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Global gridded inventories of anthropogenic emissions of sulfur and nitrogen

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Abstract. Two sets of global inventories of anthropogenic emissions of both oxides of sulfur and oxides of nitrogen for circa 1985 have been produced under the umbrella of the Global Emissions Inventory Activity (GEIA) of the International Global Atmospheric Chemistry Program. The two sets of inventories have different temporal, sectoral, and vertical resolution. Both were compiled using the same data sets; default data sets of global emissions have been refined via the use of more detailed regional data sets. This article reports on the compilation of the annual, one-vertical-level inventories, called version 1A; the inventory files are available to the scientific community via anonymous file transform protocol (FTP). Existing global inventories and regional inventories have been updated and combined on a 1° x 1° longitude/latitude grid. The resulting global anthropogenic emissions are 65 Tg S yr^{-1} and 21 Tg N yr^{-1}; qualitative uncertainty estimates have been assigned on a regional basis. Emissions of both SO_x and NO_x are strongly localized in the highly populated and industrialized areas of eastern North America and across Europe; other smaller regions of large emissions are associated with densely populated areas with developed industries or in connection with exploitation of fuels or mineral reserves. The molar ratio of nitrogen to sulfur emissions reflects the overall character of sources; its value is generally between 0.33 and 10 for industrialized and heavily populated areas but varies over a wide range for other areas. We suggest that those requiring sulfur or nitrogen emission inventories standardize on the GEIA inventories, which we believe are authoritative and which are freely available to all users by anonymous FTP.

1. Introduction

One of the most important scientific tools used in the assessment of atmospheric chemistry, air quality, and climatic conditions of the past, present and future, is the mathematical modeling of transport and transformation in the atmosphere. Such models rely in part on inventories of emissions of the pertinent species, constructed on appropriate temporal and spatial scales, and including the required chemical species. The production and evaluation of these inventories is an area of research whose importance to the accuracy of modeling results and assessment activities has come to be fully recognized.

For the oxides of sulfur (SO_x, SO_2+sulfate) and nitrogen (NO_x, $NO+NO_2$), efforts to estimate current emissions on a global scale were begun in the 1970s by *Robinson and Robbins* [1970a,b], who used fossil fuel combustion statistics and average emission factors to produce estimates of global emissions of SO_x and NO_x. Similar studies using improved data have continued since that time. As shown in Table 1a, estimates of annual anthropogenic emissions of SO_x for the same base year can differ by as much as 50%. In contrast, Table 1b shows that except in a single study [*Crutzen*, 1983] which presented a range for the NO_x emissions, global annual emissions for anthropogenic NO_x have differed by only about 15%. However, no estimates of the uncertainty associated with these emission estimates are available in the inventories.

Total global emission estimates are limited in their utility since (1) the environmental issues associated with SO_x and NO_x are partly regional in scale because of the short atmospheric lifetime of these species, and (2) the relationships of emissions to environmental effects (atmospheric concentrations, impacts on ecological systems) are nonlinear [*Liu et al.*, 1987; *Misra et al.*, 1989]. Atmospheric models used to study the transport, transformation, and removal of sulfur and nitrogen species are increasingly becoming three dimensional in structure; examples of these models are given by *Logan et al.* [1981], *Seinfeld* [1988], *Berge* [1990], *Sillman et al.* [1990], *Penner et al.* [1991], *Benkovitz et al.* [1994], and *Taylor and Penner* [1994]. In addition, policy considerations relating to emissions require information on a political entity basis. Clearly, therefore, gridded global inventories are essential from both management and scientific perspective. However, the generation of such inventories is complicated by many questions such as: Are one country's statistics comparable with another's? How can emissions from sources with pollution control devices be

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Table 1a. Estimates Of Total Global Anthropogenic Emissions of SO_x in Previous Work

Inventory Year	Emissions, Tg S yr^{-1}	Gridded	Reference
1960	46	No	[*Dignon and Hameed, 1989*]
1965	55	No	[*Möller, 1984*]
1965	65	No	[*Granat et al., 1976*]
1965	74	No	[*Cullis and Hirschler, 1980*]
1970	50	No	[*Kellogg et al., 1972*]
1970	56	No	[*Dignon and Hameed, 1989*]
1970	62	No	[*Möller, 1984*]
1970	64	No	[*Várhelyi, 1985*]
1970	73	No	[*Robinson and Robbins, 1970*]
1970	86	No	[*Cullis and Hirschler, 1980*]
1974	94	No	[*Cullis and Hirschler, 1980*]
1975	69	No	[*Möller, 1984*]
1975	94	No	[*Cullis and Hirschler, 1980*]
1976	104	No	[*Cullis and Hirschler, 1980*]
1979	64	No	[*Várhelyi, 1985*]
1980	57	5°x5°	[*Hameed and Dignon, 1988*]
1980	63	No	[*Dignon and Hameed, 1989*]
1980	78	1°x1°	[*Spiro et al., 1992*]
1980	88	No	[*Ryaboshapko, 1983*]
1985	64[b]	1°x1°	[*Dignon, 1992*]
1985[a]	90	No[c]	[*Möller, 1984*]
1986	92	5°x5°	[*Müller, 1992*]

[a] Projected.
[b] This inventory was later gridded by *Langner and Rodhe* [1991].
[c] Includes fossil fuel combustion only.

accurately related to emissions from sources without such devices? How should emissions from international voyages by ships and aircraft be estimated, assigned to specific countries, and distributed geographically? What information is available on the height at which pollutants are released? Despite recognition of these complexities, researchers have been constrained to use a variety of country-based statistics to allocate emissions onto a global grid (see Table 1). These exercises have generally relied on fuel use, population, or other surrogates to geographically distribute the emissions. A number of countries (United States, Canada, Australia, Japan, South Africa) and regions (western Europe, eastern Europe, parts of Asia) have developed their own detailed emission inventories, including spatial identification of large sources and improved surrogate statistics for spatial apportionment of diffuse sources [*Graedel et al.*, 1993]; where available, these inventories are much more reliable than those generated using surrogates.

In the present work we report on an international project to use gridded and updated regional and national inventories and surrogate information to compile improved SO_x and NO_x global inventories on a 1° x 1° longitude/latitude global grid with no vertical resolution. The results are intended to serve as a basis for modeling and related activities by the international scientific community.

2. Time Trends of Anthropogenic Emissions of SO_x and NO_x

Several estimates of historical emissions of SO_x and NO_x are available in the literature. *Dignon and Hameed* [1989] have estimated global emissions from 1860 to 1980; the emissions of both species have steadily increased over this time period. These increases are also shown in the estimates by *Möller* [1984]. Estimates for the United States from 1900 to 1980 made by *Gschwandtner et al.* [1986] show SO_x emissions peaking around 1970 and decreasing approximately 10% from 1970 to 1980, while NO_x emissions increased throught the period. Similar patterns for the United States were estimated by *Husar* [1986]. Estimates of SO_x emissions for Europe from 1880 to 1990 made by *Mylona* [1993] show steady increases until 1940, a small decrease during the World War II years, a sharp increase from 1950 to 1980, and a decrease after this year.

The emission estimates developed in this work represent conditions circa 1985. Since 1985, emissions of SO_x have decreased in certain regions of the world and increased in others. SO_x emissions in the United States have decreased about 5% since 1985 [*U.S. Environmental Protection Agency (EPA)*, 1994]. The collapse of the political and economic system in eastern Europe has caused a decrease of about 25% in sulfur emissions in this region for the period 1990 to 1993 [*Tuovinen et al.*, 1994], and emissions from this region are going to be reduced further in accordance with the Oslo protocol on sulfur emissions reduction, signed in June 1994 [*United Nations Economic Commission for Europe (ECE)*, 1995]. Flue gas cleaning in coal-fired power plants, substitution with low sulfur fuels such as natural gas, and energy conservation are the most important measures being applied. Data gathered by the *UN ECE* [1991] shows that the installation of flue gas cleaning equipment alone is responsible for a decrease of about 10% in SO_x emissions in Europe and substantial reductions in the United States and Japan. In contrast, an increase of about 10% in SO_x emissions has been reported in China since 1985 [*Kato and Akimoto*, 1992]; this increase has been related to the raising energy demands in this country, specially for electricity.

Emissions of NO_x in the United States [*U.S. Environmental Protection Agency (EPA)*, 1994] have remained fairly constant over the period 1985 to 1993. Emissions of NO_x in Europe [*Tuovinen et al.*, 1994] remained fairly constant over the period 1985 to 1990; the collapse of the political and economic system in eastern Europe has caused a decrease of about 20% in nitrogen emissions in this region for the period 1990 to 1993. Control measures for nitrogen emissions have been installed, notably for coal-fired power plants and in a form of three-way catalyst control for automobiles. However, these control measures have been largely offset by the increase in the traffic

Table 1b. Estimates of Total Global Anthropogenic Emissions of NO_x in Previous Work

Inventory Year	Emissions, Tg S yr^{-1}	Gridded	Reference
1960	12	No	[*Dignon and Hameed, 1989*]
1970	18	No	[*Dignon and Hameed, 1989*]
1970	19	No	[*Soderlund and Svensson, 1976*]
1970	20	No	[*Robinson and Robbins, 1970*]
1979	21	No	[*Logan, 1983*]
1980	12 to 20	No	[*Crutzen, 1983*]
1980	20	5°x5°	[*Hameed and Dignon, 1988*]
1980	21	2.4°x2.4°	[*Levy and Moxim, 1989*]
1985	23[a]	1°x1°	[*Dignon, 1992*]
1986	22	5°x5°	[*Müller, 1992*]

[a] All inventories compiled by Hameed and Dignon or by Dignon include fossil fuel combustion only.

volume, which in Europe also includes a substantial increase in the transport of goods by diesel-powered trucks and lorries.

3. Compilation of the GEIA Inventories of Anthropogenic Emissions of SO_x and NO_x

The results reported in this paper were produced under the umbrella of the Global Emissions Inventory Activity (GEIA), a component of the International Global Atmospheric Chemistry Program (IGAC) [*Benkovitz and Graedel*, 1992; *Galbally*, 1989]. Two GEIA study groups have compiled global inventories of both SO_x and NO_x. One set of these inventories, named version 1A, presents estimates of global, annual emissions using a single vertical level. A second set of these inventories, named version 1B, presents estimates of global emissions by season, using two-level vertical levels (surface and > 100 m), and with sectoral (industrial sectors, for example electricity generation, metal smelters, transportation) resolution. Inventories were assembled in an incremental mode; default global inventories were selected and emissions for a particular geographic area were evaluated and substituted if the data proved to be more accurate and complete. Work on compiling the two sets of inventories proceded in parallel. This article addresses the compilation of the gridded inventories of annual emissions; a complementary article addressing the compilation of seasonal inventories with two vertical levels and sectoral resolution is in preparation [*Voldner et al.*, 1995].

3.1. Supporting Inventories

On a global basis, information on anthropogenic emissions of SO_x and NO_x is consistently available based only on political units, such as countries, states, or provinces. To allocate these emissions to a gridded inventory, surrogate data available at the desired resolution must be used. Ideally, to assign emissions based on political units to a geographically based grid, emissions from major industrial installations would be assigned to their known location, and emissions from distributed sources would be assigned via the use of the most appropriate surrogate indicator (such as population, housing, minor industrial centers) available with the desired spatial resolution. Unfortunately, information on the location of major industrial installations is not available for all countries, so surrogates must be used to distribute the emissions from countries lacking such information.

The combustion of fossil fuels has been identified as the major activity contributing to emissions of these two species [*Cullis and Hirschler*, 1980; *Logan*, 1983; *Saeger et al.*, 1989; *Várhelyi*, 1985; *Wagner et al.*, 1986], with electricity generation being the sector generating the major fraction of the SO_x emissions and electricity generation and mobile sources generating the major fraction of NO_x emissions. Currently, population is the only surrogate available on a global basis with the spatial resolution needed by the GEIA inventories; therefore population has been selected as the surrogate indicator for the geographic distribution of SO_x and NO_x emissions. Use of population as a surrogate to distribute SO_x and NO_x emissions is more accurate for the transportation sector than for the electricity-generating sector, because in some countries (such as the United States, and Canada) the locations of large power plants do not coincide with the population centers they serve. However, most of these countries have generated their own gridded inventories which were used in the work described here.

Global population density maps for 1985 were prepared on a 1° x 1° longitude/latitude grid by Logan (J. Logan, Harvard University, manuscript in preparation, 1995). The population database was developed using statistical data on several levels: national, state/province/other, and urban. National populations were taken from the *United Nations* [1987] Demographic Yearbook; the seven largest countries (Australia, Brazil, Canada, China, India, the former Soviet Union, and the United States) were subdivided into their major political entities, and populations for the subdivisions were taken from the Encyclopedia Brittanica (1987). The urban fraction of the population in each country was taken from the same two sources. *Rand McNally* [1987] provided a data file giving the population and location of cities with over 50,000 inhabitants. Since the urban population calculated from the city listing was smaller than that calculated from the urban fraction for individual countries, the city populations were scaled to give the correct urban fractions. The urban population in each country or state was then assigned in the appropriate 1° x 1° grid boxes using the scaled city populations. The remaining population was distributed over the habitable areas of each country, using a digital database developed at the NASA Goddard Institute for Space Studies [*Lerner et al.*, 1988] for the purpose of locating animals. In this database each land 1° x 1° cell is labeled with one country code, chosen as the country which covers the largest fraction of the area of the cell, and assigned to use either by animals, lumbering, ice, or unused. The rural population was located in the cells designated as animal use and lumbering; in a few regions it was necessary to allow certain cells designated as "unused" to include population, based on published population maps, e.g., Times World Atlas (1990). This gridded population file has been chosen as a standard GEIA supporting inventory.

3.2. Annual Inventories of Anthropogenic Emissions of SO_x and NO_x

Combustion of fossil fuels is the major source of emissions of SO_x and NO_x, although the major emitting sectors are different [*Cullis and Hirschler*, 1980; *Logan*, 1983; *Saeger et al.*, 1989; *Várhelyi*, 1985; *Wagner et al.*, 1986]. Coal combustion, specially in large electric and heat power plants and combustion of residential fuel oil are responsible for most of the SO_x emissions, whereas mobile sources are the largest contributors to the NO_x emissions.

The sulfur content of fuels, specially in coal, is a major factor affecting SO_x emissions. This content is extremely variable, ranging from less than 1% to about 9%. The energy content is also variable, ranging from 30 MJ kg^{-1} for most of the bituminous and subbituminous coals to less than 12 MJ kg^{-1} for lignites. This means that the amount of SO_x emitted is related not only to energy consumption but also to the sulfur content and the quality of the fuel used. On average, from 5 to 15% of the sulfur in fuels used in electric- and heat-generating power plants is retained in bottom and fly ashes. Finally, various types of flue gas desulfurization (FGD) control devices are used to remove SO_x from the exhaust gases, predominantly in power plants in Australia, Canada, Japan, western Europe, and the United States.

The sulfur content of crude petroleum is also extremely variable, ranging from 1 to 2% in crudes from fields in Russia and the Middle East, to less than 0.3% in crudes from fields in the North Sea, and even lower in some crude oils from African fields. In the refining process, most of the sulfur in the crude oil may be recovered; what is not recovered remains mainly in the residual oil fraction. Hence a relatively large fraction of the SO_x is emitted from the combustion of residual oil [*Saeger et al.*, 1989; *Wagner et al.*, 1986].

The processing of sulfur-containing materials (e.g., roasting of ores, sintering plants), the production of sulfur compounds (e.g., sulfuric acid manufacturing), and the use of sulfur compounds to produce other industrial goods (e.g., cellulose production) generate large amounts of SO_x. Although most of this sulfur may be recovered, not all installations do so; the sulfur that is not recovered is released to the atmosphere. For example, in the United States where sulfur recovery is prevalent, SO_x emissions from industrial processes comprise approximately 13% of this country's total emissions [*Saeger et al.*, 1989].

Emissions of NO_x tend to be more dependent on combustion conditions, such as the size and design of furnace or engine, than on the type of fuel used. This applies specially to internal combustion engines burning gasoline or diesel oil. The high combustion temperature and long residence time of the combustion product favor the formation of NO_x from the nitrogen in the combustion air. This results in very large NO_x emissions from mobile sources, which are generally responsible for about one half of the anthropogenic emissions of NO_x in the United States [*Saeger et al.*, 1989] and Europe [*Sandnes and Styve*, 1992].

The processes generating NO_x emissions from combustion in stationary sources are also complicated and depend on the nitrogen content of the fuel, the boiler size and design, and operation-related factors (e.g., load and excess air conditions). Empirical emission factors based on measured concentrations in exhaust gases under representative conditions are commonly used to determine the NO_x emissions from stationary sources. A large fraction of the emissions used in this work were estimated based on these types of emission factors.

The GEIA inventories were compiled by selecting a global SO_x and a global No_x inventory as the "default" inventories; data available in more accurate and detailed regional inventories were used to provide more accurate estimates of emissions in the areas covered. These regional inventories covered the United States and Canada, Europe west of the Ural Mountains, major stationary sources in the Asian part of the former USSR, Australia, South Africa, and 25 countries in Asia from Afghanistan and Pakistan to the Pacific Ocean. The following sections describe each inventory that was used; diagrams of the relation of the regional and default inventories are presented in Figure 1.

3.2.1. Default global inventories. 1985 has been chosen as the base year for version 1 of the GEIA inventories of SO_x and NO_x. The global inventory compiled by *Spiro et al.* [1992] for 1980 was updated to 1985 using country specific 1985 statistics for fossil fuel use [*United Nations*, 1989], metal production [*United Nations*, 1988], and sulfur recovery [*U.S. Department of the Interior*, 1988]. This inventory was selected as the default GEIA inventory for SO_x emissions. The global inventory of emissions from fossil fuel combustion developed by *Dignon* [1992] was selected as the default GEIA inventory for NO_x emissions.

The default GEIA inventories estimated SO_x and NO_x emissions using top-down methodologies, where statistics used

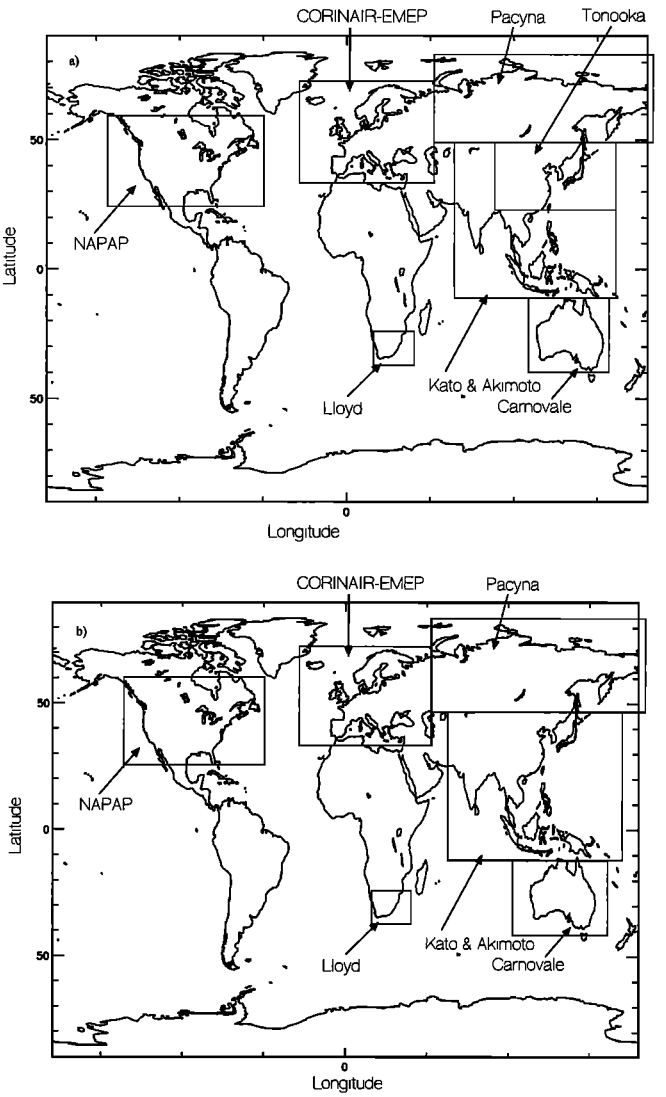


Figure 1. Regional inventories overlaid on (a) the default SO_x global inventory and (b) the default NO_x global inventory.

were assembled for larger geographic units and then distributed the corresponding smaller geographic units based on surrogates [*Benkovitz and Graedel*, 1992]. The SO_x emissions from fossil fuel use and smelting operations were estimated using the sulfur content of the fuel and activity rates (for example, the amount of fuel burned, the quantity of copper produced) at the country level [*Spiro et al.*, 1992]. SO_x emissions from smelter operations within each country were distributed spatially among major smelters in the country based on information available from economic atlases [*Spiro et al.*, 1992]. SO_x emissions from other sources were apportioned to the 1° x 1° grid based on the Logan-gridded population file. The NO_x emissions from fossil fuel combustion were estimated using a linear relationship between total fuel use by country and emissions [*Dignon*, 1992]. All NO_x emissions were apportioned to the 1° x 1° grid based on the Logan-gridded population file (J. Logan, Harvard University, manuscript in preparation, 1995).

3.2.2. United States and Canada. To date, the most complete and detailed inventories of SO_x and NO_x emissions circa 1985 for the United States and Canada to 60°N have been compiled by the National Acid Precipitation Assessment Program (NAPAP) [*Saeger et al.*, 1989]. Emissions from version 2 of the NAPAP base year 1985 inventories were

selected to replace values in the default GEIA inventories for the United States and Canada.

The United States and Canadian inventories were compiled using bottom-up methodologies (statistics are compiled for the smallest geographic units and aggregated to the larger geographic units) for large stationary sources and top-down methodologies for small stationary and mobile sources [*Saeger et al.*, 1989]. In these inventories, large stationary sources were defined as sources emitting at least "100 tons" per year of SO_x (45.4 Mg S yr^{-1}), NO_x (27.6 Mg N yr^{-1}), volatile organic compounds (VOC) (68.1 Mg C yr^{-1}), total suspended particles (90.8 Mg yr^{-1}) or carbon monoxide (38.9 Mg C yr^{-1}); detailed emissions and facility data were obtained for these sources. Emissions for smaller stationary (residential, commercial) sources and mobile sources were estimated on the county level and were calculated by multiplying the activity rate for each category by the appropriate emission factor. Activity rate data on the county level for the United States were taken from county statistics, or estimated from state statistics based on surrogate county statistics such as population and employment figures. Activity rate data for Canada were generally available from surveys compiled annually by Statistics Canada on a provincial basis.

Annual values for stationary sources were directly allocated to the GEIA 1° x 1° grid based on the coordinates of each source. County-based area sources were first allocated to the NAPAP 1/6° x 1/4° longitude/latitude grid using the population surrogate file developed by NAPAP [*Modica and Dulleba*, 1990]; the NAPAP grid was then aggregated to the GEIA 1° x 1° grid.

3.2.3. Europe. The most comprehensive inventories of European SO_x and NO_x emissions have been assembled by the Cooperative Program for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe (EMEP) [*Eliassen and Saltbones*, 1983], and the CORINAIR project of the Coordination d'Information Environnementale (CORINE) Programme established by the Commission of the European Communities (CEC) [*Bouscaren*, 1992]. Emissions from both these inventories were used for the European emissions in the GEIA inventory.

The EMEP Program is an activity of the 1979 Convention on Long-Range Transboundary Air Pollution (1995), a treaty under the auspices of the United Nations Economic Commission for Europe (UN/ECE). The EMEP inventories employ official annual emission values submitted by the countries participating in the UN/ECE convention. Reporting requirements are based on provisions in the convention and its related protocols on the reduction of sulfur emissions (1985), emissions of nitrogen oxides (1988), and emissions of volatile organic compounds (1991). The reporting procedures are defined in the Guidelines for Estimation and Reporting of Emission Data for SO_x, NO_x, NMVOCs, CH_4, NH_3, and CO in the UN/ECE region [*Pacyna and Joerss*, 1991].

The EMEP inventories currently use a polar stereographic grid of 50 x 50 km^2 actual area at 60° latitude; a 150 x 150 km^2 grid size was in use when the present work was ongoing. The 50 x 50 km^2 grid corresponds to a resolution of less than 0.5° x 0.5°; the 150 x 150 km^2 grid corresponds to a resolution of 1.5° x 1.5°. These inventories are continually updated as new information is provided by participating countries. For the GEIA 1985 inventories, emissions from the 1992 release of the EMEP inventory were used [*Sandnes and Styve*, 1992]. In this version, 77% of the member countries of the convention had reported gridded SO_x inventories for at least 1 year. For NO_x, 65% of the member countries had reported gridded emissions for at least 1 year. The procedure for updating the EMEP database generally involves scaling of the national grid distributions using the annual national totals. When no official annual totals exist, interpolation or extrapolation from known yearly values is applied [*Sandnes and Styve*, 1992]. For countries that have only submitted annual emission totals, the grid distribution is determined at the EMEP centers on the basis of available information on activity centers, population files, national statistics on activity rates, and using standard emission factors.

The overall quality of the EMEP inventory is very dependent on the reliability of the methodologies developed by each individual participant country. Although it is reasonable to expect that each country is best qualified to assess its own emission values, experience has shown that the various national activity statistics and emission inventories have evolved to meet the individual country's local needs and perspectives and that those are not necessarily the same for all member countries. Aware of this problem, the Executive Body for the Convention on Long-Range Transboundary Air pollution agreed in 1991 to the formation of a UN/ECE Task Force on Emission Inventories whose main purpose is to meet this need for collaboration, evaluation, and harmonization (i.e., insuring that the statistics and methodologies used are compatible across countries). The main objective of the task force is to prepare the atmospheric emission inventory guidebook, which should contribute to the improvement of the quality of emission data in the UN/ECE region (Europe, the United States, and Canada).

The CORINAIR project started in 1986 with the major objective to compile a coordinated inventory of 1985 atmospheric emissions, called CORINAIR85, from the 12 member states of the Commission of the European Communities (CECs): Belgium, Denmark, France, Germany, Greece, Ireland, Italy, Luxembourg, Netherlands, Portugal, Spain, and the United Kingdom. The project developed a source sector nomenclature, a handbook of default emission factors, and a computer software package for data input and the calculation of regional, sectoral, and national emissions estimates. Following completion of the CORINAIR85 inventory in 1990, the CORINAIR Group, consisting of project team and national experts, was continued to start inventories for 1990, called CORINAIR90. By this time links between the CORINAIR Group and the UN ECE/EMEP had been established, and a common source sector split had been developed. The CORINAIR90 inventories will be completed by the end of 1995. At present, as many as 19 countries are fully or partially using the CORINAIR methodology, which is available to any country [*McInnes*, 1994].

For the purposes of the GEIA inventories the CORINAIR85 data were provided in gridded format on the 1° x 1° GEIA grid, with sectoral and stack height information for major stationary sources. As previously described, the EMEP inventories were compiled and provided on a 150 x 150 km^2 polar stereographic grid; major stationary sources were not explicitly given but were grouped into a total emission from all sources discharging above 100 m within each grid cell. The domains covered by these two inventories overlap, and in view of the major stationary source detail provided by CORINAIR85, emissions from this inventory were used for the 12 CEC countries.

The EMEP inventories were transformed from areas on the polar stereographic grid to the 1° x 1° GEIA longitude/latitude grid by allocating emissions on the EMEP grid to the GEIA grid according to the fraction of the EMEP grid area which falls into each of the 1° x 1° grid cells. Since emissions in each country are assigned on a gridded basis, a portion of the emissions in a grid cell which straddles a country's borders will lie entirely outside

the country. In converting from the original grid to the GEIA grid, if a grid cell in the new system includes only this "foreign" part of the original cell, then the fraction of emissions in the foreign part will be incorrectly assigned outside the country borders. To avoid the incorrect allocation of emissions on a country basis, each country was separately regridded and incorrectly assigned emissions were reassigned to an adjacent cell associated with the appropriate country. The regridded EMEP and the CORINAIR85 inventories were combined by using the CORINAIR85 emissions in grids common to both inventories.

Information is available on the compliance of different European countries with the Helsinki and Sophia protocols for emissions reduction. The Helsinki Sulphur Protocol, signed in 1985, required from its signatory countries a 30% reduction of their 1985 sulfur emission levels by 1993. Except Ukraine, all signatory countries, which did not include all countries in Europe, have attained the reduction goal. The Sofia Nitrogen Protocol, signed in 1988, agreed to reduce emissions, so that by the end of 1994 annual nitrogen emissions should not exceed emissions in 1987. Only four countries of the signatory parties, which again did not include all countries in Europe, have not reached these "freezing-emission" levels.

For the CORINAIR90 inventory, the emission data from eastern Europe are considered to be as accurate as the emission data from western European countries. The collapse of the political and economic system in Eastern Europe has caused a decrease of about 25% in sulfur emissions and about 20% in nitrogen emissions in the region for the period 1990 to 1993. At present the emission data seem to indicate no change from 1993. However, emissions in eastern Europe could increase in the years to come due to the reactivation of closed industries in this region. New investors are maily concerned with upgrading the industrial technology, with little understanding of the need for investments in emissions control.

3.2.4. Middle East and North Africa. In areas of the Middle East and the north of Africa that are included in the EMEP modeling domain [*Sandnes and Styve*, 1992], the EMEP emission estimates were preferred over the estimates provided in the default inventories. The EMEP emissions were derived following the standard methodologies of EMEP inventories for Europe which are based on available information on population, activity rates, and emission factors. (J. Saltbones, The Norwegian Meteorological Institute, Oslo, personal communication, 1994). The EMEP emissions were apportioned to the 1° x 1° GEIA grid as described in section 3.2.3.

3.2.5. Emissions from large stationary sources in the Asian part of the former USSR. Detailed information on the locations, type of activity, and SO_x and NO_x emissions from the major stationary sources in the Asian part of the former USSR were developed by J. Pacyna. Emissions may be underestimated due to incomplete information on these sources; the underestimation could be up to 25%. To more accurately locate the emissions from these major stationary sources, emissions in each individual grid cell within the Asian part of the former USSR in the default inventories were multiplied by the ratio of (total emissions in the Asian part of the USSR in default inventories - total major stationary source emissions)/(total emissions in the Asian part of the USSR in default inventories), and the major stationary source emissions were then reintroduced in the appropriate grid cells of the GEIA inventories.

3.2.6. Asia. Inventories of SO_x and NO_x emissions for 25 Asian countries (Afghanistan, Bangladesh, Brunei, Cambodia, China, Hong Kong, India, Indonesia, Japan, N. Korea, S. Korea, Laos, Macao, Malaysia, Maldivies, Mongolia, Myanmar, Nepal, Pakistan, Phillipines, Singapore, Sri Lanka, Taiwan, Thailand, Viet Nam) were developed for years 1975, 1980, 1985, 1986 and 1987 by *Kato and Akimoto* [1992]. These inventories included emissions from transformation (electric utilities, petroleum refineries) industrial, transportation, and other sectors. Activity rates for each sector were compiled on a province and regional basis for China and India and on a country basis for all other countries. Emissions were estimated using the appropriate emission factors for each sector. Results for 1975, 1980, and 1987 were apportioned to a 1° x 1° resolution grid by *Akimoto and Narita* [1994]. The locations of the individual sources from the power production and industrial sectors were obtained and emissions were assigned to the appropriate grid cell. For the navigation and aviation sections of the transportation sector, locations of harbors and airports were used. The rest of the emissions were assigned on the basis of population, which was gridded as described by Akimoto and Narita.

The ratios of (emissions in 1985)/(emissions in 1987) for the 25 countries in the inventory were obtained from the Kato and Akimoto work. The Akimoto and Narita gridded emissions for 1987 were multiplied by the 1985/1987 ratio for each country to obtain gridded emissions for 1985.

A 1° x 1° inventory of SO_x emissions for five Asian countries, China, Japan, North Korea, South Korea and Taiwan was developed by Y. Tonooka (Institute of Behavioral Sciences, Tokyo, Japan, personnal communication, 1992). The inventory includes seasonal resolution and is compiled with detailed sectoral breakdown of sources and fuels. For the GEIA inventories the SO_x emissions from these five countries were taken from the Tonooka inventory and emissions from the rest of Asia were taken from the Akimoto and Narita inventory; the NO_x emissions for all Asian countries were taken from the Akimoto and Narita inventory.

3.2.7. South Africa. Gridded SO_x and NO_x emissions for 1985 were provided by the South African Department of National Health and Population Development (S.M. Lloyd, personnal communication, 1993). The emissions include industrial and mobile sources as well as commercial and residential coal combustion. Area sources were gridded on the basis of the distributions of surrogate data such as population and comsumption or production figures. The emissions for major stationary sources were estimated on the basis of continuous stack monitor data or stack sampling and on U.S. EPA emission factors for other sources. The emission data were provided on the 1° x 1° GEIA grid.

3.2.8. Australia. Estimates of emissions of SO_x and NO_x for 1985 in Australia were received from the Environment Protection Authority of Victoria (EPAV). Emissions from large stationary sources were inventoried individually based on data available from State Regulatory Authorities [*Horseman and Carnovale*, 1989]. Emissions from mobile, domestic, and commercial sources in Australian capital cities have been inventoried by *Farrington* [1988]. Based on detailed estimates of emissions from a large number of area sources for Australia, F. Carnovale (Environment Protection Authority of Victoria, Melbourne, Australia, personal communication, 1992) estimated the average per capita emission factor of SO_x and NO_x from diffuse anthropogenic sources. These factors were employed in conjunction with Australian population census data to estimate total anthropogenic area source emissions of mobile, domestic and commercial sources of SO_x and NO_x in 1750 1° x 1° longitude/latitude grid cells. Area and point source emission

data were provided as the sum of anthropogenic emission estimates per grid cell. Information on individual stationary sources was also. Data on NO_x emissions from some major stationary sources were not available; thus some of these emissions may be underestimated.

3.2.9. Marine and aircraft emissions. Emissions from ship traffic have not been addressed in a coherent manner in any existing inventories of SO_x and NO_x emissions. Regional inventories for the United States, Canada, Europe and Asia used for this work include emissions from national ship traffic; only the EMEP inventories include emissions from international ship traffic.

The methodologies to estimate marine emissions in the United States [*Kimbrough*, 1992] and in Canada [*Johnson et al.*, 1991] use similar algorithms. For commercial vessel emissions were estimated only for port activities; estimates are based on consumption of fuel. The commercial vessel population was assumed to be 75% motorships and 25% steamships; motorships were assumed to spend 20% of the time in port activities; steamships were assumed to spend 80% of the time in port activities. If consumption data were available by port, emissions were assigned to the corresponding county or province; the remaining emissions were assigned to ports and waterways according to tonnage handled. Emissions for time spent on international routes were not included in these inventories. For recreational vessels, all emissions were assigned to the appropriate county or province.

According to directives of the UN/ECE convention, the European countries are encouraged to include emissions from national ship traffic in the EMEP inventories. The sectors that should be included are coastal navigation (fishing, ferries, passenger ships), offshore gas and oil installations, and fishing outside territorial waters.

SO_x and NO_x emission estimates for international ship traffic were developed by MARINTEK, Norway [*Bremnes*, 1990; *Kolle et al.*, 1989]. The estimates were based on emission factors and information on ship's total bunker consumption in international trades. Emission factors for different engine types and bunker oil qualities were obtained from the latest tests and measurements and from calculations as described in the MARINTEK reports. These emission factors are significantly higher than emission factors for land-based activities mainly due to the large sulfur and nitrogen content of bunker oil. For example, the SO_2 emission factor for motorships using heavy fuel oil is 60 g S kg^{-1} fuel and for gas or marine diesel fuel this value is 20 g S kg^{-1} fuel. In contrast, SO_2 emission factors for land-based activities are 5 to 10 g S kg^{-1} for fuel oil and 2 to 4 g S kg^{-1} for diesel oil. The international sea-borne transport patterns were used for distribution of bunker consumed per year on the main international ship routes. Emissions of SO_x and NO_x were calculated and distributed over the main routes on the ocean charts. The international ship traffic in the North Sea, English Channel, and the Baltic Sea has been charted on EMEP maps; the corresponding emissions have been included in the EMEP and GEIA inventories. The world total SO_x emissions from international sea-borne transport were estimated by MARINTEK at 2.3 Tg S yr^{-1}, the corresponding NO_x emissions were estimated at 1.6 Tg N yr^{-1}.

Kato and Akimoto estimated emissions from "internal navigation" based on fuel consumption; Akimoto and Narita assigned these emissions to harbors using passenger and freight statistics whenever available. Tonooka estimated sulfur emissions from ocean vessels based on harbor arrival statistics, gross tonnage, vessel and fuel type, and emission factors. These emissions were assigned to coastal ports on the basis of the period of time spent and distance the vessels traveled in each harbor area.

In the North American inventory, emissions from aircraft were estimated relative to units of aircraft landing and takeoff cycles [*Kimbrough*, 1992]. All countries under the UN/ECE convention are required to include aircraft emissions from takeoff and landing in their national totals reported to EMEP; thus aircraft emissions in these inventories represent aircraft activities in the vicinity of airports. Kato and Akimoto estimated aircraft emissions based on fuel comsumption, but no details were presented on which aircraft activities were included in these consumption values. Emissions of NO_x, CO and VOCs from aircraft have been estimated by the NASA Atmospheric Effects of Stratospheric Aircraft Program [*Stolarski and Wesoky*, 1993]; emissions of NO_x from aircraft have also been estimated by *McInnes and Walker* [1992]. For the NASA work, emissions have been estimated using a 1° x 1° logitude/latitude grid and at 23 heights for 10 different aircraft fleet scenarios; these files are available from NASA via anonymous file transform protocol (FTP). Details on the FTP transfer can be found in the work of *Stolarski and Wesoky* [1993]. In the McInnes and Walker work, NO_x emissions have been estimated using a 7.5° longitude x a variable 5° to 10° latitude grid and at six vertical heights evenly spaced 2 km each. Discrepancies between the NO_x emissions totals in these two inventories are currently being studied. In addition, if these inventories are to be used in conjunction with the GEIA NO_x inventory described here, a careful study must be done in order to avoid double counting some of the emissions from this source category.

3.2.10. Biomass burning. In its most general definition the term biomass burning is applied to anthropogenic activities associated with savannah and praire burning, slash and burn agriculture, burning for deforestation purposes and for disposal of agricultural wastes, biomass used as fuel (such as wood, bagasse) as well as other nonanthropogenic burning of biomass such as forest wildfires. Within GEIA a study group is addressing emissions from biomass burning activities that are carried out on a large geographic scale, such as savannah and prairie burning, and burning for deforestation purposes. Therefore the work described here did not directly address emissions from these large geographic sources.

Biomass burning is not addressed consistently in the inventories used for this work. The default global SO_x inventory [*Spiro et al.*, 1992] includes emissions from the use of wood as fuel. The default global NO_x inventory [*Dignon*, 1992] does not include emissions from any biomass burning. The NAPAP inventories for the United States and Canada include emissions from the use of biomass as fuel, burning for disposal of agricultural wastes, and forest fires. No extensive burning of prairies or for deforestation is allowed in these countries. The regional inventories for Asia compiled by *Kato and Akimoto* [1992] include emissions from the use of biomass as fuel. The regional inventories for Australia include emissions from the use of biomass as fuel and emissions from anthropogenic burning activities (savannah burning, agricultural burning), but do not include emissions from forest fires and wildfires. The regional inventories for Europe compiled by CORINAIR include categories for biomass fuels (wood, charcoal, and peat), agricultural waste burning, and forest fires [*CORINAIR*, 1995], although all countries may not report emissions for some or all of these categories. The regional inventory for Europe compiled by EMEP and the South African regional inventory did not include any emissions from the biomass burning categories.

Global estimates of emissions from biomass burning activities are given by Bates et al., who estimated sulfur emissions from these sources at 2 Tg S yr^{-1} [*Bates et al.*, 1992] and *Andreae* [1991] who estimated nitrogen emissions from these sources at 9 Tg N yr^{-1}. A summary of the source types included in the default and in the regional inventories used in this work is presented in Tables 2a and 2b.

4. Results

The GEIA inventories of anthropogenic emissions of SO_2 and NO_x for 1985 most probably underestimate emissions. Although the emissions from the important source categories are included, the emissions from some of the less important source categories are incompletely represented. There are large areas, such as Latin America and most of Africa, where no regional data are available; emissions from the default inventories have been used for these areas. The emission factors used in compiling these inventories represent average conditions in these large areas and, in general, tend to underestimate emissions. Our best assessment is that the total global emissions may be underestimated by no more that 25%.

4.1. Global Totals

In the past three decades some 10 inventories of SO_x emissions from anthropogenic sources have been formulated using a variety of different base years, with total emissions ranging from 57 to 91 Tg S yr^{-1}, as presented in Table 1 and Figure 2. The present effort adopted some of the approaches used to compile those inventories but has gone considerably further in incorporating specific information available from continents and regions and by gridding the information for the use of the scientific and policy communities. The overall result is a global sulfur emissions estimate of 65.1 Tg S yr^{-1}. Recent estimates of emissions of sulfur compounds from biogenic sources [*Bates et al.*, 1992] place oceanic emissions (mostly dimethyl sulfide (DMS)) at approximately 15 Tg S yr^{-1}, terrestial emissions at approximately 0.4 Tg S yr^{-1}, emissions from biomass burning at approximately 2 Tg S yr^{-1} and volcanic emissions at approximately 9 Tg S yr^{-1}, for a total of 26.4 Tg S yr^{-1}. Volcanic emissions are located only in areas of volcanic activity, are extremely variable in time, and can be released anywhere from ground level to the stratosphere. The Bates' estimates for oceanic and terrestial sulfur emissions are at the low end of the range given by the IPCC [*Houghton et al.*, 1992],

Table 2a. Source Categories Included in the Default and in Regional Inventories of SO_x Used in This Work

Source Type	Default Inventory	United States and Canada	Europe	Middle East and North Africa	Former USSR/Asia Point Sources	Asia	South Africa	Australia
All fossil fuel combustion[a]	Y	Y	Y	Y	Y	Y	Y	Y
Noncombustion industrial processes	Y	Y	Y	Y	Y	Y	Y	Y
Marine emissions	Y(1)				NA		N	N
in port activities		Y	Y	Y		Y		
international ship traffic		N	P(1)	N		N		
Aircraft emissions	Y(1)				NA	Y(?)	N	N
landing and take-off		Y	Y					
international routes		N	N					
Biomass burning	P(2)	Y	N	N	NA	P(3)	N	Y

Y, emissions are included in inventory; (1), as part of fossil fuel combustion, no sectoral definition; (?), as part of fossil fuel, no definition on which activities are included; P, emissions from some sources are included in inventory; (1), North Sea, English Channel, and Baltic Sea shipping lanes only; (2), emissions from the use of wood as fuel; (3), emissions from the use of biomass as fuel; NA, not addressed in this inventory.

[a] Includes all fossil fuel combustion activities such as electrical generation, industrial, commercial, institutional and residential combustion processes, and transportation.

Table 2b. Source Categories Included in the Default and in Regional Inventories of NO_x Used in This Work

Source Type	Default Inventory	United States and Canada	Europe	Middle East and North Africa	Former USSR/Asia Point Sources	Asia	South Africa	Australia
Fossil fuel combustion[a]	Y	Y	Y	Y	Y	Y	Y	Y
Industrial processes	N	Y	Y	Y	Y	Y	Y	Y
Marine emissions	Y(1)				NA		N	N
in port activities		Y	Y	Y		Y		
international ship traffic		N	P(1)	N		N		
Aircraft emissions	Y(1)				NA	Y(?)	N	N
landing and take-off		Y	Y					
international routes		N	N					
Biomass burning	P(2)	Y	N	N	NA	P(3)	N	Y

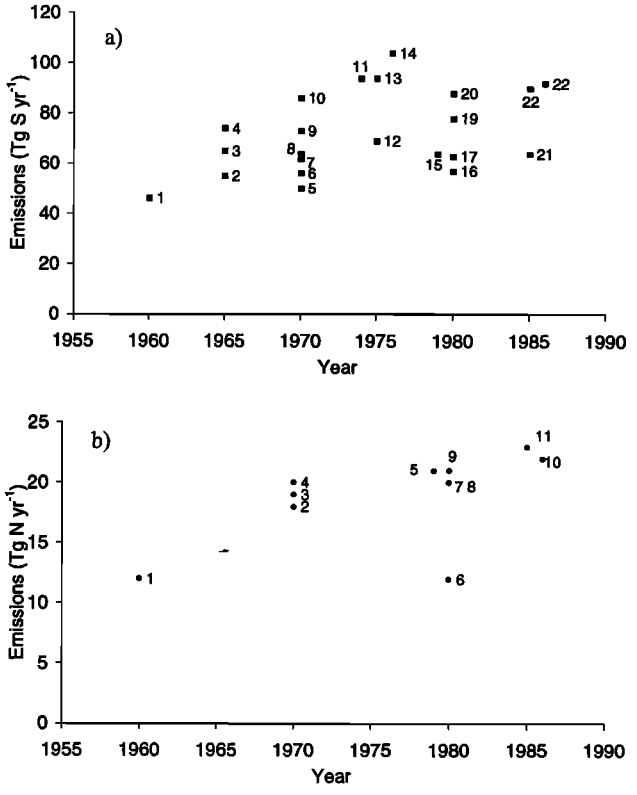


Figure 2. Previous estimates of global emissions of (a) sulfur and (b) nitrogen. The numbers indicate the relative position of the data in Tables 1 and 2.

where oceanic emissions are estimated to be in the range 10 to 50 Tg S yr^{-1} and terrestial emissions in the range 0.2 to 4 Tg S yr^{-1}.

Unlike the emissions of SO_x, in which anthropogenic sources clearly dominate, biogenic sources of NO_x may be comparable with anthropogenic sources on a global basis. For example, soil-biogenic emissions, calculated on the basis of nitrogen with no chemical speciation, are of the order of 5 Tg N yr^{-1} [*Yienger and Levy*, 1995]. They are, of course, concentrated in the temperate and tropical regions. Biomass burning emissions have been estimated at approximately 9 Tg N yr^{-1} [*Andreae*, 1991], and emissions generated by lightning, though poorly quantified, are thought to be less than 10 Tg N yr^{-1} [*Yienger and Levy*, 1995]. Thus the natural total is perhaps 15 to 20 Tg N yr^{-1}.

Anthropogenic emissions of NO_x are related almost totally to the combustion of fossil fuels [*Logan*, 1983]. In the present work, the anthropogenic NO_x inventory was constructed in parallel to the inventory for anthropogenic SO_x but used a different default inventory and the nitrogen emission estimates from the regional inventories. The overall result is a global emissions estimate of 21.0 Tg N yr^{-1}. A summary of emissions for the base inventories and for the regional inventories used in this work is presented in Table 3.

4.2. Geographic Distribution of Emissions

Plate 1 presents the geographic distributions of sulfur and nitrogen emission flux (defined as emissions per unit area per unit time) due to anthropogenic activities. Anthropogenic SO_x and NO_x emissions are distributed over approximately 14% of the 1° x 1° cells of the global grid, representing approximately 17% of the global surface area. Figure 3 presents the latitudinal distribution of the average anthropogenic emission flux of sulfur and nitrogen. As seen from these figures, SO_x and NO_x emissions are strongly localized in the highly populated and industrialized regions in eastern North America and across Europe from the United Kingdom over central Europe to the Donbas region in Russia. Outside these areas are smaller regions or hotspots with quite large emissions, either in connection with densely populated areas with developed industries or in connection with exploitation of fuels or mineral reserves.

Sources in North America and Europe (including Russia west of the Urals) contribute about 20 and 30%, respectively, to the estimated global emissions of SO_x from anthropogenic sources. Other areas of high SO_x emissions are located in the Far East, particularly in China, where large amounts of coal are being used to produce heat and electricity. Industrial emissions in central Siberia are also relatively large in east Ural, Kuznetsk, Karaganda, and the Irkutsk regions. The large emissions of SO_x in the Kola Peninsula and the Norilsk area, specially prominent in Figure 3a, are due to nonferrous metal smelting.

The spatial distribution of NO_x emissions is different from that of SO_x emissions, partly because of the strong link between motor vehicles and NO_x and partly because the SO_x emissions are linked to process emissions and fuels with high sulfur contents. Regions with large emissions of SO_x from smelters and other process industries may have relatively small emissions of NO_x.

Figure 4 summarizes the regional contributions to global anthropogenic SO_x and NO_x emissions in the northern and the southern hemispheres. Emissions in the polar region of the southern hemisphere are zero; this inventory includes no estimates of emissions from the stations in Antartica. Approximately 80% of the global sulfur emissions and approximately 84% of the global nitrogen emissions originate in the northern hemisphere midlatitudes. Because of the distribution of the landmasses the relative contributions of the tropical and midlatitude regions to emissions in the southern hemisphere are comparable. Overall, the midlatitude regions of both hemispheres contribute approximately 85% of the global sulfur emissions and approximately 88% of the global nitrogen emissions.

4.3. Nitrogen to Sulfur Ratio in Emissions

The ratio of nitrogen to sulfur emissions, calculated as (moles N emitted)/(moles S emitted), can be used as an indicator of the overall character of sources, to compare with the same ratio in deposition measurements. Plate 2 presents the distribution of the nitrogen/sulfur ratio (N/S ratio) for the GEIA inventories described in this paper. Anthropogenic emissions of both SO_x and NO_x are present (i.e., emissions of both are greater than zero) in approximately 16% of the global surface area; the N/S ratio ranges from approximately 1×10^{-4} to 6.5×10^{4}. In approximately 0.1% of the global surface area, where emissions of both SO_x and NO_x are present, the N/S ratios, are greater than 400 and in approximately 0.2% of the global surface, where emissions of both SO_x and NO_x are present, the N/S ratios are less than 0.01. These extreme values are not included in Plate 2. In general, locations with N/S ratios greater than 400 are

removed from high industrial or population areas and have low nitrogen emissions and very low sulfur emissions, representative of remote clusters of population; locations with N/S ratios less than 0.01 have high sulfur emissions and low nitrogen emissions, representative of isolated regions of smelters and other process industries.

The N/S ratio in many European countries is typically around 1; this ratio is less than 1/3 in some countries in central and eastern Europe. In industrialized and heavily populated areas of North America, N/S ratios are generally less than 1; these ratios can be around 10 where large stationary sources are located away from populated areas.

4.4. Comparison With Biogenic Emission Estimates

The results presented here and the most recent estimates of global emissions from biogenic sources show that approximately 70% of the global sulfur emissions are in the form of SO_x from anthropogenic sources, the rest are in the form of miscellaneous sulfur compounds such as DMS, H_2S, OCS, and CS_2 from biogenic sources. However, global emissions of nitrogen compounds are almost evenly split between NO_x emissions from anthropogenic sources and emissions of miscellaneous nitrogen compounds from biogenic sources. Anthropogenic emissions of SO_x and NO_x have been historically dominated by emissions from the industrialized regions of the world. However, the geographic distribution of these emissions can be expected to change in the near future, as the reduction of emissions in some regions is compensated by the increase in other regions. Economic development is a driving force in changing the quantity and spatial distribution of anthropogenic emissions of SO_x and NO_x.

5. Uncertainty

Uncertainty estimates for emission data are an important part of any inventory. Such estimates enable modelers and other users of emission data to attach appropriate significance to features of their results. Ideally, statistical estimates of uncertainty would be provided for each country or grid cell. To make such statistical estimates is difficult, even where a high level of inventory detail is available. In general, results of the measurements on which emission factors are based are not readily available; there are no objective criteria on which to base the estimation of uncertainty when missing values for emissions estimation parameters are assigned based on values from a "similar" sector, and the normal statistical methodologies which require normality and independence of parameters are not directly applicable [*Benkovitz and Oden*, 1989; *Oden and Benkovitz*, 1990].

As a guide to users of the inventories developed in this work, a qualitative uncertainty estimate was assigned by regions according to the level of regional detail in the methodology and data used to prepare the emission estimates; a scale of low, medium, and high was used. High uncertainty was assigned to the default SO_x and NO_x inventories [*Dignon*, 1992; *Spiro et al.*, 1992]. Low uncertainty was assigned to the emissions from those regions covered by the inventories of NAPAP [*Saeger et al.*, 1989], CORINAIR85 [*Bouscaren*, 1992], *Lloyd* [1993], *Carnovale* [1992], and the *Tonooka* [1993] data sets for Japan, North Korea, South Korea, and Taiwan, because of extensive data documentation and validation. The remaining data sets consist of regional inventories for much of Southeast Asia, China, India, and the former Soviet Union. In these cases some degree of information was available, specially on the size and type of the major point sources, but the information is not well validated and minor point source and area source emissions must be estimated. As a consequence, the uncertainty of these data sets is classified as medium.

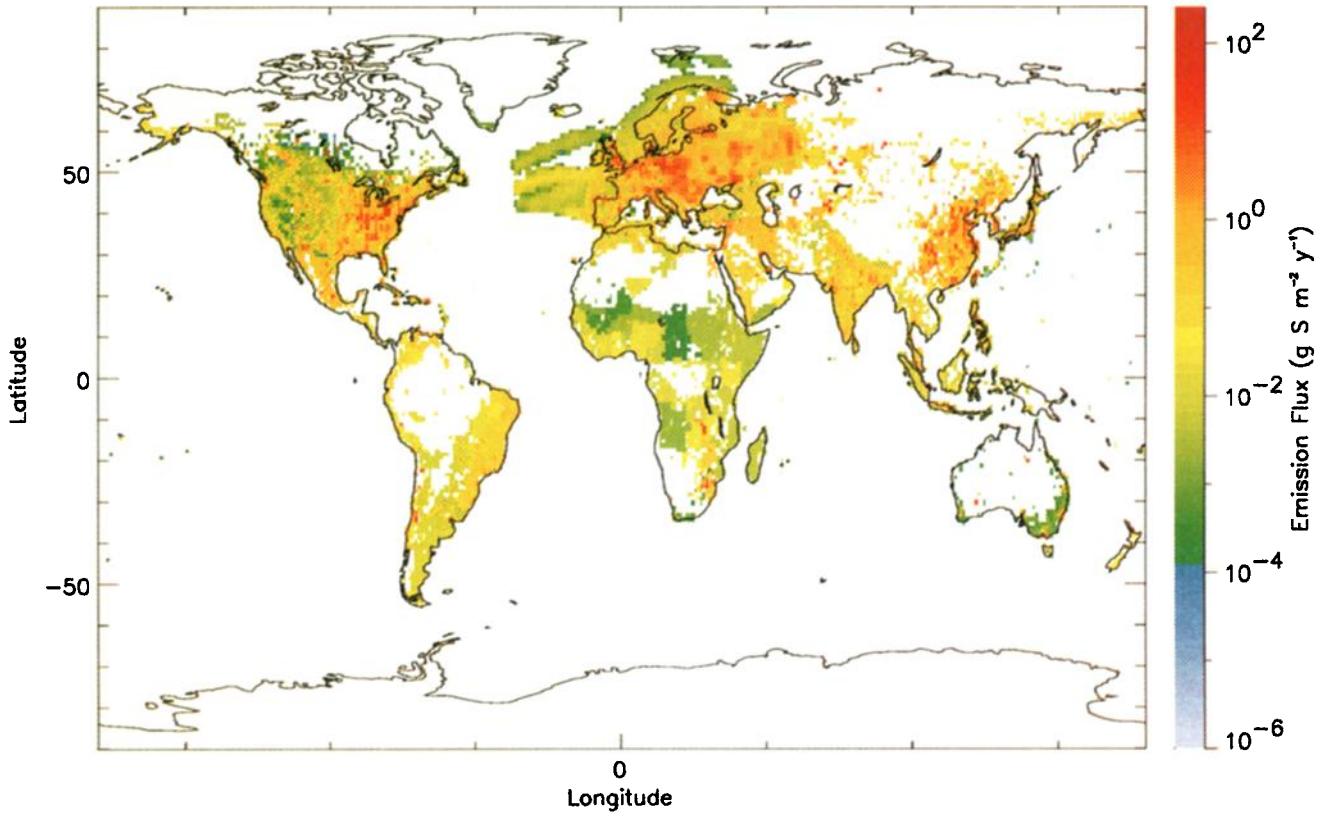
It should be pointed out here that several studies based on flue gas measurements for major stationary sources for a number of industries have been carried out from which quantitative estimates of uncertainty could be derived. However, such estimates would only apply to the source categories which were studied and in the countries and regions where operating conditions are known to match those in the studies. We feel that

Table 3. Summary of Emission Totals From the Default and in Regional Inventories Used in the Compilation of the Global Emissions Inventory Activity [GEIA] Inventories of Anthropogenic Emissions of SO_x [Tg S yr^{-1}] and NO_x [TgN yr^{-1}]

	Reference	SO_x Tg S yr^{-1}	NO_x Tg N yr^{-1}	Notes
Default inventory	Spiro	25.07		non-indust[a]
	Dignon [global]		23.44	
North America	NAPAP	12.30	6.24	
Former USSR stationary sources	Pacyna	4.85	0.50	east of 60°E
Europe, Middle East, North Africa	CORINAIR/EMEP	21.26	6.07	Russia to 60°E.
South Africa	Lloyd	0.93	0.22	
Australia	Carnovale	0.81	0.21	
25 Asian countries	Kato & Akimoto	13.04	4.20	
5 Asian countries	Tonooka	10.49		

Please note that these inventories overlap in their geographic coverage.

[a] Emissions from the following countries are not included in the total: Albania, Australia, Austria, Belgium, Bulgaria, Canada, Czechoslovakia, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, India, Ireland, Italy, Japan, North Korea, South Korea, Liechtenstein, Luxemburg, Malta, Netherlans, Norway, Poland, Portugal, Romania, former USSR/total, Spain, Sweden, Switzerland, Taiwan, United Kingdom, United States, and Yugoslavia.



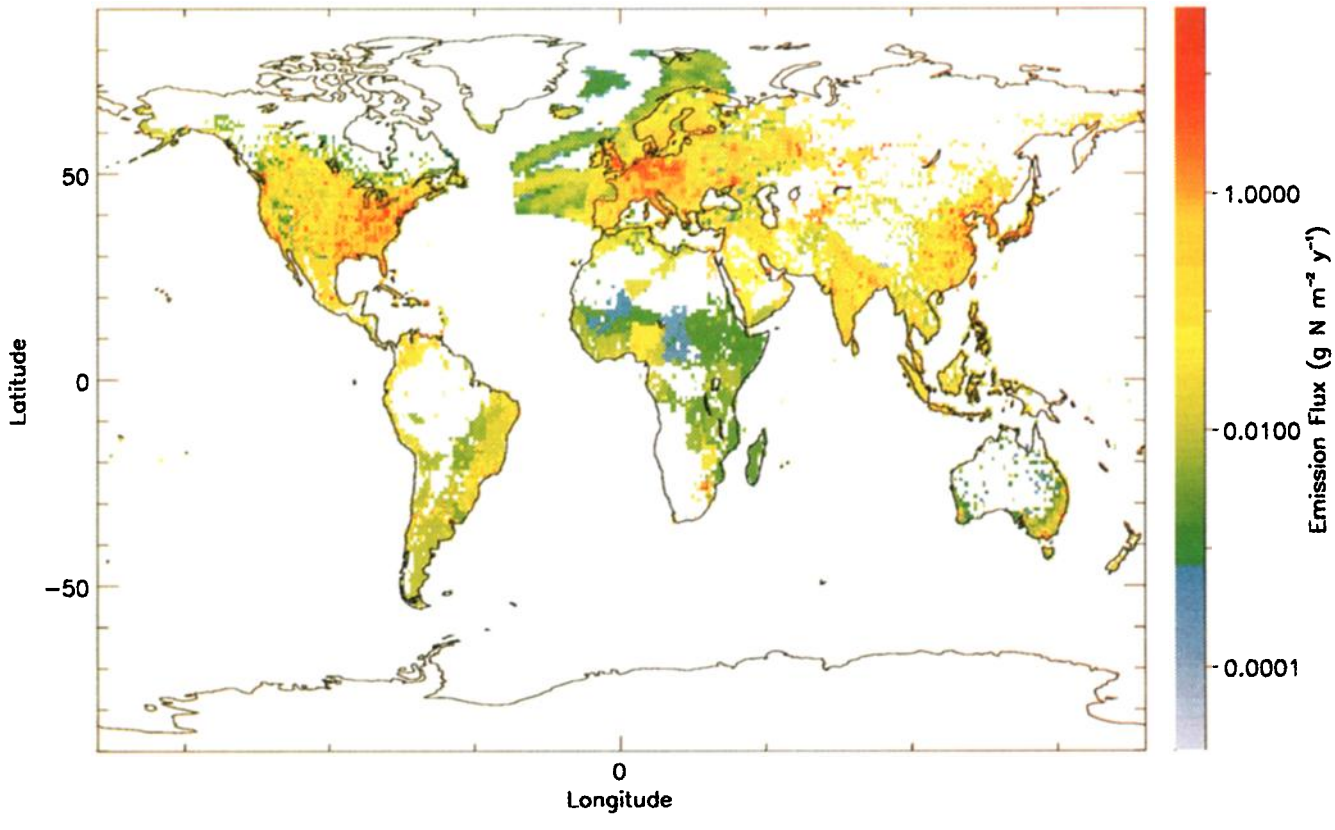


Plate 1. Geographic distribution of emission flux for (a) sulfur and (b) nitrogen. Shipping emissions over the northeastern Atlantic Ocean, the Baltic Sea, and the North Sea in the Cooperative Program for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe (EMEP) inventories for Europe were assigned to the appropriate shipping routes. Other shipping emissions have been assigned to port areas.

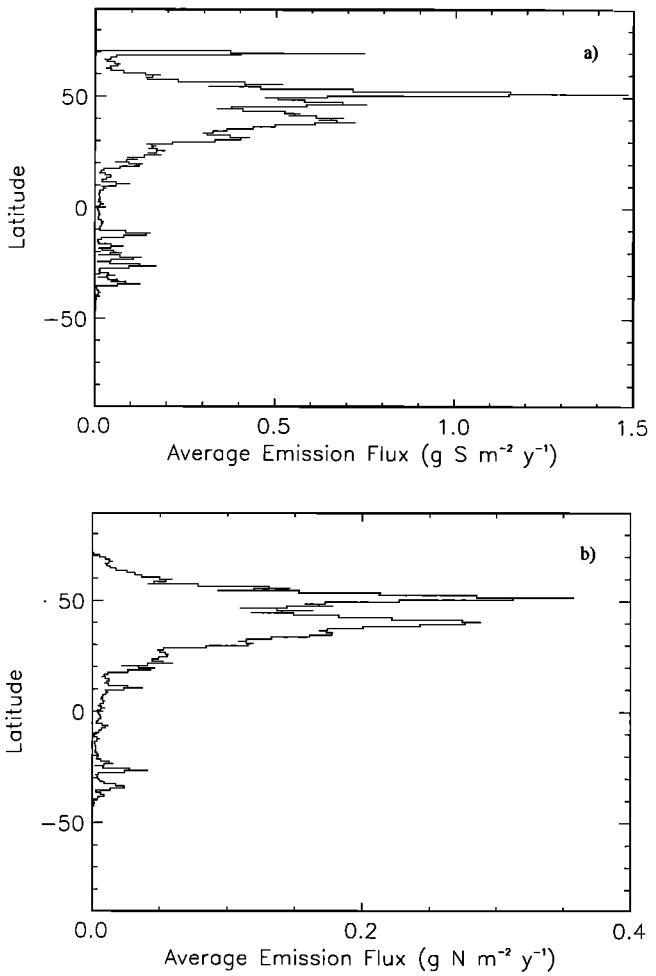


Figure 3. Latitudinal distribution of average (a) sulfur and (b) nitrogen emission flux. The increase in sulfur emissions at latitude ~68°N is due to nonferrous metal smelters in the Kola Peninsula.

this information is not complete enough to allow us at this time to assign quantitative estimates of uncertainty to the GEIA inventories.

More quantitative assessments of the uncertainty associated with emission estimates in some of the regional inventories used are available. The estimates for the United States and Canada [*Saeger et al.*, 1989] and for Europe [*Tuovinen et al.*, 1994] are believed to be within 25%, with SO_x emissions being more accurately determined than NO_x emissions. It should be admitted, however, that, to date, no verification of these estimates has been made even on regional scales. Verification procedures for emission estimates on at least regional scales are now being prepared within the UN/ECE Task Force on Emissions Inventories (J.D. Mobley, U.S. Environmental Protection Agency, personal communication, 1994).

To the uncertainty in the estimation of emissions described above, the uncertainty of the gridding process must be added. Except for smelter emissions, all other emissions in the default SO_x inventory and all emissions in the default NO_x inventory were gridded based on population as the surrogate, using the 1° x 1° file described in section 3.1. This file assigns each grid cell to the country (or water area) occupying the major fraction of the area of the cell. Thus for a particular grid cell emissions originating in countries other than the designated one must be displaced to the nearest grid cell assigned to the country of origin of the emissions. The regional data sets that were received already gridded to the 1° x 1° GEIA grid may have used additional surrogates to allocate their emissions. At this time, quantitative estimates of the uncertainties associated with the gridding procedures are not available.

Emissions in the GEIA inventories represent "typical" conditions circa 1985; that is, special events, such as breakdown of control equipment, temporary shutdown of individual sources, are not captured. The uncertainty of using the GEIA inventories to represent actual conditions for 1985 must be added to the other uncertainties described above. If a more accurate representation of actual conditions during 1985 is desired, the GEIA inventories must be examined in light of surveys or other reports that can describe exactly what occurred during the periods and in the locales of interest.

6. Summary

Version 1 of the GEIA global inventories of annual emissions of SO_x and NO_x from anthropogenic sources circa 1985, called version 1A, is now available for use by the scientific community. These inventories are distributed to a 1° x 1° longitude/latitude grid and include the most current information

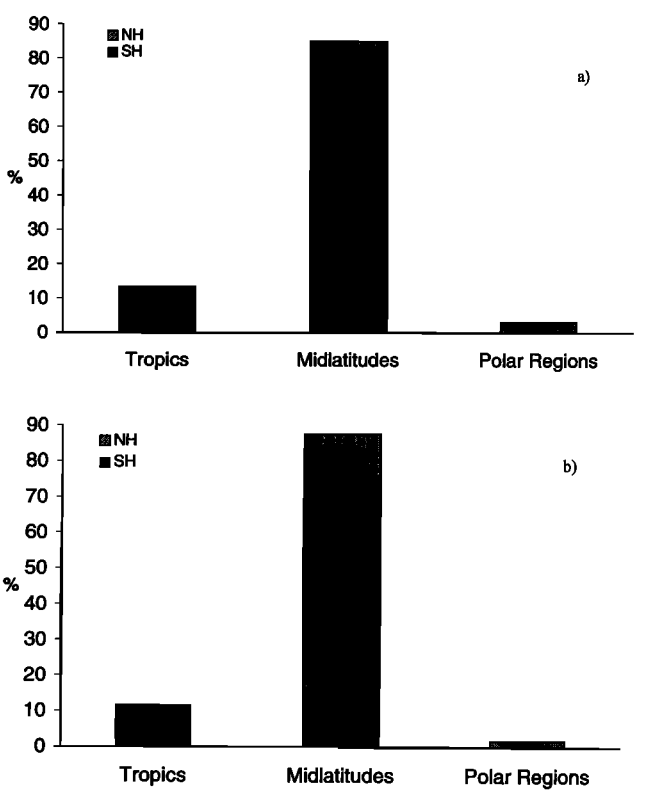


Figure 4. Summary of the regional contributions to global anthropogenic (a) SO_x and (b) NO_x emissions in the northern and the southern hemispheres. The tropical regions are defined as the latitude band from the equator to 23.5°, the midlatitudes regions are defined between latitudes 23.5° and 60°, and the polar regions are defined from latitude 60° to the poles. Emissions in the southern hemisphere polar region are zero.

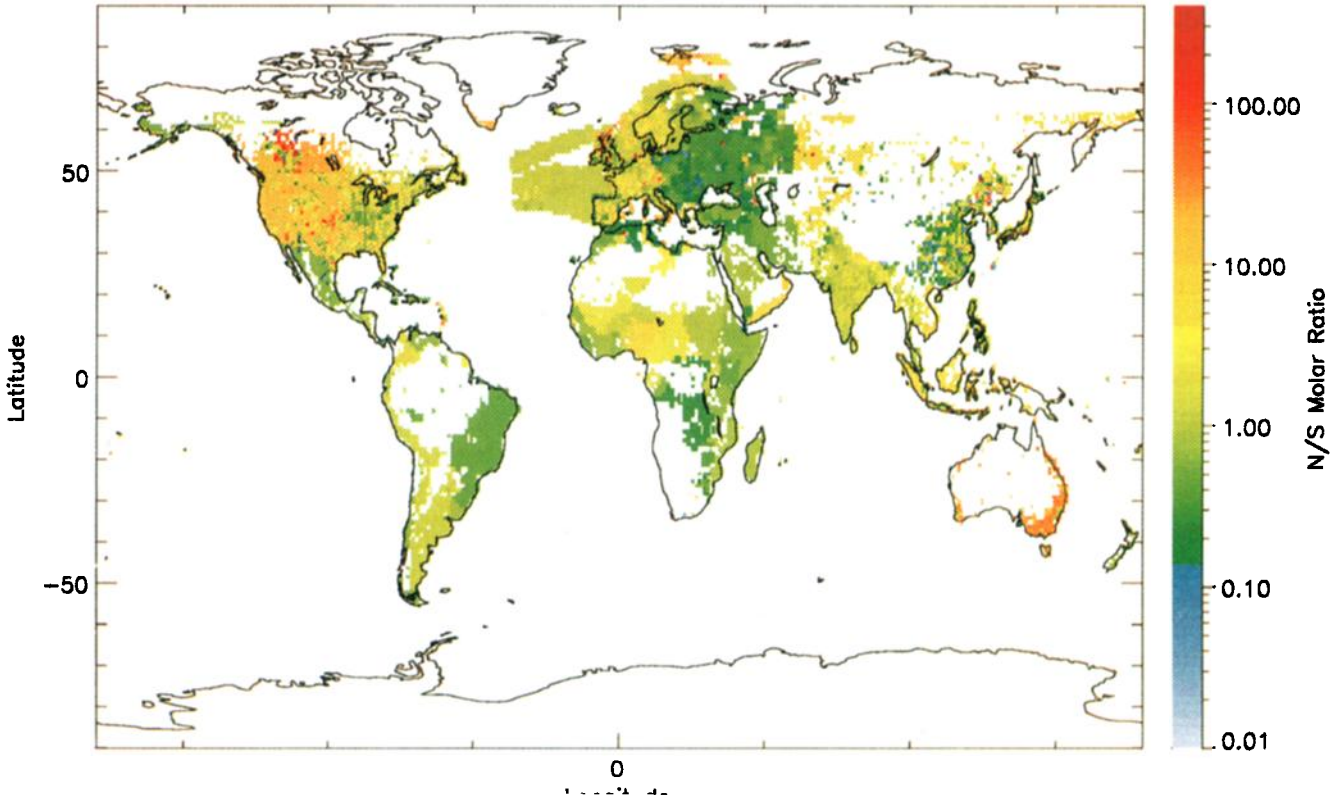


Plate 2. Ratio of nitrogen to sulfur emissions. Areas with N/S ratios < 0.01 and > 400 are not included.

Table 4. Contributors to the GEIA Version 1A Inventories of Anthropogenic SO_x and NO_x Emissions

Name	Country	Contribution
Hajime Akimoto[a]	Japan	Gridded SO_x and NO_x for 20 countries in East Asia
Frank Carnovale[b]	Australia	Gridded and point source SO_x and NO_x for Australia
Jane Dignon[c]	United States	Global gridded NO_x from fossil fuel combustion
Daniel Jacob[d]	United States	Global gridded SO_x
Nobuo Kato[e]	Japan	SO_x and NO_x for 20 countries in East Asia
S.M. Lloyd[f]	South Africa	Gridded and point source SO_x and NO_x for S. Africa
Jennifer A. Logan[d]	United States	Gridded 1° x 1° population file
Gordon McInnes[g]	UN/ECE	Area and point source SO_x and NO_x from CORINAIR
J. David Mobley[h]	United States	Gridded 1985 SO_x and NO_x from NAPAP
Hirohito Narita[i]	Japan	Gridded SO_x and NO_x for 20 countries in East Asia
Jozef Pacyna[j]	Norway	Point source SO_x and NO_x in Asian part of former USSR
Peter Spiro[k]	United States	Global gridded SO_x
Leonor Tarrasón[l]	Norway	Gridded SO_x and NO_x from EMEP
Yutaka Tonooka[m]	Japan	Gridded and point source SO_x for 5 countries in Asia

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from the NAPAP regional inventories for the United States and Canada [*Saeger et al.*, 1989], EMEP, and CORINAIR85 for Europe [*Bouscaren*, 1992; *Sandnes and Styve*, 1992], Asia [*Kato and Akimoto*, 1992; Tonooka, personal communication, 1993], Australia [*Carnovale et al.*, 1992; *Horseman and Carnovale*, 1989], and South Africa (Lloyd, personal communication, 1993). SO_x emissions for other regions of the globe were taken from the work by *Spiro et al.* [1992] updated to 1985; NO_x emissions were taken from the work by *Dignon* [1992]. Diagrams of the relationship between the default inventories and the regional inventories used are presented in Figure 1; a detailed list of contributors is presented in Table 4.

We suggest that those requiring emission inventories for these species standardize on the GEIA inventories, which we believe are authoritative and which are freely available to all users. The GEIA inventories generated by the work described here can be acquired by anonymous FTP at ncardata.ucar.edu (128.117.8.11). Please scan the README files at that location for information about the NCAR Data Support Section. The emission data reside on /pub/GEIA. Questions about data access or programming issues can be directed to Debra Hopkins, telephone (303)442-6866, e-mail hopkins@ncar.ucar.edu. Scientific questions or administrative issues can be directed to Paulette Middleton, telephone (303)442-6866, e-mail Paulette@rmii.com.

The GEIA inventory effort on anthropogenic emissions of SO_x and NO_x will not cease with the release of the version 1 inventories. These inventories will be periodically updated; however, except for correcting major errors, the frequency of updating will be chosen so that users will have ample time to perform their studies and reach their conclusions before the next version of a GEIA inventory is released. Inventories of SO_x and NO_x updated to base year 1990 are planned for release in approximately 1997. All who are interested in becoming involved in these efforts, or contributing emissions inventory information, are urged to contact any of the authors.

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