emissions from soils can be enhanced because of fertilization. We used the 1990 Global Emissions Inventory Activity (GEIA) estimate for above-canopy soil emissions compiled by *Yienger and Levy* [1995]. According to Yienger and Levy, ~2 out of 5.5 Tg N yr^{-1} present-day soil emissions result from agriculture and conversion of forests to grasslands. Using fertilizer use as an approximation for the regional development of agriculture, soil-NO_x emissions were scaled with time. Thus, following the estimate of Yienger and Levy, preindustrial soil NO_x emissions were assumed to amount to 2.7 Tg N yr^{-1}.

Country statistics for livestock are available for the period 1961–1990 from *FAO* [1991], and regional estimates for the period 1890–1960 are presented in the HYDE database, based on *Mitchell* [1992, 1993, 1995]. Animals included in this study are cattle, goats, chicken, pigs, sheep, horses, buffaloes, and camels. To derive country data on animal numbers in the period 1890–1990, we combined the FAO country figures of 1961 with the regional trends from the HYDE database. Emission factors for CH_4, N_2O, and NH_3 from animal waste are taken from *Kreileman and Bouwman* [1994] and *Bouwman et al.* [1995, 1997] and are applied to the whole period (see Tables 4a and 4b). Global emission factors for emissions by buffaloes and camels and regional emission factors for other ruminants were taken from *Gibbs and Leng* [1993].

2.4. Biomass Burning

In this study we define biomass burning as savannah burning and deforestation, although formally, organic waste burning (municipal, industrial, and agricultural) and residential biofuel burning also contribute. In this study municipal and industrial waste burning are considered to be negligible, and agricultural waste burning and biofuel burning are treated as separate source categories (see sections 2.5 and 2.1). Savannah burning and deforestation are mostly human-induced but can also be caused by lightning. A problem with determining the emissions from biomass burning is the lack of information, especially about the amount of biomass burned, which applies to both recent and historical data. A further complicating factor is that even emission factors may have changed since at present, vegetation burns more frequently than in the past; young vegetation contains more nitrogen than mature vegetation. Both savannah burning and deforestation are large sources for CO, and deforestation is a significant source for CO_2 (Table 1). Note

Table 4a. Emission Factors in Rounded Figures as Used for Agricultural Emissions

	CO_2	CO	CH_4^{-1}, ton C km^{-2}	NMVOC	SO_2	N_2O, kg N (kton fertilizer)$^{-1}$	NO_x, kg N (kton fertilizer)$^{-1}$	NH_3, kg N (kton fertilizer)$^{-1}$
			Agricultural Land					
Rice 1890–1970	...	...	34	...	...	...	...	...
Rice 1970–1990	...	...	regional[a]	...	...	...	...	...
Fertilizer	...	...	...	...	...	12,500	see description in text	*Bouwman et al.* [1997]

	CO_2	CO	CH_4^{-1}, kg C (1000 head)$^{-1}$	NMVOC	SO_2	N2O, kg N (1000 head)$^{-1}$	NO_x	NH3, kg N (1000 head)$^{-1}$
				Livestock				
Buffaloes	...	...	39,800	...	...	286	...	7160
Camels	...	...	460	...	...	350	...	870
Poultry	...	...	regional[b]	...	...	3	...	1480
Goats	...	...	regional[b]	...	...	57	...	7410
Horses	...	...	regional[b]	...	...	286	...	7160
Pigs	...	...	regional[b]	...	...	70	...	3260
Sheep	...	...	regional[b]	...	...	64	...	820
Nondairy cattle	...	...	regional[b]	...	...	regional[c]	...	6410
Dairy cattle	...	...	regional[b]	...	...	regional[c]	...	14300

that we assume that savannah burning is not a net source of CO_2 since this vegetation regrows on the timescale of a year or so.

Present-day savannah-burning emissions were taken from the compilation by *Hao et al.* [1990], which was interpolated to a 1° × 1° grid. This compilation was augmented by an estimate for savannah burning in tropical north Australia according to *Bouwman et al.* [1997]. To estimate the human contribution to savannah burning, we assumed that in the tropical wet savannah (arbitrarily defined as that exposed to an annual rainfall amount in excess of 1000 mm yr^{-1}) all biomass burning is induced by human activities (J. Lacaux, personal communication, 1998). For dry savannas we assumed that natural fires lead to half of the biomass-burning emissions.

Deforestation is related to both slash-and-burn agriculture and, especially in more recent years, large-scale logging. This presents the difficulty that whereas the first practice leads to direct trace gas emissions in the respective regions, the latter activity only partly leads to reactive trace gas emissions. In, for example, the Amazon region, deforestation was strongly related to the building of new roads, opening up the primary forest, which led to the migration of slash-and-burn farmers. Therefore we assumed the development of a rural population, for which we have a gridded database available, as an indicator for emissions resulting from deforestation fires. We compared the temporal development of these emissions with deforestation data provided by *FAO* [1991] and *Richards* [1990] and infer that as a first approximation, the relation between rural population and deforestation emissions can be used. The above mentioned approach only refers to tropical regions. We have assumed that deforestation in industrialized regions occurred primarily before 1890, and we have neglected for these regions both deforestation and afforestation activities after 1890. Clearly, our simplified approach for tropical as well as for temperate regions should be improved by other studies on global biomass burning, which were not accessible during the construction of our data set. We note, however, that at present, there is no consensus on the net flux of CO_2 from industrialized countries. Since most CO_2 flux models are process-based, one cannot easily derive national total and spatial distributions of large-scale biomass burning related to deforestation. According to *Hougthon* [1999] the nontropical countries contribute only about 25% to the global total net flux for the past 100 year period. Using an alternative method than the so-called bookkeeping methods used by Houghton and others,

Table 4b. Regions Referred to in Table 4a

	Canada	United States	Latin America	Africa	OECD Europe	Eastern Europe	FSU	Middle East	India Region	China Region	East Asia	Oceania	Japan
						CH_4, ton C km^{-2}							
Rice	34	34	29	17	34	34	34	34	30	33	30	34	34
						Livestock CH_4, kg C (1000 head)$^{-1}$							
Poultry	200	100	90	86	120	100	110	58	74	43	95	110	270
Goats	4,070	4,070	3,920	3,910	4,190	4,330	4,520	3,980	3,870	4,010	4,070	4,080	4,070
Horses	16,300	16,400	16,400	15,100	18,800	24,100	15,900	17,800	14,400	15,500	16,300	16,300	16,300
Pigs	5,360	15,300	2,970	2,500	7,640	8,180	2,570	750	1,300	4,000	6,200	30,400	12,500
Sheep	6,300	6,300	4,000	3,910	6,440	6,580	6,770	3,980	3,900	4,000	4,100	4,080	6,300
Nondairy cattle	42,700	42,100	38,300	24,900	58,500	55,800	54,500	27,300	23,000	34,200	26,200	41,800	52,400
Dairy cattle	51,100	95,200	39,000	25,600	72,300	53,500	54,400	29,200	24,600	43,600	72,500	44,400	103,000
						Livestock N_2O–N, kg N (1000 head)$^{-1}$							
Nondairy cattle	290	290	250	250	290	290	290	250	250	250	250	250	290
Dairy cattle	510	510	380	380	510	510	510	380	510	380	380	510	510

Table 5. Emission Factors in Rounded Figures as Used for Waste-Related Emissions

	CO_2, kg C GJ^{-1}	CO, kg C GJ^{-1}	CH_4, kg C GJ^{-1}	NMVOC, kg C GJ^{-1}	SO_2, kg S GJ^{-1}	N_2O, kg N GJ^{-1}	NO_x, kg N GJ^{-1}	NH_3, kg N GJ^{-1}
Agricultural waste burning	...	100,000	10,000	1,600	710	110	2,500	1,500
Landfills								
Canada			31					
United States			30					
Latin America			5					
Africa			2					
OECD Europe			13					
Eastern Europe			9					
FSU			8					
Middle East			7					
India region			1					
China region			2					
East Asia			2					
Oceania			34					
Japan			4					

DeFries et al. [1999] tried to quantify this integrated net CO_2 flux and found that regional estimates deviate considerably from the other methods, notably in the temporate regions.

Since the amount of biomass burnt at present and the spatial distribution are already highly uncertain, and in addition subject to a large interannual variability, we feel that it is not justified to add too much detail to the database. Therefore, in future work a combination of satellite retrieval and modeling may provide more reliable estimates on the present and past emissions by biomass burning.

2.5. Agricultural Waste Burning and Landfills

Agricultural waste burning is a large source of CO emissions and significant for NO_x and NMVOC emissions. Landfills are a significant source of CH_4 (Table 1). The amount of arable land is used as a proxy for historical agricultural waste burning and has been derived from the HYDE database and *Houghton et al.* [1983]. Global emission factors were applied to calculate emissions of CH_4, CO, N_2O, NH_3, NMVOC, NO_x, and SO_2 (see Table 5). N_2O emission factors were taken from *Crutzen and Andreae* [1990]; the NMVOC, CO, and CH_4 emission factors are from *Veldt and Berdowski* [1995]; the SO_2 and NO_x emission factors are from *Andreae* [1991]; and that for NH_3 is from *Bouwman et al.* [1997]. As a first approximation for landfill emissions, we assumed a linear relationship with urban population numbers [*Klein Goldewijk and Battjes*, 1997; *UN*, 1995]. Regional emission factors for CH_4 were derived from *Subak et al.* [1992].

2.6. Distribution of Emissions on a 1° × 1° Grid

The calculated country-by-country anthropogenic emissions in this study are distributed on a 1° × 1° grid using spatial allocation functions in the form of thematic maps on this grid scale. Grid maps used for distributing the per country emission for 1990 were also applied for the whole 100 year period. Obviously, this is only valid as a proxy since many human activities have spatially shifted in time, sometimes even substantially. However, since accurate historical information is lacking, this approach is a transparent method to estimate the spatial distribution of historical emissions within a country and is also used in other studies [e.g., *Andres et al.*, 1996, 1999]. Thus the method can be easily improved as more information becomes available.

For fossil fuel distribution and combustion, industrial processes, and landfills the emissions were distributed according to population density (J. Logan, personal communication, 1993). Emissions from fossil fuel production were allocated using point source maps for coal mining and for oil and gas production, respectively [*Olivier et al.*, 1996, 1999a]. In the agricultural sector, emissions from fertilizer use were allocated according to an arable land map [*Bouwman et al.*, 1995]. Emissions from rice cultivation were distributed using a wetland rice cultivation map at 5° × 5° [*Aselman and Crutzen*, 1989], while emissions from animals were distributed according to livestock density [*Lerner et al.*, 1988]. Biomass-burning emissions were allocated to a 1° × 1° grid by a deforestation and savannah-burning map [*Hao et al.*, 1990]. Finally, agricultural waste-burning emissions were distributed according to *Bouwman et al.* [1995].

3. Results

By using the activity data and emission factors as described in section 2, emissions were calculated for the period 1890–1990 by country and interpolated to a 1° × 1° longitude/latitude grid. Global emissions for the eight compounds by source category for the years 1890–1990 are presented in Plate 1 and Table 6. In Plates 2a–2d we present as an example the temporal and spatial development of NOx emissions in 1890, 1940, 1970, and 1990. The strong increases in emissions in Europe and North America are clearly visible in the 1890–1970 plots, whereas emissions in, for example, Asia become increasingly important after 1970.

3.1. Carbon Dioxide (CO_2)

According to this study the overall global anthropogenic CO_2 emissions have increased from 1.2 in 1890 to 7.9 Pg CO_2-C in 1990, which is a more than sixfold increase (Plate 1a). The contributions by biofuel and fossil fuel combustion emissions increased by factors of 2 and 13, respectively. It is clear that energy use has been the major contributor to the total anthropogenic carbon dioxide emissions throughout this century (85% in 1890 and 92% in 1990). Within the energy sector the gross share of biofuel combustion has declined from 64% in 1890 to 21% in 1990, (net: from 15 to 3% when assuming 10% unsustainable production), while the contribution of fossil fuel combustion increased conversely. CO_2 emissions caused by deforestation, fossil fuel production, and industrial activity (cement production) also show a relatively large increase, but these sectors contribute little to the total CO_2 emissions. In 1890 the regions with the largest contribution to CO_2 emissions are the United States (22%), OECD Europe (19%), China (16%), Latin America (12%), and India (11%). In 1990 the four main regions are the United States (18%), China (16%), Latin America (12%), and India (11%).

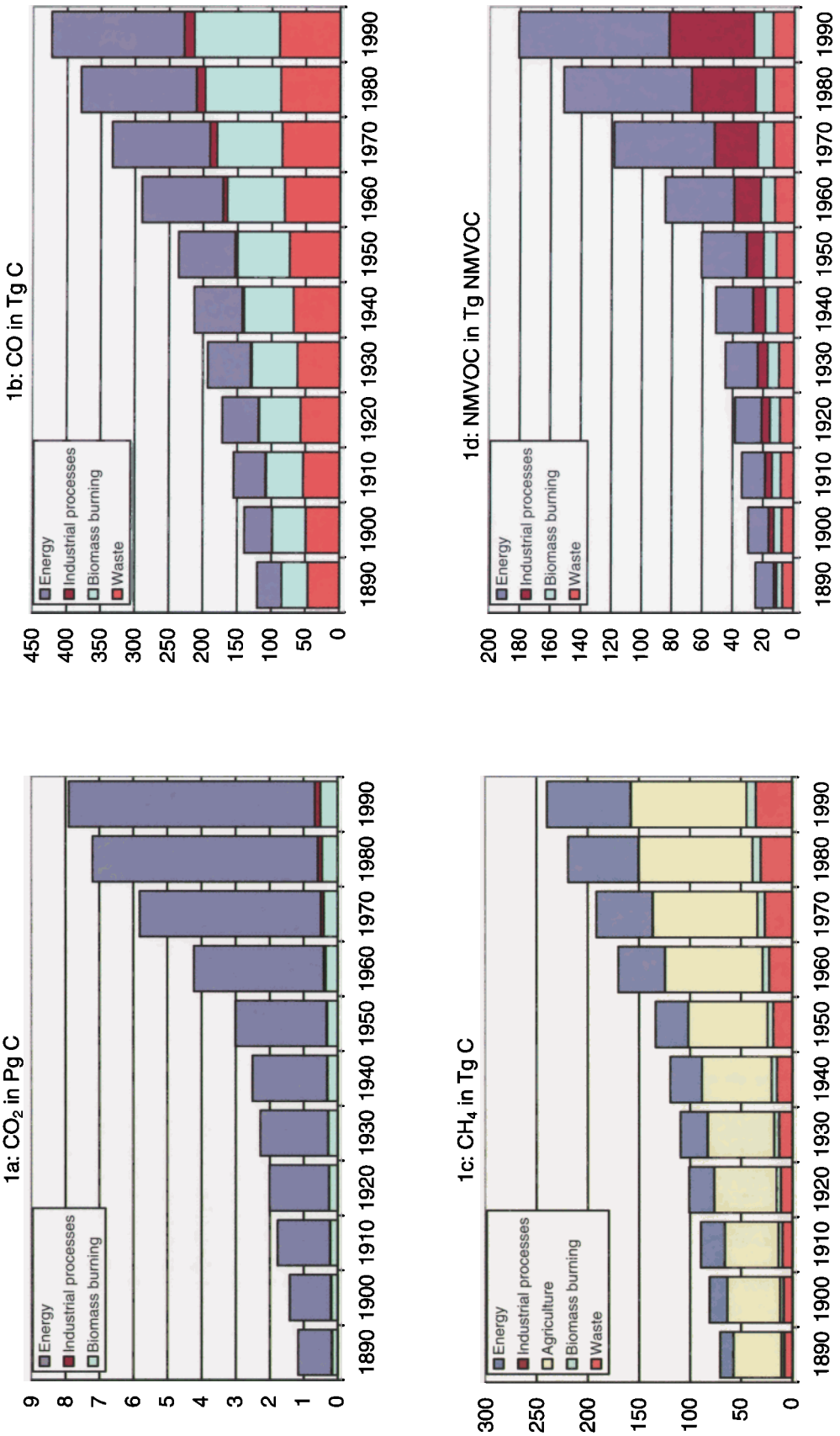

Plate 1. Estimated anthropogenic emissions in the period 1890–1990: (a) CO_2 emissions in Pg CO_2-C, (b) CO emissions in Tg CO-C, (c) CH_4 emissions in Tg CH_4-C, (d) NMVOC emissions in Tg NMVOC, (e) SO_2 emissions in Tg SO_2-S, (f) NO_x emissions in Tg NO_2-N, (g) N_2O emissions in Tg N_2O-N, and (h) NH_3 emissions in Tg NH_3-N.

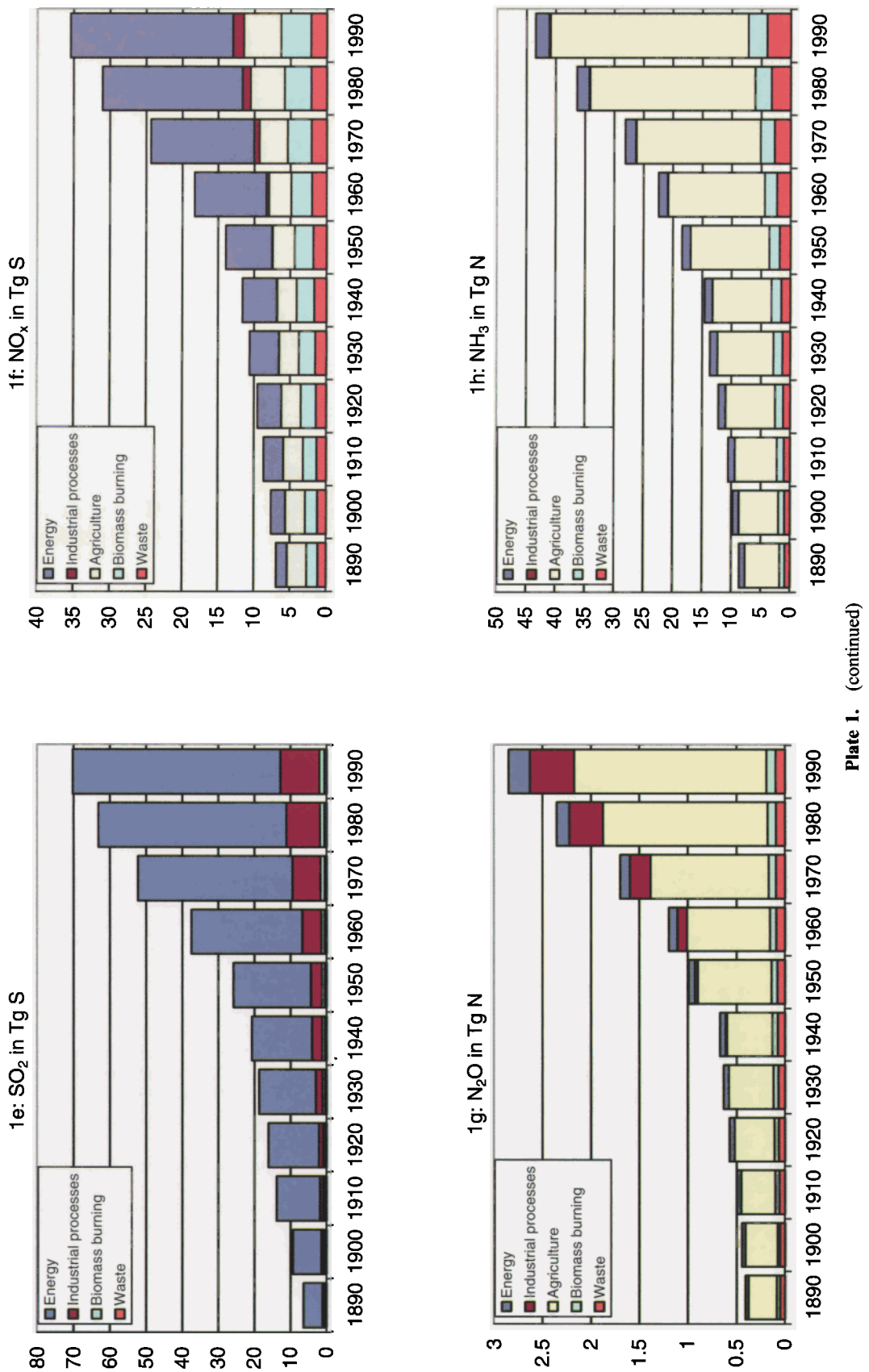

Plate 1. (continued)

3.2. Carbon Monoxide (CO)

Anthropogenic CO emissions have increased by a factor of 3, from 121 Tg C in 1890 to 423 Tg C in 1990. Main source categories in 1890 were agricultural waste burning (46 Tg, 38%), biomass burning (savannah burning and deforestation; 38 Tg, 31%), and biofuel combustion (31 Tg, 26%). Throughout the years these sources have increased by factors of 2, 2, and 3, respectively. Fossil fuel combustion has increased from a small CO source in 1890 to the largest CO emission category in 1980 and 1990 (112 Tg in 1990, 26% of the total anthropogenic CO emissions). CO emissions from industrial activities (iron and steel production) show the largest increase in 100 years (a factor of 62), but this sector does not contribute significantly to the total CO emissions (<4% in 1990). Africa, Latin America, India, and China are the regions with the largest CO emissions over the entire time period, which is remarkably different from the emissions of CO_2. This difference can be explained by the fact that waste and biomass-burning activities (mostly applied in these regions) contribute significantly to total CO emissions, while CO_2 emissions are dominated by fossil fuel combustion.

3.3. Methane (CH_4)

Global anthropogenic CH_4 emissions in the period 1890–1990 show a threefold increase, from 71 Tg C in 1890 to 240 Tg in 1990. In 1890 the agricultural emissions were the largest CH_4 source (47 Tg, 66% of the total anthropogenic emissions), with equal contributions by domestic ruminants and agricultural land (anaerobic processes in rice fields). Over the period of study the agricultural sector remained the largest CH_4 source (113 Tg in 1990, 47%). From 1950 onward, livestock was the largest contributor to the sector total. After 1960, fossil fuel production became a significant source (67 Tg in 1990, 28%), mainly caused by leakage during transmission and distribution of natural gas. Because of the dominance of methane emissions by the agricultural sector, it is no surprise that regions with important agricultural activity are the largest CH_4-emitting regions. In 1890 the regions with the largest contribution to CH_4 emissions were India (36% of total emission), OECD (15%), China (14%), and the United States (11%). India (18%), China (15%), Former Soviet Union (FSU; 15%), the United States (13%), and Latin America (11%) were the major contributors to the global CH_4 emissions in 1990.

3.4. Nonmethane Volatile Organic Compounds (NMVOC)

In the period 1890–1990, anthropogenic NMVOC emissions increased by a factor of 7, from 25 Tg NMVOC in 1890 to 181 Tg in 1990 (note that the units are Tg NMVOC). In 1890, biofuel combustion (11 Tg, 44% of total emission) and agricultural waste burning (7 Tg, 28%) accounted for 72% of the total NMVOC emissions. During the period of study there has been a shift from biofuel combustion and agricultural waste burning toward industrial and energy-related processes. In 1990, industrial processes (solvent use and iron and steel production) emit 56 Tg NMVOC, which is 31% of the total emissions. Fossil fuel combustion accounts for 42 Tg (23%). The 32 Tg from biofuel combustion and the 26 Tg from fossil fuel production account for 18 and 14%, respectively. The largest emitters in 1890 were Africa (24%), China (18%), India (16%), and Latin America (15%) related to biofuel, savannah, and agricultural waste burning. Over the period 1890–1990, energy and industrial processes increasingly contributed to NMVOC emissions, reflected in the contribution of OECD (11% in 1990) and the United States (13%) as important emitters in 1990.

3.5. Sulphur Dioxide (SO_2)

Anthropogenic SO_2 emissions have increased from 6 Tg S in 1890 to 70 Tg in 1990, an increase of a factor of 12. Throughout the period considered in this study, fossil fuel combustion remained by far the major contributor to SO_2 emissions (81% in 1890 and 81% in 1990). Over the years, emissions from industrial production (copper smelting) became a significant source of SO_2, with an increase from 0.3 Tg in 1890 (5% of the total) to 11 Tg in 1990 (16%). From 1890 to 1940, world SO_2 emissions mainly originate from the regions the United States (~3 Tg in 1890, 43% of total emission) and OECD Europe (~2 Tg in 1890, 28%). From 1940 onward the contribution of the FSU region increased, and from 1960 the emissions from the China region became important.

3.6. Nitrogen Oxides (NO_x)

The calculated NO_x emissions show an increase from 1890 to 1990 by a factor of 5 from 7 Tg N in 1890 to 35 Tg in 1990. In 1890, NO_x was mainly emitted from soils and savannah burning; in fact, these sources represent to a large degree the "natural" fraction of these emissions. Within only a few decades the emissions from fossil fuel combustion became more important, and from 1930 onward it has been the largest NO_x source, resulting in 60% of the global NO_x emissions in 1990. Africa, Latin America, and the United States together accounted for 60% of the global NO_x emissions in 1890. Later in the period of study, emissions from Europe, FSU, and China also contributed significantly to the total NO_x emission. In Plates 2a–2d we present the temporal and spatial development of NO_x emissions in 1890, 1940, 1970, and 1990. In 1890, parts of northern Europe and the east coast of the United States have emissions larger than 1000 mgN m^{-2} yr^{-1}; in the rest of the world, emissions are generally below 300 mg N m^{-2} yr^{-1}. In 1940 the regions of high emissions are still found in northern Europe and the eastern United States. The development and further spread of anthropogenic emissions in the United States and Europe are evident in 1970. In Africa and Latin America, biomass-burning emissions become more important, whereas the first signs of strong economical development in Asia become apparent. The further increase of Asian emissions is visible in 1990.

3.7. Nitrous Oxide (N_2O)

Nitrous oxide emissions increased from 0.4 Tg N_2O-N in 1890 to 2.9 Tg in 1990, which is an increase of a factor of 7 (excluding indirect emissions from deforestation). Animals were the main source of N_2O emissions until World War II, ranging from 0.3 Tg (70%) in 1890 to 0.5 Tg (70%) in 1940. In 1950, after the introduction of synthetic fertilizers, N_2O emissions from agricultural land and industry became significant. Regions with the largest N_2O emissions in 1890 were India (34%), the United States (15%), and OECD Europe (11%). In the period between 1890 and 1990, emissions from China and Latin America contributed significantly to the global budget, while the contribution of the Indian region became less dominant (17% in 1990).

3.8. Ammonia (NH_3)

Ammonia emissions increased from 9 Tg NH_3-N in 1890 to 43 Tg in 1990 (almost a fivefold increase). Throughout the period of study, NH_3 emissions are dominated by agricultural emissions. Emissions by animals account for 65% of total NH_3 emissions in 1890. In 1990, 79% of the NH_3 emissions originate from the agricultural sector, with 21 Tg from animals (~50%) and 13 Tg from agricultural land (~30%).

Although the hundred year historical emission data set is based on the EDGAR 2.0 methodology, as presented by *Olivier et al.* [1999a], some differences between our 1990 emission estimates and the 1990 emission estimates in this report are present. The use of aggregated emission factors will lead to some differences in

Table 6. Anthropogenic Emissions by Compound and Sector for the Period 1890–1990

	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990
					CO_2, Pg C						
Energy	1.0	1.2	1.5	1.7	2.0	2.2	2.7	3.8	5.3	6.6	7.2
Fossil fuel combustion	0.4	0.5	0.8	1.0	1.2	1.3	1.8	2.8	4.1	5.2	5.7
Fossil fuel production	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
Biofuel combustion[a]	0.6	0.7	0.7	0.8	0.8	0.9	0.9	1.0	1.2	1.3	1.5
Industrial processes	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.2
Biomass burning	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.4	0.4	0.5	0.5
Savannah burning	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Deforestation	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.4	0.4	0.5	0.5
Total[a]	1.2	1.4	1.8	2.0	2.3	2.5	3.0	4.2	5.8	7.2	7.9
					CO, Tg C						
Energy	35.9	40.5	46.8	52.9	62.5	70.0	82.6	118.1	143.0	169.1	194.9
Fossil fuel combustion	4.8	7.4	11.3	14.7	21.7	26.4	37.0	65.7	80.6	96.1	112.0
Biofuel combustion	31.0	33.1	35.5	38.1	40.7	43.6	45.6	52.4	62.4	73.0	82.9
Industrial processes	0.2	0.5	1.2	1.4	1.8	2.8	3.6	6.7	10.2	12.6	14.9
Biomass burning	38.4	48.8	54.1	60.4	66.8	72.1	76.2	83.4	94.9	110.6	124.4
Savannah burning	23.3	31.5	34.1	37.2	40.9	44.3	47.4	50.0	56.2	67.3	76.6
Deforestation	15.0	17.4	20.1	23.2	25.9	27.8	28.8	33.4	38.6	43.3	47.8
Waste	46.0	49.3	53.2	57.1	61.9	67.9	73.7	81.4	85.1	86.9	88.7
Agricultural waste burning	46.0	49.3	53.2	57.1	61.9	67.9	73.7	81.4	85.1	86.9	88.7
Total	120.5	139.2	155.3	171.7	193.0	212.8	236.2	289.6	333.1	379.3	422.9
					CH_4, Tg C						
Energy	13.0	17.3	23.4	24.5	26.4	30.7	31.8	45.7	55.0	68.8	81.7
Fossil fuel combustion	0.5	0.7	1.1	1.3	1.5	1.6	2.0	3.1	3.6	3.2	3.3
Fossil fuel production	8.4	12.2	17.6	18.1	19.4	23.2	23.6	35.5	42.8	55.4	66.8
Biofuel combustion	4.1	4.4	4.7	5.1	5.5	5.9	6.2	7.1	8.7	10.2	11.6
Industrial processes	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.3	0.4	0.5	0.6
Agriculture	46.8	51.5	52.4	60.7	65.0	68.2	77.5	95.3	102.3	111.2	113.1
Agricultural land	26.7	28.1	27.9	31.7	32.8	34.5	35.2	43.6	45.0	46.2	44.4
Animals	20.1	23.3	24.5	29.1	32.2	33.7	42.3	51.7	57.2	65.0	68.7
Biomass burning	3.2	3.5	3.9	4.4	4.9	5.2	5.5	6.2	7.1	8.0	9.0
Savannah burning	1.8	2.0	2.2	2.4	2.6	2.8	2.9	3.3	3.7	4.2	4.8
Deforestation	1.3	1.5	1.8	2.0	2.3	2.4	2.5	2.9	3.4	3.8	4.2
Waste	7.5	8.5	9.6	11.0	12.7	14.9	18.5	22.6	26.8	30.6	35.4
Agricultural waste burning	4.6	4.9	5.3	5.7	6.2	6.8	7.4	8.1	8.5	8.7	8.9
Landfills	2.9	3.5	4.3	5.3	6.5	8.1	11.2	14.4	18.3	21.9	26.5
Total	70.5	80.8	89.4	100.7	109.1	119.1	133.4	170.1	191.6	219.1	239.8
					NMVOC, Tg mass						
Energy	12.2	13.5	15.3	17.6	21.2	24.4	29.8	45.3	66.1	84.2	98.9
Fossil fuel combustion	0.8	1.3	2.0	2.8	4.7	6.1	8.8	17.7	25.4	34.2	41.5
Fossil fuel production	0.1	0.2	0.3	0.8	1.5	2.2	4.0	8.1	17.1	22.3	25.8
Biofuel combustion	11.3	12.1	13.0	14.0	15.0	16.1	16.9	19.5	23.6	27.7	31.6
Industrial processes	1.8	3.5	4.5	5.5	6.6	8.2	11.2	17.5	28.7	42.0	55.7
Biomass burning	3.6	5.0	5.8	6.6	7.4	8.0	8.4	9.1	10.4	11.7	12.7
Savannah burning	1.2	2.2	2.5	2.8	3.2	3.5	3.6	3.7	4.1	4.6	4.9
Deforestation	2.5	2.8	3.3	3.8	4.2	4.6	4.7	5.5	6.3	7.1	7.8
Waste	7.2	7.7	8.3	8.9	9.6	10.6	11.5	12.7	13.3	13.5	13.8
Agricultural waste burning	7.2	7.7	8.3	8.9	9.6	10.6	11.5	12.7	13.3	13.5	13.8
Total	24.8	29.7	33.8	38.5	44.9	51.2	60.8	84.6	118.4	151.5	181.0
					SO_2, Tg S						
Energy	5.3	7.9	12.0	14.0	15.7	16.6	21.4	30.8	42.8	52.0	57.5
Fossil fuel combustion	5.2	7.8	11.8	13.9	15.6	16.5	21.2	30.6	42.7	51.7	57.2
Biofuel combustion	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2
Industrial processes	0.3	0.5	0.9	1.0	1.8	2.8	3.0	5.2	7.8	9.3	10.8
Biomass burning	0.5	0.6	0.6	0.7	0.7	0.8	0.8	0.9	1.1	1.2	1.4
Savannah burning	0.4	0.4	0.5	0.5	0.6	0.6	0.6	0.7	0.8	0.9	1.0
Deforestation	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.4
Waste	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.6	0.6	0.6	0.6
Agricultural waste burning	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.6	0.6	0.6	0.6
Total	6.4	9.3	13.8	16.1	18.7	20.7	25.8	37.5	52.3	63.1	70.2
					NO_x, Tg N						
Energy	1.5	2.0	2.7	3.3	4.1	4.7	6.4	10.0	14.3	19.4	22.5
Fossil fuel combustion	0.9	1.4	2.1	2.6	3.4	3.9	5.6	9.1	13.3	18.2	21.2
Biofuel combustion	0.6	0.6	0.6	0.7	0.7	0.8	0.8	0.9	1.0	1.1	1.3
Industrial processes	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.4	0.7	1.1	1.5
Agriculture	2.7	2.7	2.7	2.7	2.7	2.7	3.0	3.1	3.9	4.7	5.2

Table 6. (continued)

	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990
Agricultural and natural land	2.7	2.7	2.7	2.7	2.7	2.7	3.0	3.1	3.9	4.7	5.2
Biomass burning	1.5	1.7	1.9	2.1	2.3	2.4	2.5	2.9	3.3	3.7	4.2
Savannah burning	1.2	1.3	1.4	1.5	1.7	1.8	1.9	2.1	2.4	2.7	3.1
Deforestation	0.3	0.4	0.5	0.5	0.6	0.6	0.7	0.8	0.9	1.0	1.1
Waste	1.1	1.2	1.3	1.4	1.5	1.7	1.8	2.0	2.1	2.1	2.2
Agricultural waste burning	1.1	1.2	1.3	1.4	1.5	1.7	1.8	2.0	2.1	2.1	2.2
Total	6.9	7.6	8.7	9.5	10.6	11.6	13.9	18.3	24.3	31.0	35.4
				N_2O, Tg N							
Energy	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.2
Fossil fuel combustion	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.2
Biofuel combustion	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1
Industrial processes	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.2	0.3	0.5
Agriculture	0.3	0.3	0.4	0.4	0.5	0.5	0.8	0.9	1.2	1.7	2.0
Agricultural land	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.4	0.8	1.0
Animals	0.3	0.3	0.4	0.4	0.5	0.5	0.6	0.7	0.8	0.9	1.0
Biomass burning	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Savannah burning	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1
Deforestation[b]	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Waste	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Agricultural waste burning	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Total	0.4	0.4	0.5	0.6	0.6	0.7	1.0	1.2	1.7	2.4	2.9
				NH_3, Tg N							
Energy	1.0	1.0	1.1	1.2	1.3	1.3	1.4	1.6	1.8	2.1	2.3
Fossil fuel combustion	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
Biofuel combustion	1.0	1.0	1.1	1.1	1.2	1.3	1.3	1.5	1.7	2.0	2.2
Industrial processes	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.2	0.2
Agriculture	5.9	6.7	7.1	8.4	9.5	10.0	13.3	16.4	21.1	28.1	33.7
Agricultural land	0.3	0.3	0.4	0.5	0.6	0.8	1.8	2.4	4.7	8.8	12.6
Animals	5.6	6.4	6.7	8.0	8.9	9.2	11.5	14.0	16.4	19.3	21.1
Biomass burning	0.9	1.0	1.2	1.4	1.5	1.7	1.8	2.1	2.4	2.8	3.2
Savannah burning	0.4	0.5	0.6	0.7	0.8	0.8	0.9	1.1	1.3	1.5	1.8
Deforestation	0.4	0.5	0.6	0.7	0.8	0.8	0.8	1.0	1.1	1.3	1.4
Waste	0.9	1.0	1.1	1.2	1.3	1.6	1.9	2.3	2.7	3.3	4.0
Agricultural waste burning	0.7	0.7	0.8	0.9	0.9	1.0	1.1	1.2	1.3	1.3	1.3
Landfills	0.2	0.2	0.3	0.3	0.4	0.5	0.7	1.1	1.4	1.9	2.7
Total	8.6	9.7	10.5	12.2	13.7	14.6	18.4	22.4	28.1	36.3	43.4

[a] Gross CO_2 from biofuel combustion, i.e.,assuming 100% unsustainable production. In practice, this value may be as low as 10%, which could reduce the net CO_2 emission from biofuels by 90%.

[b] Direct effects only. Total N_2O emissions from deforestation including indirect effects resulting from delayed emissions are estimated at about ten times the level of the direct emissions presented here.

situations where EDGAR 2.0 applied detailed emission factors. Furthermore, the spatial emissions from aircraft were not explicitly considered in this study but were partially included in the total transport sector. These emissions are mainly important for CO_2 (~2–3% of present-day fossil fuel-related emissions) and NO_x (~1.5% of present-day NO_x emissions, mainly emitted at upper tropospheric and lower stratospheric altitudes, where NO_x is very efficient in O_3 production). We recommend future users of this database to include separately the aircraft emissions.

4. Discussion

4.1. Results Compared to Other Studies

Figure 1 presents an overview of CO_2 emissions from fossil fuel combustion and cement production computed in this study and estimates by *Keeling* [1994] and *Marland et al.* [1994]. For the period 1890–1960 the global emissions calculated by Keeling and from this study are in good agreement (<5% difference). For the years after 1960 the emissions calculated by Marland et al. are somewhat higher than our estimates. This discrepancy can be partly explained from the fact that we excluded emissions from international air transport, which accounts for about 2% in 1990, and we also used different input data sets (United Nations energy data by Marland et al. versus *IEA* [1994] and *Darmstadter* [1971] energy data in our study). A detailed study by *Marland et al.* [1999] showed that for 1990 the data sets showed very little differences on a country level, generally <1%. The smallest countries, for example, in Africa, had the largest differences, but the largest contributions to uncertainties originate from the largest emitting countries, where low relative uncertainties lead to high absolute uncertainties.

Global CH_4 emissions are presented in Table 7. The results of this study are in fair agreement with the estimates of *Stern and Kaufmann* [1995]. For the years 1980 and 1990 the estimates of Stern and Kaufmann are higher than in this study. The sector emissions are different in a few cases: our estimates of energy-related emissions are 25% or higher throughout the period of study. This difference can be partly explained by the fact that biofuels are included in the energy sector, while they are not included in the Stern and Kaufmann study. Biomass-burning emissions are in good agreement for the period 1890–1960. From 1970 onward our estimate is about 30% lower. Emissions by animals are for most years comparable, with the exception of 1950 and 1960 (our numbers indicate a 10% higher emission) and 1990 for which we calculate a 15% lower emission by domestic ruminant. The emissions from agricultural land (mainly rice cultivation) calculated by Stern and Kaufmann are higher than our estimates with the

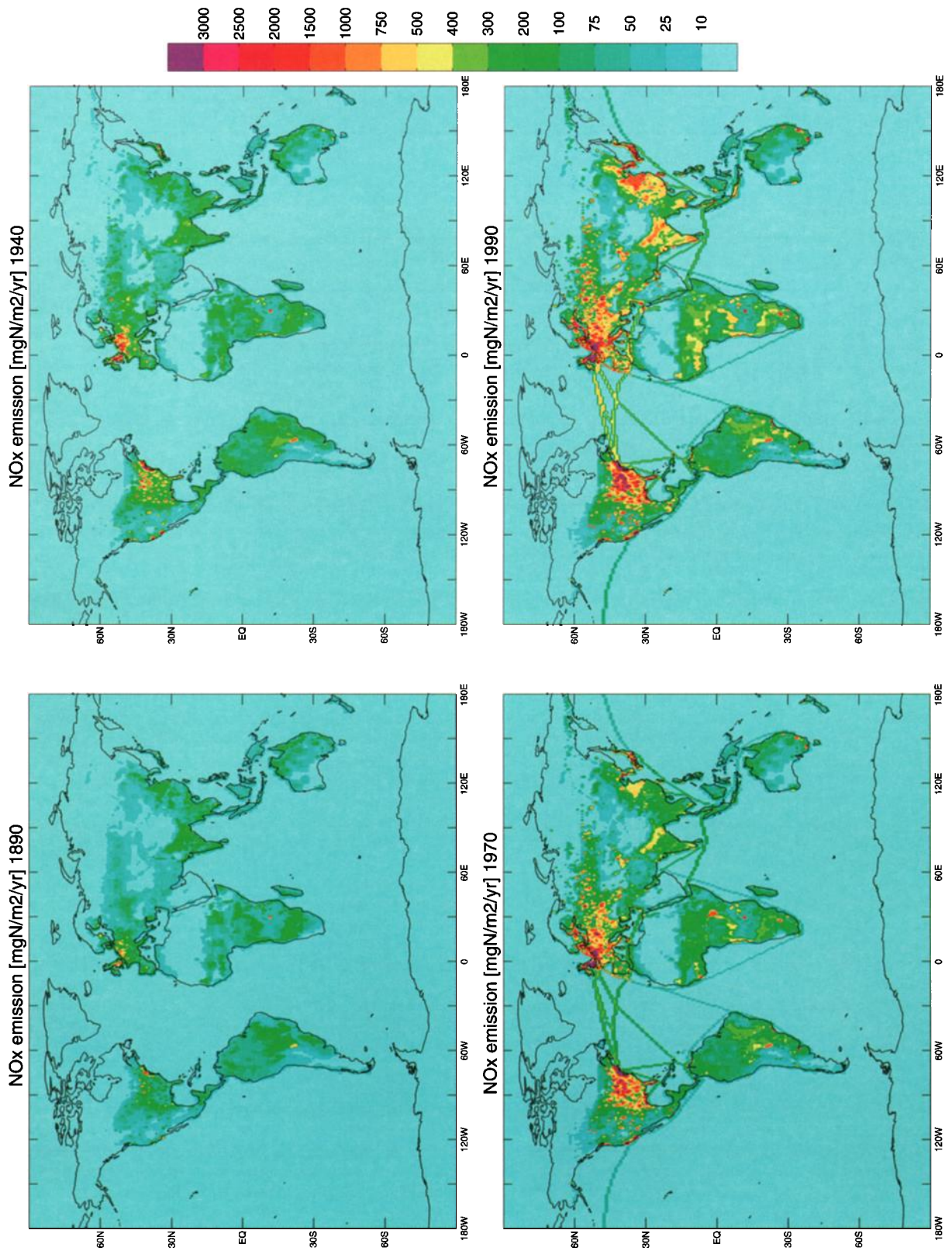

Plate 2. Spatial distribution of average NO_x emissions in (a) 1890, (b) 1940, (c) 1970, and (d) 1990. Note that emissions from ships prior to 1970 are represented through continental emissions

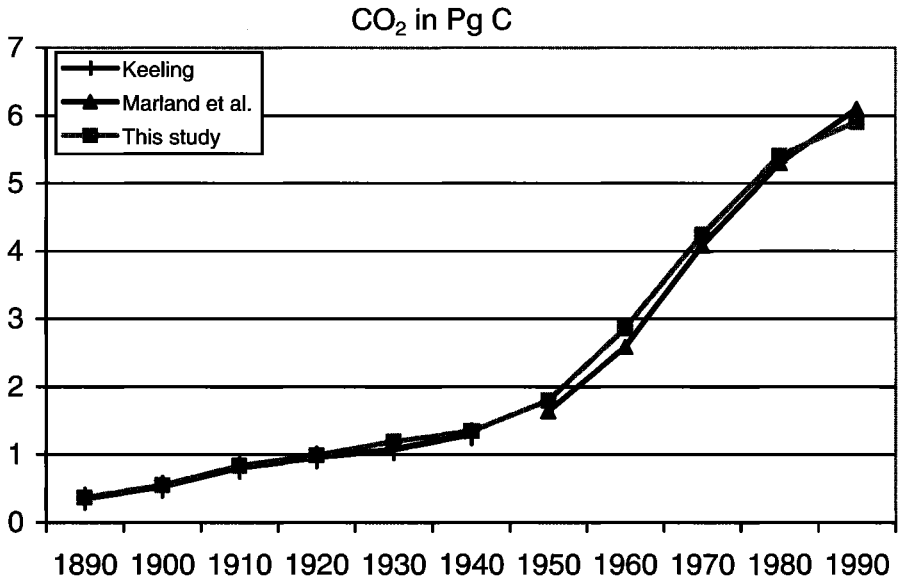

Figure 1. Fossil fuel and industrial CO_2 emissions from this study compared with *Keeling* [1994] and *Marland et al.* [1994].

difference increasing over the years from 20 to 40%. Finally, emissions from landfills are in good agreement.

We compared our SO_2 emissions with global estimates by *Stern and Kaufmann* [1996], *Örn et al.* [1996], *Lefohn et al.* [1996, 1999], *Möller* [1984] and *Ryaboshapko* [1983]. The results are presented in Figure 2. Figure 2 illustrates the apparently large uncertainty in the historical emission estimates. Furthermore, our global estimates are in the lower part of the range compared to previous studies. However, when looking at two regional studies, European SO_2 emissions according to *Mylona* [1996] and U.S. emissions from *Gschwandtner et al.* [1985], as presented in Figure 3, our emission estimates and trends are in rather close agreement. The year 1990 is an exception for which our estimates for the United States are significantly higher than the estimate by Gschwandtner et al., which is probably caused by differences in the assumed effectiveness of emission abatement. Note that Örn et al. use the Mylona and Gschwandtner et al. estimates for Europe and the United States, respectively. This indicates that regions other than Europe and the United States primarily cause the difference between our estimates and the other estimates. Discrepancies in emission estimates in these regions are probably caused by uncertainties in activity data and emission factors.

In Figure 4, global NO_x emissions for the years 1960, 1970, and 1980 are compared with *Hameed and Dignon* [1988]. Regional estimates for the United States [*Gschwandtner et al.*, 1985] are compared with our NO_x calculations for this region. Our global estimates for the years 1960, 1970, and 1980 are substantially lower, which might be related to differences in the methodology. Compared to the study by *Gschwandtner et al.* [1985], our estimates of the U.S. NO_x emissions are in close agreement for the period 1900–1960. For the period 1970–1990 our estimates are slightly different.

Comparison of our gridded historical emissions of N_2O, NH_3, and CO prior to 1970 was not possible because of a lack of comparable data sets. For comparison of the 1990 data we refer to *Bouwman et al.* [1995, 1997] and *Olivier et al.* [1996, 1999b].

4.2. Uncertainties

The results of this study are associated with significant uncertainties. Even for present-day emissions, estimates, based on relatively more reliable activity data and emissions factors, are sometimes highly uncertain (see, e.g., *Olivier et al.* [1999a, 1999b, and references therein] and for CO_2, *Marland et al.* [1999]). We refer to the various cited papers on $1° \times 1°$ grid emission studies for an extensive discussion of the variability of the emission factors, both within and between countries. Historical activity data, as presented in EDGAR and HYDE, are mostly based on studies using data from national or international statistics agencies. Although the quality of these data is difficult to assess, this information is probably the best available, with consistent source definitions across countries. Where no activity data are available, assumptions were made on processes leading to the activity or process developments influencing the activity. Evidently, this is an important source of uncertainty, also influencing the sector-weighted emission factors for fuel combustion and the historical spatial distribution of total emissions. Improvements in the activity data and, in particular, historical land use maps (e.g., biomass burning!) are needed and collaborations with other research disciplines (socioeconomic) could be helpful.

Using constant aggregated emission factors for the period 1890–1970 instead of representative emission factors for emitting processes in the past is, of course, a major source of uncertainty; however, it cannot be avoided in studying historic emissions. To our knowledge, verified emission factors prior to 1970 are hardly available; thus one can only make assumptions on the value of emission factors in the past. For example, many emission factors for fossil fuel combustion and industrial activities during the past 10–15 years (on which most emission factors in this study are based) were probably lower compared with technology used prior to 1970, in particular in the more industrialized countries. For fossil fuel combustion we tried to incorporate this by using globally uniform emission factors for combustion emissions in the pre-1970 period based on emission factors for 1990 in EDGAR 2.0 in regions without emission controls, reflecting the higher uncontrolled emission factors in less industrialized regions in 1990. *Olivier et al.* [1999a, 1999b] specified the regional average emission factors, as used for 1990 for the compounds considered here. For other activities, such as industrial processes and agriculture, this was not done as we did not have a ratio for selecting specific factors. Hence, for years prior to 1970, constant default

Table 7. CH_4 Emission Estimates From This Study Compared With *Stern and Kaufman* [1995][a]

	Energy, Tg C	Biomass Burning, Tg C	Animals, Tg C	Rice, Tg C	Landfills, Tg C	Total, Tg C
1890						
Stern and Kaufman	6	10	23	33	3	74
This study	13	8	20	27	3	71
1900						
Stern and Kaufman	9	10	25	35	3	82
This study	17	8	23	28	4	81
1910						
Stern and Kaufman	14	12	27	37	4	94
This study	23	9	24	28	4	89
1920						
Stern and Kaufman	16	11	29	38	5	99
This study	24	10	29	32	5	101
1930						
Stern and Kaufman	17	12	32	40	7	109
This study	26	11	32	33	7	109
1940						
Stern and Kaufman	20	12	35	43	9	118
This study	31	12	34	34	8	119
1950						
Stern and Kaufman	23	13	38	45	11	130
This study	32	13	42	35	11	133
1960						
Stern and Kaufman	33	18	46	51	15	162
This study	46	14	52	44	14	170
1970						
Stern and Kaufman	48	22	56	59	19	204
This study	55	16	57	45	18	191
1980						
Stern and Kaufman	56	23	67	67	24	238
This study	69	17	65	46	22	219
1990						
Stern and Kaufman	61	29	80	75	31	275
This study	82	18	69	44	26	239

[a] Biomass includes waste burning.

aggregated emission factors of the latter categories will probably lead to the underestimation of these emissions.

Another source of uncertainty is the interpolation of the emissions to a $1° \times 1°$ grid. The location of the release of pollutants to the atmosphere is dependent on the location of the activity leading to the emission. Since we used 1990 maps for population, etc., migration of people within a country, animals, and economic activities in the past are not taken into account in this study. Although this does not influence the total emissions flux on a country level, it causes additional uncertainties in the spatial emission distribution at $1° \times 1°$ resolution, which increases going back in time. A similar problem was faced by *Andres et al.*'s [1996, 1999] focusing on CO_2 emissions from fossil fuel use and cement production. The largest uncertainties may be expected in large countries with substantial internal migration (e.g., the United States) but may be less for, for example, northern Europe. To give an indication of the uncertainty in the activity data the emission factors and 1990 grid maps, a broad classification of data quality is applied (Table 8).

For activity data we use the quality ratings "acceptable," "poor," and "very poor." Data rated acceptable are based on national/international statistics or on publications where activity data are presented. Acceptable is applied here because this category of data is probably the best available at this moment as they are used and tested for many different applications. If data from national/international statistics or other publications were not sufficient for use over the entire period of study or for application to all countries or regions, we used extrapolations or assumptions based on these data to estimate these activities. This category of activity data is rated poor. For activities where no data at all were available the scaling of known information was a means to achieve an estimate of the activity in the past. The rating very poor is applied to these data.

For emission factors we used three types of emission factors: "detailed" emission factors, "regional" aggregated emission factors, and "global" averaged emission factors. Detailed emission factors are country-specific emission factors as used in the EDGAR 2.0 database. Regional aggregated emission factors are based on aggregation of detailed emission factors to a certain region-specific emission factor or on studies where only regional-specific emission factors are presented. Global emission factors are the result of aggregation of detailed and/or regional emission factors to global emission factors or are used because only one emission factor for a certain sector is known. The

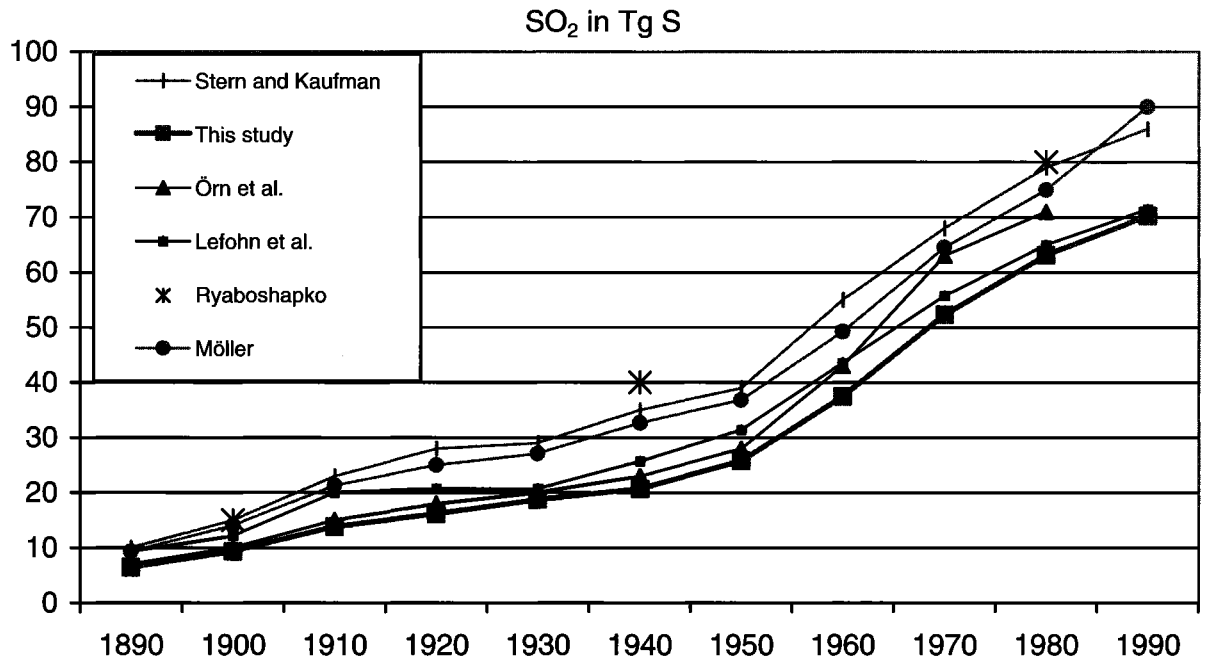

Figure 2. Comparing our estimated global SO_2 emissions with *Stern and Kaufmann* [1996], *Örn et al.* [1996], *Lefohn et al.* [1996], *Ryaboshapko* [1983], and *Möller* [1984].

nature of the inherent uncertainties associated with emission factors, the variability of the 1990 emission factor figures over different countries, and the increased uncertainty from applying 1990 emission factor figures for the more distant past result in a qualitative picture of uncertainty varying from source to source in 1990 and in all cases increasing when going farther back in the past.

The quality of spatial distributions using the 1990 grid maps for the whole 100 year period could generally be classified as acceptable or poor except for a few sources (Table 8). This refers both to the quality (applicability) of the grid maps used for distribution of 1990 emissions and to the facts that spatial distributions of emission sources within a country shift in time (e.g., because of urbanization) and that we have neglected this feature by applying

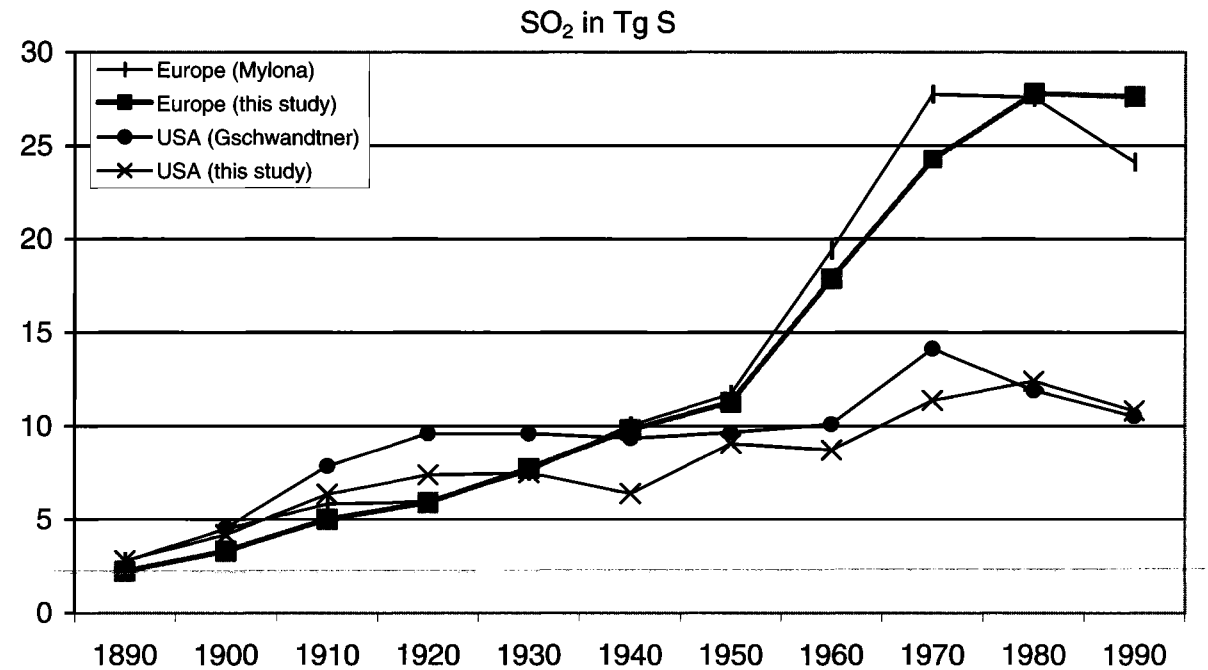

Figure 3. Comparing our estimated European SO_2 emissions with *Mylona* [1996] and U.S. emissions with *Gschwandtner et al.* [1985].

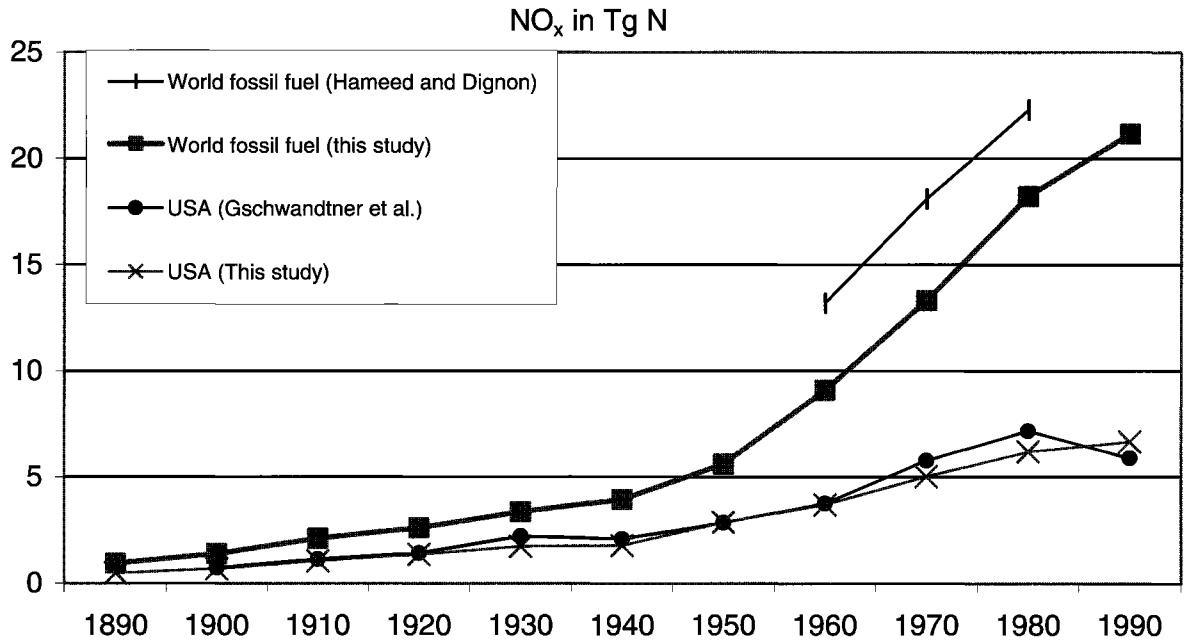

Figure 4. Comparing our estimated global NO_x emissions with *Hameed and Dignon* [1988] and U.S. emissions with *Gschwandtner et al.* [1985].

the same 1° × 1° distribution for maps for the whole 100 year period.

In particular, for biomass burning due to the simplified approach, and also for point sources like power plants and industrial process locations, we note the important large uncertainties associated with the methods used. Nevertheless, these uncertainties of the within-country distribution are partly compensated by the presumably more accurate emission estimates at the country total level, which are, in fact, the first step in locating emissions on the world map. The highest uncertainties can therefore be expected in the largest countries as well as in the gridded emissions of biomass burning.

This evaluation indicates that although the uncertainty in the gridded inventories for the distant past may be considerable at the 1° × 1° resolution (also because shifts in boundaries between countries have not been considered), it is likely that the uncertainty in the spatial distribution is smaller when the aggregated data are used at a lower resolution, for example, in models with a 5° × 5° grid spacing. The result of this data quality rating is presented in Table 8 and provides an impression of the uncertainty in the emission estimates. Very uncertain emission estimates are the result of applying very poor activity data with global aggregated emission factors. Probably the best emission estimates are based on acceptable activity data and detailed emission factors. This qualitative discussion will be used to prioritize improvements on the historical emission data. Unfortunately, we are at this point not able to give a more quantitative statement on data quality, although this appears to be very important for, for example, inverse modeling studies [e.g., *Houweling et al.*, 1999].

5. Concluding Remarks and Recommendations for Future Research

We constructed a data set of historical anthropogenic trace gas emissions for the period 1890–1990, which can be useful in trend studies of tropospheric trace gases [e.g., *Lelieveld and Dentener*, 2000; *Houweling et al.*, 1999] and also in environmental assessments. Examples are the analyses of historical contributions of regions and countries for environmental problems such as the enhanced greenhouse effect, acidification,and eutrophication. In general, the global and regional emission trend estimates show a rather good agreement with previous studies. Nevertheless, some differences in the total emission fluxes are evident, although there is no absolute reference to (in)validate the different estimates. One advantage of our data set is that greenhouse and reactive trace gas emission estimates are calculated consistently using the EDGAR system so that future improvements concerning activities and emission factors can be easily implemented. Validation and further development of the emissions inventory will rely to a large degree on the use of the data in global models and comparison of the results with field observations.

We started with 1990 data, which are relatively robust, focusing on energy/industry and including biomass burning for completeness in a simple, transparent way, and worked back in time with activity data and emission coefficients, realizing that the data quality of all aspects considered (statistics, emission factors, and country definition/spatial distribution) becomes increasingly uncertain when going farther back in time. Thus the results should be viewed as a first attempt to estimate historical emissions for a time span of 100 years in a spatial explicit way using a consistent approach, i.e., taking into account the relative uncertainties for various source categories in both present and past emission inventories. In section 4.2 several elements were identified that could be recommended for improvement of the data set: (1) improvement of estimates of uncontrolled emissions factors for 1970 for many trace gases, if possible region-specific, to reflect better the average age and maintenance levels of applied technologies; (2) inclusion of trends in emission factors for the period 1890–1970 in cases where significant shifts can be expected, for example, in the type of coal mining (surface versus underground) and rice production regimes (irrigated versus other) and in autonomous trends in emission factors, for example, for road transport; (3) improvement in procedures to estimate activity data, notably fuel consumption and fuel mix per key economic sector and the amount of biomass burning in all regions; (4) inclusion of separate data sets for air traffic and international ship traffic; (5) improve-

Table 8. Qualitative Analysis of Activity Data, Emission Factors and 1990 Grid Maps Used to Calculate Historical Emissions

Emission Source	Activity Data (Quality[a])	Emission Factors[b]								1990 Grid Map[c]	
		CO_2	CO	CH_4	NMVOC	SO_2	N_2O	NO_x	NH_3	Type	Quality
					Fossil Fuel Combustion						
Power plants	1890–1920: scaling (3)	G	G	G	G	G	G	G	···	pop	3
	1930–1960: literature, assumptions (2)										2
	1970–1990: detailed international statistics (1)										1
Domestic	as above	G	G	G	G	G, R for coal	G	G	···	pop	1
Industry	as above	G	G	G	G	G	G	G	G	pop	2
Transport	as above	G	G	G	G	G	G	G	G	pop	1
					Fossil Fuel Production						
Brown coal + hard coal	international statistics + assumptions (2)	···	···	G	···	···	···	···	···	ps	1
Oil	international statistics (1)	D	···	G	G	···	···	···	···	ps	1
Gas	international statistics (1)	···	···	G	···	···	···	···	···	ps	1
					Biofuel Combustion						
Industry + domestic	scaling of 1990 data (3)	G	G	G	G	G	G	G	G	pop	2
					Industrial Processes						
Iron	international statistics (1)	···	G	G	G	R	···	···	···	pop	2
Steel	international statistics (1)	···	R	···	R	···	···	R	···	pop	2
Copper	international statistics (1)	···	···	···	···	R	···	···	···	ps	1
Nitric acid	scaling of 1990 detailed data (2)	···	···	···	···	···	G	···	G	ps/pop	2
Adipic acid	scaling of 1990 detailed data (1)	···	···	···	···	···	G, R: United States, Canada	···	···	ps	1
Cement	1890–1960: literature (3) 1970–1990: international statistics (1)	G	···	···	···	···	···	···	···	pop	3
Solvents	scaling of 1990 data (3)	···	···	···	G	···	···	···	···	pop	2
					Agriculture						
Rice	international statistics (1)	···	···	G: 1890–1970 R: 1970–1990	···	···	···	···	···	s-s	2
Fertilizer	pre-1960: scaling (3) 1960–1990: international statistics (1)	···	···	···	···	···	G	G	G	s-s	1
Animal types	1890–1960: literature (1) 1970–1990: international statistics (1)	···	···	G, R	···	···	G,R	···	G,R	s-s	1
					Biomass Burning						
Savannah burning	educated guess (3)	···	G	G	G	G	G	G	G	s-s	2
Deforestation	scaling of 1990 data (3)	G	G	G	G	G	G	G	G	s-s	3
					Waste						
Agricultural waste burning	scaling of 1990 data (3)	···	G	G	G	G	G	G	G	s-s	1
Landfills	scaling of 1990 data (3)	···	···	R	···	···	···	···	···	s-s	2

[a] Quality ranking: 1, acceptable; 2, poor; and 3, very poor.
[b] Emission factors: G, global aggregated; R, regional aggregated; and D, detailed (country-specific).
[c] Map type: pop, population as surrogate; ps, point sources; and s-s, source-specific.

ment of grid maps for older historical years, in particular, for key maps like population, for example, by taking into account shifts due to ongoing urbanization, and like large-scale biomass burning. However, the results of the present study can be used as an a priori emission data set for atmospheric models that investigate the effect of long-term trends in the emissions of trace gases and require global emissions with an explicit spatial distribution at a model resolution lower than or equal to 1° × 1°. This database is available to the scientific community. Interested readers can contact J. van Aardenne for information on the construction of the data set and J. Olivier for distribution of the data set.

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